1 Response to Anonymous Referee #1

1.1 Anonymous Referee #1

The manuscript by Bergstrom and coworkers presents simulations of organic aerosols over Europe using the EMEP model for 2002-07 using four different treatments of organic evaporation and chemical aging, and compares them to several types of measurements. Some conclusions appear strong, such as the underestimation of residential wood combustion emissions in winter, while the relative performance of different OA aging and SOA formation model variants is less clear. This paper represents the second application of OA models based on the ”volatility basis set” (VBS) type of parametrisation to Europe, using a different host model than the previous publication. The topic of the paper is of high current interest in the research community and fits well in ACP, and the paper is of good quality. I recommend that this paper is accepted into ACP once the following major issues have been addressed.

Reply

We thank the referee for their thorough reading on the manuscript and very useful and constructive comments. We believe the paper has been much improved as a result of these comments.

1.2 Major Issues

1.2.1 Referee comment

There appears to be major terminology error in the manuscript. On P5439 the authors describe that POA emissions are treated as semivolatile (leading to evaporation of SVOC to the gas phase) and are accompanied by a mass 1.5 times larger of intermediate volatility species (IVOC). IVOCs are never in the condensed phase (as their c* is 1000 ug m-3 or larger, e.g. Robinson et al. Science 2007). IVOCs are thus NEVER part of the POA, even though their emission rates are estimated based on the POA emission rates here. IVOC material may form SOA in the gas-phase leading to particle-phase material. However the authors appear to call this material oxidized POA (OPOA). This term is almost exclusively used in the community to refer to species whose oxidation has occurred in the particle phase due to heterogeneous or multiphase reactions. Referring to SOA species as OPOA creates much confusion in the aerosol community. E.g. the following papers all discuss SOA formation from IVOC species, and ALL of them refer to this material as SOA:

(Refs omitted here...)

Similarly the evaporation of SVOC from POA is followed by SOA formation in the gasphase and condensation of this material. Thus it would also be objectively wrong to refer to this material as OPOA. The terminology in the manuscript needs to be modified to follow the proper definition of SOA. Some papers have referred to ”non-traditional SOA” when discussing SOA formed from SVOC and IVOC. This or another term that includes ”SOA” should be used to refer to this material throughout the manuscript.

Reply:

The phrasing here surrounding IVOC and POA was confusing we agree, and this has been corrected. These compounds were implemented correctly in the model. We used the term OPOA in the same way as Shrivastava et al., JGR 2008, Murphy & Pandis, 2009, and Fountoukis et al., 2011, that is, to cover all the OA formed from aged primary emissions of S/IVOCs, but we agree with the referee that this is a confusing terminology and have changed this for the revised version of the paper, using sub and superscripts to flag the source of SOA species.

1.2.2 Referee:

Emissions of non-fossil carbon from urban areas, such as from cooking sources, appear to be ignored in this paper. Multiple papers using both molecular markers and aerosol mass spectrometry point to the importance of this source, which may constitute about half of the POA in urban areas. See e.g. Schauer et al. (Atmos Environ 1996); Schauer et al. (Env Sci Tech 2000), Zheng et al. (J Air Waste Manag Assoc 2007), Allan et al. (Atmos Chem Phys 2010) , Huang et al. (Atmos Chem Phys 2010), Ham and Kleeman (Atmos Environ 2011), Sun et al. (Atmos Chem Phys 2011), Minguillon et al. (Atmos Chem Phys 2011), Mohr et al. (Atmos Chem Phys 2012). For example Ham and Kleeman (2011) state that ”Meat cooking was the largest identified source of PM(1.8) is organic carbon (OC) at the urban site” and Mohr et al. (2012) state that ”primary OA in Barcelona contains a surprisingly high fraction (59%) of non-fossil carbon.” Omitting these sources will confuse the interpretation of non-fossil carbon measurements, and this omission should at least be discussed in the manuscript.

Reply:

Yes, this should have been mentioned. The emission inventory was prepared before the importance of cooking emissions was established for European conditions, and it is still unclear how important this factor is for much of Europe. The inventory does not really include such emissions, which implies both that we should have had an extra source of OM in the model, and that the 14C comparisons need to be interpreted with caution. The inventory will be updates in future to account We have added text to the manuscript to discuss this:

It should be noted that the emission inventory used in this work has only small contributions from cooking emissions. Although cooking emissions were established many years ago as major sources of OA in Northern America (e.g. Schauer et al., 1996, 2000, Ham and Kleeman, 2011), it has been assumed in most European inventories that this is not a major source. The reasons include a lower consumption of fried meats and other food in Europe, and less outdoor cooking in general. However, Mohr et al. (2011) recently found that primary OA in Barcelona contained almost 60% of non-fossil carbon from cooking sources during March 2009.

