Responses to the Comments of Referee 1

(1) This paper describes simulations using the PMCAMx chemical transport model of concentrations of organic aerosol (OA) over Europe for a wintertime period and a summertime period, with the simulated OA concentration fields subdivided according to biomass burning POA, fossil POA, and SOA derived from biomass burning emissions and from other I/S-VOC. The purpose is both to simulate the biomass burning source of OA specifically, but also to be able to apply differential volatility distributions and aging characteristics for the biomass burning source compared with other OA sources. The time periods simulated include instances of very large localised biomass burning emissions, such as wildfires, and winter-time heating emissions in northern Europe.

This is a short and very clearly presented piece of work. Methodological approach and results are clearly described. I do not have any scientific/technical issue with the work. The work adds to the estimations of amount of OA from different origins for the European domain, where that estimation is sometimes derived from 'backward' source-receptor modelling of measurements or, as here, from 'forward' chemical-transport modelling from estimated emissions. Where this paper has some limitation is in 'ground truthing' the model simulations. The authors do provide some comparison summary statistics between their model concentrations and those derived from AMS-PMF measurements at a few sites across Europe but it can be difficult to draw conclusions from such comparisons because model and measurement data do not always represent exactly the same chemical/source entity. The authors conclude there is a potential shortcoming in emissions data for residential heating but do not undertake model sensitivities on changing the emissions.

Overall, however, I am happy to recommend this manuscript for publication as it is. I spotted only very few formatting errors:

We appreciate the positive feedback from the referee. Indeed, the evaluation of the ability of PMCAMx-SR to reproduce the biomass burning OA is necessarily limited, because of the lack of the corresponding necessary measurements. Comparisons of total OA measurements and model predictions are difficult to interpret, because there are so many OA sources. We did our best using the available estimated biomass burning OA concentrations from the analysis of the Aerosol Mass Spectrometer measurements during the period (see for example Figure 8). Improving the OA emission estimates from residential heating in Europe is a major undertaking and it is clearly beyond the scope of the present work. Such an effort is described by Denier van der Gon et al. (ACP, 15, 6503-6519, 2015). However, we clearly need to do better and this requires a good pan-European OA measurement dataset that is currently lacking. We have added some discussion about this important model evaluation issue in the revised paper.

(2) Line 112: insert "and" before "intermediate".
Done.

(3) Line 283: cite to Fig. 4a rather than generically to Fig. 4.
Corrected.

(4) Line 302: after "on March 21" add a citation to Fig. 4b.
Done.

Responses to the Comments of Referee 2

(1) The Theodoritsi and Pandis manuscript reports on the predicted sensitivity of organic aerosol (OA) mass to biomass burning emissions, using a source resolved version of the chemical transport model, PMCAMX (PMCAMX-SR). Studies such as this one are important for understanding the potential air quality and climate effects of anthropogenic and biogenic biomass burning emissions, particularly since the representation of biomass burning-derived SOA is relatively undeveloped in most chemical transport models. The inclusion of biomass-burning derived SOA, particularly when including IVOC, leads to substantial contributions to total predicted OA. This study highlights the need to better constrain biomass burning emissions inventories, including the volatility distribution, and to better understand SOA formation potentials of those emissions. This manuscript is likely to be of interest to the ACP community, and publication is recommended upon addressing the following comments.

We do appreciate the constructive comments and suggestions of the referee. We have done our best to address them and revise the paper accordingly. Our responses (regular font) follow the comments of the referee (in italics) below.

Technical Comments:

(2) It is known that simulating the spatial and temporal distribution of OA, particularly SOA, can be challenging; compensating errors can obscure model performance. In the abstract and in section 7, in addition to the absolute performance statistics, it would be useful to report the change in performance with the expanded treatment of bb-OA (POA+SOA). Weaker performance in winter could be a function of the base simulation (emissions, chemistry, and/or meteorology) and not necessarily a function of the expanded treatment of bbOA.

We agree with the referee, that the performance of a model for total OA depends on a lot of factors (multiple sources, secondary production, removal, meteorology). The PMCAMx-SR performance is a little better for OA than that of the regular PMCAMx. PMCAMx has a similar weak performance in the winter suggesting, as the reviewer states, that this is probably due to other factors and not the expanded treatment of the bbOA. We have added a new section discussing briefly the differences in performance and also the above issues.

(3) Line 58: It is recommended that it be emphasized that bbOA is added as a third category, and is not explicitly considered anthropogenic or biogenic, though bb emissions are characterized in the manuscript as anthropogenic (ag. and heating) or biogenic (wildfire).

We have clarified this point, following the reviewer's suggestion. Indeed, the model does not currently separate the anthropogenic (e.g., residential heating, agricultural burning) from the biogenic sources (wildfires) of bbOA.

(4) Line 111-113: How many model compounds are used to represent IVOCs and SVOCs, respectively? Was the SAPRC mechanism updated as part of this study? If so, the authors should provide further detail in the supplement. If not, a reference should be provided (may be the Environ reference, just needs to be moved).

In this work the IVOCs, SVOCs, and LVOCs are described with 9 volatility bins ($10^{-2} - 10^{6}$ μg m^{-3}). Different lumped compounds are used for the fresh (primary) and secondary organic compounds. The simple reactions of these compounds (one volatility bin change for each reaction with OH) have been added the original SAPRC mechanism. The size distributions of the SVOCs and LVOCs in the particulate phase are described. This information has been added to the revised paper.

(5) Line 139: Was the May et al. volatility distribution applied to all bb emissions? The use of "wood burning" here implies only residential wood burning, but it is assumed that the bb volatility distribution was applied to all three categories of bb emissions. This needs to be modified/clarified.

The May et al. volatility distribution was applied to all three categories of bb emissions. The use of the term "wood burning" in line 139 is now replaced with "biomass burning". We also clarify that the same volatility distribution is assumed for all bbOA sources.

(6) Line 146: How does partitioning within this model framework depend on aerosol composition?

Organic gas-particle partitioning depends on aerosol composition according to gas-particle partitioning absorption theory. The model assumes that the organic compounds form a single pseudo-ideal solution in the particle phase and do not interact with the aqueous phase. This is now clarified in the paper and a reference is provided.

(7) Lines 153-155: The description of the Lane et al. VBS scheme is confusing as written. Given the generally widespread use of the VBS SOA model, it might be clearer to write that SOA is represented using 4 bins, and X number of VOC precursors that are tracked separately as either aSOA-v or bSOA-v. So the number of actual model surrogates seems like it would only be 4*aSOA-v,gas + 4*aSOA-v,p +4*bSOA-v,gas +4*bSOA-v,p, and is not dependent of the number of VOC precursors (as implied by 4 surrogate SOA compounds per VOC).

We have followed the reviewer's suggestion and provided additional information to avoid confusing. Based on the original work of Lane et al. (2008a), SOA from VOCs is represented using four volatility bins (1, 10, 10^{2}, 10^{3} μg m^{-3} at 298 K). As the reviewer suggests the model uses 4 surrogate compounds for SOA from anthropogenic VOCs (aSOA-v) and another 4 for SOA from biogenic VOCs (bSOA-v). These can exist in either the gas or particulate phase so there are two variables from each. There additional surrogate compounds for the oxidation products of anthropogenic IVOCs and SVOCs. PMCAMx-SR includes additional SOA surrogate compounds from biomass burning. We have followed the suggestion of the reviewer and clarified the SOA VBS-scheme in the revised manuscript.

(8) Lines 162-170: The description of chemical aging is also somewhat confusing. It might be clearer to refer to the volatility bin, rather than "vapors" and "semi-volatile SOA". Do the POA and SOA aging reactions both result in an increase in OA mass (line 170)? Is this independent of the mass increase associated with a shift to a semi-volatile bin? Does the OA mass increase apply to the biogenic SOA aging, even though no change in volatility is assumed?

We have rephrased these sentences to clarify that the aging reactions are for the material of each volatility bin that is in the gas phase. We clarify that all these aging reactions (both POA and SOA) are assumed to reduce the volatility of the reacted vapour by one order of magnitude which is linked to an increase in OA mass by approximately 8 percent to account for added oxygen. For the biogenic SOA aging is assumed to lead to no net change of volatility and OA mass. This is also clarified now.

(9) To clarify the volatility distributions and aging, a figure such as 5-2 in the CAMx user's guide would be very helpful.
This is a good idea. We have added a new figure to the paper (Figure 2) depicting the various OA components simulated and their chemical aging reactions.

(10) Section 2.2: It is recommended that the authors consider the publication by Alvarado et al. (2015), which also evaluated volatility distributions of bb emissions. It may be beyond the scope of the manuscript to repeat the model runs using the Alvarado volatility distribution, but it would be useful to consider it in the introduction and discussion, and include it in the Figure 1 panels. The Alvarado et al. study also attempted to account for IVOC emissions not included in two published volatility distributions (including May et al.). Overall, there is significantly more mass (or higher fraction of bb-POA emissions) in the 10^5 and 10^6 bins in the subject manuscript (base case) than in Alvarado et al.
We have followed the recommendation of the reviewer and added some discussion of the Alvarado et al. volatility distribution. We also added the results of the Alvarado study in the discussion of the role of IVOCs from biomass burning in SOA formation.

(11) Also, while scaling the anthropogenic POA EF by 1.5, which gives a sum of fractions >1, has been well described in current literature, it is not clear that the same rationale applies to the biomass burning emissions used in this work. While the IVOC bins are not constrained by data and thus absent in the published VBS distributions, this is not equivalent to missing mass in the bb-POA emissions totals. It seems that some scaling of the May et al. fractions may be needed to include the IVOC bins without giving a sum of fractions >1 (e.g., as done in Alvarado et al.). This probably needs a bit more discussion/clarification in the methods, as the mass attributed to the IVOC bins has a significant effect on predictions of bb-SOA (as demonstrated by the sensitivity case). Reference: Alvarado et al., ACP, 15: 6667-6668, doi:10.5194/acp-15-6667-2015
This is often a confusing point. We have added discussion clarifying it. We do underline the difference with the Alvarado et al. emissions in the corresponding discussion. We also include the absolute emission rates for all bins in Table S1 to make sure that there is no confusion about the used input in our simulation.

