

The authors thank the three referees for their thorough and pertinent reviews, which certainly allow us to improve the paper. Below we provide answers to points raised by each of the referees.

In this response, bold black parts are direct extracts of referee comments, blue italic parts are changes made in the article and black normal texts are answers/explanations on each comment made by the referees.

Referee 1 comments:

We would like thank this referee for their pertinent remarks, in the section that follows we try to answer every question/comment raised by this referee.

Referee 1 general comments:

- 1. The fact that several aspects are addressed makes it however more difficult to present the results. This is already is also reflected in the title, which tries to summarize this but is difficult to understand and not completely correct.**

This comment has been addressed both in the title and in the text. The title has been modified to:

Biogenic SOA sensitivity to organic aerosol simulation schemes in climate projections

- 2. The order of presenting the results could also be improved, e.g. present figure 6 earlier in the manuscript to give the reader an idea of the gradients and order of magnitude, and going into validations and more detailed analyses.**

This comment has been taken into account partly, modifying the order of presenting the results in section 4, however figure 6 has been kept where it was. Section 4 has been modified according to the comment number 4 of this referee. We chose to keep figure 6 where it is since the discussion for figures 4 and 5 come before figure 6 and wed like to have a more general (averaged on sub-domains) discussions before entering into the regional changes in 2D images.

- 3. Also, since absolute concentrations are rather different for the three schemes, comparing relative differences is on the one hand a respected method, but one should be careful especially when concentrations are low and small absolute differences are exaggerated.**

Yes, this comment has been taken into account and will be specified in answering the detailed remarks below.

- 4. Structure of section 4 is not consistent with subchapters: would be easier for the reader to present section 4.3 after presentation of Europe and Mediterranean.**

Yes, we have modified the order of subsections in section 4 in order to put the spatial analysis after the discussion of the two sub-domains.

- 5. Sometimes the authors should be more precise. Below, detailed comments on the various parts of the paper will be given.**

The detailed comments have been taken into account thoroughly and explanations have been given for each one below.

- 6. The motivation to focus on the summer and on biogenic SOA is there but could also be made a bit more prominent.**

Some phrases have been added (as per suggestions by the referee #1 and referee #3) in order to address this comment. They have been mentioned in the detailed comments below.

Referee 1 Detailed comments:

- 1. L1 Organic aerosol (OA). .introduce abbreviation here.**

OA has been added to this sentence.

- 2. L6 The differences between three different schemes to simulate OA are explored. These schemes are. . . 1) a molecular scheme, 2) a standard. . .**

Modified.

- 3. L12 These changes are largest over the summer period. . .**

Modified.

- 4. L17: Absolute concentrations are different: move this sentence to l 10, before addressing the relative change, also quantify (molecular scheme gives twice as much OA as VBS scheme)**

The sentence was moved higher, before addressing the relative changes, the following part was added as well:

Absolute concentrations between different schemes are also different, the molecular scheme showing the highest concentrations between the three schemes.

- 5. P218 e.g. temperature change, land use changes and CO2 inhibition (Heald..)**

Modified.

- 6. P2119 BVOC emissions have been quantified. . .mainly to assess the future evolution of. . . Statement that it is only emerging does not reflect the date of the papers cited, and they are of the same period as the Arneth paper.**

That is true. The phrase has been modified to the following:

It is mainly to assess the future evolution of tropospheric ozone that BVOC emissions have been quantified at global scale in chemistry-climate projections (Arneth et al., 2010). Their importance for organic aerosol chemistry has also been considered in global and regional scale atmospheric models (Maria et al., 2004; Tsigaridis et al., 2007; Heald et al., 2008b), but to a lesser degree.

- 7. P3110 Impact of climate change is different per region since emissions and atmospheric composition are different per region, and depend on sensitivity of scheme to temperature changes. The description of the thermodynamics are different between the scheme. The current sentence is not precise and confusing**

The phrase has been modified to the following:

Differences induced by different schemes are also expected to vary regionally, depending on the concentration ranges encountered and ranges and changes in meteorological parameters.

- 8. P416 More information on the modelling framework in the current study is provided in..**

Modified.

9. P5 12: the domain covers latitudes 30-70°N and longitudes 40W-60E

Modified.

10. P5 132: What about the performance of SOA2p in the Mediterranean, as compared to SOAvbs and SOAmod?

A detailed comparison of these schemes in two Mediterranean sites (in Corsica and Mallorca) was performed and published in Cholakian et al, 2018. The results show that the SOA2p shows a high overestimation in the simulated concentrations compared to the two VBS based schemes, while the other two both perform quite well for the simulation of the concentration. However, when it comes to the simulation of oxidation state and origins of the observed OA, the modified VBS scheme performs better than the standard VBS scheme (which overestimates the fossil contribution and doesn't age particles as much as it should).

11. Figure 2: averaged pver 70 years of RCP scenarios (2031-2100) and averaged over 30 years for historic simulations. A-c mentioned in text, not indicated in figure

The information has been added to the figure legend.

12. P6: 110. For the perspective it would be good to mention that winter OA concentrations in cold episodes are higher that summer OA concentrations (Table 3)

It is in the data, but the passage here is about BSOA being the major source of OA in the summer months, since the study revolves around BSOA concentrations.

13. P7 125-26: would be better to stick to SOAvbs and SOAmod convention as defined on p 5. Differences in correlation are small.

The phrase has been modified to:

The correlation between observed and simulated OA concentrations for different schemes are the highest for the SOAmod, and lowest for the SOAvbs in most seasons; it should also be noted that the difference between the correlations seen for each scheme are rather small (difference of below 0.05).

14. Table in figure 3 is nearly unreadable, shoud be put as a text table outside the figure for readability. Also fonts of figure tick labels too small.

The tables on the side of the image have been moved to a separate table in the text. The size of the ticks has been modified and some general description has been added in the caption to describe the taylor diagram.

15. P8 19 : change quite a bit: be more precise

The intensities of the changes have been quantified in the lines that come below this phrase. This sentence is meant as an introductory phrase to enter into the explanation about the intensity of these changes.

16. P8 111 annual average “historical”. . . , annual average historical . . . Unit is per cm2 I assume, this is not what it states. Correct woud be molecules cm-2 yr-1.

The historical has been changed to historic and the units have been modified.

17. P8 117-20: Leave out sentence : for the Mediterranean region, there are no local emissions. The following sentence is more clear by itself.

The paragraph was changed to:

For the Mediterranean region, there are no local biogenic emissions included in the model.

18. Fig 4 Quality of graphics is poor, tick labels unreadable, too small

The size of the figure has been increased; the labels have been changed as well.

19. P913 Important sentence, could be combined with your reference to Cholakian (2019) in the section on choice of years, and maybe included in the introduction, depending on how you see this as a motivation and how this is the result of the present study. Would help the reader to get this statement very clear in an earlier part of the paper.

The sentence was moved to the choice of years section. The following sentences were added to the abstract and the conclusion:

The study focuses on BSOA since the contribution of this fraction of BSOA is more important in both historic and future scenarios (40 to 78% for different schemes in historic simulations).

Since BSOA contributes to an important degree to the total concentration of OA, the focus of this article is the evolution of BSOA concentrations in different schemes in future climatic projections.

20. P10 13 The statement can be related to seasonality in biogenic emissions.

That is correct, the following phrase was added to the aforementioned line to highlight that fact:

Highest relative changes occur for august for all schemes (+133%, +168% and +333% for SOA_{2p}, SOA_{vbs} and SOA_{mod} respectively) resulting from the seasonal profile of BVOC emissions.

21. P10 116: What do you mean by distribution of origins and volatility bin aspects?

The sentence sounds a bit ambiguous, what was meant is each scheme has its own way of simulating the origins of formed OA (because of difference in aerosol formation reactions) as well as the volatility of simulated OA. For example, the VBS scheme has a bin-per-bin distribution. The sentence is modified to:

Since the schemes behave differently both in contribution of different origins in the formation of OA as well as volatility distribution of OA, it is interesting to compare these two aspects in the tested schemes.

22. P11 18: relative contribution of HOA becomes less, since in your scenario you did not change anthropogenic emissions, but more BSOA is formed.

That is what was meant by the second part of the phrase ‘probably since more BSOA formation happens in future scenarios’. More explanation has been added to the phrase:

The relative contribution of HOA decreases in all schemes since the anthropogenic emissions are kept constant in future simulations and the relative contribution of SOA increases in the future.

23. P12 Fig 6: State in caption that these are summer averages.

The caption has been modified.

24. P12 l8 The maximum change is found in the summer period, reaching a maximum of . . . These large differences are for areas with very low concentrations, so the small increase in absolute numbers is blown up. A few words should be devoted to this aspect, to indicate where the most relevant increases are found. Although Baltic sea and North Sea are identified as region with larges changes, I think that the relevance is larger for central Europe where a small relative increase implies substantially higher absolute SOA concentrations. When you put section 4.3 after section 4.4 you could easily include the Mediterranean as a focus area in this discussion and make it the ultimate summary.

Putting section 4.4 before section 4.3 is quite logical, the modification has been made in the paper. In regards to smaller differences being amplified when looking at relative changes, this occurs mostly over the ocean, so a possible solution would have been using a land-sea mask. However, since the discussion is around the Mediterranean area, this idea was disregarded. Therefore, some sentences were added in order to address this fact:

It is important to keep in mind that some of these differences occur in areas with low concentrations of BSOA, which can lead to large relative changes despite of only small absolute ones. This mostly occurs in the oceanic regions of the domain. When a land-sea mask is used, the maximum changes occur on the British Isles, Scandinavian area and the central Europe.

25. P13 l 15-18: Part on PM10 not relevant for this paper

That is true, the phrase is meant to highlight the differences between the two different areas. The remark about PM₁₀ not being relevant to the paper is true, therefore the phrase was changed to the following:

There are major differences between the concentrations of different aerosol components over the Mediterranean area compared to continental Europe.

26. P13 l 27 : difference, not change

Modified.

27. P14: Figure 7: differnces between dots and squares only visible when enlarging on computer screen, not when one prints the paper.

This point was raised by referee #3 as well. The shapes of the points were changed in order to illustrate the differences more vividly.

28. P14 section 5: In climate projections, not only the temperature changes but also the circulation patterns (and even differently for different global climate models). This might be the reason for the different directions in the lines for the two periods. Also, when looking at a smaller domain, this change in circulation may become more relevant instead of averaging it out over a large domain. In particular over the Mediterranean without sources of isoprene/terpenes, changes in transport patterns are important.

This is true and it's a pertinent remark. Circulation patterns change between the two periods for each simulated year. However, since the average of 5 years of simulations are compared for each period, it is likely that the changes caused by the circulation pattern changes of a specific year are averaged out

especially since the years have been chosen in a way to represent low/high temperature maxima for historic/future periods respectively. The following phrase has been added in the article in order to mention this point:

Before entering into the discussion around sensitivity to temperature changes, it is important to keep in mind that the circulation patterns can change between the historic and future periods. Although averaging of 5 years of simulations likely filters out part of the noise in these patterns, this could also affect BSOA concentrations in addition to temperature changes, especially since the Mediterranean area since its remote with respect to sources.

29. P15 1 4 correpondence of what to what?

The sentence has been changed to:

In contrast, for SOA2p and SOAvbs, less BSOA is formed with a temperature increase as could be expected from the correspondence of the temperature with BVOC emissions.

30. P16 Conclusionss: reasoning could be slightly more precise. Your results show that BSOA changes due to climate change are highly sensitive to the SOA scheme used, and that none of th BSOA schemes here matches the observations, which shows the importance of further development of more accurate SOA schemes. When the SOA scheme is truly accurate, a good temperature dependence would be implicit, since you would like to use it over cold areas like Scandinavia as well as warmer areas like the Mediterranean.

These are both good points, the following phrases were added to the conclusion to emphasize these remarks:

In conclusion, our study suggests that the BSOA concentrations changes are highly sensitive to climate change and the scheme used for their simulation. The changes reported until now for future scenarios are highly uncertain, both on absolute and on relative scale. On a relative scale, these changes might be higher with OA schemes that include formation of non-volatile SOAs (up to a factor of two).

Future work is necessary in still developing more accurate organic aerosol schemes, not only in terms of absolute concentrations simulated, but also with respect to their temperature sensitivity. The three schemes used in this study can accurately simulate the concentrations of OA each for a specific season and for a specific region, while none of the schemes seem to be able to do so for the whole domain. Therefore, more research is necessary in order to develop OA simulation schemes that are able to represent the concentrations of OA accurately and the temperature sensitivity of this species on a regional scale.

Referee 2 comments:

In this manuscript, Arineh Cholakian et al. apply the CHIMERE CTM to the European domain and the Mediterranean region. They present the differences in BSOA by comparing 5 years in the past (historical) and 5 years in the future. The choice of the years aimed to maximize the differences between future and historic simulations regarding the change in temperature. The authors explored three schemes (i) a molecular single -step oxidation scheme, (ii) a standard VBS scheme with anthropogenic SVOC aging only and (iii) a modified VBS scheme containing functionalization, fragmentation and formation of non -volatile SOA for all SVOC species. The year 2013 was used in order to evaluate the schemes for the European region.

We would like to thank referee #2 for their pertinent and thorough comments, in the section that follows we will address every point raised by this referee.

Referee 2 major comments:

1. A clarification between absolute and relative changes and annual and summer period should be made for the whole manuscript.

The text is modified in order to reflect the nature of the comparison wherever necessary.

2. For the entire manuscript it is not clear if the authors refer to PM₁₀ or PM_{2.5} BSOA mass concentration. Please clarify.

Since most of BSOA concentrations is included in the PM₁ fraction, this is the fraction that has been used in this article. In the scheme validation part of the paper the fraction used for the simulated BSOA corresponds to what the measurement is for (either PM₁ or PM_{2.5}, as explained in the next point). The following sentence has been added to the beginning of the “Analysis of the simulations”:

It is important to keep in mind that from this section on, whenever BSOA concentrations are discussed the PM₁ fraction of this species has been used.

