6.03 AEROSOL MODELLING WITH CAMX4 AND PMCAMX

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INTRODUCTION

In spite of several studies published on aerosols in recent years, there still exists significant lack of knowledge of the relevant processes, especially in the formation of secondary organic aerosols (SOA). Models that simulate the transport and transformation of aerosols and gases can contribute to improve the understanding of these processes. In recent years several air quality models have been upgraded to include aerosol dynamical processes such as coagulation, nucleation, evaporation and condensation [*Ackermann et al.,* 1998, *Schell et al.,* 2001, *Griffin et al.,* 2003, *Zhang et al.,* 2004]. However, a model intercomparison study showed that a more complex model approach to the aerosol problem does not automatically lead to better results in a 3-dimensional application (*Hass et al.,* 2003). In this study, two air quality models with different complexity were applied to two domains using the same input data to investigate the capabilities, advantages and disadvantages.

MODELS

The first model is the latest version of CAMx (Comprehensive Air Quality Model with extensions) which is called CAMx4 (*Environ,* 2003a). It has a 1-atmosphere approach for gaseous and particulate air pollution modelling. Aqueous sulphate and nitrate formation in cloud water is calculated using RADM aqueous chemistry algorithm (*Chang et al.,* 1987). Partitioning of condensable organic gases to secondary organic aerosols (SOA) to form a condensed organic solution phase is performed by the semi-volatile equilibrium scheme called SOAP (*Strader et al.,* 1998). There are 4 SOA classes depending on the gaseous precursors. One class (SOA4) represents the SOA formed from the biogenic precursors, the other three are anthropogenic (SOA1 and SOA2 are produced from TOL and XYL with different aerosol yields, SOA3 is produced from PAR, OLE and CRES). ISORROPIA thermodynamic module is used to calculate the partitioning of inorganic aerosol constituents between gas and particle phases (*Nenes et al.,* 1998). Particle sizes are static. Primary particles are modelled as fine and/or coarse particles whereas secondary species are modelled as fine particles.

The second model used in this study is PMCAMx which is still under development and testing (*Environ,* 2003b). PMCAMx contains so-called full science aerosol algorithms. It is a more complete aerosol model than CAMx4, but it is more demanding as well. Aerosol dynamics and particle size distribution with a sectional approach are treated. There are 10 fixed size sections. PMCAMx contains a complete aqueous chemistry mechanism (Variable Size Resolution Model) developed by the Carnegie Mellon University. The two models have similar gas phase (CBMIV/SAPRC99), inorganic (ISORROPIA) and organic (SOAP) aerosol treatment. The main differences include the particle size distribution and aerosol dynamics (see Table 1). The aqueous chemistry mechanisms are also different.

Table 1. Comparison of CAMx4 and PMCAMx models

	CAMx4	**PMCAMx**
approach	1-atmosphere	full-science PM model
gas-phase mechanism	CBMIV/SAPRC99	CBMIV/SAPRC99
inorganic aerosol module	ISORROPIA	ISORROPIA
organic aerosol module	SOAP	SOAP
aqueous chemistry	RADM	VSRM
particle size	fine/coarse	10-sectional
cpu time for appl.1 (dec)	18273	38982
cpu time for appl.2 (linux)	6560	20302

Application1: The first study was performed over a domain covering northern Italy, for the period 12-13 May 1998. The horizontal resolution was 3 km x 3 km and there were 8 layers up to about 3000 m agl. The meteorological fields for this period were calculated by the SAIMM meteorological model (*SAI,* 1995). The same input data from the modelling study using the previous CAMx version 3.10 (*Andreani-Aksoyoglu et al.,* 2004) were used for this application.

