

Interactive comment on “Evaluation of a three-dimensional chemical transport model (PMCAMx) in the European domain during the EUCAARI May 2008 campaign” by C. Fountoukis et al.

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(1) This paper presents a modelling application in which PMCAMx is applied to a one month period in May 2008, with a focus on the modelling of organic aerosol (OA). The paper is interesting in that it applies a code which has been previously used in very different conditions (e.g. Mexico) to the European situation. Indeed, it seems to be the first application of PMCAMx to Europe. The model is compared to AMS measurements of sulphate, nitrate and ammonium as well as OA. Of particular interest is the comparison with aircraft measurements, since previous studies have suggested

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that OA models significantly underpredict airborne OA.

We would also like to note that the parameters used in the OA module of PMCAMx in the current application are the same as those in its previous applications in the Eastern US and in Mexico City (Lane et al., 2008a; b; Murphy et al., 2009; 2010; Tsimpidi et al., 2010; 2011)

(2) The paper is interesting in several respects, but I think it has some significant shortcomings also, and that major revisions are needed before I could recommend it for publication. A general problem with the paper is that no attention is paid to the major uncertainties inherent in OA modelling. The danger of getting apparently good answers for the wrong reasons is clear, yet here the authors present one setup of the model and suggest that the model evaluates well. I encourage the authors to explore these uncertainties in more detail. The result would be a much more robust paper.

We agree with the reviewer's statement regarding the uncertainties inherent in OA modeling. Our paper evaluates the model predictions of all fine PM components trying to avoid an exclusive focus on the OA concentration fields. There have been extensive sensitivity analyses of the same module presented by Lane et al. (2008a, b), Murphy et al. (2009, 2010) and Tsimpidi et al. (2010; 2011). We do recognize though that this is one of the most interesting aspects of this work, so we have added a series of sensitivity tests following the reviewer's suggestions (see following comments). These tests address the effect of chemical aging parameters, enthalpies of evaporation, etc.

The results of the previous sensitivity tests of the OA module are cited and described briefly in the revised paper. We have added descriptions of a series of sensitivity tests of the OA module in a new Sensitivity Analysis section.

Main comments:

(3) One particular problem is that the BVOC emissions used are very high, and no

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justification is presented for their use. Table 2 suggests VOC emissions of ca. 2.9 Tg for the month of May, coming from the MEGAN inventory. Emissions in May are usually about 10 percent or less of annual emissions, so an annual equivalent would be ca. 30 Tg/yr. This amount seems greatly in excess of any European estimate. For example, Simpson et al. (1999) estimated EMEP-area isoprene emissions to be 4.6 Tg/yr, monoterpene emissions to be 3.9 Tg/yr, 8.4 Tg/yr together. From the NATAIR project Curci et al. (2009) reported 3.2 Tg/yr isoprene and 5.4 Tg/yr monoterpenes for the year 2001. The recent most recent estimate I know is that of Keenan et al. (2009) who found annual EU15(+2) emissions of 0.97 TgC/yr for isoprene, and 1.03 TgC/yr for monoterpenes, so SOA-precursor emissions of BVOC of ca. 2 TgC/yr. These estimates were largely consistent with previous estimates by Arneth et al. (2008) and Simpson et al. (1999). Of course, the EU-15+2 area is smaller than that covered by PMCAMx, but the agreement for EU15+2 suggests that the Simpson et al 1999 estimate is still valid.

Large discrepancies for MEGAN in Europe have been reported before – Rinne et al (2009) showed that MEGAN predicted 240 Gg/yr of isoprene for Finland, whereas the latest estimate from Finnish experts suggested just 15 Gg/yr. How then can the authors explain such a huge BVOC emission? Presumably the use of one of the inventories developed specifically for Europe would have led to a significant underprediction of the organic aerosol, primarily through the reduced BSOA formation but also through the reduced condensational mass for all OA components.