Also, a table showing the country, longitude/latitude, duration, type and altitude of measurements used in this study has been added here. Station names refer to EBAS names, the measurement is not present in EBAS if a station name isn't given.

Country	Station name	Longitude	Latitude	Altitude	Type	Duration	Resoluition
Switzerland	CH0001G	7,99	46,55	3578,0m	PM1	9mo	1h
Switzerland	CH0002R	6,94	46,81	489,0m	PM2.5	1yr	1d
Switzerland	CH0005R	8,46	47,07	1031,0m	PM2.5	1yr	1d
Switzerland	CH0033R	8,93	46,16	203,0m	PM1	18w	1h
Cyprus	CY0002R	33,06	35,04	520,0m	PM2.5	1yr	1d
Czech republic	CZ0003R	15,08	49,57	535,0m	PM2.5	1yr	1d
Germany	DE0002R	10,76	52,80	74,0m	PM2.5	1yr	1d
Germany	DE0003R	7,91	47,91	1205,0m	PM2.5	1yr	1d
Germany	DE0007R	13,03	53,17	62,0m	PM2.5	1yr	1d
Germany	DE0008R	10,77	50,65	937,0m	PM2.5	1yr	1d
Germany	DE0044R	12,93	51,53	86,0m	PM10	1yr	1d
Spain	ES0001R	-4,35	39,55	917,0m	PM2.5	1yr	1d

Spain	ES0009R	-3,14	41,28	1360,0m	PM2.5	1yr	1d
Spain	--	3,03	39,84	15,0m	PM1	3mo	1h
Spain	ES1778R	2,35	41,77	700,0m	PM1	6mo	1h
Finland	FI0050R	24,28	61,85	181,0m	PM1	9mo	1h
France	FR0009R	4,63	49,90	390,0m	PM2.5	1yr	6d
France	FR0013R	0,18	43,62	200,0m	PM2.5	1yr	6d
France	FR0030R	2,95	45,77	1465,0m	PM2.5	1yr	2d
France	--	9,38	42,97	520,0m	PM1	3mo	1h
France	--	2,15	48,71	156m	PM2.5	1yr	1d
Greece	GR0002R	25,67	35,32	250,0m	PM10	1yr	1d
Ireland	IE0031R	-9,90	53,33	10,0m	PM1	41w	1h
Italy	IT0004R	8,63	45,80	209,0m	PM2.5	1yr	1d
Netherlands	NL0644R	4,92	51,97	1,0m	PM2.5	1yr	1d
Norway	NO0002R	8,25	58,39	219,0m	PM1	1yr	1w
Norway	NO0039R	8,88	62,78	210,0m	PM1	1yr	1w
Norway	NO0056R	11,08	60,37	300,0m	PM2.5	1yr	1d
Poland	PL0005R	22,07	54,15	157,0m	PM10	1yr	1d
Sweden	SE0011R	13,15	56,02	175,0m	PM10	1yr	1d
Sweden	SE0012R	17,38	58,80	20,0m	PM10	1yr	1d
Slovenia	SI0008R	14,87	45,57	520,0m	PM2.5	1yr	1d

3. P7 line 15. The observations are filter-based or online measurements; PM10, PM2.5, PM1?

The observations are mostly PM_{2.5}, in some cases PM₁. None of the sites include PM₁₀ measurements. For each case (PM_{2.5} or PM₁) the corresponding fraction from the simulations have been used. The following phrase has been added to the paper to include this explanation:

The used measurements are mostly PM_{2.5}, in some cases PM₁. None of the sites include PM₁₀ measurements. For each type of measurement (PM_{2.5} or PM₁) the corresponding fraction from the simulations have been used.

4. P7 line 24. The authors should explain the correlation. A coefficient of determination (R²) should be used instead of correlation coefficient (R).

The R values have been all modified to R².

5. P7 line 25-27. “The three schemes perform reasonably well according to the criteria introduced by Boylan and Russell (2006), with the values for all the schemes falling into in zone 1 for both mean fractional bias and mean fractional error.” Please provide more information about the criteria by Boylan and Russell (2006). Provide more information about Taylor diagram.

The aforementioned reference fixes a model performance goal for PM with a mean fractional bias (MFB) and a mean fractional error (MFE) smaller than equal to +50% and ±30%, respectively. The model performance criterium is achieved when the 2 aforementioned statistic metrics are less than or equal to +75% and ±60% respectively. The author explains that the performance goals presented in this reference are close to the best the model can achieve, while performance criteria are defined as an acceptable performance for the model. In other words, in order to show the accuracy of a model, the

performance criteria have to be met, while the performance goals show the optimal performance of the model. The following information and values of MFB and MFE have been added to the paper:

The goal for these two metrics according to the aforementioned reference is less than or equal to $\pm 30\%$ and $+50\%$ and the criteria is less than or equal to $\pm 60\%$ and $+75\%$ respectively. The MFB values for the three schemes are -19.7% , 16.5% and 26.9% , while MFE shows 47.9% , 51.1% and 47.2% for SOA2p, SOAvbs and SOAmod respectively. Performance goals are met for nearly for all schemes, with a slight exceedance for MFE and the SOAvbs scheme, still meeting the criterium.

The Taylor diagram summarizes several statistic information in one plot: The Correlation coefficient, root-mean-square (RMS) difference between observations and simulations and the standard deviation ratio can be seen. More information about the construct of this diagram is given in Taylor, 2001. The standard deviation is read on the radii of the quadrant, the correlation coefficient on the outside rim of the circle and the RMS on the demi-circles centered around the normalized standard deviation. For the Taylor diagram the following information has been added:

The Taylor diagram in figure 3 shows the comparisons of different stations to simulations for each scheme (black for SOA2p, green for SOAmod and red for SOAvbs). The Taylor diagram summarizes several statistic information in one plot: The Correlation coefficient, root-mean-square (RMS) difference between observations and simulations and the standard deviation ratio can be seen. More information about the construct of this diagram is given in Taylor, 2001. As seen in this diagram, there is a high variability in the simulation of different stations, where some stations are better represented by the model than the others, which might be because of the geographical placement (altitude, types of emissions in said location, etc.) of stations or because of the nature of the station (urban, rural, etc.).

- 6. P7 line 27-29. “Each one of the schemes performs better for a specific period; modified VBS in summer, CHIMERE standard scheme during winter, and the standard VBS scheme showing average performance during the whole year.” This result is not clear in Figure 3 or 4. Please provide which statistical metric is used for this statement.**

This fact is shown in the tables initially connected to figure 3 and now presented separately as table 1. Looking at table 1, it is seen that for example for the summer period, the SOAmod scheme shows the highest correlation of determination, while SOA2p shows the lowest bias for this season. For spring SOA2p shows the highest R^2 , while SOAvbs shows the lowest bias. For winter and autumn, the performance of the schemes is quite similar. Annually, SOAvbs presents a similar correlation to SOA2p while showing the lowest bias in general. This has been added to the text as well:

Looking at table 1, it is seen that for example for the summer period, the SOAmod scheme shows the highest correlation of determination, while SOA2p shows the lowest bias for this season. For spring SOA2p shows the highest R^2 , while SOAvbs shows the lowest bias. For winter and autumn, the performance of the schemes is quite similar. Annually, SOAvbs presents a similar correlation to SOA2p while showing the lowest bias in general.

- 7. Figures 4 and 5. The authors, currently present together European sub-domain and Mediterranean Sea sub-domain. It would be better if they split the two regions, as Mediterranean is discussed in the next chapter. In Fig.4 keep only a, b and c, and add from Fig. 5 the EUR-related figures. The same applies for MED-related ones, keep d, e and f and MED form Fig.5.**

The two images have been modified according to this comment.

8. P8 lines14-15. Please provide which correlation the authors are referring to.

Here the correlation coefficient has been used. We have changed the correlation coefficients to correlation of determination in order to be consistent:

The correlation of determination between historic isoprene and terpene emissions is 0.6 and 0.63 while this correlation is 0.65 and 0.57 for the future simulations.

9. P9 line 3-4. “We address results for BSOA, as it makes the major contribution to OA during summer (between 40 and 78% for different schemes in the historic scenario)” In which figure is this shown?

This has not been shown in any of the figures, it is a value calculated using the simulations. However, since the majority of non-fossil OA comes from BSOA, this percentage can be implicitly seen in figure 5. The phrase “according to our simulation results for the historic period with differences schemes, not shown in figures” has been added to this statement in order to remove the confusion.

10. P9 lines 5-6. “while for SOA2p an increase of +94% is calculated, this percentage raises to +135% for SOAvbs and +189% for SOAmod” These numbers do not correspond to Fig.4 c1 and summer season.

These values correspond to annual averages, not the summer period. They are not shown in figure 4. It has not been mentioned in the manuscript to which period these values refer to, so we added this information:

while for SOA2p an annually averaged increase of +94% is calculated, this percentage raises to +135% for SOAvbs and +189% for SOAmod for the same period.

11. P10 lines 5-8. The numbers given in the manuscript are not consistent to figure 4 c.1. Please confirm the right one.

Scripting problem, the values in the figure 4 are correct, the values in the text have been changed to the right values:

Summer shows the maximum relative increase (+113%, +155% and +262% for SOA2p, SOAvbs and SOAmod respectively) and winter the lowest one in all schemes (+31.1%, +26.2% and +20.5% for SOA2p, SOAvbs and SOAmod respectively). For autumn and spring SOA2p and SOAmod show similar and intermediate changes while SOAvbs shows higher differences (+59.6%/+40.3%, +79.9%/+60.0% and +57.3%/+50.0% for SOA2p, SOAmod and SOAvbs respectively for autumn/spring).

12. P10 lines 11-13. The numbers given in the manuscript are not consistent to figure 4 c.2. Please confirm the right one.

Same issue as for the point above, the values have been changed:

In July, when the BSOA concentration reaches its maximum, the percentage of change in the future is high as well (+115%, +151% and +243% for SOA2p, SOAvbs and SOAmod respectively). Highest relative changes occur for August for all schemes (+111%, +165% and +356% for SOA2p, SOAvbs and SOAmod respectively)

13. P10 line 25. How were calculated these percentages? How are they linked to Fig. 5?

They represent the relative increase of the non-fossil part of the OA concentration between the future and historic simulations for each scheme on an annually averaged basis. The phrase has been modified to include the fact that the relative changes for annual averages are discussed:

All schemes show a relative increase in the contribution of non-fossil sources in the future on an annually averaged basis (10%, 17% and 22% of increase for the non-fossil partition between future and historic simulations for SOA2p, SOAvbs and SOAmod respectively).

14. P10 line 25-26. “SOA2p indicates a higher increase in nonfossil contribution compared to other schemes.” This cannot be stated unless the actual concentrations are shown. Please clarify if the authors are referring to the percentage of the increase.

It refers to absolute values, but as the referee indicates the absolute values haven't been shown so, to avoid confusion, the phrase has been removed.

15. P11 line 4-5. From Fig. 5 HOA is not that much different between SOA2p and SOAmod schemes. In contrast, SVOOA is a lot higher in the SOA2p than SOAmod. Please clarify if the authors are referring to actual concentrations. If yes, please provide a figure.

The specified line and page do not correspond to the given sentence. Figure 5 shows relative changes for historic and future simulations. The text has been edited in order to assure that the fact that relative comparisons are meant to be discussed comes through more clearly.

16. P11 lines 5-6. A more in-depth explanation of why the LVOOA is underestimated is needed. A suggestion to the authors would be the addition of a distribution of simulated organic aerosol (OA) in volatility bins for each scheme.

While this is a great idea, the volatility bin distributions have been discussed in our previous work (Cholakian et al, 2018), and we would like to refrain from adding additional images only to show the formation of LVOOA to this article, especially since the oxidation state does not vary much for the Mediterranean area and the variations in oxidation state in the European area are already visible in figure 4-d. The requested image has been added to this response in order to include what the referee wanted to see. Also, for the sake of being more coherent, the images 4 and 5 have been modified to show HOA and SOA (sum of LVOOA and SVOOA) since SOAvbs does not form LVOOA particles and whatever is formed stays in the SVOOA section. The entire section has been modified, here are the modifications that pertain to this comment:

Figure 4-d shows a simplified distribution for the OA in different schemes: SOA and HOA (hydrocarbon-like organic aerosol) presenting the freshly emitted primary OA. Figure 4-d shows that the predicted distribution between HOA and SOA is different for the three schemes. SOA2p indicates a smaller contribution of SOA and a larger one from HOA compared to SOAvbs and SOAmod schemes. This is because POA emissions in SOA2p are considered non-volatile, while they are volatile in VBS schemes. The relative contribution of HOA decreases in all schemes in the future scenario, since the anthropogenic emissions are kept constant, and the concentration SOA increases. However, the decrease in the relative contribution of HOA is stronger for the SOAmod scheme, since it shows a higher relative increase in the formation of BSOA in future scenarios.

We have also added the following phrase in section 5:

Indeed, the SOAmod scheme shows 80% of the OA mass in the non-volatile bins, while the SOAvbs and the SOA2p schemes only shows respectively around 10% and 20% in these bins.

17. P11 lines 8-9. “The contribution of HOA in future scenarios becomes less compared with historic simulations, probably since more BSOA formation happens in future scenarios”. Please verify if this applies for the actual concentration of HOA, as from Fig.5 the HOA contribution slightly changed between the historic and future simulations.

As it was mentioned by referee #1, the relative contribution of HOA becomes less since the relative contribution of LVOOA increases and the anthropogenic emissions don't change between historic and future simulations, by design of our study the absolute concentration of HOA does not change significantly, although some climatic response is seen for HOA as well. The following phrase has been added to the sentence:

The relative contribution of HOA decreases in all schemes since the anthropogenic emissions are kept constant in future simulations and the relative contribution of SOA increases in the future.

18. P12 lines 5-7. “However, the predicted increase for the future is higher for SOAvbs and SOAmod (figure 6, second column), reaching a maximum of 300% increase for the SOAmod scheme.” These numbers are not shown in Fig. 6 column 2. Is the second column referring to annual or summer period?