Editorial Comments:

(12) In general, it is recommended that the authors check carefully for use of abbreviations. In many instances, an abbreviation is introduced but then not used consistently throughout the manuscript (e.g., organic aerosol (OA) in section 2.1). In a few cases, an abbreviation is introduced but not defined (e.g., AMS line 100).

We have reviewed all the abbreviations used in the manuscript and made the corresponding corrections.

(13) Line 16: Oxidation products of the bbOA? Or of bb emissions? If the latter, sentence needs revision.
We have revised this sentence. These are the oxidation products of biomass burning emissions.

(14) Line 22: Suggest removing “same” before contribution. It is a little confusing as written.
Corrected.

(15) Line 50: What does “their” refer to?
It refers to the organic compounds. We have rephrased this sentence.

(16) Lines 54 and 57: Suggest using “or” rather than “and”, to indicate OA can be primary or secondary and of anthropogenic or biogenic origin.
Done.

(17) Lines 217-232: The discussion about the emissions is a bit unorganized. Are the anthropogenic biomass burning emissions from a source other than GEMS or the Pan-European inventory? If not, recommend to add “including anthropogenic biomass burning emissions” (line 203 or 210). Line 218-219 is then not needed. It is also recommended to move line 217 to the previous paragraph in which the other emissions inventories are described (likely before the introduction of MEGAN).
We have made the corresponding corrections to organize better the discsussion about the emissions used in this study.

(18) Italicize variables in equations.
Done.

Simulation of the chemical evolution of biomass burning organic aerosol

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Abstract

The chemical transport model PMCAMx was extended to investigate the effects of partitioning and photochemical aging of biomass burning emissions on organic aerosol (OA) concentrations. A source-resolved version of the model, PMCAMx-SR, was developed in which biomass burning emissions and their ~~organic aerosol (bbOA) and its~~ ~~the~~ oxidation products ~~of biomass burning emissions~~ are represented separately from the other OA components. The volatility distribution of biomass burning OA (bbOA) and its chemical aging were simulated based on recent laboratory measurements. PMCAMx-SR was applied to Europe during an early summer (1-29 May 2008) and a winter period (25 February-22 March 2009).

During the early summer, the contribution of biomass burning (both primary and secondary species) to total OA levels over continental Europe was estimated to be approximately 16%. During winter the ~~same~~ contribution was nearly 47% due to both extensive residential wood combustion, but also wildfires in Portugal and Spain. The intermediate volatility compounds (IVOCs) with effective saturation concentration values of 10^5 and 10^6 μg m^{-3} are predicted to contribute around one third of the bbOA during the summer and 15% during the winter by forming secondary OA. The uncertain emissions of these compounds and their SOA formation potential require additional attention. Evaluation of PMCAMx-SR predictions against aerosol mass spectrometer measurements in several sites around Europe suggests reasonably good performance for OA (fractional bias less than 35% and fractional error less than 50%).

The performance was weaker during the winter suggesting uncertainties in the residential heating emissions and the simulation of the resulting bbOA in this season.

1 Introduction

Atmospheric aerosols, also known as particulate matter (PM), are suspensions of fine solid or liquid particles in air. These particles range in diameter from a few nanometers to tens of micrometers. Atmospheric particles contain a variety of non-volatile and semi-volatile compounds including water, sulfates, nitrates, ammonium, dust, trace metals, and organic matter. Many studies have linked increased mortality (Dockery et al., 1993), decreased lung function (Gauderman et al., 2000), bronchitis incidents (Dockery et al., 1996), and respiratory diseases (Pope, 1991; Schwartz et al., 1996; Wang et al., 2008) with elevated PM concentrations. The most readily perceived impact of high particulate matter concentrations is visibility reduction in polluted areas (Seinfeld and Pandis, 2006). Aerosols also play an important role in the energy balance of our planet by scattering and absorbing radiation (Schwartz et al., 1996).

Organic aerosol (OA) is a major component of fine PM in most locations around the world. More than 50% of the atmospheric fine aerosol mass is comprised of organic compounds at continental mid-latitudes and as high as 90% in tropical forested areas (Andreae and Crutzen, 1997; Roberts et al., 2001; Kanakidou et al., 2005). ~~Despite their importance of the organic compounds, ~~T~~t~~here are many remaining questions regarding the~~ir~~ identity, chemistry, lifetime, and in general fate of ~~these ~~organic compounds, despite their atmospheric importance. OA originates from many different anthropogenic and biogenic sources and processes and has been traditionally categorized into primary OA (POA) which is directly emitted into the atmosphere as particles ~~and ~~or secondary OA (SOA) that is formed from the condensation of the oxidation products of volatile (VOCs), intermediate volatility (IVOCs), and semivolatile organic compounds (SVOCs). Both POA and SOA are usually characterized as anthropogenic (aPOA, aSOA) ~~and ~~or biogenic (bPOA, bSOA) depending on their sources. Biomass burning OA (bbOA)~~ compounds~~ ~~-~~is~~are~~ treated separately from the other anthropogenic and biogenic OA components in this work. ~~classified separately due to the difficulty in assigning them to either anthropogenic (e.g., residential heating, agricultural burning) or biogenic (wildfires)~~

~~sources. In this work we define biomass burning OA (bbOA) as the sum of bbPOA and bbSOA following the terminology proposed by Murphy et al. (2014).~~

Biomass burning is an important global source of air pollutants that affect atmospheric chemistry, climate, and environmental air quality. In this work, the term biomass burning includes wildfires, prescribed burning in forests and other areas, residential wood combustion for heating and other purposes, and agricultural waste burning. Biomass burning is a major source of particulate matter, nitrogen oxides, carbon monoxide, volatile organic compounds, as well as other hazardous air pollutants. Biomass burning contributes around 75% of global combustion POA (Bond et al., 2004). In Europe, biomass combustion is one of the major sources of OA, especially during winter (Puxbaum et al., 2007; Gelencser et al., 2007).

Chemical transport models (CTMs) have traditionally treated POA emissions as non-reactive and non-volatile. However, dilution sampler measurements have indicated that POA is clearly semi-volatile (Lipsky and Robinson, 2006; Robinson et al., 2007; Huffman et al., 2009a, 2009b). The semi-volatile character of POA emissions can be described by the volatility basis set (VBS) framework (Donahue et al., 2006; Stanier et al., 2008). The VBS is a scheme of simulating OA accounting for changes in gas-particle partitioning due to dilution, temperature changes, and photochemical aging. The third Fire Lab at Missoula Experiment (FLAME-III) investigated a suite of fuels associated with prescribed burning and wildfires (May et al., 2013). The bbOA partitioning parameters derived from that study are used in this work to simulate the dynamic gas-particle partitioning and photochemical aging of bbOA emissions. In this work we define bbOA as the sum of bbPOA and bbSOA following the terminology proposed by Murphy et al. (2014).

A number of modeling efforts have examined the contribution of the semi-volatile bbOA emissions to ambient particulate levels using the VBS framework. For example, Fountoukis et al. (2014) used a three dimensional CTM with an updated wood combustion emission inventory distributing OA emissions using the volatility distribution proposed by Shrivastava et al. (2008). However, this study assumed the same volatility distribution for all OA sources. This volatility distribution is not in general representative of biomass burning emissions since it was derived based on experiments using fossil fuel sources (Shrivastava et al., 2008). Volatility distributions of wood smoke have been measured by Grieshop et al. (2009a) and May et al. (2013) covering the volatility range up to approximately 10^4 μg m^{-3} (at 298 K). Alvarado et al. (2015) in a modelling study stressed the importance of the emissions of the rest of the IVOCs (at 10^5 and 10^6 μg m^{-3}) and attempted to constrain the corresponding chemistry using observations from a biomass burning plume from a prescribed fire in California

The main objective of this study is to develop and test a CTM treating biomass burning organic aerosol (bbOA) emissions separately from all the other anthropogenic and biogenic emissions. This extended model should allow at least in principle more accurate simulation of OA and direct predictions of the role of bbOA in regional air quality. The rest of the manuscript is organized as follows. First, a brief description of the new version of PMCAMx (PMCAMx-SR) is provided. The source-resolved version of PMCAMx (PMCAMx-SR) treats bbOA emissions and their chemical reactions separately from those of other OA sources. The details of the application of PMCAMx-SR in the European domain for a summer and a winter period are presented. In the next section, the predictions of PMCAMx-SR are evaluated using aerosol mass spectrometer (AMS) measurements collected in Europe. Finally, the sensitivity of the model to different parameters is quantified.

2 PMCAMx-SR description

PMCAMx-SR is a source-resolved version of PMCAMx (Murphy and Pandis, 2009; Tsimpidi et al., 2010; Karydis et al., 2010), a three-dimensional chemical transport model that uses the framework of CAMx (Environ, 2003) and simulates the processes of horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, gas, aqueous and aerosol-phase chemistry. The chemical mechanism employed to describe the gas-phase chemistry is based on the SAPRC mechanism (Carter, 2000; Environ, 2003). The version of SAPRC currently used includes 211 reactions of 56 gases and 18 radicals. The SAPRC mechanism has been updated to include gas-phase oxidation of semivolatile organic compounds (SVOCs), and intermediate volatility organic compounds (IVOCs). In this work the IVOCs~~,~~ and SVOCs are described with 9 volatility bins ($10^{-2} - 10^6$ μg m^{-3} at 298 K). Different surrogate species~~lumped~~ ~~compounds~~ are used to represent~~for~~ the corresponding fresh (primary) and the secondary organic compounds. –The chemical ~~simple~~ reactions

reactions of these compounds parameterized as ~~(~~one volatility bin reduction~~change~~ during~~for~~ each reaction with OH~~)~~ have been added to the original SAPRC mechanism. Three detailed aerosol modules are used to simulate aerosol processes: inorganic aerosol growth (Gaydos et al., 2003; Koo et al., 2003), aqueous phase chemistry (Fahey and Pandis, 2001), and secondary organic aerosol (SOA) formation and growth (Koo et al., 2003). The above modules use a sectional approach to dynamically track the size evolution of each aerosol component across 10 size sections spanning the diameter range from 40 nm to 40 μm.

