

Regional-scale modeling of organic aerosol composition in Europe: Insights from comparison with aerosol mass spectrometer factor analysis

C. Fountoukis¹, A.G. Megaritis^{1,2}, K. Skylakou^{1,2}, P.E. Charalampidis³, C. Pilinis³, and S.N. Pandis^{1,2,4}

¹Institute of Chemical Engineering Sciences (ICEHT), Foundation for Research & Technology-Hellas (FORTH), Patras, Greece

²Department of Chemical Engineering, University of Patras, Patras, Greece

³Department of Environment, University of the Aegean, Mytilene, Greece

⁴Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

Keywords: PMCAMx, Regional air quality, Positive Matrix Factorization, AMS data

Presenting author email: cfountoukis@iceht.forth.gr

Recently, the volatility basis set framework (VBS) has been implemented in several Chemical Transport Models (CTMs) improving predictions of organic aerosol (OA) concentrations (Fountoukis et al., 2011). However, uncertainties still exist in these models regarding the volatility distribution of the primary OA emissions, the chemical aging of the OA or errors in either the anthropogenic or biogenic emissions.

The increasing availability of Aerosol Mass Spectrometer (AMS) field measurements analyzed by Positive Matrix Factorization (PMF) provides an opportunity to further evaluate and constraint CTMs based on their ability to predict concentrations of the different OA components. In this work we apply PMCAMx-2008 over Europe (Fountoukis et al., 2011) during 3 periods representative of different seasons, and use an extensive set of AMS measurements (15 datasets in total) to evaluate the model and (using PMF results) gain more insight into the sources and physical and chemical transformations of OA, as well as identify strengths and limitations of our current OA modeling framework.

In this study we simulate 3 periods (1–29 May 2008, 25 February – 24 March 2009, and 15 September – 17 October 2008) during which 3 intensive measurement campaigns took place as part of large-scale projects of the EU. During all three campaigns, AMS measurements of aerosol mass composition were performed at several sites in Europe while further analysis of the OA composition was performed using PMF. The modeling domain covers a 5400×5832 km² region in Europe with 36×36 km grid resolution and 14 vertical layers extending up to approximately 6 km in height with a surface layer depth of 55 m.

Fig. 1 shows a comparison of predicted vs. observed PM₁ oxygenated OA (OOA) from all three measurement datasets. The overall agreement between AMS measurements and model predictions is encouraging. The OA formation during the late spring and autumn period is mostly dominated by enhanced photochemical activity while during the winter period by direct primary emissions. Predictions of fresh primary OA (POA) are the highest during the winter period ranging from 0.5 to 1.5 μg m⁻³ on average, with peak values up to 10 μg m⁻³ at urban centers. On average, the model predicts that oxygenated OA contributes 93% to

total OA during May, 87% during winter and 96% during autumn with the rest comprising fresh POA.

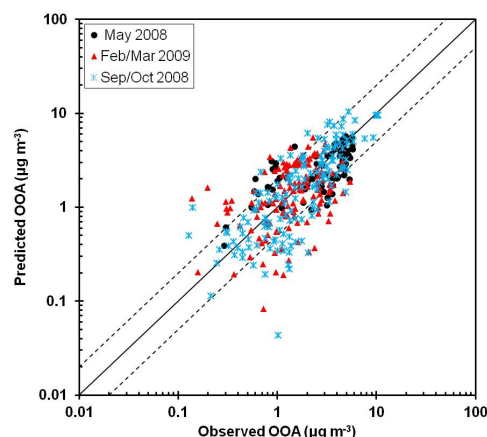


Figure 1. Comparison of predicted vs. observed PM₁ oxygenated organic aerosol from 3 measurement periods. Each point corresponds to a daily average value. Also shown the 1:1, 2:1 and 1:2 lines. Observed data represent AMS measurements. The slope of the line for OA is 0.893 ($R^2 = 0.49$).

Oxidized POA and OOA from intermediate volatility organic compounds (IVOCs) is predicted to be a major OA component with a contribution of ~50% to total OOA in all three periods. Biogenic SOA (BSOA) comprises around one third of OOA during May and around 40% during the other two periods. In each one of the simulation periods, the spatial pattern of concentrations for OPOA, ASOA and BSOA is quite similar, while the fresh POA pattern is different indicating differences in sources and production mechanisms. Inferences regarding the nature of LV-OOA and SV-OOA measured components and their representation in the model will be discussed.

This work was funded by the European Community's 7th Framework Programme EU project PEGASOS (contract 265307).

Fountoukis, C., et al. (2011) Evaluation of a three-dimensional chemical transport model (PMCAMx) in the European domain during the EUCAARI May 2008 campaign, *Atmos. Chem. Phys.*, 11, 10331–10347.