All features shown in figure 6 are for the summer period. We have added a statement to make this clear in the figure caption, and we have also emphasized this fact in the text where we present figure 6. The phrase is changed to the following:

reaching an average of 290% increase over the whole domain for the SOAmod scheme.

19. P12 Figure 6. Please clarify the second column if it is from summer period. Add the summer indication also in the caption for columns first and third. A suggestion would be to add the corresponding figure 6 for the annual simulation.

Again, figure 6 is solely for the summer period. We have added that the values are for the summer period, and we have also emphasized this fact in the text where we present figure 6. An annual figure for the same image, although an excellent idea, adds another image to an article already overcharged by images, so the authors will like to respectfully keep the image that is shown and not add another one.

20. P13 lines 22-24. The authors state that BSOA in SOAmod scheme presents the highest change. This is not true for the actual concentrations of BSOA, as SOA2p shows the highest absolute change. Also, regarding the relative change this applies only during summer period.

That's true for the Mediterranean area, this has been changed in the text to the following:

For BSOA relative changes, SOAmod still shows the largest relative change in the summer period compared to historic simulations (76%, 75% and 127% for SOA2p, SOAvbs and SOAmod respectively), but the differences between schemes are less pronounced in the Mediterranean area.

21. P13 lines 29-30. The statement is not true according to Fig. 5. The fossil sources are the major contributors only in the case of SOAvbs.

We agree. The phrase was modified to the following, to make a point about on larger fossile contributions in the med. area as compared to the European arean :

As for the European are, the contribution of non-fossil sources in the future scenarios also increases in the Mediterranean area, but still contributes less than over the European area.

22. P14 Figure 7. Please use the same axis for the two cases. Also, it is not clear where the cycles and cubes are in the figures. The use of the coefficient of determination (R²) instead of correlation coefficient (R) is advised.

The remark about using R² instead of R was taken into account and the figure was modified accordingly. The shapes of the points were changed to add a bit of a clearer view to the figure. The limits of the axis were not changed, since the two subdomains present very different axis limits both in terms of temperature and BSOA concentration/BVOC emissions which results in big empty spaces in both plots especially since the plots are on a logarithmic scale. The plot already being a bit complicated to read, we would like to keep the axis limits the way they are.

23. P15 line 22. Change the corresponding numbers according to the main manuscript.

The line has been modified to the following:

The results show that the change in concentration indicated by the SOAmod scheme is stronger especially for summertime, showing a difference of +113%, +156% and +263% for SOA2p, SOAvbs and SOAmod respectively, for the European area.

24. P16 line 4. Change the corresponding numbers according to the main manuscript. Currently are not consistent.

The line has been changed to the following:

the changes for this region are stronger in the SOAmod scheme as well (76%, 75% and 127% for SOA2p, SOAvbs and SOAmod respectively for summer).

Referee 2 minor comments:

1. P2 line 12. Remove commas after the dots in reference.

Modified.

2. P2 line 31. Remove comma after the dot in reference.

Modified.

3. P2 line 32. Define CHIMERE.

As far as I know, CHIMERE does not have a definition, it's a specific name given to the model.

4. P2 line 34. Remove all the dots in µg.m-3

Modified.

5. P2 line 35. Remove comma after the dot in reference.

Modified.

- 6. P5 line 12. Remove commas after the dots in reference.**

Modified.

- 7. P5 line 31. Add a dot after Cholakian.**

Modified.

- 8. P7 line 12. Define ECMWF.**

Modified.

- 9. P7 line 13. Define MEGAN.**

Modified.

- 10. P8 lines 11-12. Remove dots in molecules.cm2**

Modified.

- 11. P8 Figure 3. Check the order of the months. June-July-August are before March-April-May. Please magnify the table. Replace bials with bias.**

Modified.

- 12. P9 Figure 4. Line2. Please add next to historic simulations, also future simulations.**

Modified.

- 13. P10 line 12. Change august to August.**

Modified.

- 14. P12 Figure 6. Remove dots from units.**

Modified.

- 15. P15 line 6. Change VBS mod to SOAmod.**

Modified.

Referee 3 comments:

In this work, Cholakian et al. used three different organic aerosol simulation schemes in order to identify how they impact the calculated OA load on future climate projections. They found significant differences on the calculated biogenic SOA projections over Europe between the three OA schemes; highlighting the uncertainties that still exist on OA calculations. This study is of definite interest to the organic aerosol modeling community by contributing towards the understanding of the source of uncertainty between OA schemes (e.g., highlighting the role of temperature sensitivity). Overall, the manuscript is very well written and the presentation is clear. Therefore, I recommend this study for publication. Below are a few minor comments to be considered prior to publication.

We would like to thank referee #3 for their pertinent remarks for this paper, we have answered these comments point by point in the section that follows.

Referee 3 specific comments:

1. **Title: I believe the manuscript focuses a lot on biogenic SOA, therefore is better to replace the general term “particulate matter” with biogenic SOA. Furthermore, the manuscript presents the sensitivity of BSOA concentrations on the OA scheme used and not vice versa as the title implies. I suggest to consider revising the title.**

Yes, that is true. The title of the article has been changed to:

Biogenic SOA sensitivity to organic aerosol simulation schemes in climate projections

2. **Page 1 lines 7-8: the word “formation” is unnecessarily repeated two times in the sentence**

Yes, the second formation has been omitted from the sentence.

3. **Page 1 lines 7-8: This sentence is not clear. I assume you mean the temperature differences.**

I do not understand which sentence is referred here, probably the wrong line/page number is given in the comment?

4. **Page 2 line 1: Tsimpidi et al. (2017, doi: 10.5194/acp-17-7345-2017) is also a nice recent study that emphasizes the large uncertainty of OA formation.**

Reference added.

5. **Page 2 line 3: Lelieveld et al. (2015, doi:10.1038/nature15371) also highlight the adverse effects of OA on human health due to their increased toxicity.**

Reference added.

6. **Page 2 lines 22-25: The scheme of Pankow (1994, doi: 10.1016/1352-2310(94)90093-0) should be included in the discussion here**

Pankow (1994) is not an actual OA simulation scheme, but an OA gas-particle partitioning scheme which is also used in the CHIMERE model in order to distribute the OA into gaseous/particulate phases. It would be also appropriate to cite Odum (1996) in this part (the basis of the 2-product scheme) which has more bearing to what is being discussed. Both references have been added to this section:

Odum (1997) suggested a two-product scheme, where he calculated yields of production of OA from VOCs from laboratory data. He concluded that two virtual semivolatile organic

compounds were sufficient to represent the formation of OA. Following the partitioning theory of Pankow (Pankow, 1994), these species are distributed between the aerosol and gas phases. Pun and Seigneur, (2007) suggested a molecular single-step oxidation scheme for the formation of SOA, based on the Odum scheme.

- 7. Page 4 1st paragraph: More information is needed for the simulations conducted by the global models and WRF (e.g., which RCPs were used, which is the simulation period, etc. ?) Furthermore, are all the links between the models offline?**

The following information have been added to this paragraph:

The WRF simulations were prepared for the EURO-CORDEX project (Jacob et al., 2014) and use representative concentration pathways (RCPs, Meinshausen et al., 2011 ; van Vuuren et al., 2011) for future simulations. The EURO-CORDEX climatic runs were performed for the period of 1976—2005 for historic simulations and 2031—2100 for future scenarios, for RCP2.6, RCP4.5 and RCP8.5. A detailed analysis of these runs is provided in Vautard et al. (2014) and Jacob et al. (2014). In this work, the RCP8.5 runs are used for a selection of years (section 2.3). Anthropogenic emissions (base year 2010) are taken from the ECLIPSEv4a inventory (Amann et al., 2013; Klimont et al., 2013; Klimont et al., 2017), and the biogenic emissions calculated with by the MEGAN model (Guenther et al., 2006). The coupling of all these models with the CHIMERE model is done in an offline fashion, except for MEGAN which is directly coupled with CHIMERE.

- 8. Page 5 lines 18-21: It has been also shown that the aging of BSOA does not lead to any net changes on its mass concentration due to a balancing effect between fragmentation and functionalization (Murphy et al., 2012, doi: 10.5194/acp-12-10797-2012)**

As far as we've seen, that is the case when BSOA aging is considered with both functionalization and fragmentation processes active. If the fragmentation processes are inactive in the scheme, the aging causes a large amount of overestimation in the concentration of BSOA. This is mentioned in Murphy et al. (2012): "The detailed functionalization case overpredicted OA concentrations at all sites and underpredicted O:C ratios considerably" (taken from Murphy et al, 2012, conclusions) and Lane et al. (2008): "Including the aging reactions for SOA compounds causes a significant increase in the total predicted SOA concentrations". Also, we have seen the same results (as in an overestimation of BSOA when only aging and functionalization are considered) in our previous study (Cholakian et al. 2018).

- 9. Page 5 lines 22: Actually, the standard VBS scheme assumes that fragmentation and functionalization processes result in a net average decrease in volatility for SOA. Therefore, even if it does not simulate explicitly the fragmentation process, it has taken into account its effects on the SOA volatility changes.**

The sentence is modified to say that fragmentation is at least not considered explicitly.

Since the standard VBS scheme does not include fragmentation processes explicitly (when molecules break into smaller and more volatile molecules in the atmosphere)...

- 10. Page 5 lines 27: Can you briefly discuss the main differences between the standard and the modified VBS schemes (e.g., aging rate constants, changes on volatility and oxygen atoms added after each reaction step, etc.?)**

Reaction rates for the common species (and generations) do not change between schemes, reaction rates for new species and generations is taken from Shrivastava et al. (2013). The aging processes are all turned on in the modified VBS scheme, two more oxidation generations are added to POAs. BSOA oxidation generations are kept the same (one generation of oxidation). The formation of non-volatile SOA is added to all the SOA oxidized species (excluding POA), forming a nonvolatile SOA which cannot return to the gaseous phase. The same fragmentation fractions reported by Shrivastava et al. (2015) are used without any change. This same explanation has been added to the text at the specified part as well. The following paragraph has been added to the article:

Reaction rates for the common species (and generations) do not change between these two schemes, reaction rates for new species and generations is taken from Shrivastava et al. (2013). The aging processes are all turned on in the modified VBS scheme, two more oxidation generations are added to POAs. BSOA oxidation generations are kept the same (one generation of oxidation). The formation of non-volatile SOA is added to all the SOA oxidized species (excluding POA), forming a nonvolatile SOA which cannot return to the gaseous phase. The same fragmentation fractions reported by Shrivastava et al. (2015) are used without any change.

11. Page 7 lines 22: Can you comment on why all schemes significantly fail to reproduce the observed OA concentrations during winter?

It has been documented that the VBS and the molecular scheme both tend to underestimate the concentration of formed OA in urban areas (Bergström et al., 2012, Ciarelli et al., 2016). This is mostly because of sources lacking in the emissions inventories, especially for the residential sector (biomass burning). Since wintertime OA formation comes to a high degree from this source (Louvaris et al., 2017; Roig Rodelas et al., 2019), it is logical for the wintertime OA to be underestimated. However, the goal of the paper is changes of BSOA in future scenarios and this underestimation does not affect the results of our study.

12. Page 8 lines 11: Add “the” before “average”. Furthermore, the emission units do not seem correct. You need “amount time-1 area-1”. It would be better to report the emissions in Tg/yr for the whole domain.

The modifications have been made and emissions unit has been fixed. However, since the output unit in the model for these emissions is in the unit mentioned in the article ($\text{molecules cm}^{-2} \text{s}^{-1}$), we would prefer to keep it the way it is.

13. Page 9, Figure 4: The figure caption states that these are BSOA but the figure legend has SOA (i.e., SOA2p, SOAvbs, SOAmod)

Yes, the figures are for BSOA. The SOA2p, SOAvbs and SOAmod are the names given to each scheme (as explained in section 2.2) and do not mean that the SOA is shown.

14. Page 9 lines 6-9: I found this sentence long and confusing

Yes, the sentence seems to be too long and confusing and has been modified:

These changes show that the climate impact on changes of BSOA in the future might be underestimated until now on a relative scale. This is because most of the future simulations performed in order to explore climate impact use a two-product or a molecular single step scheme for the simulation of SOA. However, our study shows that using a VBS based scheme increases the climate induced effect on the change in BSOA concentration in the future.

15. Page 10 lines 12: use capital A for August

Modified.

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\documentclass[acp]{copernicus} % for 2-column papers
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\begin{document}
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\title{Biogenic SOA sensitivity to organic aerosol simulation schemes in climate
projectionsSensitivity of organic aerosol simulation scheme on particulate matter concentrations
in climate projections}
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\runningtitle{Importance of organic aerosol simulation scheme in climate projections}
\runningauthor{A.~Cholakian et al.}
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\begin{abstract}
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Organic aerosol (OA) can have important impacts on air quality and human health because of its large contribution to atmospheric fine aerosol, and its chemical composition, including many toxic compounds. Simulation of this type of aerosol is difficult since there are many unknowns in its nature, and mechanism and processes involved in its formation. These uncertainties become even more important in the context of a changing climate, because different mechanisms, and their representation in atmospheric models, imply different sensitivities to changes in climate variables. ~~Organic aerosol can have important impacts on air quality and human health because of its chemical composition and its large contribution to the atmospheric fine aerosol. Simulation of this aerosol is difficult since there are many unknowns in the nature, mechanism and processes involved in the formation of these aerosols. These uncertainties become even more important in the context of a changing climate, because different mechanisms, and their representation in atmospheric models, imply different sensitivities to changes in climate variables.~~ In this work, the effects caused by using different schemes to simulate OA are explored. Three schemes are used in this work: 1) a molecular scheme, 2) a standard volatility basis set (VBS) scheme with

anthropogenic aging and 3) a modified VBS scheme containing functionalization, fragmentation and formation of non-volatile SOA formation for all semi-volatile organic compounds (SVOCs). 5 years of historic and 5 years of future simulations were performed using the RCP8.5 climatic scenario. The years were chosen in a way to maximize the differences between future and historic simulations. The study focuses on BSOA since the contribution of this fraction of BSOA among OA is major in both historic and future scenarios (40 to 78% for different schemes in historic simulations). Simulated OA and BSOA Absolute concentrations between with different schemes are also different, the molecular scheme showing the highest concentrations among the three schemes. The comparisons show that for the European area, the modified VBS scheme shows the highest relative change between future and historic simulations, while the molecular scheme shows the lowest (a factor of two lower). These changes are ~~maximized-largest~~ over the summer period for biogenic SOA (BSOA) because the higher temperatures increase terpene and isoprene emissions, the major precursors of BSOA. This increase is partially off-set by a temperature induced shift of SVOCs to gas phase. This shift is indeed scheme dependent, and it is shown that it is the least pronounced for the modified VBS scheme including a full suite of aerosol aging processes, comprising also formation of non-volatile aerosol. For the Mediterranean Sea, without BVOC emissions, the OA changes are less pronounced and, at least on an annual average, more similar between different schemes. ~~Absolute concentrations between different schemes are also different.~~ Our results warrant further developments in organic aerosol schemes used for air quality modelling to reduce their uncertainty, including sensitivity to climate variables (temperature).

