Sensitivity of organic aerosol simulation scheme on biogenic organic aerosol concentrations in climate projections

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Abstract. 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 aerosols. 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
5   simulate OA are explored. Three schemes are used in this work: a molecular scheme, a standard volatility basis set (VBS) scheme with anthropogenic aging and 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 comparisons show that for the European area, the modified VBS scheme shows
10   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 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
15   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).

20   # 1   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). 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) make them an important subject to study.

Considering that modelling OA already contains important uncertainties, the uncertainties become even more important for

5 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 temperature and land use changes, CO_2 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

10 depending on the factors that were considered in each study. For example, Heald et al., (2009) explored the effects of land use change and CO_2 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. (2018b), found an increase of 52% for isoprene for the period of 2031-2100

15 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 error sources.

It is mainly to assess the future evolution of tropospheric ozone that BVOC emissions have been quantified at global scale in

20 chemistry-climate projections (Arneth et al., 2010). Their importance for organic aerosol chemistry is only emerging in global and regional scale atmospheric models (Maria et al., 2004; Tsigaridis et al., 2007; Heald et al., 2008b). Several different types of OA simulation schemes can be used in chemistry-transport models (CTMs). 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

25 et al., 2007). This scheme presents two major versions: 1-dimensional (1D) and 2-dimentional (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., 2018a), the use of 2D-VBS is less frequent because of its even more challenging numerical needs.

30 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) 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$\mu g.m^{-3}$ for the summer period of 2013), fossil/non-fossil distribution and oxidation level of OA (Cholakian et al., 2018a).

35 Besides, Lannuque et al., (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 review).

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.

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 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 follow: 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.

2 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,
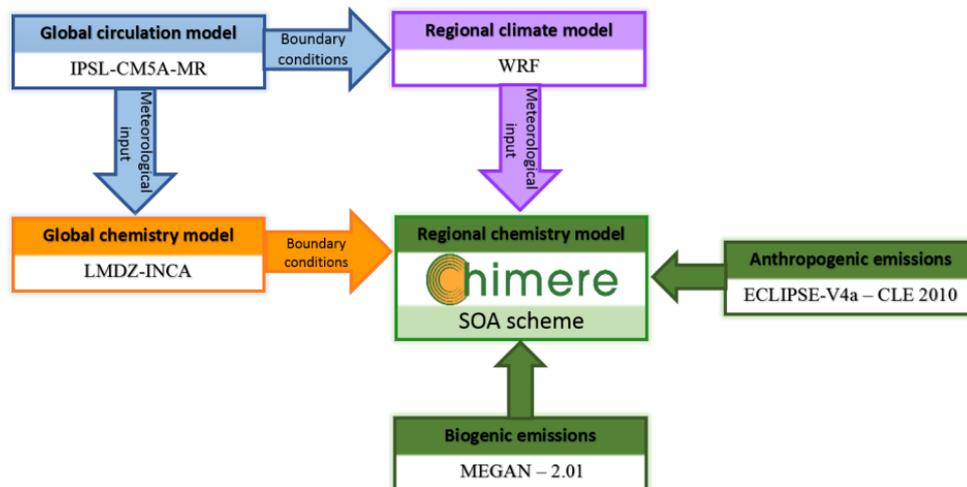
Figure 1. Simulation chain used for this study: the focus of this work is the SOA scheme inside the regional chemistry transport model.

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. Anthropogenic emissions are taken from the ECLIPSEv4a inventory (Amann et al., 2013; Klimont et al., 2013; Klimont et al., 2017), and the biogenic emissions

5 are provided by the MEGAN model (Guenther et al., 2006). 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 this modeling framework is provided in Colette et al., 2013; 2015.

2.1 CHIMERE chemistry transport model

The CHIMERE chemistry transport model has been widely used in different parts of the world (Carvalho et al., 2010; Hodzic
10 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
15 used with a range of $40nm$ to $40\mu m$. 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 latitudes and -40–60 longitudes).

2.2 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 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., (2013), Lemaire et al., (2016) and Cholakian et al. (2019) 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 to $10^6 \mu g.m^{-3}$ 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 to $1000 \mu g.m^{-3}$. 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 (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) 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).