On the other hand, Barcelona is a very large (population 5 million) Mediterranean city with very strong urban impacts on air quality, and is not necessarily typical of the rest of the Europe. Szidat et al. (2009) found that both the water soluble and water-insoluble (WINSOC) fractions of organic carbon sampled in and around Gothenburg in Sweden showed similar fractions of modern carbon. As cooking emissions should be associated primarily with the WINSOC fraction, this similarity was interpreted to suggest that cooking was not of major importance even in the centre of this moderately sized (500 000 inhabitants) city. Thus, the question of the importance of cooking emissions in the European inventory is still open, but obviously one that warrants urgent attention. The addition of cooking emissions to the inventory would of course raise urban OA levels, and also affect the interpretation of source-apportionment data based upon 14C measurements.

1.2.3 Referee:

P5438-L14; the DHvap value chosen is too low. Such low values are appropriate for models with a few lumped species. However models with volatility bins every order of magnitude in c* should use higher and more realistic enthalpies of vaporization. See Donahue et al. (Env Sci Tech 2006) for a discussion of this topic. The argument that Fountoukis results did not find a strong sensitivity to this parameter is not so relevant here, as those authors only simulated the month of May, while the current study spans the whole annual cycle and thus a much wider range of ambient temperatures. Also the partitioning of semivolatile species as the air rises and cools in the atmosphere is very sensitive to this parameter.

Reply

This is partially a misunderstanding caused by an unclear formulation on page 5438. We agree with the referee that realistic ΔH_{vap} should be used and we have in fact used realistic (VBS-bin-dependent) values for the species treated with the large VBS-basis set (the POA/S/IVOC species). For these we have used the same ΔH_{vap} values as Robinson et al., 2007. This was described on page 5439-L11-14 of the manuscript (but referring to Shrivastava et al., 2008).

The low effective ΔH_{vap} (based on the parametrisation by Pathak et al., 2007) was only used for the species treated with the small basis set with 4 lumped species (BSOA and traditional ASOA) in the same way as done in several studies by other groups (e.g., Lane et al., 2008; Murphy and Pandis, 2009; Farina et al., 2010). We will reformulate this to make it clear in the revised manuscript:

The temperature dependence of the gas-particle partitioning is taken into account by using the Clausius-Clapeyron equation to calculate the saturation concentrations, along with the enthalpy of vapourisation, ΔH_{vap}. In principle, ΔH_{vap} should vary across the VBS bins, with higher values for the lower C* values (Epstein et al. 2010). In this study we use the ΔH_{vap}-values from Robinson et al., 2007, for the nine-bin VBS used for the primary emissions (values vary from 64 kJ/mole for the most volatile to 112 kJ/mole for the least).

The VBS-parametrisation of SOA yields from Pathak et al., 2007, used a constant effective ΔH_{vap}=30kJ/mole for the four-bin VBS. This value was selected to reproduce the observed temperature dependence of the smog chamber aerosol yields and accounts for various temperature effects on the SOA yields. Here we use this effective ΔH_{vap} for the SOA from VOC (similar to e.g. Lane et al., 2008a,b; Murphy and Pandis, 2009; Farina et al. 2010).

1.2.4 Referee:

The 'aging' of SOA from some biogenic and anthropogenic VOCs as used here leads to unrealistically large yields. Since the semivolatile species continue to age until they completely reside in the particle-phase (at OA concentrations of a few ug m-3), we can estimate the final yields from Table 3 as 100% for aromatics under high NOx and 120% under low NOx. Similarly high yields around 100% are obtained for terpenes. Those yields are extraordinarily high and are inconsistent with current understanding of the oxidation of those species. In particular there is no experimental support for such extremely high yields in the chamber laboratory literature, as recent reevaluations find yields on the range 5-30% under atmospheric concentration levels (see e.g. Ng et al. Atmos Chem Phys 2007a, 2007b). Chamber studies are uncertain and could underestimate the yields somewhat, but not by such a large factor. Although the authors did not come up with the aging parameterizations that they are using, the fact that they produce yields 3-20 times larger than observed in chamber studies needs to be mentioned in the paper. If such a mechanism results in SOA levels that compare well with measurements it is most likely due to cancellation of errors, as this extra SOA must be compensating for precursors with similar emission footprints and formation timescales that are missing from the model.