\end{abstract}

\introduction

Organic aerosol (OA) is an important fraction of fine particulate matter (PM) concentrations. Its production results from both primary emissions of organic aerosols, as well as secondary formation from semi-volatile or polar precursor gases in the atmosphere. The mechanisms and pathways of secondary organic aerosol (SOA) formation are in general highly uncertain (Hallquist et al., 2009, [Tsimpidi et al., 2017](#)). Yet, the importance of the concentrations of OA in the atmosphere (Jimenez et al., 2009) and their adverse effects on human health (Mauderly and Chow, 2008, [Lelieveld et al. 2015](#)) make them an important subject to study.

Considering that modelling OA already contains important uncertainties, the uncertainties become even more important for future climate scenarios which account for climate change. These future scenarios present an important number of uncertainties, both due to climate related parameters, but also due to the description of how they act on specific processes. As an example, biogenic volatile organic compound (BVOC) emissions, which are the main precursors of biogenic SOA (BSOA), can be affected by e.g. temperature and land use changes, CO₂ inhibition (Heald et al., 2008a) among other factors. Many studies have addressed the effects of these parameters on the BVOC emissions, and a high variability was found in BVOC emissions depending on the factors that were considered in each study. For example, Heald et al. (2009) explored the effects of land use change and CO₂ inhibition on the emission of BVOCs and they found a 130% of isoprene emission increase in 2100 compared to 2000, while Pacifico et al. (2012) and Hantson et al. (2017) show 70% and 41% increase for isoprene for the same years with different parameters. Langner et al. (2012) compares four different models for the European region reporting an isoprene increase in the range of 21%-26%. Cholakian et al. (2019b), found an increase of 52% for isoprene for the period of 2031-2100 compared to 1976-2005 because of only temperature change for Europe, amounting to a 12% increase in BSOA concentrations.

In addition, for the formation of anthropogenic SOA (ASOA), future urbanization, anthropogenic emission and wood burning emission changes can be mentioned as possible factors. Each one of these parameters represents an uncertainty, which, when coupled with the inherent uncertainty in the simulation of OA, can present important sources of error-sources.

It is mainly to assess the future evolution of tropospheric ozone that BVOC emissions have been quantified at global scale in chemistry-climate projections (Arneth et al., 2010). Their importance for organic aerosol chemistry is only emerging has also been considered in global and regional scale atmospheric models (Maria et al., 2004; Tsigaridis et al., 2007; Heald et al., 2008b), but to a lesser degree. Several different types of OA simulation schemes can be used in chemistry-transport models (CTMs). Odum (1997) suggested a two-product scheme, where he calculated yields of production of OA from VOCs from laboratory data. He concluded that two virtual semi-volatile organic compounds were sufficient to represent the formation of OA. Following the partitioning theory of Pankow (Pankow, 1994), these species are distributed between the aerosol and gas phases. Pun and Seigneur, (2007) suggested a molecular single-step oxidation scheme for the formation of SOA, based on the Odum scheme. ~~Pun and Seigneur, (2007) suggested a molecular single-step oxidation scheme for the formation of SOA.~~ Another approach is the volatility basis set (VBS) scheme, which includes different volatility bins and aging of semi-volatile species lowering their volatility (Donahue et al., 2006; Robinson et al., 2007). This scheme presents two major versions: 1-dimensional (1D) and 2-dimensional (2D) VBS. 1D-VBS distributes semi-volatile organic compounds (SVOCs) into different bins with regards to their volatility (Robinson et al., 2007). A 2D-VBS scheme, takes into account the oxygen to carbon (O/C) ratio as well as the volatility (Donahue et al., 2011; Donahue et al., 2012). While 1D-VBS has been tested extensively in different CTMs (i.e Lane et al., 2008; Hodzic and Jimenez, 2011; Zhang et al., 2013; Cholakian et al., 2018*), the use of 2D-VBS is less frequent because of its even more challenging numerical needs. Other variations of the 1D-VBS have been also used for observation-simulation comparisons, each one adding some variables to the basic VBS scheme or building upon its framework. For example, Shrivastava et al., ~~(2015)~~ (2015) adds fragmentation and formation of nonvolatile SOA mechanisms to the basic 1D-VBS scheme. This scheme was implemented into the CHIMERE CTM and tested for the Mediterranean region with good results in terms of concentration (correlation of 0.55 and a bias of $-0.68 \mu\text{g m}^{-3}$ for the summer period of 2013), fossil/non-fossil distribution and oxidation level of OA (Cholakian et al., 2018*). Besides, Lannuque et al., ~~(2018)~~ (2018) provide a new parameterization for the VBS scheme by using a box model based on the GECKO-A modelling tool, which was afterwards implemented in CHIMERE and tested for the European continent, showing a good correspondence between modeled and measured OA (Lannuque et al. 2019, in prep).

In addition, the sensitivity of OA schemes to thermodynamic parameters could show large differences due to different processes considered or due to the differences in the parameterization. The formation and partitioning of particulate OA can show various degrees of dependency to temperature in different OA schemes. Therefore, the sensitivity of organic aerosol to climate change, affecting these thermodynamic parameters (mainly temperature), also depends on the OA scheme used. To our knowledge, this issue has not yet been addressed in a dedicated work. In most future scenarios, a two-product scheme is used for the simulation of SOA. However, other schemes, such as different variations of the VBS scheme, could better represent the more complex characteristics of SOA, such as, for example, its oxidation state.

~~Differences induced by different schemes are also expected to vary regionally, depending on the concentration ranges encountered and ranges and changes in meteorological parameters. It is also interesting to take into account that the thermodynamic changes in each scheme can be different depending on the region for which the simulations are performed, since various areas in the world can show different sensitivity to climate change.~~ In this study, we focus on the European continent and the Mediterranean basin. The Mediterranean basin, is one of the most sensitive regions to climate change, which makes it important and at the same time interesting to study. However, not much focus has been given to the Mediterranean in the literature, especially for the western side of this basin (Giorgi, 2006). For this reason, the ChArMEx project was put into place, in order to study the current chemical characteristics of the atmosphere of the Mediterranean region and its changes in future scenarios.

In this study, future OA concentrations under a climate change scenario will be quantified using different OA schemes. Three OA simulation schemes are compared, namely (i) a ~~two-product~~two-product scheme, (ii) a VBS scheme with anthropogenic aging and (iii) a modified VBS scheme including fragmentation and nonvolatile SOA formation. A representative concentration pathway climatic scenario (RCP) has been used. RCP8.5 has been chosen in order to maximize future changes and to get a clear climate change related signal in our study.

The paper is organized as follows: Section 2 explains the modeling framework for this work. An evaluation of the three schemes against measurements is provided in section 3, while section 4 presents results for the different scenarios. Conclusions are presented in section 5.

\section{Simulations}

The modelling framework in this study utilizes a chain of models, covering the different compartments of the atmosphere, a global circulation model and a global chemistry transport model providing meteorological and chemical conditions of the atmosphere respectively (figure 1). In order to down-scale the output provided by the global models a regional climate model and a regional chemistry transport model are used (figure 1). Global circulation data is provided by IPSL-CM5A-MR (Taylor et al., 2012 ; Dufresne et al., 2013 ; Young et al., 2013), while the LMDZ-INCA (Hauglustaine et al., 2014) global chemistry transport model, using simulations from global circulation model as meteorological input, provides boundary conditions for the regional chemistry transport model (CTM). The boundary conditions include inputs for organic carbon as well. The global circulation model also provides boundary conditions for the regional climate model, WRF (Weather Research and forecasting, Wang et al., 2015), which, in return provides meteorological input fields for the regional CTM, CHIMERE (Menut et al., 2013). The WRF simulations were prepared for the EURO-CORDEX project (Jacob et al., 2014) and use representative concentration pathways (RCPs, Meinshausen et al., 2011 ; van Vuuren et al., 2011) for future simulations. The EURO-CORDEX climatic runs were performed for the period of 1976—2005 for historic simulations and 2031—2100 for future scenarios, for RCP2.6, RCP4.5 and RCP8.5. A detailed analysis of these runs is provided in Vautard et al. (2014) and Jacob et al. (2014). In this work, the RCP8.5 runs are used for a selection of years (section 2.3). Anthropogenic emissions (base year 2010) are taken from the ECLIPSEv4a inventory (Amann et al., 2013; Klimont et al., 2013; Klimont et al., 2017), and the biogenic emissions are ~~provided~~bycalculated with the MEGAN model (Model of Emissions of Gases and Aerosols from Nature, Guenther et al., 2006). The coupling of all these models with the CHIMERE model is done in an off-line fashion, except for MEGAN which is directly coupled with CHIMERE. Since the focus of this article is on the SOA scheme changes in the regional CTM, only this model will be

discussed in further detail. More information on the modeling framework [in the current study](#) is provided in Colette et al., 2013; 2015.

[\subsection{CHIMERE chemistry transport model}](#)

The CHIMERE chemistry transport model has been widely used in different parts of the world (Carvalho et al., 2010; Hodzic and Jimenez, 2011), especially in Europe (Zhang et al., 2013 ; Petetin et al., 2014; Colette et al., 2015; Menut et al., 2015 ; Rea et al., 2015), for both forecasting and analysis purposes. It provides a wide range of capabilities; if input information such as anthropogenic/biogenic emissions, meteorological conditions are given, it can simulate an exhaustive list of atmospheric components. Different chemistry schemes are available in the model, in the case of our simulations, the MELCHIOR2 scheme (Derognat et al., 2003) is used, containing around 120 reactions. A sectional logarithmic aerosol size distribution of 10 bins is used with a range of 40 nm to $40\text{ }\mu\text{ m}$. The aerosol module in CHIMERE includes different chemical and physical processes such as gas/particle partitioning, coagulation, nucleation, condensation, as well as dry and wet deposition. The chemical speciation contains EC (Elemental Carbon), sulfate, nitrate, ammonium, SOA/SVOC species, dust, salt and PPM (primary particulate matter other than ones mentioned above). More information on the SOA scheme will be provided in the next section. The simulation domain covers the whole Europe with a resolution of 0.44° , the domain used in all the simulations are all the same [\(the domain approximately covers 30-70°N and 40W-60E\)](#).

[\subsection{OA schemes used for the simulations}](#)

The CHIMERE model has three SOA simulation schemes with different levels of complexity, all based on a molecular single-step oxidation scheme. In our base simulations, the medium complexity scheme is used (Bessagnet et al., 2008). In this scheme, lumped volatile organic compounds (VOCs) can react and form classes of organics with reduced volatility, i.e. SVOCs. Once formed, the model distributes these species between the gaseous and particulate phases according to the mixing theory of Pankow (Pankow, 1987). The yields for the formation of SOA are taken from Odum et al., (1997), Griffin et al., (1999) and Pun and Seigneur (2007). This scheme is referred to as the SOA2p scheme here after. A large database of historic and future simulations exists for this scheme, for three RCPs (RCP2.6, RCP4.5 and RCP8.5), each containing 70 years of simulation (2031 – 2100) and 30 years (1976 – 2005) of historic simulations. These scenarios are discussed and compared in Colette et al., (2013a), Lemaire et al., (2016) and Cholakian et al. (2018b) in more detail.

The VBS approach was developed as a general framework to account for the semi-volatile character of organic matter and to allow for changes in volatility over time. In VBS schemes, the SVOCs are partitioned into bins according to their saturation concentrations. Aging processes included by transferring species from one volatility bin to another (Robinson et al., 2006). This scheme was implemented into CHIMERE and tested for Mexico City (Hodzic and Jimenez, 2011) and the Paris region (Zhang et al., 2013). Nine volatility bins with saturation concentrations in the range of $0.01\text{ }\mu\text{ g m}^{-3}$ to $10^6\text{ }\mu\text{ g m}^{-3}$ are taken into account and the emissions of SVOC and IVOC (Intermediate Volatility Organic Compounds) are distributed into these bins using the aggregation proposed by Robinson et al. (2007). Four volatility bins are used for ASOA and BSOA ranging from $1\text{ }\mu\text{ g m}^{-3}$ to $1000\text{ }\mu\text{ g m}^{-3}$. Since the aging processes of biogenic SOA were reported to overestimate the BSOA concentrations in CTM runs for North America (Robinson et al., 2007; Lane et al., 2008) and the [Mediterranean sea](#) [Mediterranean Sea](#) (Cholakian et al., 2018a), these processes are not taken into

account in this work. Gaseous-particulate partitioning is treated following Raoult's law and depends on total organic aerosol concentrations.