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.
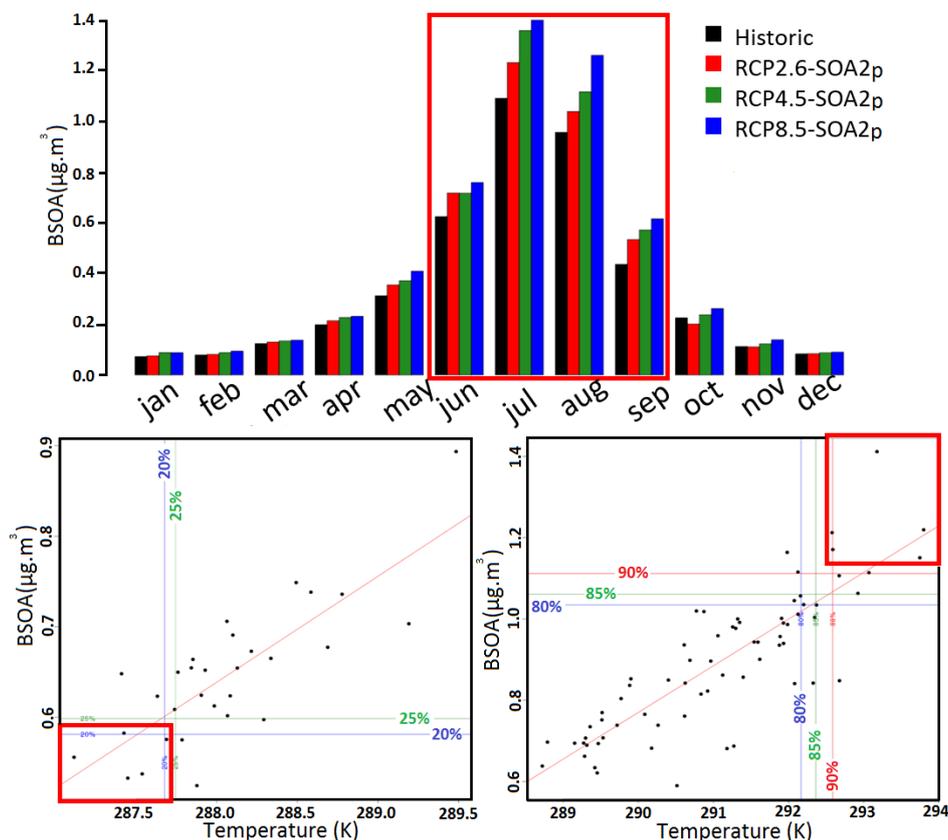
Figure 2. Monthly BSOA concentrations in different RCP scenarios, averaged over 70 years of simulations (upper-panel). BSOA for the June to September period was plotted against temperature (lower panel). Percentiles both for BSOA and temperature are also shown. Years with the lowest temperature and lowest 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.

2.3 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).

5   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

10   and OA component over Europe as also discussed in Cholakian et al. (2018b).

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.

3 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, http://www.ceip.at), meteorological fields are generated using the ECMWF 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 (http://ebas.nilu.no/, last accessed:). 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 (http://mistrals.sedoo.fr/ChArMEx/,last accessed:) 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.

Results of these comparisons are shown in figure 3. Regarding the concentration of OA, the modified VBS scheme shows slightly more bias ($-0.65 \mu g.m^{-3}$ compared to $0.42 \mu g.m^{-3}$ and $0.64 \mu g.m^{-3}$ for SOAvbs and SOA2p respectively) for the summer period. In fact, all compared schemes underestimate the winter period (-1.45, -1.67 and $-0.63 \mu g.m^{-3}$ for SOAmod, SOAvbs and SOA2p respectively). The annual biases for the three schemes are -0.91, -0.4 and $0.14 \mu g.m^{-3}$ for SOAmod, SOAvbs and SOA2p respectively. The correlation between observed and simulated OA concentrations for different schemes are the highest for modified VBS scheme, and lowest for the standard VBS scheme in most seasons. 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. 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. 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.
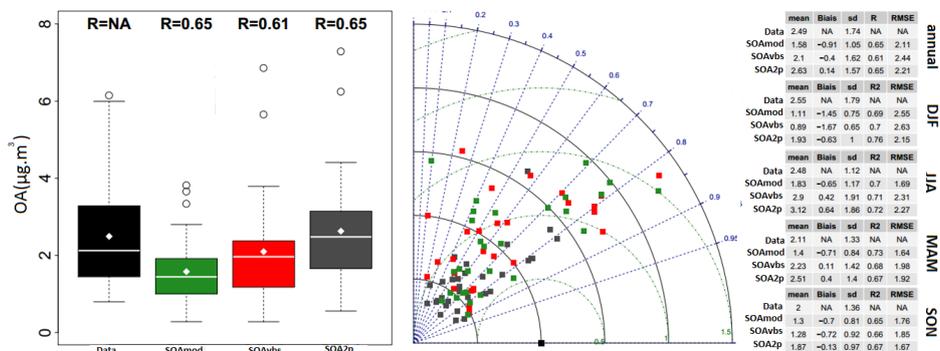
Figure 3. Observation-simulation comparisons for different schemes. Average concentration boxplots and correlations (averaged temporally for all stations) are shown on the left side. On each boxplot, the white diamond and the white line show the average and the median for each series respectively. The upper and lower lines show the 90% and 10% percentile respectively, the circles after the lines showing outlier values. A Taylor diagram is shown in the middle, color code for individual points is the same as on the left side points. The table on the right side shows statistical information for different seasons.