Answer: The yields we use are taken from Lane et al. (2008), and are those which underpin much of the evaluation work with VBS done in North America and Europe (Fountoukis et al.), but yes, these high potential yields are a well-known problem with the simple VBS approach. As the referee points out, yields approaching 100% are possible in theory, but usually such high yields are in practice not reached (or rarely). As seen in the maps (or European measurements), most ambient OA values are of order 1-10 $\mu\text{g m}^{-3}$, so only SVOC compounds with C*=1-10 are really contributing much to OA. For the aromatics, the largest production of SVOC is for the C*=100, C*=1000 $\mu\text{g m}^{-3}$ classes, which require 2-3 oxidation generations before they contribute to OA. This is possible in some areas in the summer, in which case we agree that we may well be getting too much SOA for the wrong reasons.

We do share the referees scepticism that these high yields are possible, but this variant of the VBS approach is still valuable in that it represents the extreme high end of what is possible with current VBS methods. It is for example valuable to demonstrate that even with the high yields and aging associated with the PAA approach, the model still underpredicts observed OA and SOA. These results suggest that either BVOC emissions have to be higher in the model (e.g. with the inclusion of sesquiterpenes, or simply more monoterpenes), or that other mechanisms are needed to obtain a higher condensation of the semi-volatile gases.

With regard to yields from smog-chamber studies, these are usually restricted in time. Even though e.g. Ng et al. (2007, cited by referee) used unusually long time-scales of up to 20 hours in their experiments, most yield data are based upon shorter experiments. Ambient aerosol often has had many days of photo-oxidation, and in much more complex conditions. The study of ambient AMS data by Ng et al. (2010) concluded that usually smog-chamber data generate so-called SV-OOA, whereas ambient aerosol had much lower volatility, denoted LV-OOA.

Interestingly, Donahue et al. 2005 pointed out that yields could approach near-unity after several generations, and at that time they believed that there was insufficient data to eliminate the possibility; indeed it was even consistent with some atmospheric observations (ibid.). More recently, this ‘zombie’ effect (all the high C^* bins marching towards low volatility, Donahue’s terminology) is avoided in models through the 2-D VBS framework (Jimenez et al., 2009) which can account for fragmentation. Experimental work by e.g. Henry and Donahue (2012) suggests photolysis as one contributor to increasing volatility, but also suggests that the behaviour is complex.

We have added text to summarise the above points in the discussion section of the paper, with increased emphasis on the assumptions used.

1.3 Other Topics

1. The paper relies too much on tables, which makes some sections quite difficult to follow (e.g. the comparison with source apportionment studies does not have a single figure and is quite difficult to read). I strongly recommend that most of the tables are moved to the Supp info and the information is presented instead in graphical form.

   Answer: We agree that too much information was tabular in the original manuscript. We have now created Figures from much of this, and the new manuscript will refer largely to these Figures. We believe the whole manuscript has benefited greatly from this change. (Tables will be kept in the Supplementary material.)

   We will also restructure the source apportionment section to make it easier to read/follow.

2. The paper is inconsistent in the use of the terms of residential combustion, wood burning, biomass burning, vegetation burning... For example in P5447-L26 the term ’biomass burning’ is used to refer to wintertime residential heating, when this term is most frequently used to refer to wildfires in the literature. I strongly recommend defining unique terms to refer to residential combustion vs open burning (the latter comprising wildfires and agricultural fires) early in the paper, and sticking to those later on, to reduce confusion.

   Answer: We agree with this suggestion and will change to a more consistent terminology.

3. P5430; it appears that the EMEP model does not represent the aerosol size distribution explicitly? Is a constant distribution prescribed for deriving aerosol deposition velocities, wet deposition etc? This may be described in previous papers but should be summarized in a couple of sentences here since it is important for understanding the current results.

   Answer: Yes, basically we use two modes, although our definitions of particle-size depend a little on the compound. The present version of model is mainly designed to calculate PM_{10} and $PM_{2.5}$ mass closure (i.e. concentrations and chemical composition), which over the last decade has been the highest priority within the EMEP/LRTAP Convention framework. More details are given in the EMEP model documentation paper (Simpson et al., 2012). We will add some sentences to make this clear.

4. P5431-L12; the range of deposition velocities calculated here for the higher aldehydes should be given, so that the relative effect of deposition of semivolatile organics calculated in this paper can be compared against that of future studies. Since these may depend on season and location, it may be useful to provide maps of average deposition velocities for summer and winter in the supplementary information.

