

# An Aerosol Modelling Study of Winter and Summer Periods in Switzerland

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In this paper, we discuss the application of the MM5/CAMx model system under winter and summer conditions in 2006 over the complex terrain of Switzerland. The focus of the study is on the formation and transport of particulate matter (PM). Both measurements and model results indicate that the main components of the winter aerosols are particulate nitrate and organic aerosols. The meteorological model seems to predict the meteorological parameters reasonably well except during periods with low-wind speed. The results suggest that organic aerosols are the most dominant species in summer and they are mostly secondary. In general, CAMx predicts the concentrations of inorganic aerosols correctly while organic aerosols are underestimated. Possible causes for the underestimation might be the lack of wood-burning emissions and missing processes leading to secondary organic aerosols like oligomerization which are not yet treated by the model.

## 1. Introduction

Quantitative measurements of aerosols in Switzerland indicate that organic aerosols are the major components of the aerosol composition both in summer and winter [Lanz, *et al.*, 2007; Lanz, *et al.*, 2008]. Additional measurements using  $^{14}\text{C}$  as tracer show that biogenic emissions in Zurich contribute about 60 and 27 % to organic carbon (OC) in summer and winter, respectively [Szidat, *et al.*, 2006]. Recent measurements suggest that wood-burning emissions might also contribute significantly to the aerosol concentrations, especially in winter. Understanding the partitioning behaviour of semi-volatile species between the gas and aerosol phases can help us to predict how changes in anthropogenic and biogenic activity will influence the formation of aerosols in the atmosphere. Modelling secondary organic aerosol (SOA) formation is among the most demanding aspects associated with atmospheric organic photo-oxidation. The current models often underestimate SOA concentrations [Cousin, *et al.*, 2005; Volkamer, *et al.*, 2006; Zhang, *et al.*, 2004]. In Switzerland which has a very complex terrain, there are very few model studies on aerosols [Andreani-Aksoyoglu, *et al.*, 2003; Andreani-Aksoyoglu, *et al.*, 2008]. Recent experimental evidence for oligomerization reactions in organic aerosols indicated the need to readdress the current assumptions in models about the partitioning of oxidation products in the gas and the particle phase [Dommen, *et al.*, 2006; Kalberer, *et al.*, 2004]. A recent model study by [Morris, *et al.*, 2006] showed that including mechanisms such as polymerization, SOA formation from isoprene and sesquiterpenes, led to increased SOA yields.

## 2. Methodology

In this study, the 3-dimensional photochemical model CAMx (Comprehensive Air Quality Model with Extensions, version 4.40) was applied with 3 nested domains as shown in Fig. 1 [Environ, 2006]. The coordinate system was Lambert Conic Conformal. The resolutions of the three domains were 27, 9 and 3 km, respectively with 14  $\sigma$ -layers in a terrain-following coordinate system. The model top was set at  $\sigma = 0.55$ , which corresponds to a geometric layer top at sea level of about 7000 m. The simulations started always at 0000 UTC on the first day and ended at 2400 UTC on the last day of the months January and June in 2006. There are detailed aerosol mass spectrometer (AMS) measurements available in Zurich during January 2006 and in Payerne in June 2006. We apply the meso-scale model MM5 [PSU/NCAR, 2004] as meteorological driver for the air quality model CAMx. MM5 was initialized by assimilated data of the COSMO-7 forecast model of MeteoSwiss. The four-dimensional meteorological data assimilation was conducted using balloon soundings and COSMO-7 upper level data. The emission inventory was prepared by compiling European and Swiss anthropogenic emissions from various data sources, as described in [Keller, et al., 2008]. Emissions of PM<sub>2.5</sub> and PM<sub>10</sub> of 9 source categories were obtained from INFRAS and Meteotest for the reference year 2000. Using land use and meteorological data, biogenic emissions were calculated by means of temperature and irradiance dependent algorithms [Andreani-Aksoyoglu and Keller, 1995]. In the biogenic emission inventory, the most abundant species are monoterpenes, which are emitted mainly by Norway Spruce and fir trees. Less abundant is isoprene, emitted by oak trees and pasture, mainly in the southern part of Switzerland. Initial and boundary conditions were obtained from the output of similar periods generated by the global model MOZART [Horowitz, et al., 2003]. In this study, the CBM-IV mechanism with the extensions for aerosol modelling was used [Gery, et al., 1989]. Calculations of aerosols with  $d < 2.5 \mu\text{m}$  were performed using the fine/coarse option of the aerosol module.