4 Analysis of the simulations

The presentation 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

5 parameters will be performed for the Mediterranean sub-domain including their origins and the oxidation state.

4.1 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

10 of the years was done to maximize future temperature changes, the differences between future and historic simulations are quite remarkable. For the European region, average "historical" isoprene emissions are $1.3 \times 10^{11} molecules.cm^2$ and average historical terpene emissions are $3 \times 10^{10} molecules.cm^2$. 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

15 correlation between historic isoprene and terpene emissions is 0.85 and 0.6 while this correlation is 0.91 and 0.7 for the future simulations.

For the Mediterranean region, 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 sub-domain only contains cells with the sea without any land cells, i.e. with zero biogenic emissions.
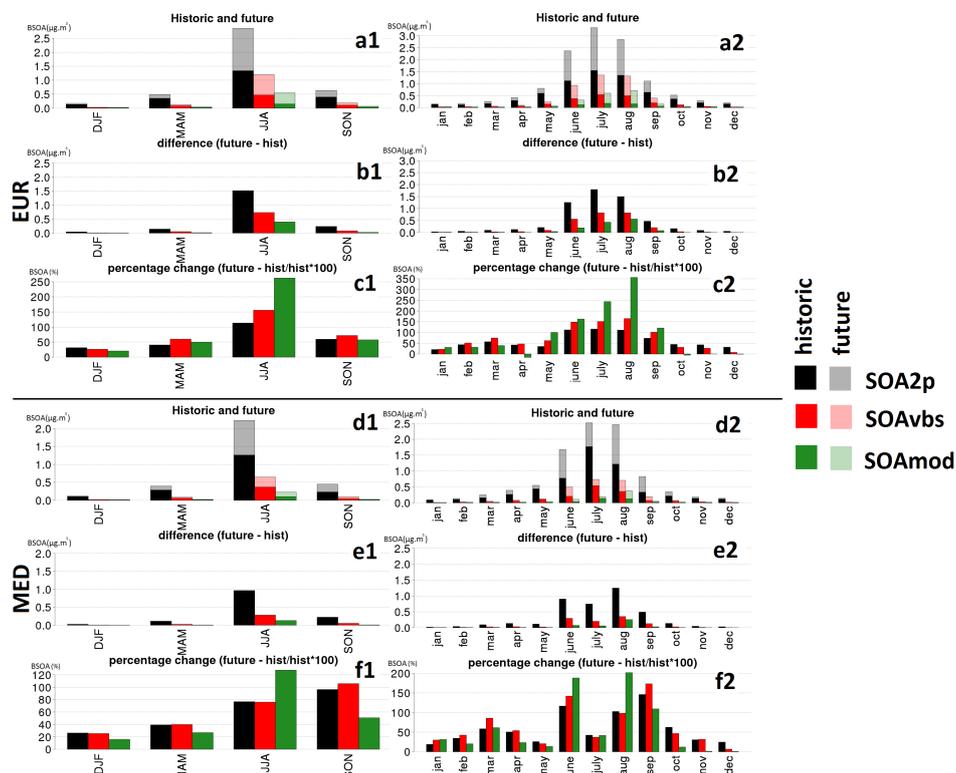
Figure 4. Seasonal (first column) and monthly (second column) averaged concentrations of BSOA for 5 years of simulations. Figures a, b and c are for the European sub-domain and d, e and f for the Mediterranean sub-domain: a1, a2, d1 and d2 → historic simulations. b1, b2, e1 and e2 → Absolute changes in future scenarios compared to historic simulations (future – historic), c1, c2, f1 and f2 → Relative changes in future scenarios.