2.1 Organic aerosol modelling

PMCAMx-SR simulates ~~organic aerosol~~ OA based on the volatility basis set (VBS) framework (Donahue et al., 2006; Stanier et al., 2008). VBS is a unified scheme of treating ~~organic aerosol~~ OA, simulating the volatility, gas-particle partitioning, and photochemical aging of organic pollutant emissions. PMCAMx-SR incorporates separate VBS variables and parameters for the various OA components based on their source.

2.1.1 Volatility of primary emissions

PMCAMx-SR assumes that all primary emissions are semi-volatile. According to the VBS scheme, species with similar volatility are lumped into bins expressed in terms of effective saturation concentration values, C^*, separated by factors of 10 at 298 K. POA emissions are distributed across a nine-bin VBS with C^* values ranging from 10^{-2} to 10^6 μg m^{-3} at 298 K. SVOCs and IVOCs are distributed among the 1, 10, 100 μg m^{-3} C^* bins and 10^3, 10^4, 10^5, 10^6 μg m^{-3} C^* bins respectively. Table 1 lists the generic POA volatility distribution proposed by Shrivastava et al. (2008) assuming that the IVOC emissions are approximately equal to 1.5 times the ~~primary organic aerosol~~ POA emissions (Robinson et al., 2007; Tsimpidi et al., 2010; Shrivastava et al., 2008). This volatility distribution is used in PMCAMx-SR for all sources with the exception of biomass~~wood~~ burning. In the original PMCAMx this volatility distribution is also used for biomass~~wood~~ burning emissions.

The partitioning calculations of primary emissions are performed using the same module used to calculate the partitioning of all semi-volatile organic species (Koo et al., 2003). This is based on absorptive partitioning theory and assumes that the bulk gas and particle phases are in equilibrium and that all condensable organics form a pseudo-ideal solution (Odum et al., 1996; Strader et al., 1999). Organic gas-particle partitioning is assumed to depend on temperature and aerosol composition. ~~Organic gas-particle partitioning depends on aerosol composition according to gas-particle partitioning absorption theory.~~ The portioning model assumes that the organic compounds form a single pseudo-ideal solution in the particle phase and do not interact with the aqueous phase (Strader et al., 1999). The Clausius-Clapeyron equation is used to describe the effects of temperature on C^* and partitioning. Table 1 also lists the enthalpies of vaporization currently used in PMCAMx and PMCAMx-SR. All POA species are assumed to have an average molecular weight of 250 g mol^{-1}.

2.1.2 Secondary organic aerosol from VOCs

Based on the original work of Lane et al. (2008a), SOA from VOCs (SOA-v) is represented using four volatility bins (1, 10, 10^2, 10^3 μg m^{-3} at 298 K). The model uses 4 surrogate compounds for SOA from anthropogenic VOCs (aSOA-v) and another 4 for SOA from biogenic VOCs (bSOA-v). These can exist in either the gas or particulate phase so there are two model variables ~~from~~for each volatility bin. A~~There are a~~dditional surrogate compounds and thus model variables are used to keep track of~~for~~ the oxidation products of anthropogenic IVOCs (SOA-iv) and SVOCs (SOA-sv). PMCAMx-SR includes additional SOA surrogate compounds to simulate the oxidation productions of the biomass~~from biomass~~ burning emissions.~~Following Lane et al. (2008a), the SOA VBS-scheme uses four surrogate SOA compounds for each VOC precursor with 4 volatility bins (1, 10, 100, 1000 μg m~~$^{-3}$~~) at 298 K. Anthropogenic (aSOA-v) and biogenic (bSOA-v) components are simulated separately.~~ aSOA components are assumed to have an average molecular weight of 150 g mol^{-1}, while bSOA species 180 g mol^{-1}. Laboratory results from the smog-chamber experiments of Ng et al. (2006) and Hildebrandt et al. (2009) are used for the anthropogenic aerosol yields.

2.1.3 Chemical aging mechanism

All OA components are treated as chemically reactive in PMCAMx-SR. Anthropogenic SOA components resulting from the oxidation of SVOCs and IVOCs (aSOA-sv, aSOA-iv) ~~S-SOA, I-SOA and V-SOAVapors resulting from the~~

~~evaporation of POA~~ are assumed to react with OH radicals in the gas phase with a rate constant of $k = 4 \times 10^{-11}$ cm^3 molec^{-1} s^{-1} resulting in the formation of ~~oxidized OA~~lower volatility aSOA. ~~These reactions are assumed to lead to an effective reduction of volatility by one order of magnitude.~~ Semi-volatile anthropogenic aSOA-v components are ~~also~~ assumed to react with OH in the gas phase with a rate constant of $k = 1 \times 10^{-11}$ cm^3 molec^{-1} s^{-1} ~~for anthropogenic SOA~~ (Atkinson and Arey, 2003). All these aging reactions ~~(both POA and SOA)~~ are assumed to reduce the volatility of the reacted vapo~~u~~r by one order of magnitude, which is linked to an increase in OA mass by approximately 7.5% to account for added oxygen. Biogenic SOA (bSOA-v) aging is assumed to lead to zero net change of volatility and OA mass (Lane et al., 2008b). ~~Each reaction is assumed to increase the OA mass by 7.5% to account for added oxygen.~~

2.2 PMCAMx-SR enhancements

In PMCAMx-SR, the fresh biomass burning organic aerosol (bbOA) and its secondary oxidation products (bbSOA) are simulated separately from the other POA components. The May et al. (2013) volatility distribution is used to simulate the gas-particle partitioning of fresh bbOA. This distribution includes surrogate compounds up to a volatility of 10^4 μg m^{-3}. This means that the more volatile IVOCs, which could contribute to SOA formation, are not included. To close this gap, the values of the volatility distribution of Robinson et al. (2007) are used for the 10^5 and 10^6 μg m^{-3} bins (Table 1). The sensitivity of PMCAMx-SR to the IVOC emissions added to the May et al. (2013) distribution will be explored in a subsequent section. The effective saturation concentrations and the enthalpies of vaporization used for bbOA in PMCAMx-SR are also listed in Table 1. The new bbOA scheme requires the introduction of 36 new organic species to simulate both phases of fresh primary and oxidized bbOA components. The rate constant used for the chemical aging reactions is the same as the one currently used for all POA components and has a value of $k = 4 \times 10^{-11}$ cm^3 molec^{-1} s^{-1}. The volatility distributions of bbOA in PMCAMx and PMCAMx-SR are shown in Fig. 1a. The volatility distribution implemented in PMCAMx-SR results in less volatile bbOA for ambient OA levels (a few μg m^{-3}) (Fig. 1b). ~~Figure 2 is a~~A schematic representation of the ~~proposed~~ organic aerosol ~~framework~~module ~~implemented in~~of ~~PMCAMx and~~ PMCAMx-SR is shown in

Figure 2. ~~depicting the various OA components simulated and their chemical aging reactions.~~

3 Model application

PMCAMx-SR was applied to a 5400×5832 km^2 region covering Europe with 36×36 km grid resolution and 14 vertical layers extending up to 6 km. The model was set to perform simulations on a rotated polar stereographic map projection. The necessary inputs to the model include horizontal wind components, temperature, pressure, water vapor, vertical diffusivity, clouds, and rainfall. All meteorological inputs were created using the meteorological model WRF (Weather Research and Forecasting) (Skamarock et al., 2005). The simulations were performed during a summer (1-29 May 2008) and a winter period (25 February-22 March 2009). In order to limit the effect of the initial conditions on the results, the first two days of each simulation were excluded from the analysis.

Anthropogenic and biogenic emissions in the form of hourly gridded fields were developed both for gases and primary particulate matter. Anthropogenic gas emissions include land emissions from the GEMS dataset (Visschedijk et al., 2007) and also emissions from international shipping activities. Anthropogenic particulate matter mass emissions of organic and elemental carbon are based on the Pan-European Carbonaceous Aerosol Inventory (Denier van der Gon et al., 2010) that has been developed as part of the EUCAARI project activities (Kulmala et al., 2009). All relevant significant emission sources, including anthropogenic biomass burning emissions from agricultural activies and residential heating, are included in the two inventories. Day-specific wildfire emissions were also included (Sofiev et al., 2008a; 2008b). Emissions from ecosystems were calculated offline by MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006). The marine aerosol emission model developed by O'Dowd et al. (2008) has been used to estimate mass fluxes for both accumulation and coarse mode including the organic aerosol fraction. Wind speed data from WRF and chlorophyll-a concentrations are the inputs needed for the marine aerosol emissions module.

~~Day-specific wildfire emissions were also included (Sofiev et al., 2008a; 2008b). Anthropogenic sources of wood combustion include residential heating and agricultural activities.~~ The gridded emission inventories of bbOA species for the two modeled periods are shown in Fig. ~~2~~3. During the early summer simulated period wildfires were responsible for 60% of the bbOA emissions, agricultural waste burning for 15% and residential wood combustion for 25% (Table 2). Details about the OA emission rates from agricultural activities are provided in the Supplementary Information (Fig. S1). During winter residential combustion is the dominant source (63%). The wintertime wildfire emissions in the inventory, approximately 3,000 tn d^{-1}, are quite high especially when compared with the corresponding summer value which is 1,700 tn d^{-1}. The spatial distribution of OA emission rates from wildfires during 25 February-22 March 2009 is provided in the Supplementary Information (Fig. S2). Analysis of fire counts in satellite observations used for the development of the inventory suggests that some agricultural emissions have probably been attributed to wildfires. All bbOA sources are treated the same way in PMCAMx-SR so this potential misattribution does not affect our results.