Application 2: The second study was carried out in a domain covering Switzerland and some parts of the neighbouring countries for the period 4-7 August 2003. The model domain encloses 650 km in the west-east direction and 450 km in the north-south direction with a horizontal resolution of 9 km. In the vertical direction 10 layers with varying heights were used, lowest being 30 m above ground. The model top is at about 4000 m above ground. For this application, meteorological data were calculated using MM5 meteorological model (PSU/NCAR, 2004). Initial and boundary conditions were extracted from the output of the European model REM3, provided by the Free University of Berlin. The emission inventory of gaseous species for Europe was based on an inventory provided by the Free University of Berlin. The emissions in Switzerland were compiled from various sources. The reference year is 2000.

In both studies, CAMx4 and PMCAMx were used without nesting. There were no primary particle emissions in the emission inventories. CAMx4 results refer to PM2.5 whereas PMCAMx calculates the aerosol concentrations for each of the 10 size bins between 0.04 and 40 μm . The sum of the first 6 size bins in PMCAMx which corresponds to PM2.5, was compared with CAMx4 results.

RESULTS AND DISCUSSION

Application 1: Comparisons of two model results with the measurements of secondary inorganic aerosols performed in Verzago, 35 km north of Milano, shows that the diurnal variations of the models are similar (Figure 1). In general, PMCAMx predicted lower concentrations than CAMx4 and CAMx4 results are closer to the observations. Since both models use the same inorganic aerosol module ISORROPIA and the same gas-phase mechanism, similar model results are expected. However, it should be kept in mind that the gas-phase chemistry of PMCAMx is based on an earlier version (CAMx 3.01) and it will be updated to the latest version in the future (Environ, 2003b). Improvements in the latest gas-phase mechanism in CAMx4 may lead to differences in the aerosol concentrations calculated by the two models. Another difference is in the aqueous chemistry mechanism. VSRM used in PMCAMx takes into account the size-dependent differences in pH.

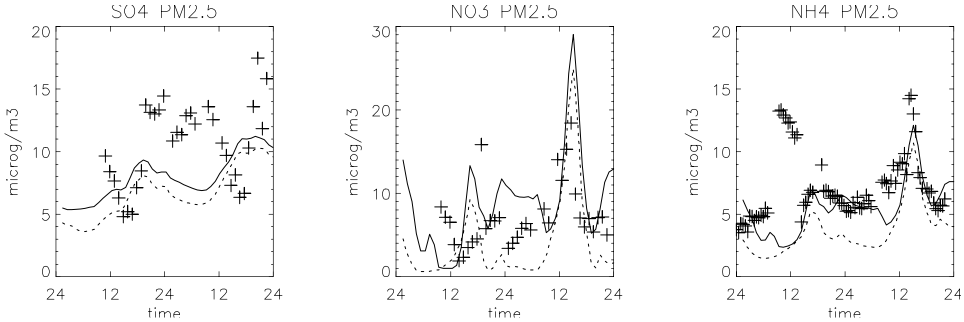

Figure 1. Diurnal variation of measured (+) and predicted concentrations (µg/m^3) by CAMx4 (solid line) and PMCAMx (dotted line) of particulate SO_4^{2-}, NO_3^- and NH_4^+ in Verzago.

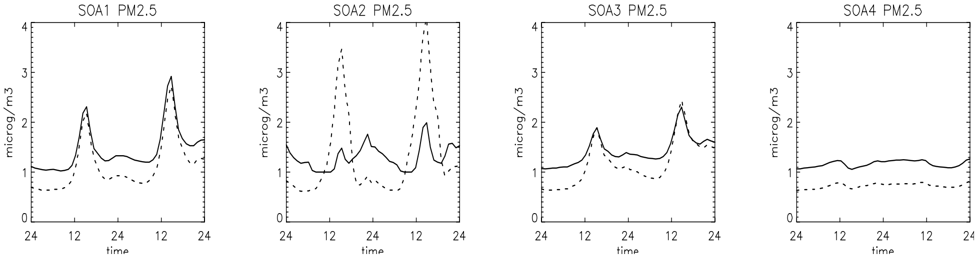

Figure 2. Diurnal variation of predicted concentrations (µg/m^3) of 4 SOA species by CAMx4 (solid line) and PMCAMx (dotted line) in Verzago.