This is a valid concern due to an incomplete description of the biogenic inventory. The 2.9 Tg BVOC emission value presented in Table 2 includes not only isoprene and monoterpenes, but other VOCs (OVOCs) as well. The emissions of isoprene and monoterpenes during May 2008 used in these simulations are actually much lower, approximately 1.37 Tg (892 Gg of isoprene and 474 Gg of monoterpenes). The remaining 1.5 Tg of OVOCs is mostly methanol, which does not contribute to the formation of SOA. We have changed Table 2 to avoid any confusion related to the above.

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Analyzing the seasonality of the available emissions from the MEGAN model, we estimated annual emissions of approximately 10 Tg (isoprene and monoterpenes) certainly a lot lower than the 30 Tg assumed by the reviewer and very close to the value reported by Simpson et al. (1999). The seasonal variability of the predicted VOC emissions were in reasonable agreement with the predictions of Steinbrecher et al. (2009) and Karl et al. (2009).

Another important factor is that the summer of 2008 and especially May of 2008 were relatively warm periods for Europe. We have also analyzed the NATAIR VOC emissions for May 2003. The original NATAIR grid covered an area similar to the PMCAMx grid. The NATAIR isoprene and monoterpene emissions were found to be 330 Gg and 640 Gg respectively, 0.97 Tg in total for the month of May of 2003. This value is close to the 1.37 Tg emissions used in our inventories. Once more the observed variations could be attributed mainly to different meteorological conditions and are within the range of uncertainty reported in the literature (Karl et al., 2009).

Regarding the article of Rinne et al (2009), the 240 Gg/yr were not predicted by the MEGAN model. These estimates are based on the algorithms of the Guenther et al. (1995). However MEGAN deviates from the original G95 approach, since it includes various refinements of the G95, geographically distributed base emission factors and also includes dependence of the emissions with the past 24 to 240 hour temperature and light conditions (Guenther et al. 2006). According to Rinne et al (2009) the estimated isoprene emissions for Finland are much lower (160 Gg/yr), while in our biogenic inventory (and for different meteorological conditions) we estimated approximately 105 Gg/yr. For the specific time period of May 2008 the estimated MEGAN VOC emissions for Finland are comparable to estimates of Karl et al. (2009).

(4) *It is hard to see the justification for not aging BSOA. It is clear that previous PM-CAMx models have not done this, but there is plenty of evidence that BSOA do age (see e.g. Hallquist et al, 2009, Jimenez et al., for some citations). The recent review of*

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AMS data by Ng et al. 2010 clearly shows this, also noting that chambers data usually only explore the initial stages of oxidation.

This is an important issue that clearly deserves additional discussion. There is no question that the biogenic SOA components undergo additional chemical reactions in the atmosphere (chemical aging). These reactions are probably taking place in both the gas and particulate phases and involve both functionalization and fragmentation reactions. These changes in the chemical composition of the biogenic SOA have been documented by the studies mentioned by the reviewer. The question here is how these reactions affect the BSOA aerosol concentration and to what degree they are captured by the yields currently used by models like PMCAMx. For example, combinations of functionalization and fragmentation may lead to significant changes in chemical composition but small changes in the overall OA concentration (or SOA yield).

We have tried to address these questions from the 3D-CTM perspective in previous publications. Lane et al. (2008a) concluded that simulating BSOA aging assuming a gas-phase rate constant with OH equal to $4 \times 10^{-12} \text{ cm}^{-3} \text{ molecule}^{-1} \text{ s}^{-1}$ results in significant overprediction of the observed OA in rural areas (e.g., in the southeast US) dominated by BSOA. Murphy and Pandis (2009) showed that PMCAMx, assuming that the effect of the chemical aging of the BSOA on its concentration is negligible, was able to reproduce well the summertime OA concentration field in the Eastern US. Murphy and Pandis (2010) explored a series of aging scenarios showing that the current aging scheme (increasing ASOA yields and constant BSOA yields) resulted in the best performance. Simulating the BSOA aging reactions in the same way as the ASOA reactions or neglecting aging reactions resulted in significant error and bias in the model predictions. Lowering the aging rate constant of the ASOA to $2.5 \times 10^{-12} \text{ cm}^{-3} \text{ molecule}^{-1} \text{ s}^{-1}$ and assuming the same reactions for the BSOA (intermediate aging case in that study) resulted in almost acceptable model performance but still worse than that of the base case. Based on these results we have kept the basecase parameters of Murphy and Pandis (2009) for the present study.