4 PMCAMx-SR testing

To test our implementation of the source-resolved VBS in PMCAMx-SR we compared its results with those of PMCAMx using the same VBS parameters. For this test we used in PMCAMx-SR the default PMCAMx bbOA partitioning parameters shown in Table 1 as proposed by Shrivastava et al. (2008). In this way both models should simulate the bbOA in exactly the same way, but PMCAMx-SR describes it independently while PMCAMx lumps it with other primary OA. The differences between the corresponding OA concentrations predicted by the two models were on average less than 10^{-3} $\mu g\ m^{-3}$ (0.03%). The maximum difference was approximately 0.03 $\mu g\ m^{-3}$ (0.6%) in western Germany. This suggests that our changes to the code of PMCAMx to develop PMCAMx-SR did not introduce any inconsistencies with the original model. The small differences are due to numerical issues in the advection/dispersion calculations.

5 Contribution of bbOA to PM over Europe

In this section the predictions of PMCAMx-SR for the base case simulations during 1 - 29 May 2008 and 25 February - 22 March 2009 are analysed. Figure ~~3~~ 4 shows the PMCAMx-SR predicted average ground-level $PM_{2.5}$ concentrations for the various OA components for the two simulated periods.

The POA from non-bbOA sources will be called fossil POA (fPOA) in the rest of the paper. fPOA levels over Europe were on average around 0.1 μg m^{-3} during both periods (Figs. ~~3a~~ 4a and ~~3b~~4b). However, their spatial distributions are quite different. During May, predicted fPOA concentrations are as high as 2 μg m^{-3} in polluted areas in central and northern Europe but are less than 0.5 μg m^{-3} in the rest of the domain. These low levels are due to the evaporation of POA in this warm period. For the winter period peak fPOA levels are higher reaching values of around 3.5 μg m^{-3} in Paris and Moscow. fPOA contributes approximately 3.5% and 6% to total OA in Europe during May 2008 and February-March 2009 respectively. bbPOA concentrations have peak average values 7 μg m^{-3} in St. Petersburg in Russia and 10 μg m^{-3} in Porto in Portugal during summer and winter respectively (Figures ~~3c~~ 4c and ~~3d~~4d). During the summer bbPOA is predicted to contribute 5% to total OA, and its contribution during winter increases to 32%. The average predicted bbOA concentrations over Europe are 0.1 μg m^{-3} and 0.8 μg m^{-3} during the summer and the winter period respectively.

The SOA resulting from the oxidation of IVOCs (SOA-iv) and evaporated POA (SOA-sv) has concentrations as high as 1 μg m^{-3} in central Europe and the average levels are around 0.3 μg m^{-3} (13% contribution to total OA) during summer (Fig. ~~3e~~4e). During winter the peak concentration value was a little less than 0.5 μg m^{-3} in Moscow in Russia and the average levels were approximately 0.1 μg m^{-3} (5.5% contribution to total OA) (Fig ~~3f~~4f). The highest average concentration of bbSOA-sv and bbSOA-iv (biomass burning SOA from intermediate volatility and semi-volatile precursors) was approximately 1 μg m^{-3} in Lecce in Italy during summer and 3.5 μg m^{-3} in Porto during winter. During May bbSOA is predicted to contribute 11% to total OA over Europe and during February-March 2009 its predicted contribution is 15%. The average bbSOA is 0.3 μg m^{-3} during summer and approximately 0.4 μg m^{-3} during winter (Figs. ~~3g~~ 4g and ~~3h~~4h). During the summer, the remaining 67% of total OA is biogenic SOA (52%) and anthropogenic SOA (15%), and in winter of the remaining 41% of total OA, 36% is biogenic and 5% is anthropogenic SOA (not shown).

In areas like St. Petersburg in Russia predicted hourly bbOA levels exceeded 300 μg m^{-3} due to the nearby fires affecting the site on May 3-5 (Fig. ~~4~~5a). For these extremely high concentrations most of the bbOA (90% for St. Petersburg) was primary with the bbSOA contributing around 10%. The spatiotemporal evolution of bbPOA and bbSOA during May 1–6 in Scandinavia and northwest Russia is depicted in Figure ~~5~~6. A series of fires started in Russia on May 1, becoming more intense during the next days until May 6 when they were mostly extinguished. bbSOA, as expected, follows the opposite evolution with low concentration values in the beginning of the fire events (May 1) and higher values later on. The bbSOA production increases the range of influence of the fires.

In Majden (FYROM) fires contributed up to 25 μg m^{-3} of bbOA on May 25-26. The bbSOA was 15% of the bbOA in this case (Fig. S3). Fires also occurred in south Italy (Catania) and contributed up to 52 μg m^{-3} of OA on May 15-17. During this period the bbSOA was 13% of the bbOA (Fig. S3). Paris (France) and Dusseldorf (Germany) were further away from major fires but were also affected by fire emissions during most of the month (Fig. S3). The maximum hourly bbOA levels in these cities were around 5 μg m^{-3}, but bbSOA in this case represents according to the model around 35% of the total bbOA in Paris and 55% in Dusseldorf.

During the winter simulation period, there were major fires during March 20-22 in Portugal and northwestern Spain. The maximum predicted hourly bbOA concentration in Porto (Portugal) exceeded 700 μg m^{-3} on March 21 (Fig. 5b). During the same 3 days in March the average levels of bbPOA in Portugal and Spain were 9 μg m^{-3} and their contribution to total OA was 62%. bbPOA was 80% of the total bbOA during March 20-22 in the Iberian Peninsula.

6 Role of the more volatile IVOCs

We performed an additional sensitivity simulation where we assumed that there are no emissions of more volatile IVOCs (those in the 10^5 and 10^6 μg m^{-3} bins). The partitioning parameters used in this sensitivity test are shown in Table 1. The emissions rates for each volatility bin during the two modeled periods are provided in the Supplementary Information (Table S1). The absolute emissions assigned to the lower volatility bins are approximately the same for both simulations. More specifically, during May 2008, the emission rates of LVOCs (10^{-2}, 10^{-1} μg m^{-3} C^* bins) and SVOCs (10^0, 10^1, 10^2 μg m^{-3} C^* bins) are 530 and 1050 tn d^{-1} respectively for the base-case run and 580 and 1160 tn d^{-1} respectively for the sensitivity run. During February-March 2009, the emission rates of LVOCs and SVOCs are 2100 and 4100 tn d^{-1} respectively for the base-case run and 2300 and 4500 tn d^{-1} respectively for the sensitivity run. The base case simulation assumes higher emissions in the upper volatility bins of the IVOCs (10^3, 10^4, 10^5, 10^6 μg m^{-3} C^* bins) which can be converted to bbSOA. During summer, the emission rate of IVOCs is 4460 tn d^{-1} in the base-case run and 1160 tn d^{-1} in the sensitivity test. During winter, the emission rate of IVOCs is 17400 tn d^{-1} in the base case and 4500 tn d^{-1} in the sensitivity test.

The base case and the sensitivity simulations predict practically the same bbPOA concentrations in both periods (Fig. ~~6~~7) as expected based on the emission inventory. During summer, the average absolute change of bbPOA in Europe is around 10% (corresponding to 0.01 μg m^{-3}) (Fig. ~~6a~~7a). The average difference in bbSOA is significantly higher and around 60% (0.2 μg m^{-3} on average) due to the higher IVOC emissions of the base case simulation. The atmospheric conditions during this warm summer period (high temperature, UV radiation, relative humidity) lead to high OH concentrations and rapid production of bbSOA.

During winter, the average absolute change for both bbPOA and bbSOA in Europe is approximately 0.1 μg m^{-3} (Fig. ~~6b~~ 7b and ~~6f~~7f). These correspond to 15% change for the primary and 25% for the secondary bbOA levels. The maximum difference for average bbPOA is approximately 5 μg m^{-3} and for bbSOA around 1.5 μg m^{-3} both in northwestern Portugal. However, during the fire period (March 20-22) in Spain and Portugal the maximum concentration difference between the two cases was 20 μg m^{-3} for bbPOA and 7 μg m^{-3} for bbSOA.

Figure ~~7~~ 8 shows the total bbOA (sum of bbPOA and bbSOA) during both periods. Higher bbOA concentrations are predicted in the base case simulation due to the higher bbSOA concentrations from higher IVOC emissions. During summer the contributions of the biomass burning IVOC oxidation products to total bbOA exceed 30% over most of Europe, while during winter these components are important mostly over Southern Europe and the Mediterranean (Fig. S4).

~~Alvarado et al. (2015) also evaluated the volatility distribution of bb emissions by using the Aerosol Simulation Program (ASP) to simulate the chemical evolution of SOA within a young biomass burning smoke plume sampled over California in November 2009. SOA formation is simulated using the VBS scheme as proposed by Robinson et al. (2007) and the gas-phase chemistry implemented for organic compounds was RACM2. The volatility distribution for the POA was taken from the wood smoke study of Grieshop et al. (2009a) and implies that most of the total mass of the organic compounds species is in the aerosol phase which leads to OA overestimation. However, Grieshop et al. (2009a) was only able to measure species~~

~~with a saturation mass concentration of 10^4 μg m^{-3} or less. To account for the unidentified IVOCs an additional organic mass was included in the $C^*=10^5$ μg m^{-3} and $C^*=10^6$ μg m^{-3} which lead to a reduction of the OA formed. Alvadaro et al. (2015) concluded that the unidentified IVOCs are mainly more volatile ($C^*=10^6$ μg m^{-3}).~~

7 Comparison with field measurements

In order to assess the PMCAMx-SR performance during the two simulation periods the model's predictions were compared with AMS hourly measurements that took place in several sites around Europe. All observation sites are representative of regional atmospheric conditions.

The PMF technique (Paatero and Tapper, 1994; Lanz et al., 2007; Ulbrich et al., 2009; Ng et al., 2010) was used to analyze the AMS organic spectra providing information about the sources contributing to the OA levels (Hildebrandt et al., 2010; Morgan et al., 2010). The method classifies OA into different types based on different temporal emission and formation patterns and separates it into hydrocarbon-like organic aerosol (HOA, a POA surrogate), oxidized organic aerosol (OOA, a SOA surrogate) and fresh bbOA. Additionally, factor analysis can further classify OOA into more and less oxygenated OOA components. Fresh bbOA can be compared directly to the PMCAMx-SR bbPOA predictions, whereas bbSOA should, in principle at least, be included in the OOA factors. The AMS HOA can be compared with predicted fresh POA. The oxygenated AMS OA component can be compared against the sum of anthropogenic and biogenic SOA (aSOA, bSOA), SOA-sv and SOA-iv, bbSOA and OA from long range transport.