4.2 European region

4.2.1 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 increase of +94% is calculated, this percentage raises to +135% for SOAvbs and +189% for SOAmod. This change in intensity shows that the climate impact on changes of BSOA in the future might have been underestimated until now on a relative scale, since many of the future simulations performed to see climate impact use two-product or molecular single step schemes for the simulation of SOA, while 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 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.

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.

5   Summer shows the maximum increase (+122%, +149% and +244% for SOA2p, SOAvbs and SOAmod respectively) and winter the lowest one in all schemes (+28%, +28% and +22% for SOA2p, SOAvbs and SOAmod respectively). For autumn and spring SOA2p and SOAmod show similar and intermediate changes while SOAvbs shows higher differences (+56%/+39%, +56%/+45% and +73%/+60% 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 in almost all months for all schemes during

10   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 (+125%, +137% and +216% for SOA2p, SOAvbs and SOAmod respectively). Highest relative changes occur for august for all schemes (+133%, +168% and +333% for SOA2p, SOAvbs and SOAmod respectively). For SOAmod, a decrease is seen for some months in the future scenarios (-11%, -1.6% and -0.45% for April, October and November respectively).

4.2.2 Changes in the origin and volatility state of OA

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. In the model, the fossil/non-fossil repartition is not a direct output. However, since the surrogate species for different sources are present in the outputs, the fossil/non-fossil repartition can be easily calculated. ASOA is in the fossil fraction (neglecting a small fraction due to bio-fuel) and BSOA in the non-fossil fraction. For carbona-

20   ceous 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 in the fossil fraction mainly because it takes into account the aging of anthropogenic SVOCs and not the biogenic SVOCs. On the other hand, 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

25   future (79%, 74% and 84% increase in non-fossil contribution for SOA2p, SOAvbs and SOAmod in future scenarios). SOA2p indicates a higher increase in nonfossil 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 that are kept the same and the biogenic emissions of these species increase with increasing temperature.

30   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 ($C^* \leqslant 0.1 \mu g.m^{-3}$), SVOOA to semi-volatile oxidized OA ($C^* \geqslant 1 \mu g.m^{-3}$) 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 measure-
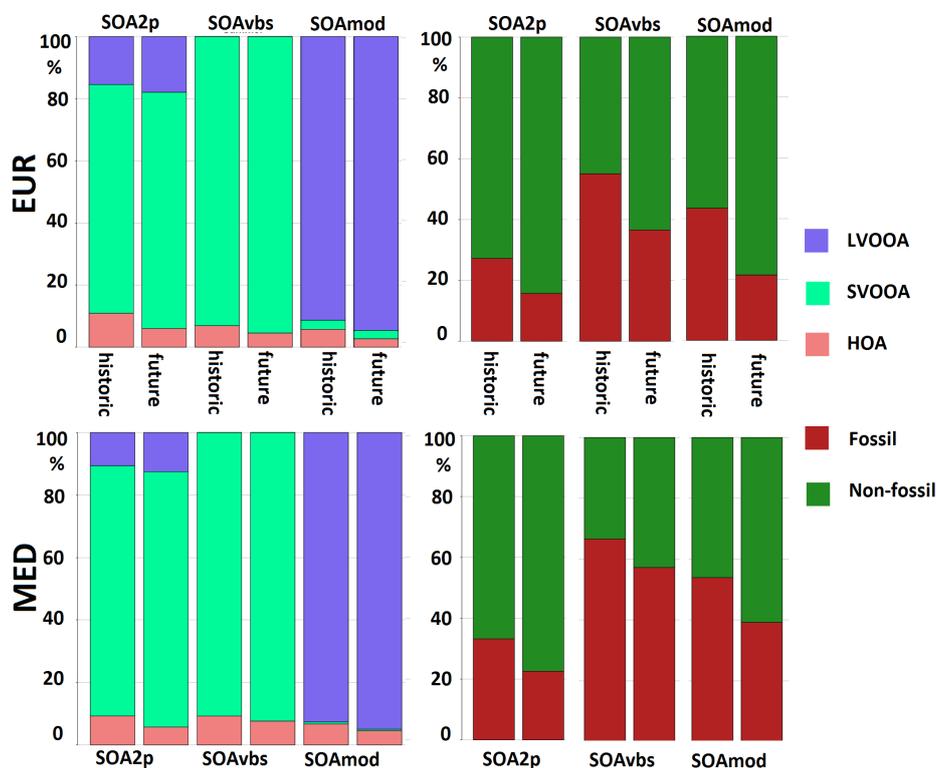
Figure 5. Oxidation state (first column) and origins (second column) of EUR (first row) and MED (second row). Lighter colors show future scenarios and darker colors the historic simulations.