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Following the reviewer's suggestion however, we have added the description of a sensitivity test in our analysis, in which aging of both aSOA and bSOA (using the same rate constant) is included. Aging of the bSOA becomes a major OA source in this case resulting in significant overpredictions of the OA. For example, the average OA in Cabauw is overpredicted in this bSOA aging case by more than 40 percent, while in Finokalia it is overpredicted by 60 percent. We have added a discussion of these results to the revised manuscript.

(5) *Why is the same enthalpy of vaporisation used for all the bins? It has been well established that less volatile compounds should have higher values of ΔH (Donahue et al., 2006, Epstein et al., 2010). What are the implications of the choices made for these comparisons?*

In this first application of PMCAMx to Europe, we have kept the same parameters in the OA module as in our previous applications in the Eastern US and Mexico City. We should clarify first that the model does not use the same enthalpy of vaporisation for all the POA and OPOA surrogate species. These POA and OPOA components have vaporization enthalpies that vary with volatility, consistent with Murphy and Pandis (2009). On the other hand a volatility-independent ΔH_{vap} is used for the SOA components. This should be viewed as an effective average ΔH_{vap} that has been derived by fitting the few experimental measurements (for example Pathak et al., 2007).

Following the reviewer's suggestion we have done 2 more (sensitivity) simulations with ΔH_{vap} values of 50 and 75 kJ/mol for aSOA and bSOA, to check the sensitivity of our results to this parameter. For the first sensitivity test ($\Delta H_{\text{vap}} = 50 \text{ kJ/mol}$) the model predicted OA mass slightly increases (by less than 4 percent) in all altitude bins. Increasing ΔH_{vap} to 75 kJ/mol, results in an increase of OA mass by approximately 7 percent in the first km altitude while the OA concentration increase ranges from 9 to 12 percent for higher altitudes. This is consistent with Murphy et al. (2011) who showed that for the same period, surface concentration predictions are not very sensitive to the

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choice of SOA vaporization enthalpy.

(6) *This paper is an application of PMCAMx to Europe, and so one would have expected the introduction to present some details of what is known already. The AMS instrument is given a lot of attention, but much of what is known in Europe stems from filter measurements. We actually have now a pretty good basic idea of the main sources of OA in Europe, arising from the 14C work of Szidat et al. 2006, Gelencser et al. 2007, or numerous other papers.*

We have added a paragraph in the introduction discussing the conclusion of some of the field campaigns and corresponding measurements in Europe.

(7) *One would also have expected at least a mention of other SOA modelling studies in Europe. Both Simpson et al. (2007) and Bessagnet et al. (2008) have dealt with SOA modelling over Europe in much more detail. How does the present modelling exercise compare with these?*

We have added a paragraph in the introduction providing additional discussion of the results of other OA modeling studies in Europe.

(8) *p14189. The discussion of POA is confusing. Is OPOA supposed to be part of the emissions, or just another secondary component? If the latter, then clarify. Why is "aged organic aerosol" counted as POA? Which label is that given? What about the IVOC emissions mentioned in Shrivastava et al., 2008 (these significantly alter the amount of organics available for OPOA)?*

The description of POA and its components has been rewritten to avoid confusion. OPOA (Oxidized Primary Organic Aerosol) is clearly secondary. It includes all non-traditional oxidized OA components (considering as traditional the VOC products). Therefore OPOA here includes material emitted in all VBS bins that has been oxidized

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through gas-phase chemistry and has been transferred to the particulate phase. The "aged organic aerosol" coming from the boundaries (BC-OA) is expected to consist of both SOA and oxidized POA. We have changed the text here to make it clearer to the reader. For the IVOC emissions we followed the approach of Tsimpidi et al. (2010) and Shrivastava et al. (2008), in which IVOCs are emitted together with the primary OA. Because of the difficulty in measuring mass in this volatility range (it remains largely in the gas phase, $C^* > 1000 \mu\text{g}/\text{m}^3$), emission inventories do not include these compounds. We thus add an additional 1.5 times the original POA mass emission rate to the intermediate volatility organic gas emission rate following several past studies (Robinson et al., 2007; Murphy and Pandis, 2009; Hodzic et al., 2010; Tsimpidi et al., 2010). The IVOCs can be oxidized by OH and may go to lower volatility and condense. The resulting OA is oxidized and is grouped here. We have now clarified these in the text.