PMCAMx-SR performance is quantified by calculating the mean bias (MB), the mean absolute gross error (MAGE), the fractional bias (FBIAS), and the fractional error (FERROR) defined as:

$$MB = \frac{1}{n}\sum_{i=1}^{n}\left(P_i - O_i\right) \qquad MAGE = \frac{1}{n}\sum_{i=1}^{n}\left|P_i - O_i\right|$$

$$FBIAS = \frac{2}{n}\sum_{i=1}^{n}\frac{P_i - O_i}{P_i + O_i} \qquad FERROR = \frac{2}{n}\sum_{i=1}^{n}\frac{\left|P_i - O_i\right|}{P_i + O_i}$$

where P_i is the predicted value of the pollutant concentration, O_i is the observed value and n is the number of measurements used for the comparison. AMS measurements are available in 4 stations (Cabauw, Finokalia, Melpitz and Mace Head) during 1-29 May 2008 and 7 stations (Cabauw, Helsinki, Mace Head, Melpitz, Hyytiala, Barcelona and Chilbolton) during 25 February-23 March 2009.

During May 2008 a bbPOA factor was identified based on the PMF analysis of the measurements only in Cabauw and Mace Head. In the other two sites (Finokalia and Melpitz) PMCAMx-SR predicted very low average bbPOA levels (less than 0.1 μg m^{-3}), so its predictions for these sites can be viewed as consistent with the results of the PMF analysis. Figure ~~8~~ 9 shows the comparison of the predicted bbPOA by PMCAMx-SR with the observed values in Cabauw. The average AMS-PMF bbOA was 0.4 μg m^{-3} and the predicted average bbPOA by PMCAMx-SR was also 0.4 μg m^{-3}. The mean bias was only -0.01 μg m^{-3}. The model however tended to overpredict during the first 10 days and to underpredict during the last week. In Mace Head PMCAMx-SR predicts high bbOA levels during May 14 – 15, but unfortunately the available measurements started on May 16. During the last two weeks of the simulation the model predicts much lower bbOA levels (approximately 0.35 μg m^{-3} less) than the AMS-PMF analysis. The same problem was observed in Cabauw suggesting potential problems with the fire emissions during this period.

During winter the model tends to overpredict the observed bbOA values in Barcelona, Cabauw, Melpitz, Helsinki and Hyytiala. On the other hand, the model underpredicts the bbOA in Mace Head and Chibolton by approximately 0.3 μg m^{-3} on average. The prediction skill metrics of PMCAMx-SR (base case and sensitivity test) against AMS factor analysis during the modelled periods are also provided in the Supplementary Information (Tables S2-S5). These problems in reproducing wintertime OA measurements were also noticed by Denier van der Gon et al. (2015) and suggest problems in the emissions and/or the simulation of the bbOA during this cold period with slow photochemistry.

Given that bbOA contributed on average less than half of the total OA during the summer, the performance (fractional bias and error) for OA of PMCAMx-SR (both for the base case and the sensitivity test) was quite similar to that of the original PMCAMx (Table S6). The performance of the sensitivity test was a little better suggesting that the bbSOA production from the corresponding IVOCs could be overpredicted. However, this can be also due to other sources of error in the model.

The situation was similar during the winter. There was a small reduction in the already small fractional bias but overall the performance of PMCAMx-SR for OA and OOA were quite similar to that of PMCAMx (Table S7). This however suggests that the errors in the OA predictions are not due to the new treatment of bbOA but rather to other errors that are also present in the original model.

8 Conclusions

A source-resolved version of PMCAMx, called PMCAMx-SR was developed and tested. This new version can be used to study independently specific ~~organic aerosol~~ OA sources (~~eg.~~e.g. diesel emissions) if so desired by the user. We applied PMCAMx-SR to the European domain during an early summer and a winter period focusing on biomass burning.

The concentrations of bbOA (sum of bbPOA and bbSOA) and their contributions to total OA over Europe are, as expected, quite variable in space and time. During the early summer, the contribution of bbOA to total OA over Europe was predicted to be 16%, while during winter it increased to 47%. Secondary biomass burning OA was predicted to be approximately 70% of the bbOA during summer and only 30% during the winter on average. The production of bbSOA increases the range of influence of fires.

The IVOCs emitted by the fires can be a major source of SOA. In our simulations, the IVOCs with saturation concentrations $C^{*}=10^{5}$ and 10^{6} μg m^{-3} contributed approximately one third of the average bbOA over Europe. The emissions of these compounds and their aerosol forming potential are uncertain, so the formation of bbSOA clearly is an importance topic for future work.

PMCAMx-SR was evaluated against AMS measurements taken at various European measurement stations and the results of the corresponding PMF analysis. During the summer the model reproduced without bias the average measured bbPOA levels in Cabauw and the practically zero levels in Finokalia and Melpitz. However, it underpredicted the bbPOA in Mace Head. Its performance for oxygenated organic aerosol (OOA) which should include bbSOA together with a lot of other sources was mixed: overprediction in Cabauw (fractional bias +42%), Mace Head (fractional bias +34%), and Finokalia (fractional bias +23%) and underprediction in Melpitz (fractional bias -14%).

During the winter the model overpredicted the bbPOA levels in most stations (Cabauw, Helsinki, Melpitz, Hyytiala, Barcelona), while it underpredicted in Mace Head and Chibolton. At the same time, it reproduced the measured OOA concentrations with less than 15% bias in Cabauw, Helsinki, and Hyytiala, underpredicted OOA in Melpitz, Barcelona, and Chibolton and ovepredicted OOA in Mace Head. These results both potential problems with the wintertime emissions of bbPOA and the production of secondary OA during the winter.

Data availability. The data in the study are available from the authors upon request (spyros@chemeng.upatras.gr).

Author contributions. GNT conducted the simulations, analysed the results, and wrote the paper. SNP was responsible for the design of the study, the synthesis of the results and contributed to the writing of the paper.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This study was financially supported by the European Union's Horizon 2020 EUROCHAMP–2020 Infrastructure Activity (Grant agreement 730997) and the Western Regional Air Partnership (WRAP Project No. 178-14).

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Table 1. Parameters used to simulate POA and bbPOA emissions in PMCAMx-SR.

C^* at 298 K ($\mu g\ m^{-3}$)	10^{-2}	10^{-1}	10^{0}	10^{1}	10^{2}	10^{3}	10^{4}	10^{5}	10^{6}
POA									
Fraction of POA emissions[1]	0.03	0.06	0.09	0.14	0.18	0.30	0.40	0.50	0.80
Effective Vaporization Enthalpy ($kJ\ mol^{-1}$)	112	106	100	94	88	82	76	70	64
bbPOA (Base Case)									
Fraction of POA emissions	0.2	0.0	0.1	0.1	0.2	0.1	0.3	0.50	0.80
Effective Vaporization Enthalpy ($kJ\ mol^{-1}$)	93	89	85	81	77	73	69	70	64
bbPOA (Sensitivity Test)									
Fraction of POA emissions	0.2	0.0	0.1	0.1	0.2	0.1	0.3	-	-
Effective Vaporization Enthalpy ($kJ\ mol^{-1}$)	93	89	85	81	77	73	69	-	-

[1] This is the traditional non-volatile POA included in inventories used for regulatory purposes. The sum of all fractions can exceed unity because a large fraction of the IVOCs is not included in these traditional particle emission inventories.

Table 2. Organic compound emission rates (in tn d^{-1}) over the modeling domain during the simulated periods.

	Emission rate (tn d^{-1})
1 – 29 May 2008	
Wildfires	1,700
Residential	700
Agriculture - waste burning	300
25 February – 22 March 2009	
Wildfires	3,000
Residential	6,000
Agriculture - waste burning	320