Since the standard VBS scheme does not include fragmentation processes (when molecules break into smaller and more volatile molecules in the atmosphere) explicitly and the formation of non-volatile SOA (when SOA, after their formation, become irreversibly non-volatile and therefore cannot be oxidized further), these processes were added to the basic VBS scheme following Shrivastava et al. (2011; 2013; 2015). Another change made to the VBS scheme was to include an interpolation between high-NO_x and low-NO_x regimes (Carlton et al., 2009). Reaction rates for the common species (and generations) do not change between these two schemes, reaction rates for new species and generations is taken from Shrivastava et al. (2013). The aging processes are all turned on in the modified VBS scheme, two more oxidation generations are added to POAs. BSOA oxidation generations are kept the same (one generation of oxidation). The formation of non-volatile SOA is added to all the SOA oxidized species (excluding POA), forming a nonvolatile SOA which cannot return to the gaseous phase. The same fragmentation fractions reported by Shrivastava et al. (2015) are used without any change.

Both the standard VBS without biogenic aging (referred to as SOAvbs scheme here-after) and the modified VBS including fragmentation and formation of non-volatile aerosol (referred to as SOAmod scheme here-after) schemes are presented in more detail and compared to experimental data in the western Mediterranean area in Cholakian et al. (2018a). In the aforementioned work, it was concluded that these two schemes can reproduce the levels of concentration of organic aerosols in the Mediterranean basin successfully in regards to concentration of OA, while oxidation state and fossil/non-fossil repartition is better represented in SOAmod.

subsection{Choice of years}

The SOAvbs and the SOAmod schemes are both numerically very resource-consuming, therefore, only 10 years of simulations for each scheme were performed. In order to choose the appropriate years for the simulation, ~~an~~ existing long-period sets of simulations were used, containing 30 years of historic simulations (1976-2005) and 70 years of future scenarios (2031-2100). We address results for BSOA, as it makes the major contribution to OA during summer (between 40 and 78% for different schemes in the historic scenario, according to our simulation results for the historic period with differences schemes, not shown in figures).

The simulations were performed using the previous version of CHIMERE (chimere-2013b, Menut et al 2013), the SOA2p scheme and the RCP8.5 scenario. This dataset was used to choose five years of simulations in the historical and future periods each, with the aim to maximize both the temperature and SOA differences between historic and future scenarios. Figure 2-a shows the monthly average of BSOA concentrations in different RCP scenarios, showing that the production of BSOA reaches its maximum in the period of 4 months of June, July, August and September. During these months BSOA is the major SOA and OA component over Europe as also discussed in Cholakian et al. (2019b).

Figure 2-a also displays that the differences of historic and future simulations reaches its maximum for RCP8.5 simulations. The concentration of BSOA and the temperature both in historic and RCP8.5 simulations show a strong positive correlation as seen in figure 2-b and 2-c, each point representing the average of the four months mentioned previously for one year.

For historic simulations, the years representing the lowest temperature and BSOA concentrations are used, which correspond to years 1980, 1981, 1984, 1985 and 1986, while for future scenarios

the years with the highest temperature and BSOA concentrations are used corresponding to years 2087, 2092, 2093, 2095 and 2098.

\section{Scheme validation}

The three schemes show high variability when simulating the concentration and characteristics of OA, therefore, we performed an evaluation to investigate their performances. The schemes are compared to observations for the year 2013 during which an abundance of observational data is available. A year-long simulation for the year 2013 was performed for each of the schemes. The inputs used in these simulations are the same: anthropogenic emissions are taken from EMEP (European Monitoring and Evaluation Programme, [\url{http://www.ceip.at}](http://www.ceip.at)), meteorological fields are generated using the ECMWF ([European Centre for Medium-Range Weather Forecasts](#)) input data (Berrisford et al., 2011), biogenic emissions are provided by MEGAN (Guenther et al., 2006) and boundary and initial conditions are taken from LMDZ-INCA (Hauglustaine et al., 2014).

The observations are mostly accessed from the EBAS database ([\url{http://ebas.nilu.no/}](http://ebas.nilu.no/), [T{\o}rseth et al., 2012](#)). The used measurements are mostly $PM_{2.5}$, in some cases PM_1 . For each type of measurement ($PM_{2.5}$ or PM_1) the corresponding fraction from the simulations has been used. In some cases, data was provided by the lead investigator for a specific station, and the measurements for the two stations of Corsica and Mallorca have been added using the ChArMEX ([\url{http://mistrals.sedoo.fr/ChArMEX/}](http://mistrals.sedoo.fr/ChArMEX/)) campaign measurements. In total, 32 stations are compared to simulations. Bear in mind that for some of these stations the available data covers a shorter period than one year, or they present weekly measurements rather than daily observations. The list of stations with information about each station and the type of measurement is provided in appendix A.

Results of these comparisons are shown in figure 3 and the statistic information is shown in table 1. Regarding the concentration of OA, the modified VBS scheme shows slightly less a stronger bias ($-0.644 \mu g \cdot m^{-3}$ compared to $0.42 \mu g \cdot m^{-3}$ and $0.1 \mu g \cdot m^{-3}$ for SOAvbs and SOA2p respectively) for the summer period. In fact, all compared schemes underestimate the winter period (-1.45 , -1.67 and $-10.63 \mu g \cdot m^{-3}$ for SOAmod, SOAvbs and SOA2p respectively). The annual biases for the three schemes are -0.91 , -0.4 and $-0.6514 \mu g \cdot m^{-3}$ for SOAmod, SOAvbs and SOA2p respectively. The correlation of determination between observed and simulated OA concentrations for different schemes are the highest for the modified VBS scheme SOAmod, and lowest for the standard VBS scheme SOAvbs in most seasons; it should also be noted that the difference between the correlations seen for each scheme are rather small (difference of below 0.05). The Taylor diagram in figure 3 shows the comparisons of different stations to simulations for each scheme (black for SOA2p, green for SOAmod and red for SOAvbs). The Taylor diagram summarizes several statistic information in one plot: The correlation coefficient, root-mean-square (RMS) difference between observations and simulations and the standard deviation ratio can be seen. More information about the construct of this diagram is given in Taylor, 2001. As seen in this diagram, there is a high variability in the simulation of different stations, some stations are better represented by the model than the others. This might be because of the geographical placement (altitude, types of emissions in said location, etc.) of stations or because of the nature of the station (urban, rural, etc.).

The three schemes perform reasonably well according to the criteria introduced by Boylan and Russell (2006), with the values for all the schemes falling into zone 1 for both mean fractional bias (MFB) and mean fractional error (MFE). The goal for these two metrics according to the aforementioned reference is less than or equal to $\pm 30\%$ and $+50\%$ and the criteria is less than or equal to $\pm 60\%$ and $+75\%$ respectively. The MFB values for the three schemes are -19.7% , 16.5% and 26.9% , while MFE shows 47.9% , 51.1% and 47.2% for SOA2p, SOAvbs and SOAvbs respectively. Thus, performance goals are met nearly for all schemes, with a slight exceedance for MFE and the SOAvbs scheme, still meeting the criterium. Each one of the schemes performs better for a specific period; modified VBS in summer, CHIMERE standard scheme during winter, and the standard VBS scheme showing average performance during the whole year. Looking at table 1, it is seen that for example for the summer period, the SOAmod scheme shows the highest correlation of determination, while SOA2p shows the lowest bias for this season. For spring SOA2p shows the highest R^2 , while SOAvbs shows the lowest bias. For winter and autumn, the performance of the schemes is quite similar. Annually, SOAvbs presents a similar correlation to SOA2p while showing the lowest bias in general. The types of stations have not been filtered in the current study, therefore, all stations, including urban, semi-rural or rural have been included for the comparisons. This could be responsible for part of the observed negative bias. As a conclusion, all three schemes correspond to the performance goals and/or criteria of Boylan and Russell (2006), albeit they show important, and spatially and seasonally dependent, differences with observations. Thus, the three schemes will be retained for the following analysis with equal confidence.

\section{Analysis of the simulations}

The ~~presentation analysis~~ of the simulations will be presented in the next two sub-sections. First, the changes in BVOC emissions are discussed. Subsequently, the results for the European continent regarding concentration, origins and oxidation state will be presented, ~~and a general comparison of the spatial distribution will be done for different schemes.~~ Finally, an analysis of these parameters will be performed for the Mediterranean sub-domain including their origins and the oxidation state. Finally, a general comparison of the spatial distribution will be performed for different schemes. It is important to keep in mind that from this section on, whenever BSOA concentrations are discussed the PM_{10} fraction of this species has been used.

\subsection{Changes in biogenic emissions}

The changes in biogenic emissions are important in the context of this work, since they are highly dependent to temperature changes. For the simulations presented in this work, the biogenic emissions do not change between different schemes, however they change quite a bit between historic and future simulations because of temperature increase in the future. Since the choice of the years was done to maximize future temperature changes, the differences between future and historic simulations are quite remarkable. For the European region, the average "historical" historic isoprene emissions are $1.3 \times 10^{11} \text{ molecules cm}^{-2} \text{ yr}^{-1}$ and average historical terpene emissions are $3 \times 10^{10} \text{ molecules cm}^{-2} \text{ yr}^{-1}$. An increase of 88% and 82% for isoprene and terpenes is seen respectively in the future scenarios in response to an average temperature increase of 5.5°C . For the summer period, the biogenic emission increase raises to 93% and 92% for isoprene and terpenes for a temperature increase of 6.4°C (figure 6). The correlation of determination between historic isoprene and terpene emissions is 0.685 and 0.63 while this correlation is 0.6591 and 0.577 for these future simulations.

For the Mediterranean ~~region~~Sea, there are no local biogenic emissions included in the model. ~~A land-sea mask was used to separate the Mediterranean Sea from the other parts of the domain; therefore, our “Mediterranean Sea” labeled domain only contains cells with sea without any land cells, i.e. with zero biogenic emissions.~~

\subsection{European region}

\subsubsection{Changes in BSOA concentration}

~~We address results for BSOA, as it makes the major contribution to OA during summer (between 40 and 78% for different schemes in the historic scenario).~~ BSOA concentrations in future scenarios are predicted to increase in all the schemes. However, the intensity of this increase is scheme dependent: while for SOA2p an annually averaged increase of +6194% is calculated, this percentage ~~raises to +13580% for SOAvbs and +98189% for SOAmod~~ for the same period. ~~These changes~~this change in intensity shows that the climate impact on changes of BSOA in the future might ~~have been~~be underestimated until now on a relative scale. ~~This is because since~~ most of the future simulations performed in order to see explore climate impact use a two-product or a molecular single step schemes for the simulation of SOA. ~~A, while~~ However, our study shows that using a VBS based scheme increases the climate induced effect on the change in BSOA concentration in the future. Reasons for this behavior will be discussed in section 5. However, we would like to ~~notice~~ emphasize that changes are maximized by the choice of the RCP8.5 scenario and the years chosen for the simulations in this work. Also, it should be noted that there are important differences in absolute concentrations between different schemes (see above). -

There is a strong seasonality for the BSOA production. The seasonal changes for BSOA are seen in figure 4-a1, 4-b1 and 4-c1 for historic simulations, the absolute difference between future and historic simulations, and their relative changes respectively. Summer shows the maximum relative increase (+11322%, +15549% and +26244% for SOA2p, SOAvbs and SOAmod respectively) and winter the lowest one in all schemes (+31.128%, +26.228% and +20.522% for SOA2p, SOAvbs and SOAmod respectively). For autumn and spring SOA2p and SOAmod show similar and intermediate changes while SOAvbs shows higher relative differences (+59.66%/+40.339%, +79.956%/+60.045% and +57.373%/+50.060% for SOA2p, SOAmod and SOAvbs respectively for autumn/spring).

For monthly results, as seen in figure 4-a2, 4-b2 and 4-c2 there is an increase (both in relative and absolute values) in almost all months for all schemes during the year, but the intensity of this increase changes for different months. In July, when the BSOA concentration reaches its maximum, the percentage of change in the future is high as well (+11525%, +15137% and +243216% for SOA2p, SOAvbs and SOAmod respectively). Highest relative changes occur for August for all schemes (+11133%, +1658% and +35633% for SOA2p, SOAvbs and SOAmod respectively) resulting from the seasonal profile of BVOC emissions. For SOAmod, a decreased change is seen for some months in the future scenarios (-11%, -1.6% and -0.45% for April, October and November respectively).

\subsubsection{Changes in the origins and volatility state of OA}

Figure 4-d shows a simplified distribution for the OA in different schemes: SOA and HOA (hydrocarbon-like organic aerosol) presenting the freshly emitted primary OA. Figure 4-d shows that the predicted distribution between HOA and SOA is different for the three schemes. SOA2p indicates a smaller contribution of SOA and a larger one from HOA compared to SOAvbs and

SOAmod schemes. This is because POA emissions in SOA2p are considered non-volatile, while they are volatile in VBS schemes. The relative contribution of HOA decreases in all schemes in the future scenario, since the anthropogenic emissions are kept constant, and the concentration SOA increases. However, the decrease in the relative contribution of HOA is stronger for the SOAmod scheme, since it shows a higher relative increase in the formation of BSOA in future scenarios. Since the schemes behave differently both in distribution of origins as well as volatility bin aspects, it is interesting to compare these two aspects in the tested schemes. However, since the surrogate species for different sources are present in the outputs, the fossil/non fossil repartition can be easily calculated. ASOA is considered to be in the fossil fraction (neglecting a small fraction due to bio-fuels) and BSOA in the non fossil fraction. For carbonaceous aerosol, residential/domestic uses are considered as non fossil as they are mostly related to wood burning (Sasser et al., 2012). When comparing the simulated fossil/non fossil fraction, some differences are observed. The SOAvbs scheme predicts more SOA in the fossil fraction mainly because it takes into account aging of anthropogenic SVOCs and not of biogenic SVOCs. On the contrary, the SOAmod scheme takes into account the aging for both biogenic and anthropogenic SVOCs, therefore it simulates more in the non fossil compartment. All schemes show an increase in the contribution of non fossil sources in the future (79%, 74% and 84% increase in non fossil contribution for SOA2p, SOAvbs and SOAmod in future scenarios). SOA2p indicates a higher increase in non fossil contribution compared to other schemes. As already discussed, a strong seasonality is seen for this factor as well. The contribution of non fossil sources becomes much higher in summer (figure 5), when BVOC emissions are largely abundant. The increase in the contribution of non fossil sources is logical since the anthropogenic emissions of OA precursors are kept the same and the biogenic emissions of these species increase with increasing temperature.