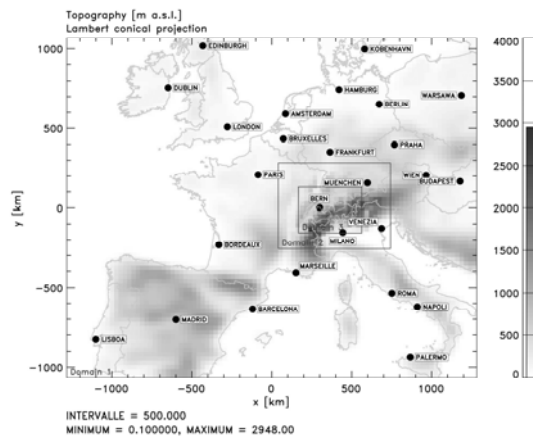


Figure 1: Topography of 3 model domains with 27, 9 and 3 km resolutions, respectively.

### 3. Results and Discussions

#### 3.1 January 2006

Wind speed, precipitation, temperature as well as the measured and modelled CO mixing ratio in Zurich are shown in Fig. 2. The period can be divided into four sections: I) 1-6 January: temperatures above zero and moderate wind speed. II) 6-17 January: colder and foggy, temperatures below zero and low wind speed. III) 17-23 January: some precipitation, temperatures above zero and higher wind speed. IV) 23-31 January: mixed conditions with variable wind speed and temperatures. The comparison of measured and modelled CO concentrations helps to understand the performance of the meteorological model. The agreement between model and measurements is better during the first and the third period when the wind speed was higher. On the other hand, the model underestimates the CO concentrations under low wind conditions (period II). A similar behaviour was observed for the inorganic and organic aerosols (Fig. 3). In addition to the insufficient performance of the meteorological model during the second period, there might be also other reasons causing underestimation such as lack of wood burning emissions, and/or oligomerization mechanism for SOA formation. Both measurements and model results suggest that the main components of the winter aerosols in Zurich are particulate nitrate and organic aerosols (Fig. 4).

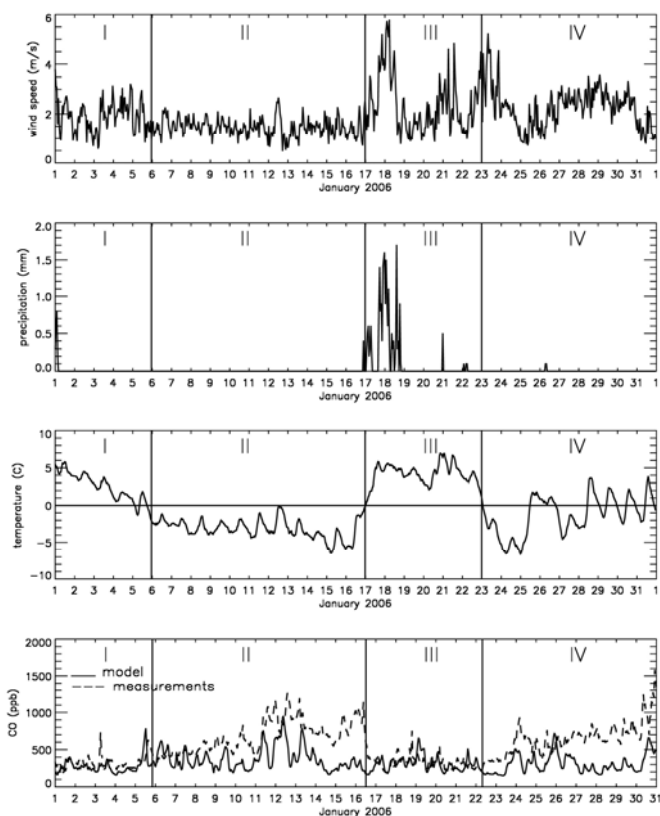


Figure 2: Measured wind speed (m/s), precipitation (mm) and temperature (C) as well as the comparison of measured and modelled CO concentrations (ppb) in Zurich (NABEL station) during January 2006.

According to model predictions (not shown), primary organic aerosols (POA) dominate the organic aerosol composition (87%). Secondary organic aerosols (SOA) on the other hand, were predicted to be mainly biogenic. The fraction of modelled elemental carbon (EC) is higher than the measured one.