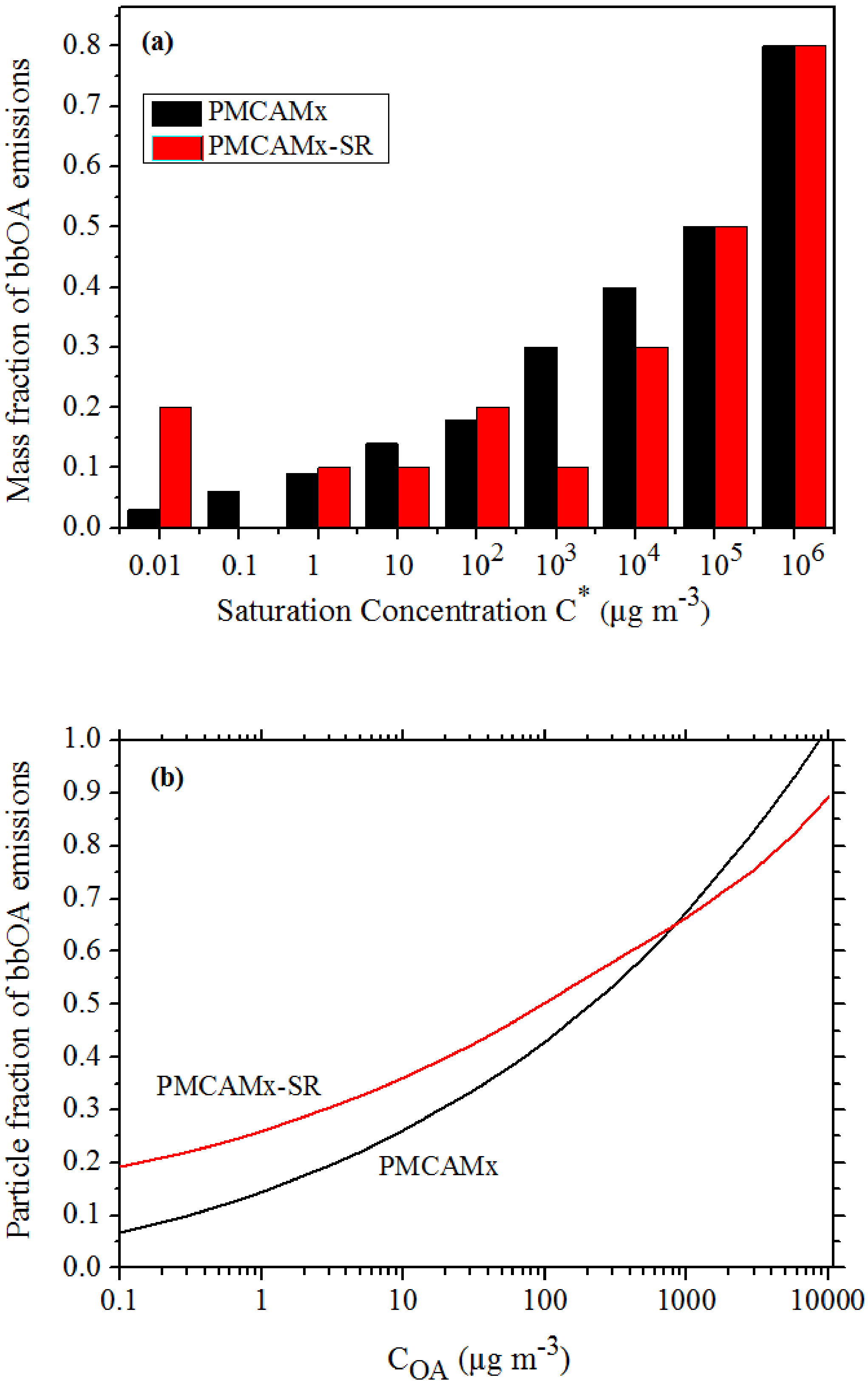
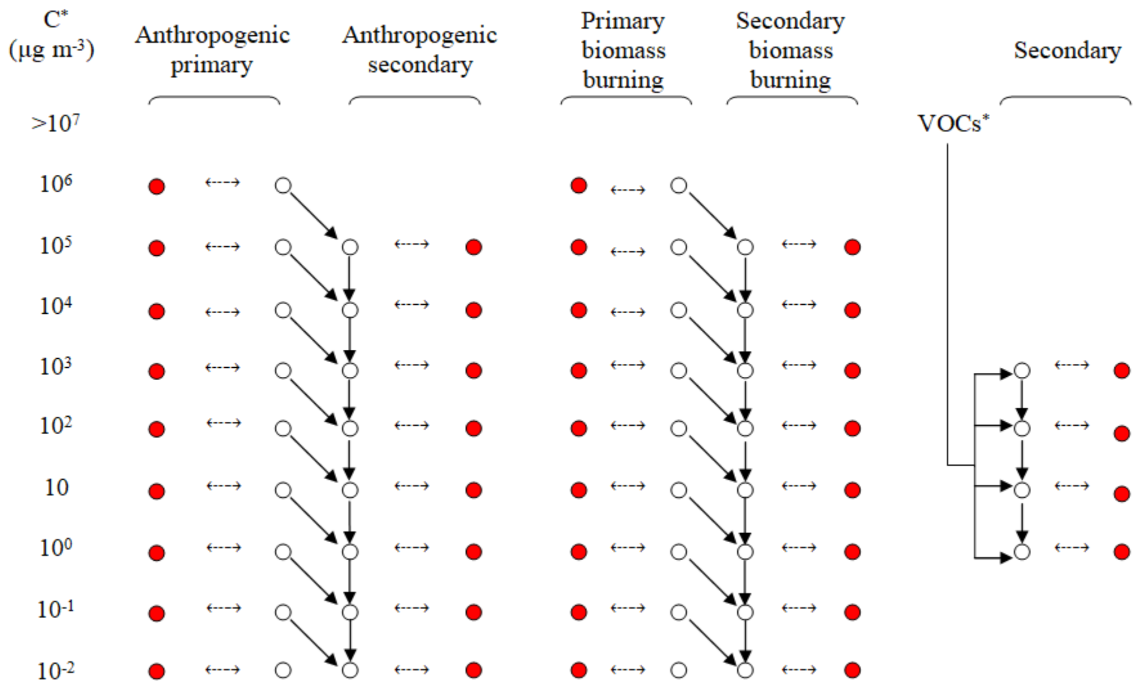


Figure 1. (a) Volatility distribution of bbOA in PMCAMx and PMCAMx-SR. (b) Particle fractions of bbOA emissions as a function of OA concentration at 298 K.



*For this application it is assumed that SOA formed by biogenic VOCs does not participate in aging reactions.

● Particle phase
○ Gas phase
←--→ Gas – particle partitioning
→ Aging reaction

Figure 2. Schematic of the ~~proposed~~ organic aerosol VBS-based ~~framework~~modeling scheme in ~~PMCAMx and~~ PMCAMx-SR.

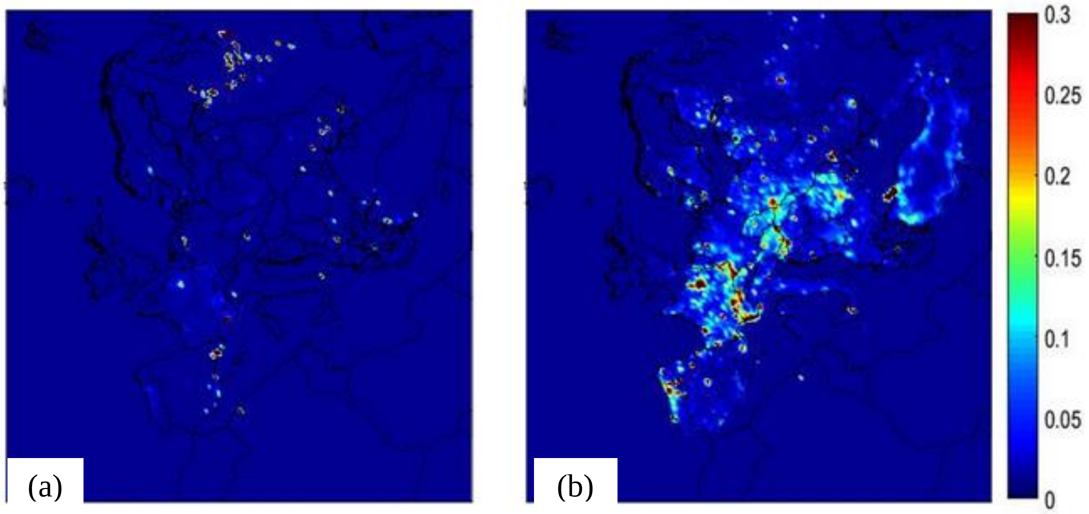


Figure ~~2~~3. Spatial distribution of average biomass burning OA emission rates (kg d^{-1} km^{-2}) for the two simulation periods: (a) 1-29 May 2008 and (b) 25 February-22 March 2009.

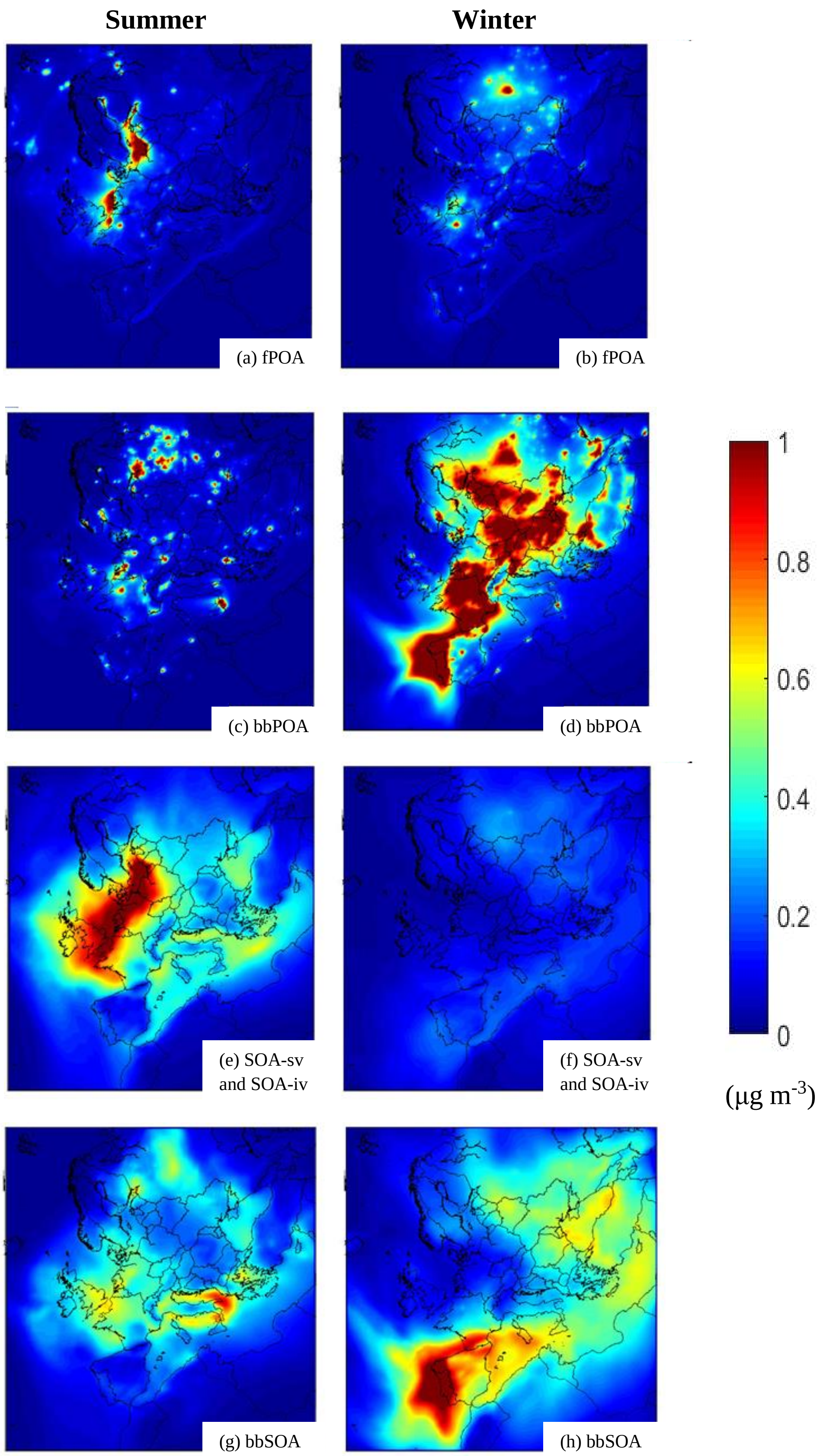


Figure ~~3~~4. PMCAMx-SR predicted base case ground – level concentrations of PM$_{2.5}$ (a-b) fPOA, (c-d) bbPOA, (e-f) SOA and (g-h) bbSOA, during the modeled summer and winter periods.