The OA oxidation state calculated for different schemes is also compared, using the definitions given to different groups of species in regards to volatility presented in Donahue et al. (2012). In this comparison, LVOOA corresponds to low volatility oxidized OA ($C^{*} \leq 0.1 \mu\text{g.m}^{-3}$), SVOOA to semi volatile oxidized OA ($C^{*} \leq 1 \mu\text{g.m}^{-3}$) and HOA to hydrogen like almost un oxidized OA (primary OA regardless of their saturation concentration). In other words, LVOOA represents aged OA, SVOOA to freshly formed SOA, and HOA primary OA (POA). In the case of all these schemes, a comparison was done for the measurements during the ChArMEx campaign for the summer of 2013 (Cholakian et al. 2018a), results show that SOAmod corresponds very well to the repartition observed in the measurements regarding to oxidation state and obtained by positive matrix factorization (Paatero and Tapper, 1994), thus SOAmod will be taken here as a reference. Figure 5 shows that the predicted oxidation state of OA, is different for the three schemes. SOA2p indicates much less LVOOA and much more HOA compared to SOAmod, because POA emissions in SOA2p are considered non-volatile. SOAvbs does not form particles aged enough to be considered as LVOOA, because aging of biogenic SVOCs is not taken into account. In addition, the formation of anthropogenic LVOOA is taken into account, but has a minor effect compared to the biogenic one. Thus, LVOOA is underestimated in this scheme and SVOOA overestimated. The contribution of HOA in future scenarios becomes less compared with historic simulations, probably since more BSOA formation happens in future scenarios. This is seen especially for SOA2p for which the HOA participation is more pronounced. The contribution of LVOOA becomes higher in future as well, since the volatilization of this class of organic compounds is less affected by higher temperatures than that of SVOOA.

The schemes behave differently in contribution of different origins in the formation of OA as well, therefore it is interesting to compare this aspect in the tested schemes. Since surrogate

species for different sources are present in the outputs, the fossil/non-fossil repartition can be easily calculated. ASOA is considered to be in the fossil fraction (neglecting a small fraction due to bio-fuels) and BSOA in the non-fossil fraction. For carbonaceous aerosol, residential/domestic uses are considered as non-fossil as they are mostly related to wood burning (Sasser et al., 2012). When comparing the simulated fossil/non-fossil fraction, some differences are observed. The SOAvbs scheme predicts more SOA in the fossil fraction mainly because it takes into account aging of anthropogenic SVOCs and not of biogenic SVOCs. On the contrary, the SOAmod scheme takes into account the aging for both biogenic and anthropogenic SVOCs, therefore it simulates more in the non-fossil compartment. All schemes show a relative increase in the contribution of non-fossil sources in the future on an annually averaged basis (10%, 17% and 22% of increase for the non-fossil partition between future and historic simulations for SOA2p, SOAvbs and SOAmod respectively). As already discussed, a strong seasonality is seen for this factor as well. The contribution of non-fossil sources becomes much higher in summer (figure 4-e), when BVOC emissions are largely abundant. The increase in the contribution of non-fossil sources is logical since the anthropogenic emissions of OA precursors are kept the same and the biogenic emissions of these species increase with increasing temperature.

\subsection{Mediterranean region}

While the differences between the schemes for the European area are important to explore in future scenarios, we also focus on the Mediterranean region because of several reasons: high sensitivity to climate change, high burden of OA (and PM in general, Lin et al., 2012; Lin et al., 2014) and also high temperatures in the area. Because of these reasons, we perform a similar analysis as in the previous section. As explained before, a land-sea mask has been used in order to separate the Mediterranean Sea, therefore the analysis explained below regards only the Sea without any land surface cells.

\subsubsection{Changes in BSOA concentration}

There are major differences ~~between the partitioning of PM₁₀ into different aerosol components~~between the concentrations of different aerosol components—over the Mediterranean area compared to continental Europe. For example, the concentrations of salt and dust particles are higher, for the former because of the marine environment and for the latter because of the North African dust emissions which are transported to the Mediterranean area. On the contrary, the concentrations of nitrate and BSOA are lower than in the continental area; in the case of nitrate particles, because of higher temperatures its formation is less efficient than it is in continental Europe, and for the BSOA because of lack of emission sources over the marine environment. The differences seen for BSOA concentrations in different schemes are presented in figure 54 (panels 54-a1d, 54-b1e and 54-c1 for seasonal results and 5-a2, 5-b2 and 5-c2 for monthly results†). The behavior of different schemes in regards to differences between historic and future simulations differs between the Sea and the continental area. For BSOA relative changes, SOAmod still shows the largest relative change in the summer period compared to historic simulations (762%, 753% and 12781% for SOA2p, SOAvbs and SOAmod respectively), but the differences between schemes are less pronounced—in the Mediterranean area.

\subsubsection{Changes in the origin and volatility state of OA}

For all three schemes, the contribution of fossil sources to OA is slightly larger for the Mediterranean sub-domain than for Europe (figure 5-e). The reason for this change-difference is the fact that there are local fossil OA formation sources in the Mediterranean Sea, i.e. shipping emissions, while OA originating from non-fossil sources are not directly emitted in this area and

are transported from outside. While the contribution of non-fossil sources increases in the future scenarios, fossil sources ~~still are still the major contributors~~ contribute more in the Mediterranean area compared to the European area, relative to non-fossil sources.

Both for the historic and the future simulations, the ~~oxidation state~~HOA/SOA distribution does not change considerably in the Mediterranean area as compared to the European area (figure 5-d).

subsection{Spatial distribution of future changes}

Figure 6 shows the concentration of BSOA in different schemes (in $\mu\text{g m}^{-3}$ first column), the percentage of differences between historic and future simulations (second column), concentrations of isoprene and mono-terpenes and temperature for all schemes in the third column and the changes of these parameters in future scenarios in the fourth column. All the panels in figure 6 show the summer period. The concentration of BSOA in SOA2p simulations is much higher than that of SOAvbs and even more so than that in SOAmod at the lower end. However, the predicted increase for the future is higher for SOAvbs and SOAmod (figure 6, second column), reaching an average of 290% increase over the whole domain for the SOAmod scheme. These increases are most pronounced over Scandinavia for SOAvbs and for central Europe and Scandinavia for SOAmod. The maximum change happens in the summer period, reaching a maximum of 700% for SOAmod for areas around the British Isles and around 500% in central Europe, while the differences for SOA2p simulations only show a maximum of 70% and 200% increase for annual and summer averages respectively, for the same area. This fact might suggest that the increase of BSOA concentrations due to climate change might be highly underestimated in future scenarios.

Despite the strong regional variations in the concentrations simulated by different schemes (figure 6, column 1), the geographic shape of the differences between historic and future scenarios (figure 6, second column) stay similar for all schemes, showing a maximum in the band between North and Baltic Sea. It is important to keep in mind that some of these differences occur in areas with low concentrations of BSOA, which can lead to large relative changes despite of only small absolute ones. This mostly occurs in the oceanic regions of the domain. When a land-sea mask is used, the maximum changes occur on the British Isles, Scandinavian area and the central Europe.

Figure 6 also shows the spatial distribution of temperature increase is correlated with that of BSOA increases (for all the schemes). There is an exception for the Mediterranean area, where absolute temperatures are high, but the concentration of BSOA is low, mainly because biogenic precursors of BSOA are not emitted in this area.

section{Sensitivity of different schemes to temperature changes}

Figure 7 shows the logarithm of normalized concentrations of BSOA for EUR and MED sub-domains plotted against temperature, for the summer period, using daily average values for each scheme for the five considered summer periods. Dashed lines correspond to linear least-square fits for historic simulations and full lines for future scenarios. BVOC emissions have been added to the plot as well. Normalization of the data has been done by a division by the average of each set of simulations, then the natural logarithm of this ratio is calculated. It is important to bear in mind that as mentioned before, for future scenarios the years with highest temperature and highest BSOA aerosol concentrations are chosen. For the historic scenarios the years with lowest temperature and lowest BSOA aerosol concentrations are chosen, which explains the high difference between historic and future simulations (figure 7). Before entering into the discussion

around sensitivity to temperature changes, it is important to keep in mind that the circulation patterns can change between the historic and future periods. Although averaging of 5 years of simulations likely filters out part of the noise in these patterns, this could also affect BSOA concentrations in addition to temperature changes, especially since the Mediterranean area since its remote with respect to sources.

As seen in figure 7, there is a high correlation between BVOC emissions and temperature throughout all the seasons (shown here for summer), showing an exponential behavior with temperature. The relationship between BVOCs and temperature is reported also for the Mediterranean basin, though the emissions of these species in this area are negligible. Accordingly, the correlation is lower over this area.

When looking at the different schemes, the regression lines show some differences for the future period. Interestingly SOAmod shows a slope rather similar to that of BVOC, while slopes are lower for the SOA2p and SOAvbs. Thus, for SOAmod, the temperature induced increase in BVOC fully affects BSOA. In contrast, for SOA2p and SOAvbs, less BSOA is formed with a temperature increase as could be expected from the correspondence of the temperature with BVOC emissions. This negative sensitivity of BSOA formation normalized by BVOC emissions is due to a shift of SVOC species to the gas phase for increasing temperature, as has been mentioned before. Apparently, this effect is much less pronounced or absent for SOAVBSmod, probably because it includes, contrary to the other two schemes, formation of non-volatile SOA. Indeed, the SOAmod scheme shows 80% of the OA mass in the non-volatile bins, while the SOAvbs and the SOA2p schemes only shows respectively around 10% and 20% in these bins. These results suggest that the parameterization of OA schemes might lead to different sensitivity in prediction of the OA load with respect to the variations in the temperature. The same tendencies are observed for the historic period; however they show a lower intensity because of the lower general temperature ranges.

\section{Conclusions}

In this study, we presented the effect of different OA simulation schemes -on -future aerosol projections due to climate change. For this purpose, three schemes have been used, a molecular single-step oxidation scheme (SOA2p), a standard VBS scheme with anthropogenic SVOC aging only (SOAvbs) and a modified VBS scheme containing functionalization, fragmentation and formation of non-volatile SOA for all SVOC species (SOAVBSmod). These schemes were evaluated for the European region for the year 2013. Although showing differences with observations, each one of OA schemes performs within accepted error ranges. Since VBS schemes are numerically demanding, only 10 years of simulations could be performed for each scheme. In order to maximize the differences between future and historic simulations, the RCP8.5 scenario was used. For the future scenarios, years where the temperature and the BSOA concentration were both at their maximum were chosen, while, for the historic simulations, 5 years with the lowest temperature and BSOA concentrations were selected. Indeed, climate change induced modifications were shown to affect especially the BSOA fraction of organic aerosol. Since BSOA contributes to an important degree to the total concentration of OA, the focus of this article is the evolution of BSOA concentrations in different schemes in future climatic projections.

The results show that the change in concentration indicated by the SOAmod scheme is stronger especially for summertime, showing a difference of +113%, +155% and +262% ~~-122%, -149% and -244%~~ for SOA2p, SOAvbs and SOAmod respectively, for the European area. These changes are mostly due to increased BSOA formation, which is the major SOA fraction during summer.

Previous studies investigated the changes in BSOA concentrations for future scenarios using a two-product scheme for the simulation of SOA. Thus, our suggestion is that the relative variation in SOA concentrations predicted with such schemes might be underestimated.

The reason for the augmentation of BSOA concentrations due to climate change in future scenarios is because of the high dependency to BVOC emissions (which are major precursors of the formation of BSOA in summer/warm periods) to temperature. In a future climate, with the increase of temperatures values, the emissions of BVOCs might increase, and in our easecase, they were predicted to increase by 88% for terpenes and 82% for isoprene (over the European domain). The effect on BSOA formation is tempered by the fact that higher temperatures favor the transition of semi-volatile organic material in the gas phase. This effect is much more pronounced for SOA2p and the SOAvbs schemes than for the SOAmod scheme, which is the only scheme in our study including aging of biogenic SVOCs and the formation of non-volatile SOA. The sensitivity of the SOAVBSmod scheme to temperature is the lowest, and its relation to BVOC emissions the most linear.

The differences were analyzed for the Mediterranean area as well, since organic aerosol and BSOA are transported to this area from continental Europe. While the concentrations in the Mediterranean and changes for future climate are lower for BSOA in general compared to the European area, the changes for this region are stronger in the SOAVBSmod scheme as well (76%, 75% and 127% ~~80%, 79% and 120%~~ for SOA2p, SOAvbs and SOAmod respectively for summer).

In conclusion, -our study suggests that the BSOA concentrations changes are highly sensitive to climate change and the scheme used for their simulation. The changes reported until now for future scenarios ~~could be~~ are highly uncertain, both on absolute and on relative scale. On a relative scale, ~~they~~ these changes might be higher with OA schemes including that include formation of non-volatile SOAaerosol (up to a factor of two).

Future work is necessary in still developing more accurate organic aerosol schemes, not only in terms of absolute concentrations simulated, but also with respect to their temperature sensitivity. The three schemes used in this study can accurately simulate the concentrations of OA each for a specific season and for a specific region, while none of the schemes seem to be able to do so for the whole domain. Therefore, more research is necessary in order to develop OA simulation schemes that are able to represent the concentrations of OA accurately and the temperature sensitivity of this species on a regional scale.