ments during the ChArMEx campaign for the summer of 2013 (Cholakian et al. 2018), 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.
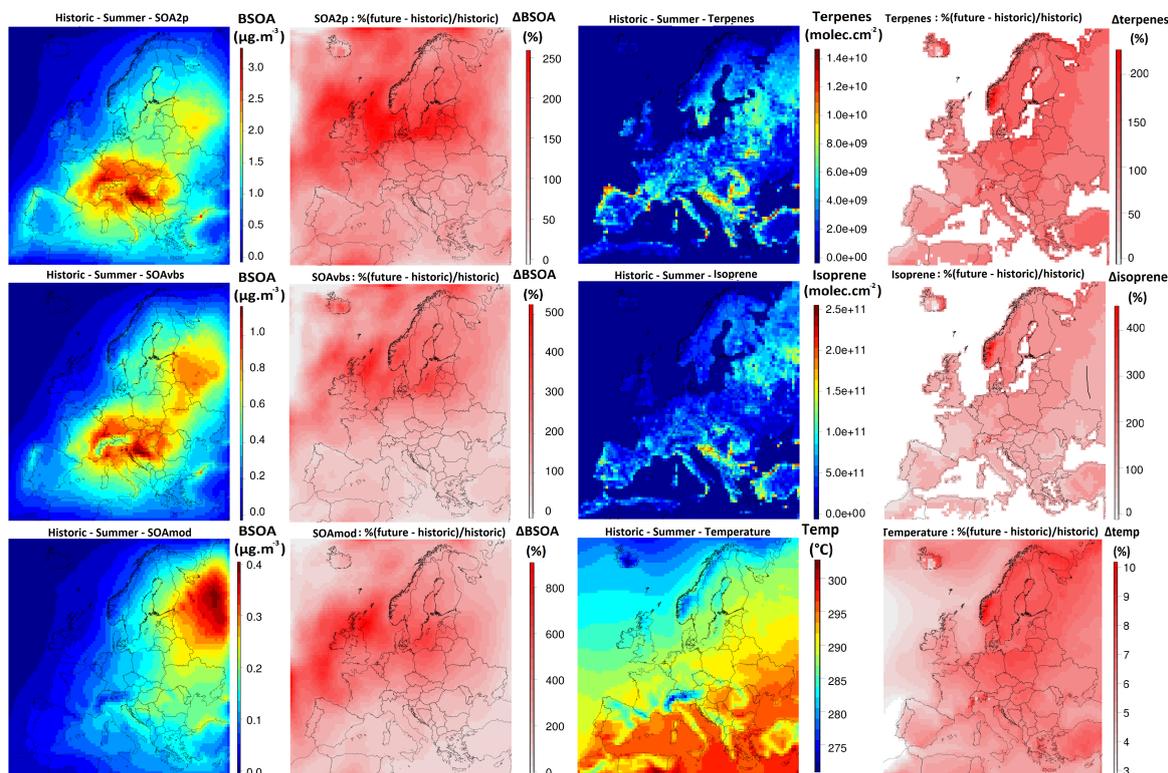
Figure 6. Concentrations of BSOA in historic (first column, $\mu g.m^{-3}$) and their future changes (second column, %(future – historic)/historic) for all three scenarios (SOA2p, SOAvbs and SOAmod in first, second and third rows respectively). Third column shows the emissions of mono-terpenes and isoprene ($molecules.cm^{-2}$, first and second row) and temperature (K, third row) and the changes of each one of these parameters is seen in fourth column (%(future – historic)/historic)). Bear in mind that emissions of BVOCs and the temperature do not change between different schemes. Also, scale for each plot is different.

4.3 Spatial distribution of future changes

Figure 6 shows the concentration of BSOA in different schemes (in $\mu g.m^{-3}$ 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. 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 a maximum of 300% increase 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 maxima 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 these regional variations, differences between historic and future scenarios (figure 6, second column) is similar for all schemes, showing a maximum in the band between North and Baltic Sea.