(9) *p 14190 and Table 1. The assumption that half of BC-OA is OPOA and half BSOA contradicts radiocarbon evidence from many studies. Where do the assumed values in Table 1 come from? By the way, what does "North" mean here. Is geographical north used, or the top edge of Fig. 2. If the latter, it looks more like east to me. Why should that boundary have more BC-OA (and hence BC-OPOA) than the other directions?*

For the BC-OA, we chose to follow the suggestions of Kanakidou et al. (2005) and the results of Farina et al. (2010). Clearly the magnitude or variation of this split are quite uncertain for the area of interest. The boundary conditions of Table 1 are based on measured background concentrations in sites close to the boundaries of the domain (e.g. Zhang et al., 2007; Seinfeld and Pandis 2006). "North" here represents the top edge of our modeling domain as it is depicted in Figure 1. We have changed the wording in Table 1 to avoid any confusion. As mentioned above, the BC-OA values are based on measured background concentrations. This is now added in the text.

(10) *p14191. The description of the emissions is rather vague. What was used for the*

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speciation of European VOC emissions (SAPRC is a model system, not an inventory).

We used the speciation scheme proposed by Visschedijk et al. (2007), which is compatible with the utilized EUCAARI anthropogenic emissions inventory. In this work, the standard chemical species of the SARPC system were adopted. We have corrected this point to avoid any confusion.

(11) *p14196. The discussion of OA components refers to Northern hemispheric studies, why? There have been many AMS studies over the years at European sites, even before the EUCAARI data became available. Why aren't the AMS components (SV-OOA, LV-OOA, HOA) used more here - I would have expected to see time-series.*

We plan to conduct a thorough comparison with the results of the PMF analysis as this will be an important additional test of the model. Unfortunately the PMF analysis has not been finalized and the results have not been published. Our intention is to perform a detailed comparison with all the PMF results (summer and winter) as soon as the complete final dataset becomes available.

(12) *The comparison with the aircraft is indeed interesting, but presumably sensitive to the enthalpy values used. I would have liked to have seen some sensitivity analysis on this, as I assume that a more realistic VBS setup might have produced a quite different vertical profile. (Of course, this depends on how the VBS bins are populated.)*

We added 2 more sensitivity runs with ΔH_{vap} values of 50 and 75 kJ/mol for aSOA and bSOA, to check the sensitivity of our results to this parameter. For the first sensitivity test ($\Delta H_{vap} = 50$ kJ/mol) the model predicted OA mass slightly increases (by less than 4 percent) in all altitudes. Increasing ΔH_{vap} to 75 kJ/mol, results in an increase of OA mass by approximately 7 percent in the first 0-1 km altitude while the OA concentration increase ranges from 9 to 12 percent for higher altitudes. This is now discussed in the sensitivity analysis section.

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(13) *The paper highlights that the Mediterranean is the only area where sulphate is higher than OA, but strangely doesn't try to explain this finding. At first I assumed that the large volcanic emissions from Italy would account for this, but Table 2 suggests no volcanic SO₂ emissions. Connected to the finding that SO₄ > OA in this region, why is OA so low? The Mediterranean should have lots of BVOC emissions, which should produce lots of BSOA. Presumably the model is allowing the OA to evaporate, but it would be good to see this explained.*

These higher sulfate levels in the Eastern Mediterranean area are due to the high photochemical activity in the area during this period, the high emissions of sulfur dioxide in the Balkans and Turkey and the long-range transport of sulfur dioxide and the resulting sulfates originating from industrial areas of eastern Europe (Sciare et al., 2003; Lelieveld et al., 2002; Ganor et al., 2000; Mihalopoulos et al., 1997) plus additional contributions of sulfur dioxide from shipping and DMS from phytoplankton. The intense sunlight and photochemistry in this area convert rapidly the transported sulfur dioxide from less photochemically active areas to sulfate. The moderate OA levels are due to moderate emission rates and also existence of some of the OA components in the gas phase due to the higher temperatures in the region. We have added text to the revised paper to explain these predictions better.