\section{Appendix A}

<u>Country</u>	<u>Station name</u>	<u>Longitude</u>	<u>Latitude</u>	<u>Altitude</u>	<u>Type</u>	<u>Duration</u>	<u>Resoluion</u>
<u>Switzerland</u>	<u>CH0001G</u>	<u>7.99</u>	<u>46.55</u>	<u>3578,0m</u>	<u>PM1</u>	<u>9mo</u>	<u>1h</u>
<u>Switzerland</u>	<u>CH0002R</u>	<u>6.94</u>	<u>46.81</u>	<u>489,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>
<u>Switzerland</u>	<u>CH0005R</u>	<u>8.46</u>	<u>47.07</u>	<u>1031,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>
<u>Switzerland</u>	<u>CH0033R</u>	<u>8.93</u>	<u>46.16</u>	<u>203,0m</u>	<u>PM1</u>	<u>18w</u>	<u>1h</u>
<u>Cyprus</u>	<u>CY0002R</u>	<u>33.06</u>	<u>35.04</u>	<u>520,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>
<u>Czech republic</u>	<u>CZ0003R</u>	<u>15.08</u>	<u>49.57</u>	<u>535,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>
<u>Germany</u>	<u>DE0002R</u>	<u>10.76</u>	<u>52.80</u>	<u>74,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>
<u>Germany</u>	<u>DE0003R</u>	<u>7.91</u>	<u>47.91</u>	<u>1205,0m</u>	<u>PM2.5</u>	<u>1yr</u>	<u>1d</u>

Germany	DE0007R	13,03	53,17	62,0m	PM2.5	1yr	1d
Germany	DE0008R	10,77	50,65	937,0m	PM2.5	1yr	1d
Germany	DE0044R	12,93	51,53	86,0m	PM10	1yr	1d
Spain	ES0001R	-4,35	39,55	917,0m	PM2.5	1yr	1d
Spain	ES0009R	-3,14	41,28	1360,0m	PM2.5	1yr	1d
Spain	--	3,03	39,84	15,0m	PM1	3mo	1h
Spain	ES1778R	2,35	41,77	700,0m	PM1	6mo	1h
Finland	FI0050R	24,28	61,85	181,0m	PM1	9mo	1h
France	FR0009R	4,63	49,90	390,0m	PM2.5	1yr	6d
France	FR0013R	0,18	43,62	200,0m	PM2.5	1yr	6d
France	FR0030R	2,95	45,77	1465,0m	PM2.5	1yr	2d
France	--	9,38	42,97	520,0m	PM1	3mo	1h
France	--	2,15	48,71	156m	PM2.5	1yr	1d
Greece	GR0002R	25,67	35,32	250,0m	PM10	1yr	1d
Ireland	IE0031R	-9,90	53,33	10,0m	PM1	41w	1h
Italy	IT0004R	8,63	45,80	209,0m	PM2.5	1yr	1d
Netherlands	NL0644R	4,92	51,97	1,0m	PM2.5	1yr	1d
Norway	NO0002R	8,25	58,39	219,0m	PM1	1yr	1w
Norway	NO0039R	8,88	62,78	210,0m	PM1	1yr	1w
Norway	NO0056R	11,08	60,37	300,0m	PM2.5	1yr	1d
Poland	PL0005R	22,07	54,15	157,0m	PM10	1yr	1d
Sweden	SE0011R	13,15	56,02	175,0m	PM10	1yr	1d
Sweden	SE0012R	17,38	58,80	20,0m	PM10	1yr	1d
Slovenia	SI0008R	14,87	45,57	520,0m	PM2.5	1yr	1d

\dataavailability{Access to the data used in this article is restricted to registered users of the ChArMEx project.

The data are available on the project website (<http://mistrals.sedoo.fr/ChArMEx/>), last access: 17~June~201) and it should be used following the data and publication policies of the ChArMEx

project;http://mistrals.sedoo.fr/ChArMEx/Data-Policy/ChArMEx_DataPolicy.pdf}. [The data used for the scheme validation part of the study was downloaded from EBAS \(<http://ebas.nilu.no/>\)](#).

\competinginterests{The authors declare that they have no conflict of interest. }

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Table 1

Table 1 Statistic information for different schemes in regards to measurements. In this table are provided the average for the data and each scheme, the bias, standard deviation and RMSE (all in $\mu\text{g m}^{-3}$). R^2 shows the correlation of determination for in scheme. Figure 3 shows the taylor diagram and the boxplots for these comparisons.

		Average	Bias	Standard deviation $\mu\text{g.m}^{-3}$	RMSE	R^2
Annual	Data	2.49		0.36		
	SOA2p	1.84	-0.64	1.40	2.21	0.50
	SOAvbs	2.1	-0.4	1.62	2.44	0.51
	SOAmod	1.58	-0.91	1.05	2.11	0.55
DJF	Data	2.55		1.79		
	SOA2p	0.91	-1.63	0.50	2.47	0.51
	SOAvbs	0.89	-1.67	0.65	2.63	0.51
	SOAmod	1.11	-1.45	0.75	2.55	0.56
MAM	Data	2.11		1.33		
	SOA2p	1.88	-0.23	1.3	1.81	0.56
	SOAvbs	2.23	0.11	1.42	1.98	0.50
	SOAmod	1.4	-0.71	0.84	1.64	0.49
JJA	Data	2.48		1.12		
	SOA2p	2.56	0.1	1.71	2.06	0.51
	SOAvbs	2.9	0.42	1.91	2.31	0.54
	SOAmod	1.83	-0.65	1.17	1.69	0.58
SON	Data	2		1.36		
	SOA2p	1.16	-0.84	0.81	1.81	0.47
	SOAvbs	1.28	-0.72	0.92	1.85	0.47
	SOAmod	1.3	-0.7	0.81	1.76	0.45

Figure 1

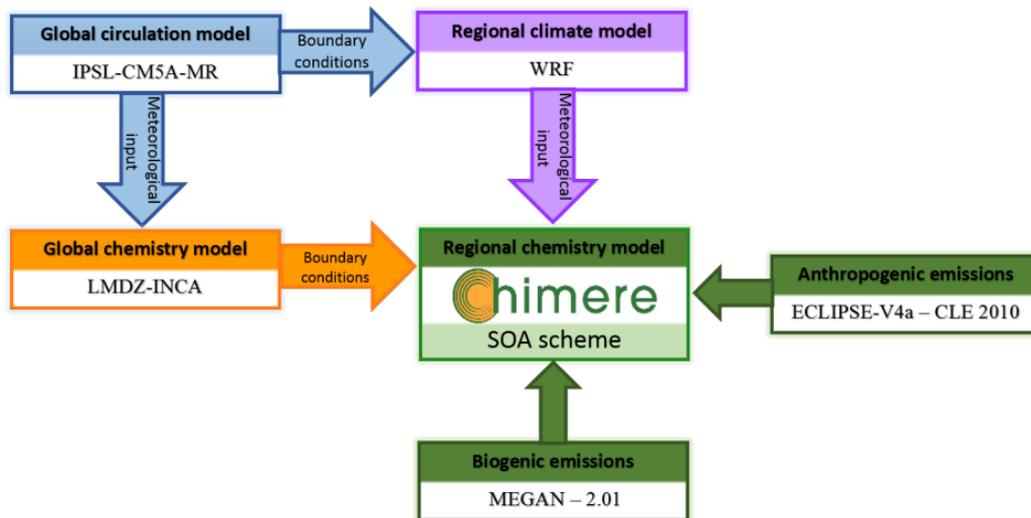


Figure 1. Simulation chain used for this study: the focused of in this work is the SOA scheme inside the regional chemistry transport model.

Figure 2

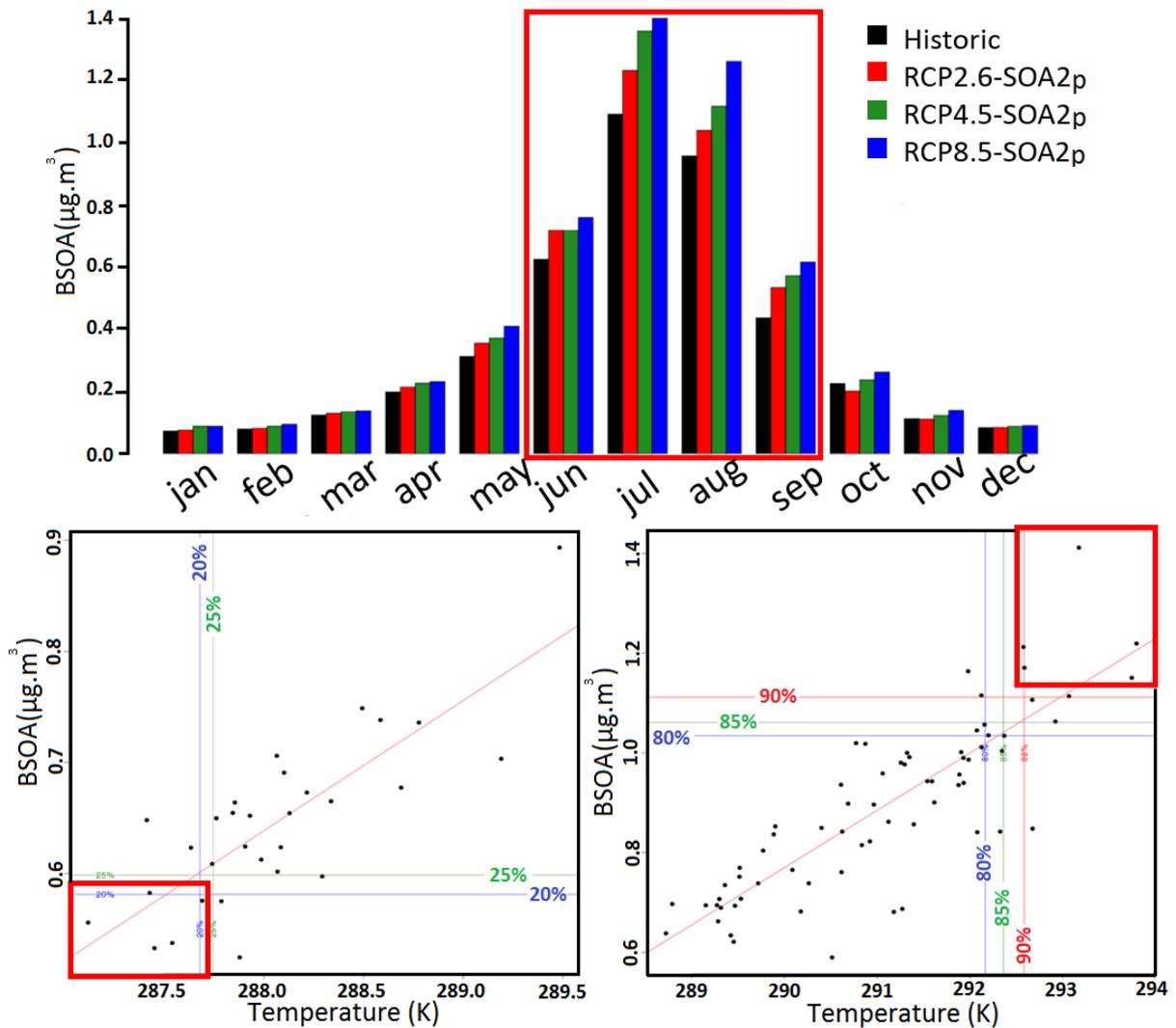


Figure 2. Monthly BSOA concentrations in different RCP scenarios, averaged over 70 years of simulations for future scenarios and 30 years for historic simulations (upper panel). BSOA for the June to September period highlighted in the upper panel was plotted against temperature and years with the lowest temperature and lowest BSOA concentrations for historic simulations are shown in the lower panel on the left side. Those with the highest temperature and BSOA concentrations for future scenarios are shown on the right side.

Figure 3

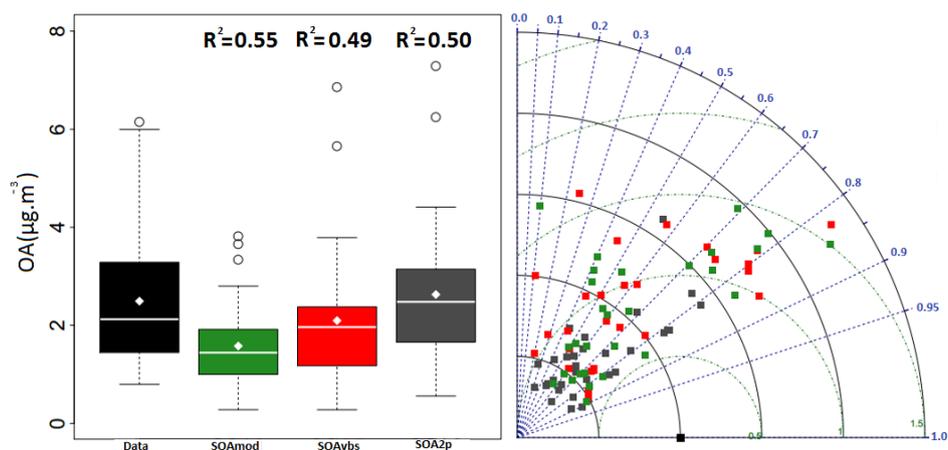


Figure 3. Observation-simulation comparisons for different schemes: The boxplots are presented for the data and the schemes for the annual averages, the correlation of determination for each scheme is shown above each boxplot. The point on the boxplot shows the average value. Each point in the Taylor diagram shows one station while each color shows one scheme (black: SOA2p, red: SOAvbs and green: SOAmod). Table 1 shows statistic information for observed data and the three studied schemes.