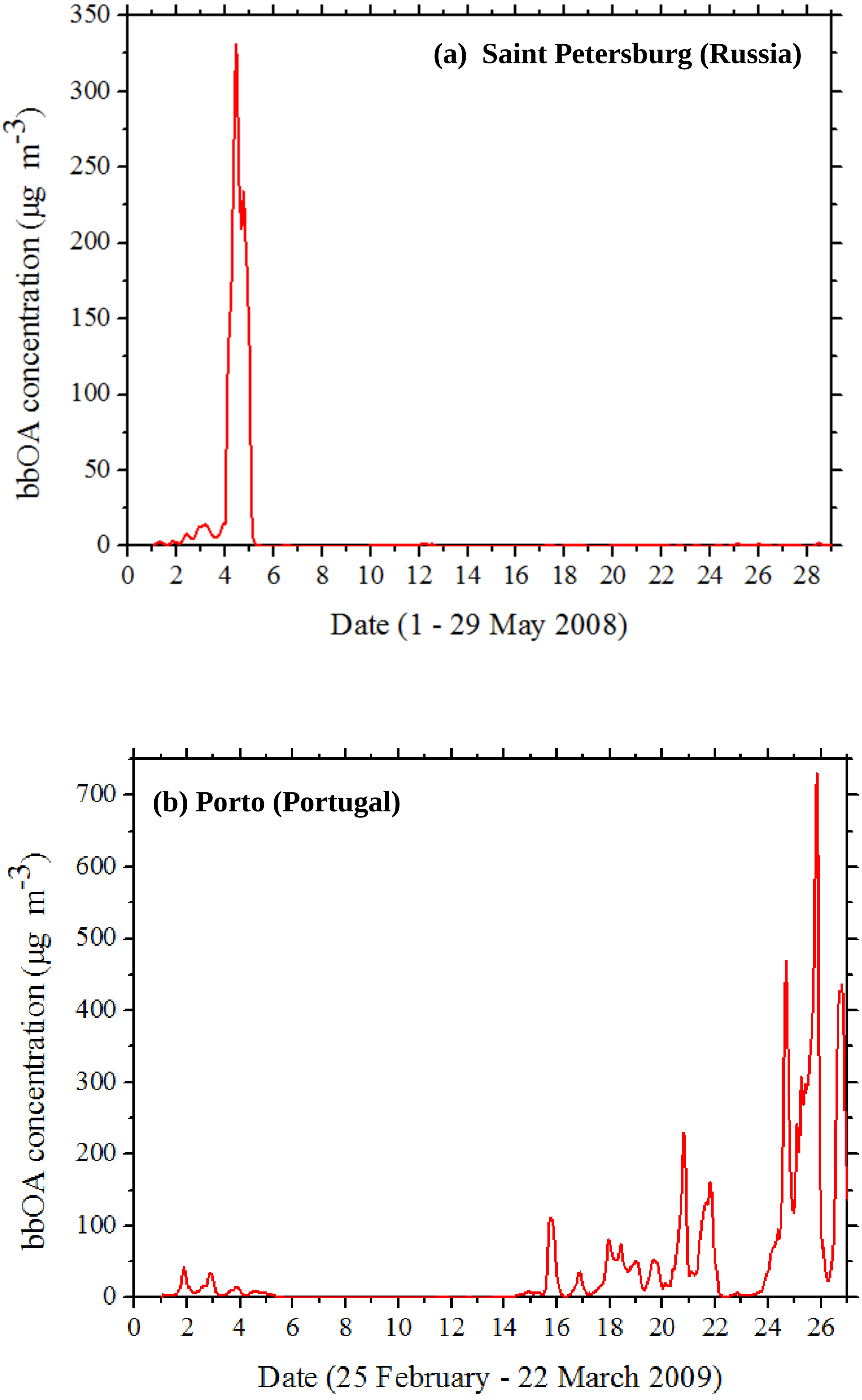


Figure 45. Timeseries of $PM_{2.5}$ bbOA concentrations in (a) Saint Petersburg in Russia during 1-29 May 2008 and in (b) Porto in Portugal during 25 February-22 March 2009.

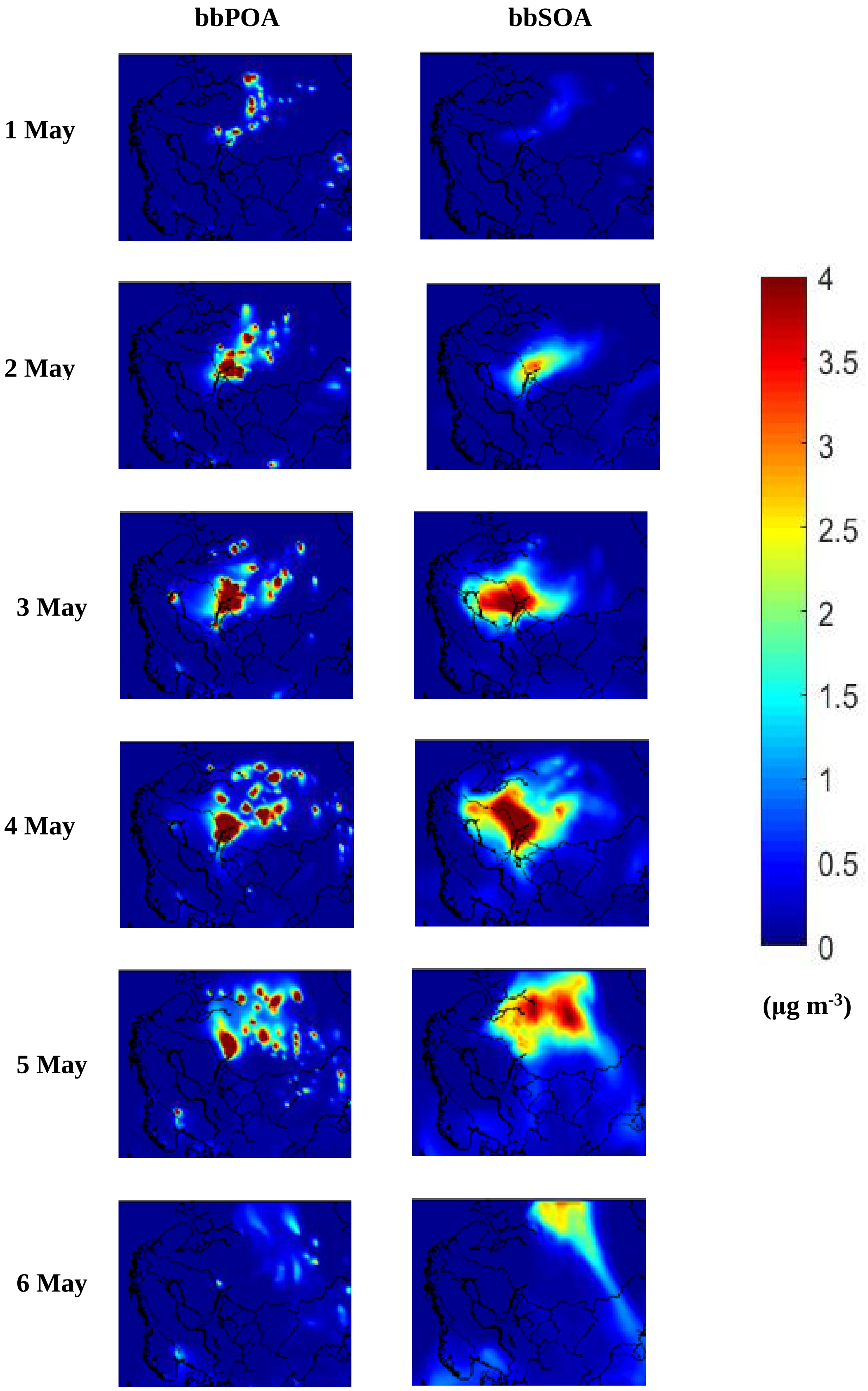


Figure ~~5~~6. PMCAMx-SR predicted base case ground – level concentrations of $PM_{2.5}$ bbPOA and bbSOA, during 1 – 6 May 2008 in the Scandinavian Peninsula and Russia.