(14) *Details:*

(a) *p14186, line 14. SOA is not emitted – rephrase.*

Corrected.

(b) *p14187. Just to be sure, is the model used in this paper identical to the PMCAMx-2008 model, or has anything changed?*

The model version used in this study (PMCAMx-2008) is the same with the one in Murphy and Pandis (2009). This is now stated explicitly in the text.

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(c) p 14190, line 12. *Rotated polar stereographic project - is this the EMEP grid?*

In PMCAMx (as in CAMx) the grid projection may be selected as Cartesian (fixed physical distance coordinates on a flat plane) or curvi-linear geodetic (following the curved surface of the Earth). The Cartesian options include Universal Transverse Mercator (UTM), Lambert Conic Conformal (LAMBERT), and Rotated Polar Stereographic (POLAR). We have chosen the POLAR one. The domain used is close but not quite the same as the EMEP grid.

(d) p14191. *Oxidation of sesquiterpenes is said to be based upon Griffin et al. (1999). What do the authors mean by this?*

This refers to the yields but the reference here is rather confusing. We have deleted this sentence to avoid any confusion. As stated in page 14189, the SOA yields used in PMCAMx-2008 are based on the NO_x-dependent stoichiometric yields of Lane et al. (2008).

(e) p14197. *Give references for the statements concerning Mace Head sea-salt. Cabauw can also be affected by sea-salt - is coarse mode nitrate not a problem there?*

We have added a reference for the Mace Head sea salt measurements (Dall'Osto et al., 2010). Cabauw and Mace Head have both significant sea-salt levels but quite different levels of ammonia and nitric acid and are therefore in different chemical regimes. In Cabauw, ammonium nitrate is dominating the inorganic part of fine aerosol as can be seen from the measured (and predicted) values in Table 3. This, results in ammonium nitrate formation in the fine mode, which the bulk equilibrium assumption has no problem reproducing. In Mace Head however, the major inorganic component in the fine mass is sulfate and there is significant competition between the fine and coarse modes for the available nitrate. This is a chemical regime that cannot be simulated accurately using the bulk equilibrium approach. This interesting point is now discussed in the revised manuscript.

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(f) p 14198. *Line 11 - missing words at end?*

The typo has been corrected.

(g) p 14210. *Table 2 - are the sea-salt emissions for PM1? Is NO_x reported as NO₂? Be explicit.*

The sea-salt emissions (as well as all other particulate emissions) in Table 2 are for PM₁₀. NO_x is reported as NO+NO₂. This is now explained in the manuscript.

(h) *Mention how the model deals with dry/wet deposition for OA components on gas and particle phase.*

For the wet deposition the basic model in PMCAMx is a scavenging coefficient approach in which the local rate of concentration change within or below a precipitating cloud is equal to the product of the concentration of a pollutant and its scavenging coefficient. The scavenging coefficient is determined differently for gases and particles, based upon relationships described by Seinfeld and Pandis (2006). For the dry deposition, PMCAMx determines a deposition velocity for each land use type, for each given species, particle size, and grid cell, and then linearly combines them according to the fractional distribution of land use. The deposition flux is used as the lower boundary condition in the vertical diffusion algorithm. For the gas phase species, the resistance model of Wesely (1989) is used, while for aerosol the resistance approach of Slinn and Slinn (1980), as implemented in UAM-AERO (Kumar et al., 1996), has been adopted in PMCAMx. An effective Henry's Law constant, which affects vapor removal processes, is assumed 2700 M atm⁻¹ for all gas-phase OA species (Murphy and Pandis, 2008). This information has been added to the manuscript.

(g) *The Figure quality could be improved. Add slope, corr.coeff. to Fig. 7.*

We have improved the quality of the figures. The slope and correlation coefficient for

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Fig. 7 have been added to the text.

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