Figure 4

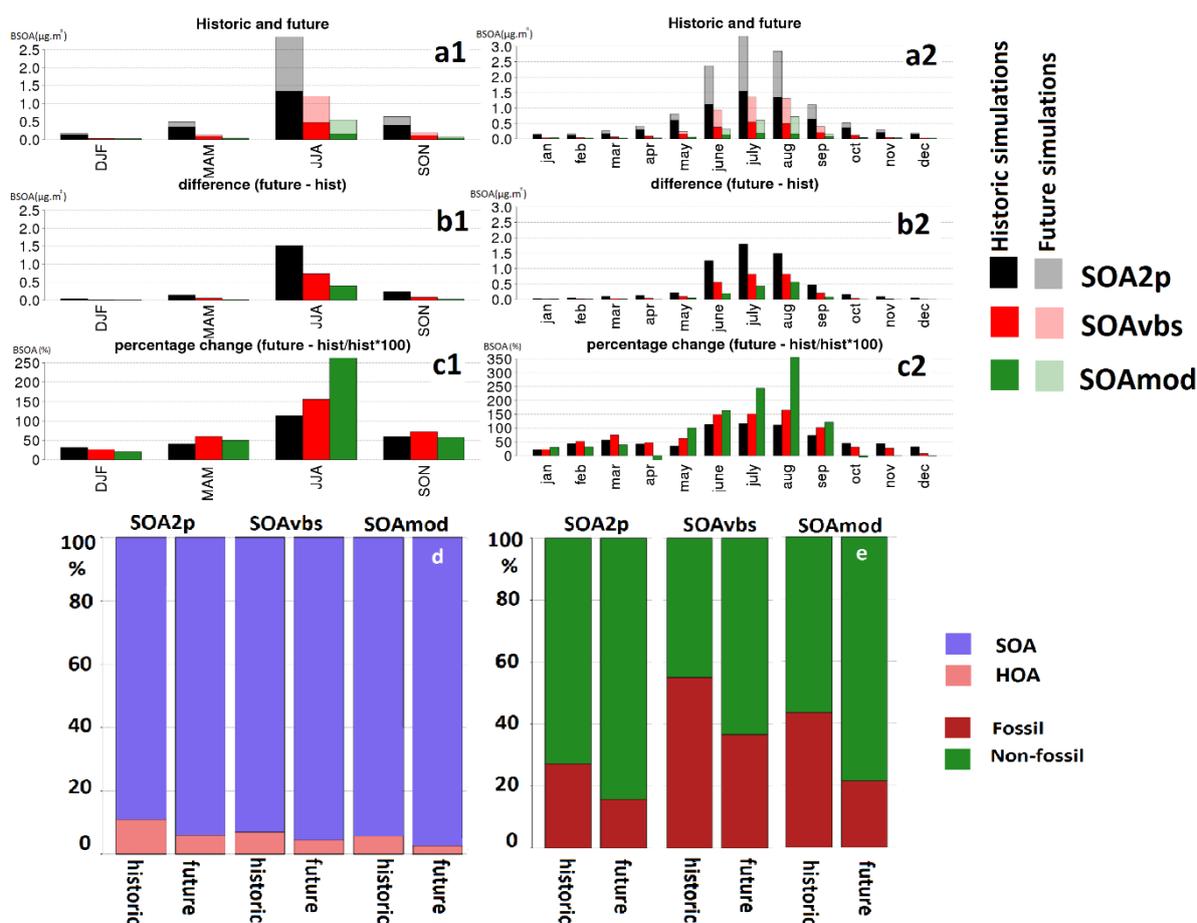


Figure 4. Seasonal and monthly averaged concentrations of BSOA for 5 years of simulations and the oxidation state and origins of OA for the European sub-domain. a1, a2 \rightarrow historic simulations. b1, b2 \rightarrow Absolute changes in future scenarios compared to historic simulations (future – historic), c1, c2 \rightarrow Relative changes in future scenarios. d, e \rightarrow Oxidation state and origins respectively. Lighter colors show future scenarios and darker colors the historic simulations.

Figure 5

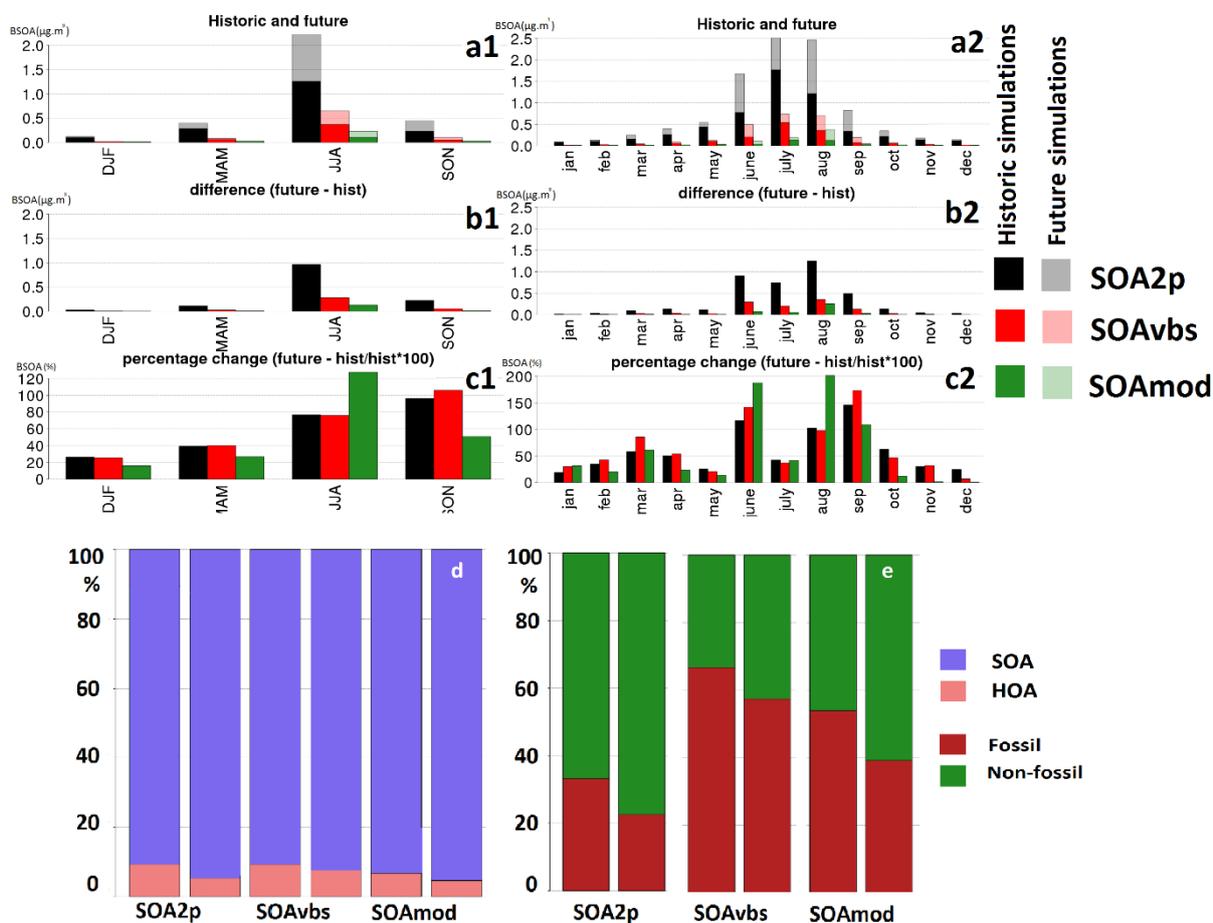


Figure 5. Seasonal and monthly averaged concentrations of BSOA for 5 years of simulations and the oxidation state and origins of OA for the Mediterranean sub-domain. a1, a2 \rightarrow historic simulations. b1, b2 \rightarrow Absolute changes in future scenarios compared to historic simulations (future – historic), c1, c2 \rightarrow Relative changes in future scenarios. d, e \rightarrow Oxidation state and origins respectively. Lighter colors show future scenarios and darker colors the historic simulations.

Figure 6

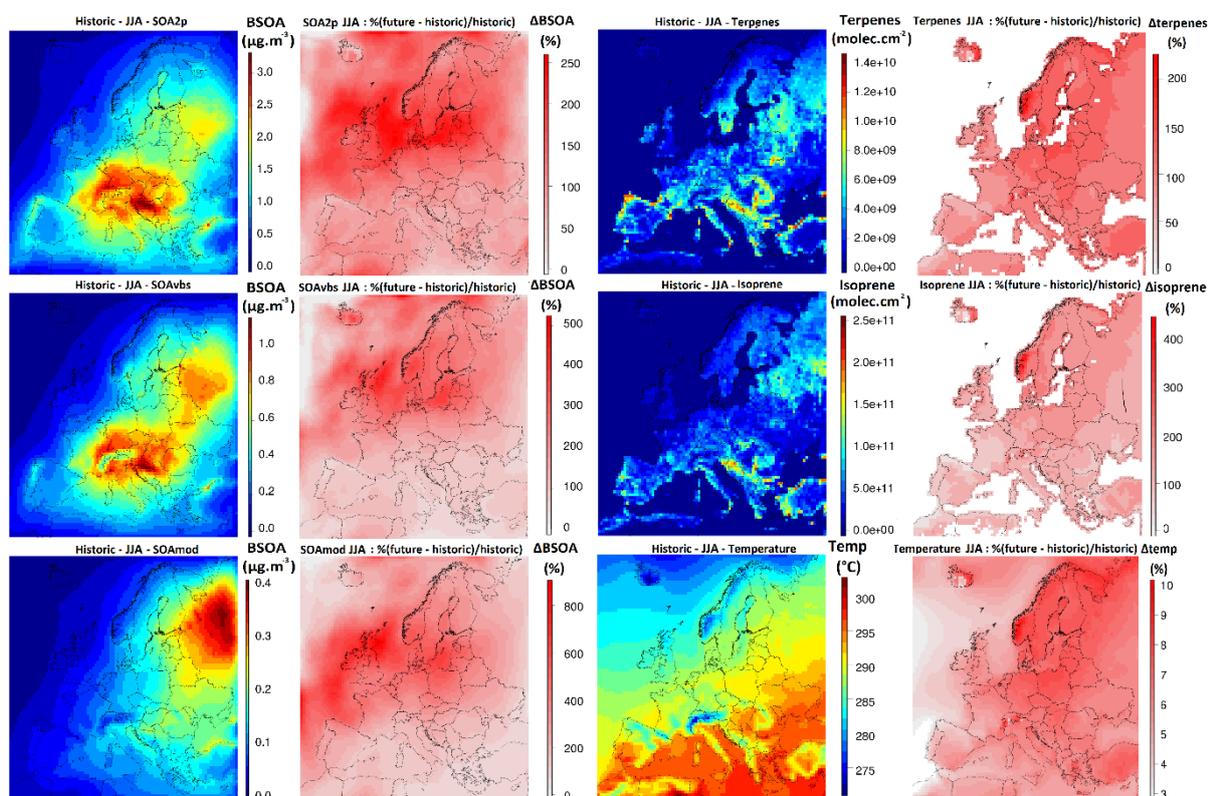


Figure 6. Averaged summer changes in concentrations of BSOA in historic (first column, $\mu\text{g m}^{-3}$) and their future changes (second column, $\%(\text{future} - \text{historic})/\text{historic}$) for all three scenarios (SOA2p, SOAvbs and SOAmod in first, second and third rows respectively). Third column shows the emissions of mono-terpenes and isoprene ($\text{molecules cm}^{-2} \text{ yr}^{-1}$), first and second row) and temperature (K, third row) and the changes of each one of these parameters is seen in fourth column ($\%(\text{future} - \text{historic})/\text{historic}$). Bear in mind that emissions of BVOCs and the temperature do not change between different schemes, scale for each plot is different and all the figures are for the summer period.

Figure 7

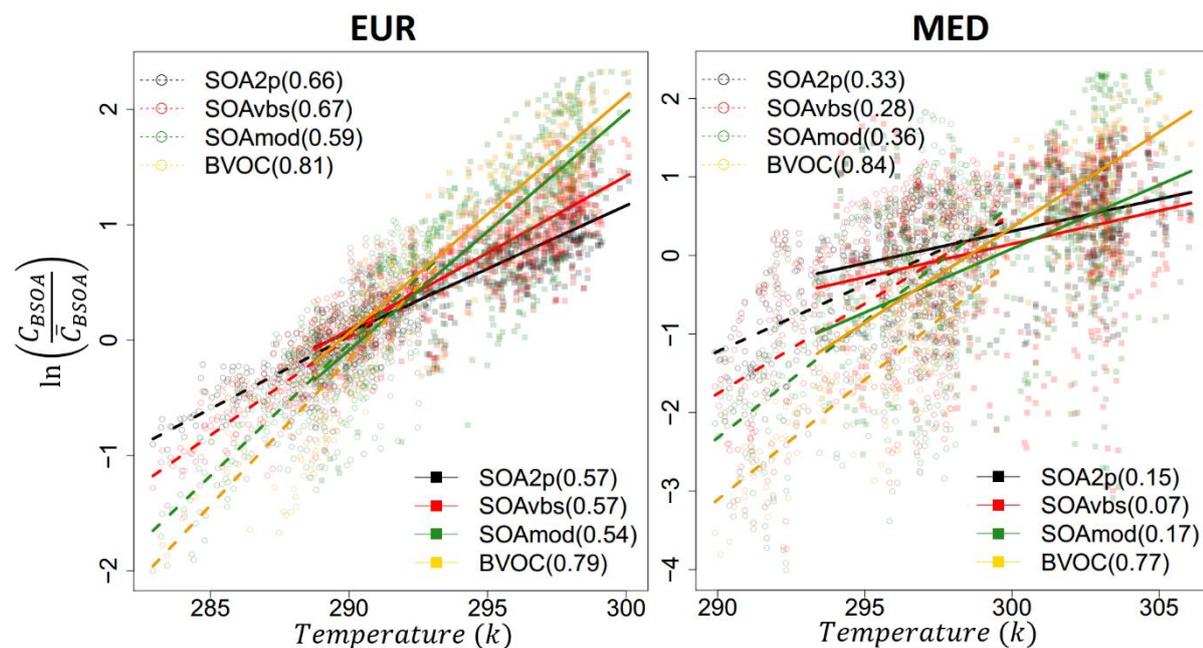


Figure 7. Normalized (divided by the average) concentrations of BSOA versus temperature for the summer period (SOA2p in black, SOAvbs in red and SOAmod in green). Each point represents one day of simulation. Empty circles/dashed lines show the historic period, while filled rectangles/filled lines show the future scenarios. The points concerning BVOCs have been added as well (in gold). The regression lines are exponential. The correlation coefficients for each of the schemes are reported in the legend. Emissions of BVOCs are kept constant between different schemes. A different scale is used for each sub-domain to facilitate the comprehension of the panel.