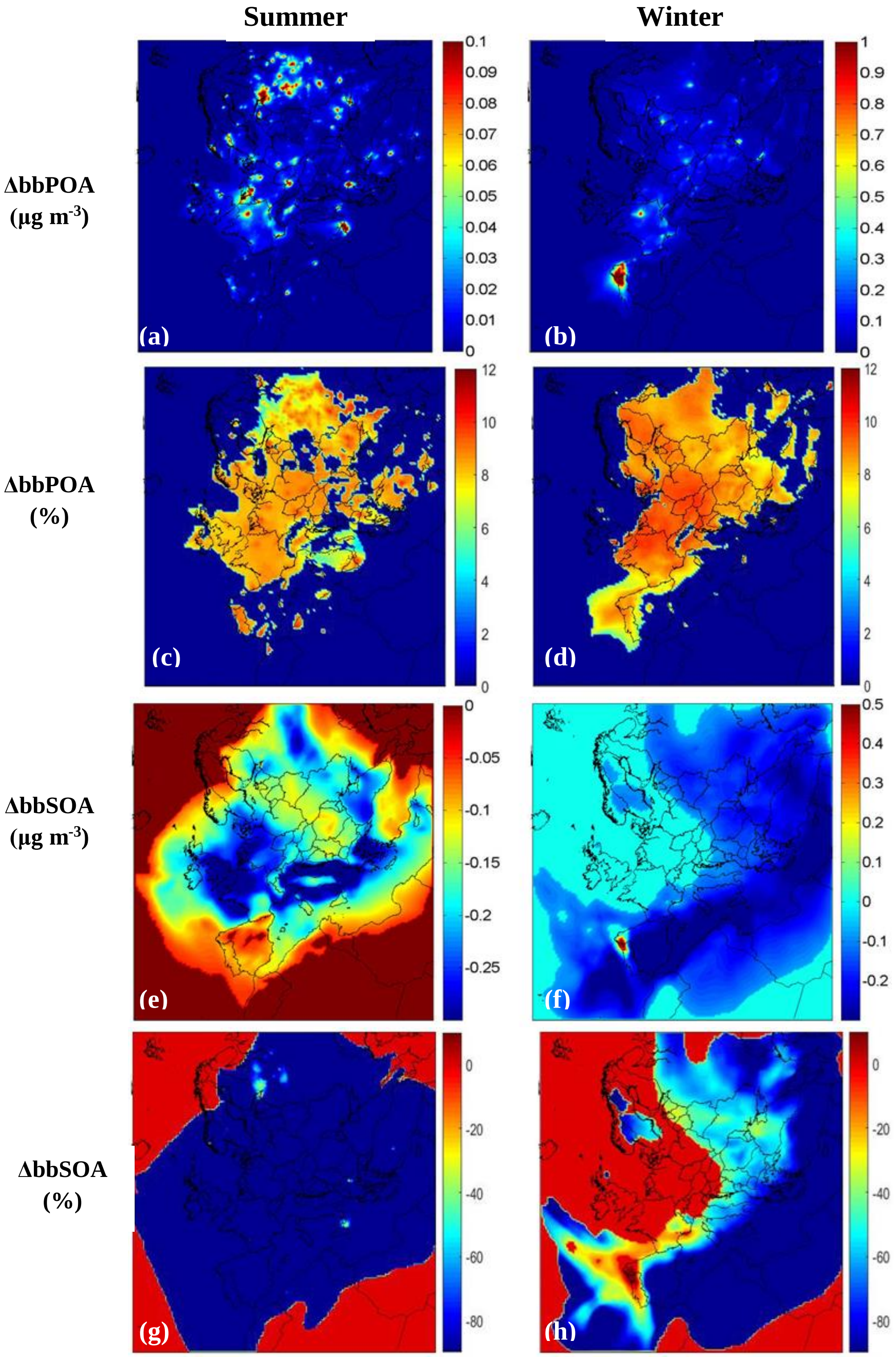


Figure ~~6~~7. Average predicted absolute ($\mu g\ m^{-3}$) difference (Sensitivity Case – Base Case) of ground-level $PM_{2.5}$ (a-b) bbPOA and (e-f) bbSOA concentrations from PMCAMx-SR base case and sensitivity simulations during the modeled periods. Also shown the corresponding relative (%) change of ground-level $PM_{2.5}$ (c-d) bbPOA and (g-h) bbSOA concentrations during the modeled periods. Positive values indicate that PMCAMx-SR sensitivity run predicts higher concentrations.

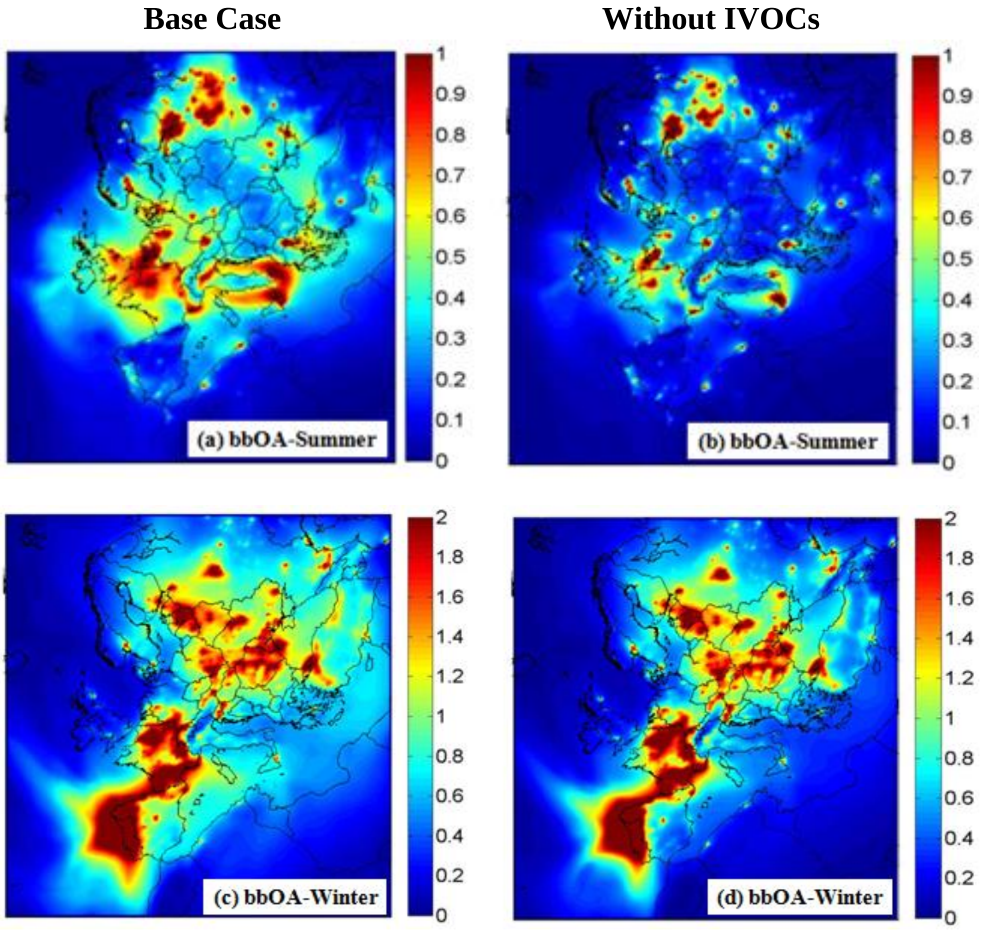


Figure ~~7~~8. Predicted ground-level concentrations of $PM_{2.5}$ total bbOA (μg m^{-3}) during the modeled summer (a-b) and the modeled winter (c-d) period. The figures to the left are for the PMCAMx-SR base case simulation while those to the right for the low-IVOC sensitivity test.

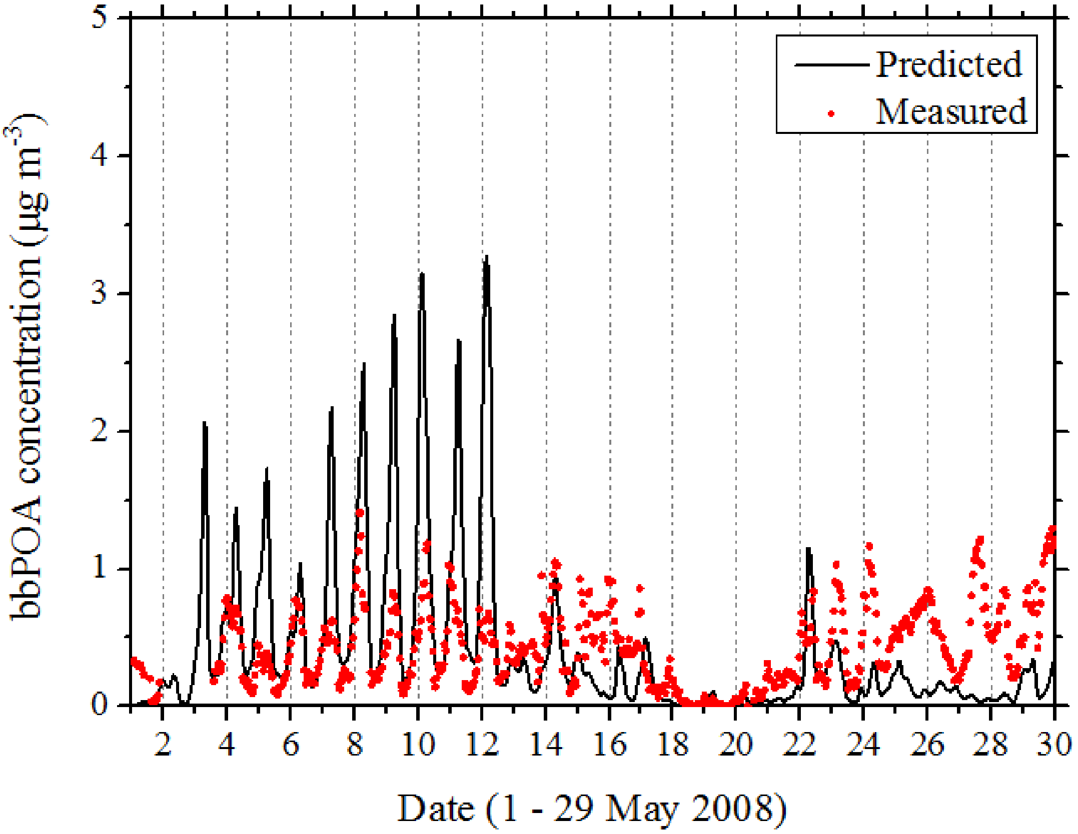


Figure ~~8~~9. Comparison of hourly bbPOA concentrations predicted by PMCAMx-SR with values estimated by PMF analysis of the AMS data in Cabauw during 1-29 May 2008.