Both models have 4 secondary organic aerosol classes. SOA4 refers to the aerosols produced from the biogenic precursor gases whereas SOA1, SOA2 and SOA3 have anthropogenic origin. In Verzago, biogenic SOA is about 20% of the total predicted SOA. PMCAMx predicts in general lower levels for all SOA classes (Figure 2). Afternoon levels of SOA1 and SOA3 are the same for both models. On the other hand, model results of SOA2, which is the aerosol product of toluene and xylene, with aerosol yield higher than SOA1, look quite different. Since the same aerosol parameters are used in both models, discrepancies are either due to differences in CBMIV versions or aerosol dynamical processes in PMCAMx. The calculated secondary organic aerosol concentrations are difficult to validate because there are no direct measurements. Estimations based on black carbon and total organic carbon measurements yielded an SOA range of 4 –5 µg/m^3 for daily average (*Andreani-Aksoyoglu et al.*, 2004). The average of calculated SOA values in Verzago are 5.4 for CAMx4 and 4.4 for PMCAMx.

Application 2: The two models over the Swiss domain are compared only with each other because of lack of measurements of aerosol species (Figure 3). Secondary aerosol concentrations are much lower in Switzerland than in northern Italy except biogenic SOA. The biogenic contribution to SOA which is more than 80% is therefore quite different from northern Italy. There are significant differences between the inorganic aerosol concentrations calculated by the models. PMCAMx results need to be analyzed further. On the other hand, SOA formation in both models is similar (Figure 3). Higher levels of SOA3 predicted by PMCAMx are probably due to the difference in gaseous precursor concentrations**.**

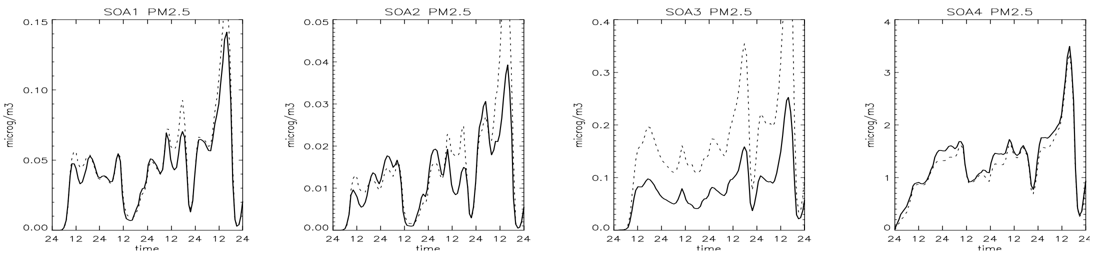

Figure 3. Diurnal variation of predicted concentrations(μg/m3) of SOA by CAMx4 (solid line) and PMCAMx (dotted line) during 4-7 August 2003 in Tänikon.

CONCLUSIONS

On the basis of two applications, the results of two models with different complexity, CAMx4 and PMCAMx, are comparable. Discrepancies are most probably due to different aqueous chemistry and aerosol dynamics. There can also be differences in the two versions of gas-phase mechanisms. In the first application in northern Italy, CAMx4 predictions for inorganic aerosols match observations better than PMCAMx, in spite of lesser complexity. Computer time required by PMCAMx is about 2-3 times higher than by CAMx4. Although PMCAMx has the capability of calculating the aerosol species in various size sections, as long as there are no measurements of speciated aerosols with high resolution of time and space, it is difficult to validate the results. Most of the particle measurements in Europe consists of PM10 only. For regulatory purposes and control strategies, CAMx4 is already good enough at the moment. It is believed that it will be really worth using more complex models such as PMCAMx when aerosol species with smaller size (not only PM10) are measured with higher resolution in time and space. It needs to be mentioned here that the concept of equilibrium of the partitioning between gas phase and aerosol organics is not valid anymore considering newest publications by Jang et al. (2002) and Kalberer et al. (2004). Due to polymerization reactions in the aerosol, more organics can partition into the aerosols compared to prediction of equilibrium models.

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