5   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 temperatures are high, but the concentration of BSOA is low, mainly because biogenic precursors of BSOA are not emitted in this area.

4.4   Mediterranean region

While the differences between the schemes for the European area are important to explore in future scenarios, we also focus
10   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.

4.4.1   Changes in BSOA concentration

15   There are major differences between the partitioning of PM_{10} into 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 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
20   because of lack of emission sources over the marine environment. The differences seen for BSOA concentrations in different schemes is presented in figure 4 (panels 4-d, 4-e and 4-f). The behavior of different schemes in regards to differences between historic and future simulations differs between the Sea and the continental area. For BSOA changes, SOAmod still shows the largest change compared to historic simulations (72%, 73% and 81% for SOA2p, SOAvbs and SOAmod respectively), but the differences between schemes are less pronounced in the Mediterranean area.

25   #### 4.4.2   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). The reason for this change 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 are still the major
30   contributors.
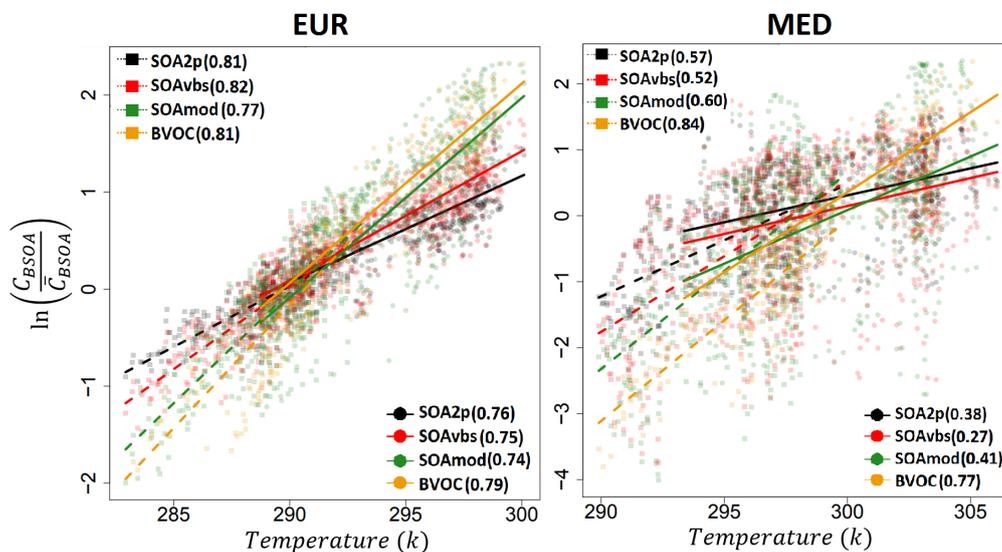
Figure 7. Normalized (divided by the average) concentrations of BSOA versus temperature for the summer period. The points concerning BVOCs have been added as well. Squares show the concentrations regarding the hisoric period and the circles the future period. The regression lines are exponential. The 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 to facilitate the comprehension of the panel.

Both for the historic and the future simulations, the oxidation state does not change considerably in the Mediterranean area as compared to the European area.

5 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. BVOCs 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).

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. 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 VBSmod, probably because it includes, contrary to the other two schemes, formation of non-volatile SOA. 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.

6 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 (VBSmod). 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.

The results show that the change in concentration indicated by the SOAmod scheme is stronger especially for summertime, showing a difference of 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 case 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 VBSmod 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 VBSmod scheme as well (80%, 79% and 120% for SOA2p, SOAvbs and SOAmod respectively for summer).

5   In conclusion, our study suggests that the BSOA concentrations changes reported until now for future scenarios could be highly uncertain, both on absolute and on relative scale. On a relative scale, the changes might be higher with OA schemes including formation of non-volatile aerosol (up to a factor of two). Future work is necessary in developing more accurate organic aerosol schemes, not only in terms of absolute concentrations simulated, but also with respect to their temperature sensitivity.

10   *Author contributions.* ArC, AuC and MB designed the experiment. ArC and AuC performed the simulations, and ArC carried out the post-processing of aforementioned simulations. Article reduction was performed by ArC, and all authors contributed to the text, interpretation of the results and review of the article.

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