   Answer: The subject of dry deposition of semivolatile organics is of course very important, but not what we wanted to focus on in this paper. Further, the deposition treatment is just one uncertainty among many, and we cannot document every aspect of the OA modelling without making the paper excessively large. However, we have added some text to summarise the range of velocities of these organics.

5. P5433-L2; the recent study of Cubison et al. (Atmos Chem Phys 2011) summarizes observations of net SOA formation from forest fires and concludes that SOA formation leads to a small net amount of SOA (equivalent to 20% of the POA), with substantial variability across locations. I suggest citing this study since it provides stronger support for the limited SOA formation from wildfires, compared to the Bessagnet modeling study. Note that applying the S/IVOC VBS mechanism to wildfire emissions may lead to a great overestimation of SOA formation from these emissions, since the amount of SOA formed will be about 200-300% of the POA emissions.

   Answer: We have added a sentence to the manuscript:

   Cubison et al. (2011), summarising the results of a number of studies, also suggested that on average SOA formation from forest fires was relatively small, about 20% of POA, although with substantial variability.

6. P5436-L25; the units of concentration are written as μgm^{-3}, which is incorrect. Either μg m^{-3} or μg/m^3 should be used throughout the paper.

   Answer: Corrected. We have used the superscript version, with spacing ($\mu\mathrm{g\ m^{-3}}$) throughout. (This is proper SI and ACP practice)

7. P5437-L4; the location that has been studied most intensively with VBS-type models is Mexico City, with at least 9 such publications by 5 different research groups (Dzepina et al., Atmos Chem Phy 2009; Hodzic et al. Atmos Chem Phys 2010a; Tsimpidi et al. Atmos Chem Phys 2010; Hodzic et al. Atmos Chem Phys 2010b; Shrivastava et al. Atmos Chem Phys 2011; Dzepina et al. Env Sci Tech 2011; Li et al. Atmos Chem Phys 2011; Hodzic et al. Geo Mod Dev 2011; Tsimpidi et al. Atmos Chem Phys 2011) and it has been more rigorously tested there due to the availability of both ground and aircraft observations

during MILAGRO and related projects. I was surprised to find no mention of any of those studies here or a comparison later between those results and the current paper.

Answer: We had not included text concerning the Mexico-city simulations in the first manuscript as the conditions there (climate, population and emissions density) are so far removed from those of Europe, in particular in areas near the sites where we have data available. Still, we acknowledge that such studies do represent a valuable knowledge-base with regard to VBS modelling. We have added text about some of these studies in the revised version.

8. P5437-L19; my immediate reaction was that the specified background concentration is unrealistically high. This is verified by the authors later in the paper. It is important to base the background concentration used in future studies on studies at locations such as Mace Head or during research cruises or flights in the Atlantic Ocean. Those studies are not very numerous but they do exist. I also suggest being careful with the marine OA parametrisation as there appears to be a wide diversity of results and it is not clear that the factors that drive the emission are well understood. In my opinion it is more important to compare the model results at remote locations with existing measurements in order to constrain the background OA.

   Answer: We agree with the referee that as far as possible the model background OA needs to be constrained by measurements. The choice of the background OA used in this work is the same as in Simpson et al. (2007), and was largely based on earlier measurements at Mace Head (e.g. Cavalli et al. 2004, Kleefeld et al. 2002), the Azores (Pio et al. 2007) and at other remote locations (Heintzenberg 1989). We will add these references to the paper.

   However, it is clear from results presented here (Figs. 6,7), and from recently available AMS measurements, that this background assumption needs to be re-visited. Future studies will indeed include a revised set of background assumptions, although we are not yet sure which approach will be the most realistic.

9. P5437-L27; I am not aware of any studies showing that adsorption of semivolatile OC species to EC or dust particles makes a non-negligible contribution to OC concentrations, or of any other model that would account for this effect, and no literature reference is provided to support that point. OA partitioning is thought to be dominated by absorptive, and not adsorptive, partitioning (see for example Seinfeld and Pankow, Ann. Rev. Phys. Chem. 2003). Thus there is no basis to assume that the model should underestimate OC due to ignoring that process. If the authors want to keep this statement they should provide credible literature references that support its importance, otherwise this statement is unsupported and just causes confusion.