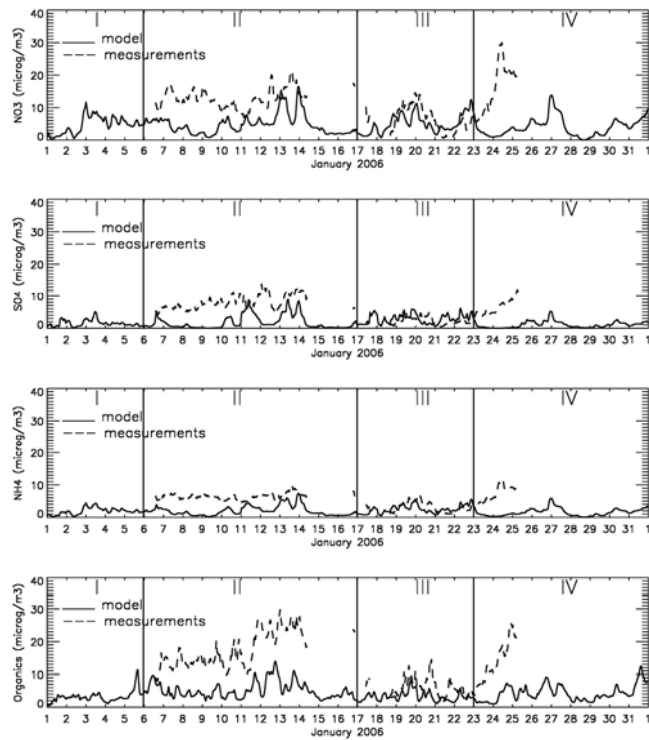


Figure 3: Comparison of modelled ( $PM_{2.5}$ ) and measured ( $PM_{1}$ ) (AMS) inorganic and organic aerosols ( $\mu\text{g}/\text{m}^3$ ) in Zurich during January 2006.

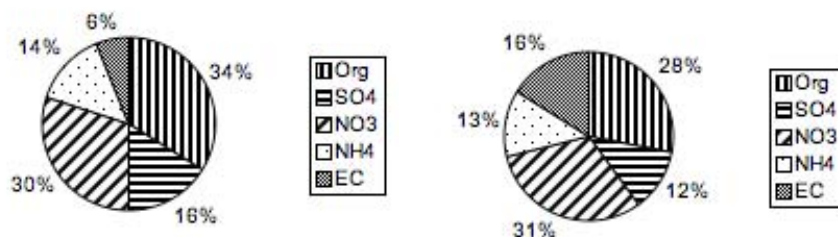


Figure 4: Fractional composition of winter aerosols in Zurich from measurements ( $PM_{1}$ ) (left) and model simulations ( $PM_{2.5}$ ) (right).

### 3.2 June 2006

The measured meteorological parameters during the simulated period June 2006 in Payerne are shown in Fig. 5. The temperature increased during the first half of the month. There was a moderate to strong wind until 12<sup>th</sup> of June. During that period, measured and modelled CO concentrations agree reasonably well with each other (see Fig. 5). The wind speed decreased during the next 5 days, and model-measurement agreement became worse. There was some precipitation during the second half of June and wind speed was variable. This figure suggests that the performance of MM5 was reasonably good except the low wind situation between 12 and 17 June. One can see a similar trend in the results of aerosols (Fig. 6). The model underestimates both inorganic and organic aerosol concentrations during the low wind period (12-17 June). Model predictions for the first 12 days seem to be good for sulphate and ammonium during the first week, but higher for particulate nitrate that needs to be investigated. On the other hand, the results look different for the organic aerosols. The AMS data match model results during the first week, but later they increase and become about 3 times higher than the modelled quantities during 12-17 June. In addition to the problems of meteorological model for the low-wind period, some other causes might lead to underestimation. The SOA formation from other sources (sesquiterpenes) and oligomerization are likely to be some of the reasons.

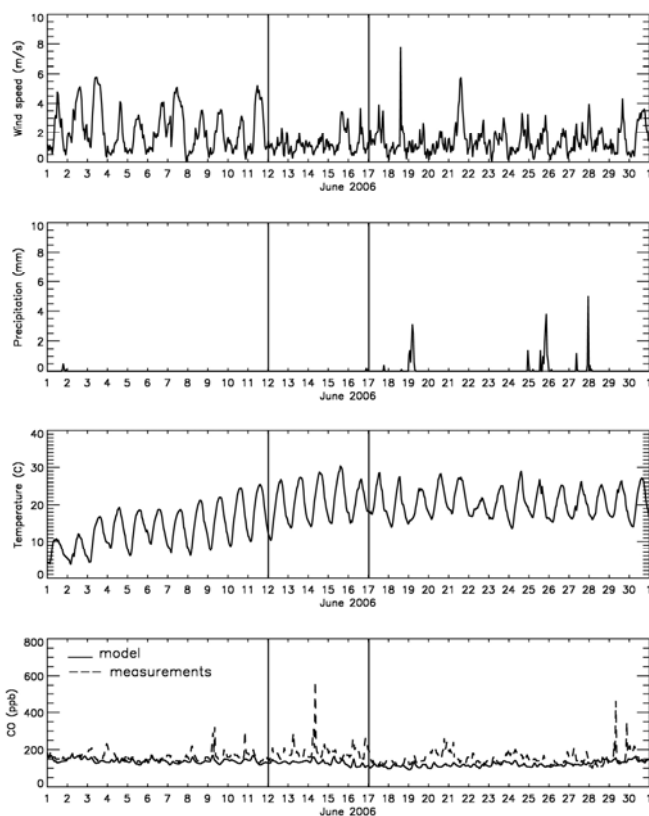


Figure 5: Measured wind speed (m/s), precipitation (mm) and temperature (C) as well as the comparison of measured and modelled CO concentrations (ppb) in Payerne (NABEL station) during June 2006.

