

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.

D. Simpson (Referee)

david.simpson@chalmers.se

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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

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as well as OA. Of particular interest is the comparison with aircraft measurements, since previous studies have suggested that OA models significantly underpredict airborne OA. 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.

Main comments:

1. One particular problem is that the BVOC emissions used are very high, and no 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% 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

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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.

2. It is hard to see the justification for not aging BSOA. It is clear that previous PMCAMx 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 AMS data by Ng et al. 2010 clearly shows this, also noting that chambers data usually only explore the initial stages of oxidation.
3. 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?
4. 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.
5. One would also have expected at least a mention of other SOA modelling studies

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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?

6. 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)?
7. p14190 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?
8. p14191. The description of the emissions is rather vague. What was used for the speciation of European VOC emissions (SAPRC is a model system, not an inventory).
9. 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.
10. The comparison with the aircraft is indeed interesting, but presumably sensitive to the enthalpy values used. I would have like to have seen some sensitivity analysis on this, as I assume that a more realistic VBS setup might have produced a

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quite different vertical profile. (Of course, this depends on how the VBS bins are populated.)

11. 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.

Details:

- p14186, line 14. SOA is not emitted - rephrase
- p14187. Just to be sure, is the model used in this paper identical to the PMCAMx-2008 model, or has anything changed?
- p14190, line 12. Rotated polar stereographic project - is this the EMEP grid?
- p14191. Oxidation of sesquiterpenes is said to be based upon Griffin et al. (1999). What do the authors mean by this?
- 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?
- p14198. Line 11 - missing words at end?
- p14210. Table 2 - are the sea-salt emissions for PM₁? Is NO_x reported as NO₂? Be explicit.

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- Mention how the model deals with dry/wet deposition for OA components on gas and particle phase.
- The Figure quality could be improved. Add slope, corr.coeff. to Fig. 7

Extra references

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