   Answer: Adsorption may be important in areas with very high EC/OC ratios (e.g., diesel soot dominated places, road tunnels, e.g., Roth et al. 2005). However, as the referee correctly points out, adsorptive partitioning to EC is unlikely to have an important effect on the gas-particle partitioning of OM for atmospheric aerosols in general. Since we do not resolve locally EC-polluted 'hot-spots' in our regional scale model, and we assume a fairly high concentration of background OC, we do not expect to have high EC/OC ratios in the model. Thus, we agree that our statement about underestimation of particle phase OC due to the fact that we ignore the impact of EC on partitioning may lead to confusion. We will remove the two sentences in the revised version of the paper.

10. P5444-L9-11; this is too speculative, the volatility distributions of the current VBS and former Kam schemes should be compared directly in the supp. Info, so that firmer conclusions can be reached.

    Answer: Unfortunately, we cannot directly compare these. The Kam-2X scheme is much more complex than the VBS schemes, in that many gas-phase reactions are involved - there is no simple yield matrix. Further, the EMEP model version used for Kam-2X has not been maintained since we moved to the VBS approach, so we cannot do simple side-by-side comparisons of the results. We believe though that the sentence as given is still valid, and it is important to draw attention to the sensitivity of SOA schemes to vapour pressure assumptions.

11. P5444-L13-14; here there is a cryptic reference to "boundary layer physics issues." Has the PBL used in the EMEP model been evaluated against measurements, especially in winter? Winter inversion heights are difficult to predict, and this could also play a role in the winter OC discrepancies. I gather from the manuscript and lack of referencing of this point that this comparison has not been done; however it is critical before further progress can be made on the winter residential burning emissions (in future publications).

    Answer: We should have said more explicitly dispersion issues. As the referee says, winter-time inversions are a possible source of problems, although typically the winter-time underestimates for OA are far higher than those for NO_2, a pollutant which also has major near-ground sources, and whose emissions and chemistry we know far better than for OA compounds. Indeed, comparisons of NO_2 show that model performance does not degrade as much between summer and winter as do the OA compounds seen here. For example, at the Ispra site, mean NO_2 for June 2006 was underpredicted by 11% (r=0.68) whereas in January 2007 the model underpredicted by 31%. Worse, but not dramatically so. (These time-periods have been the subject of intensive study, Aas et al. 2012). Further comments on similar issues can be found in our reply to referee #2.

    About the mixing height, and vertical dispersion, Jeričevič et al. (2010) developed methods in comparison with observations and 2nd-order turbulence models which were later adopted for the EMEP model. Only one station (Cabauw in the Netherlands) had data which allowed an examination of low (<200m) mixing heights though. For all models such wintertime mixing conditions represent a significant challenge.

12. P5445-L23: Supp Info figures are out of order compared to their introduction in the manuscript, please re-order them.

    Answer: Fixed

13. P5446-L24; an emission algorithm for fungal PBAP has been proposed by Heald and Spracklen (Geophys Research Lett 2009). I suggest that the authors adapt this algorithm to future simulations to start to incorporate the impact of this source.

    Answer: We have added a reference to this study in the paper and will consider adoption in future studies.

14. P5447-L1; it seems that R depends strongly on geographical location. A map of R in the sup info could be useful.

    Answer: We have replaced most of the Tables in the text with Figures, and in some of these we have illustrated the spatial variation in both concentrations and R.

15. P5479, Fig 3: the labels of the figures do not correspond to the 4 variants of the model described in the paper.

    Answer: Corrected.

16. P5457-L16; it is not clear that slower oxidation will lead to a larger influence of long range transport. Gas-phase semivolatile species dry deposit faster than submicron particles, so in the absence of wet deposition non-volatile particles should be transported farther than oxidized semivolatile species.

    Answer: We agree that the issue is more complex than indicated in the formulation on P5457. In order to avoid a complex discussion, we will just remove the sentence starting on line 14 (about the increased importance of LRT when the emissions are treated as partially IVOCs).

17. P5457-L21; the fact that the aging of aromatics and terpenes is not realistic and leads to too high yields should be mentioned here (i.e. the model performance is likely improved for the wrong reasons when using this SOA ’aging’).

    Answer: Yes, we will add a comment here.

18. P5482 and later; the crosses are difficult to see in these figures, I recommend changing the color and potentially the shape of those to make the figures more readable. E.g. open red or blue triangles may offer more contrast with the colors used for the model results. Also the figures appear to have been squished vertically which leads to distortion of the horizontal vs vertical font. Please reformat to publication-quality figures.

    Answer: These figures will be updated according to the suggestions from Referee #2

19. Typos etc P5427-L7; no need for dash between AMS and data
    P5428-L8: ’non-refractive’ should be ’non-refractory’
    P5451 L23-24: awkward wording

    Answer: Typos corrected and sentence reformulated.

Extra references

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