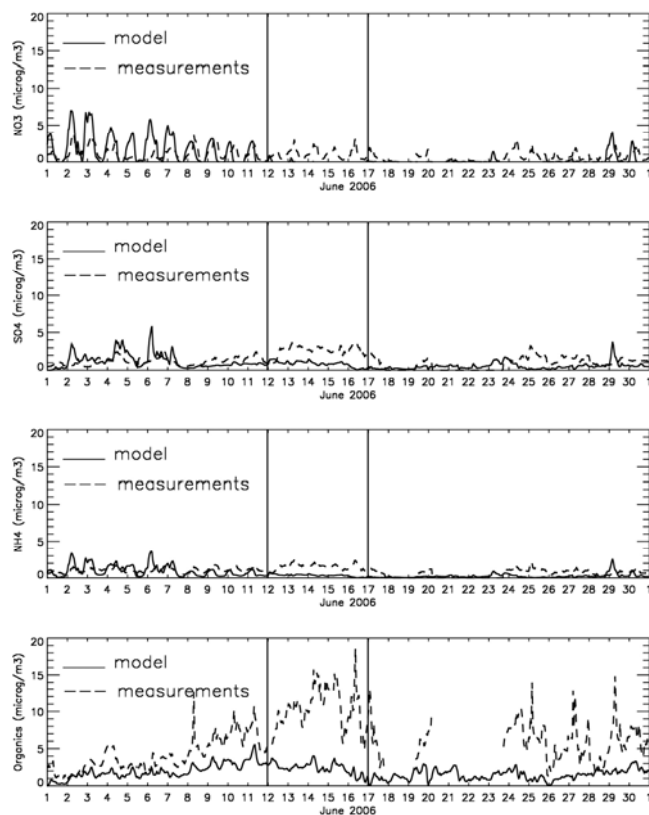


Figure 6: Comparison of modelled ( $PM_{2.5}$ ) and measured ( $PM_{1}$ ) inorganic and organic aerosols ( $\mu\text{g}/\text{m}^3$ ) in Payerne during June 2006.

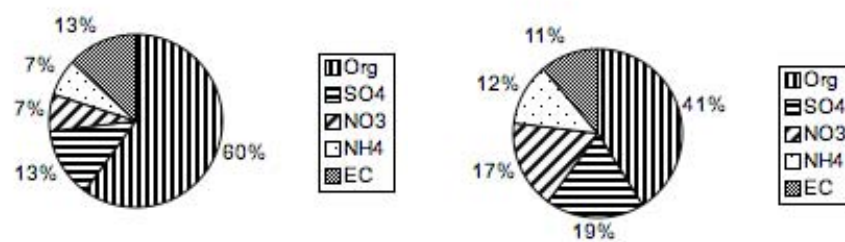


Figure 7: Fractional composition of summer aerosols in Payerne from measurements ( $PM_{1}$ ) (left) and model simulations ( $PM_{2.5}$ ) (right).

As seen in Fig. 7, organic aerosol fraction is underestimated while particulate nitrate is overestimated. SOA dominates the organic fraction of aerosols by 60% and it is mainly

biogenic (not shown). This is in agreement with  $^{14}\text{C}$  measurements carried out in northern Switzerland [Szidat, Jenk, Synal, Kalberer, Wacker, Hajdas, Kasper-Giebl and Baltensperger, 2006].

#### 4. Conclusions

In this study we investigate the formation and distribution of particles (PM<sub>2.5</sub>) in Switzerland in winter and summer 2006 by means of the air quality model CAMx. The results suggest that the meteorological model MM5 can predict the meteorological parameters reasonably well during the studied periods except during the low-wind situations. Both measurements and model simulations show that the main components of winter aerosols are particulate nitrate and organic aerosols. The fraction of POA is higher than SOA in Zurich. In summer, organic aerosols dominate the aerosol composition and they are mainly secondary. The comparison of model results with the data from the aerosol mass spectrometer (AMS) measured at Payerne during June 2006 shows an underestimation of organic aerosols. The model results suggest that secondary organic aerosols formed from the biogenic precursors (monoterpenes) can be more important than the SOA produced from the anthropogenic precursors both in winter and summer, although the absolute concentrations are much higher in summer. We are going to continue the following projects in the near future: The improvement of the meteorological modelling under low-wind conditions, implementation of wood-burning and sesquiterpene emissions, and simulations with a new SOA module including the polymerization mechanism for SOA.

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