MODELLING OF AIR QUALITY WITH CAMx: A CASE STUDY IN SWITZERLAND

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(Received July 2002; accepted 15 December 2002)

Abstract. The three-dimensional Eulerian model CAMx (*C*omprehensive *A*ir Quality *M*odel with E*x*tensions) was applied for the first time to simulate both gaseous and particulate photochemical air pollution in Switzerland during July 28–30, 1993. The meteorological input data were prepared using the Systems Applications International Mesoscale Model (SAIMM). The CAMx model results were compared with the measurements carried out at ground level and from airborne measuring platforms within the frame of the Swiss POLLUMET research programme. In general, the CAMx performance for gaseous species was found to be better than that of the previously used Urban Airshed Model (UAM) and the Variable Grid Urban Airshed Model (UAM-V). The most significant improvement for the gaseous species is in the prediction of HNO_3 concentrations, due to the inclusion of aerosol chemistry. Aerosol species such as NO_3^-, NH_4^+, SO_4^{2-}, and secondary organic aerosols were calculated in one particle size range (0.04–2.5 μm) and compared with a few measurements available. Although July 29 was reasonably well simulated, overestimated wind speeds by SAIMM for July 30 caused a too fast transport of pollutants. Similarly to measurements, significant spatial correlation of the secondarily formed aerosols with ozone and formaldehyde is found in the afternoon.

Keywords: aerosols, CAMx, photochemical modelling, Pollumet

1. Introduction

Ozone is a secondary air pollutant causing damage on plants and human health and is produced by chemical reactions of nitrogen oxides (NO_x) and volatile organic compounds (VOC) in the presence of sunlight. Since ozone formation is a highly non-linear and complex process, simple reduction of its precursor emissions does not necessarily provide lower ozone concentrations (Seinfeld and Pandis, 1998). The correlation between the emission rates of species released into the atmosphere and the resulting concentrations and load patterns due to transport, dispersion, chemical transformation and deposition can be obtained by chemical dispersion models. Model calculations require a reliable dataset of measured parameters for simulating various processes in the atmosphere and for validating the results.

In this study, the air quality in Switzerland was simulated by the 3-dimensional photochemical model CAMx, and the results were evaluated using the measurements performed over the Swiss Plateau in July 1993 within the pollution and met-

Water, Air, and Soil Pollution: Focus **3:** 281–296, 2003.

eorology (POLLUMET) programme (Neininger and Dommen, 1996). So far there have been only a few 3-dimensional modelling studies about the Pollumet period, and they all included only gas phase chemistry (Kuebler *et al*., 1996, Perego, 1999, Andreani-Aksoyoglu *et al*., 2001). In this work, we performed the calculations with the CAMx model (ENVIRON, 2000) for the first time including both trace gases and aerosol chemistry.

2. Measurements

The POLLUMET research programme focused on the production, transport and distribution of ozone and other photo-oxidants over the Swiss Plateau and the neighbouring Alpine valleys. The Pollumet Observation Period (POP) covered the period from July 1 to August 31, 1993 with a special observation period (SOP) on July 29 and 30. During this field experiment extensive readings of air pollutants not routinely measured, were taken, for example H_2O_2, HNO_3, NH_3, H_2CO, NO_y, PAN, hydrocarbons, particulate NO_3^- and NH_4^+ at ground level and/or from airborne measuring platforms. Details of the measurements are given by Neininger and Dommen (1996).

3. Modelling Systems

3.1. Meteorological model

The meteorological data needed as input for the photochemical model, were calculated with the SAI Mesoscale Model (SAIMM) which is based on the hydrostatic model MM2 (SAI, 1995). The basic meteorological quantities required by SAIMM are time and space dependent soundings and surface measurements of pressure, temperature, relative humidity, wind speed and direction. The model uses geometric terrain following coordinates. The model domain is 470 $\times$ 385 km with 5 $\times$ 5 km horizontal resolution. There are 19 layers up to 9000 m above sea level. The bottom of the lowest level is the topographic surface and the top of the uppermost level is at constant height above sea level. Hence, the thickness of a given vertical layer generally varies within the domain and is smaller for higher topographic altitudes. Furthermore, the model is driven by data of the Swiss Model (SM) of the Swiss Meteorological Institute, MeteoSwiss. The simulated meteorological quantities are 'nudged' towards the experimental data and the SM fields to obtain a better agreement. Details of the SAIMM application were described in Keller *et al*. (2002).

3.2. PHOTOCHEMICAL MODEL

The Comprehensive Air Quality Model with extensions (CAMx) is an Eulerian photochemical grid model that allows for integrated assessment of gaseous and particulate air pollution over many scales ranging from urban to super-regional (ENVIRON, 2000). CAMx simulates the dispersion, chemical reactions, and removal of pollutants in the lower troposphere by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids.

CAMx contains both CBM-IV (Gery *et al.*, 1989) and SAPRC99 (Carter, 1996) mechanisms. The updated CBM-IV chemical mechanism, which was used in this study, has been recently modified to include aerosol chemistry (ENVIRON, 2000). In addition to the original version, the updated CBM-IV contains a second olefin species to account for biogenic olefins and a condensible organic gas species. The CAMx aerosol chemistry routine calculates the transformations of SO_2 to sulfate via aqueous reactions, condensible organic carbon to aerosol organic carbon, gaseous HNO_3 to aerosol nitrate, gaseous NH_3 to aerosol ammonium and sodium nitrate formation. There is no particle size distribution modelling in the latest model version yet, however one can choose a representative size range for each aerosol species. In this study, the particle size range for these secondary aerosol species has been chosen as 0.04–2.5 μm, as indicated by experimental results (see e.g. Hüglin 2000, Baltensperger 2001).

The CAMx model was applied to simulate the air quality in Switzerland during the period of July 28–30, 1993. The model domain covers an area which is 370 km in the west-east and 285 km in the north-south direction. The horizontal resolution was 5 $\times$ 5 km, and there were 8 vertical layers with varying heights from 50 to 3000 m above ground. Meteorological input data such as wind fields, humidity, pressure, temperature and vertical exchange coefficients were obtained from the SAIMM output. The anthropogenic emissions were compiled from various data sources and include emissions from traffic, industrial processes, and residential areas (BUWAL, 1995). The biogenic emissions were included in the emission inventory as well (Andreani-Aksoyoglu and Keller, 1995). Boundary conditions and initial conditions of gaseous chemical species were derived both from measurements and previous model runs.

4. Results and Discussion

The first day (July 28) was used to initialize the model, therefore only the results of July 29 and 30 are discussed in this paper. In general, there was a weak westerly wind on July 29. On both days clear skies and high temperatures prevailed, but the wind direction changed in the afternoon of July 30, blowing from south, and there were clouds in some parts of the model domain. The wind fields in the lowest model layer (top at 50 m) simulated by SAIMM at 1700 CEST (central European summer

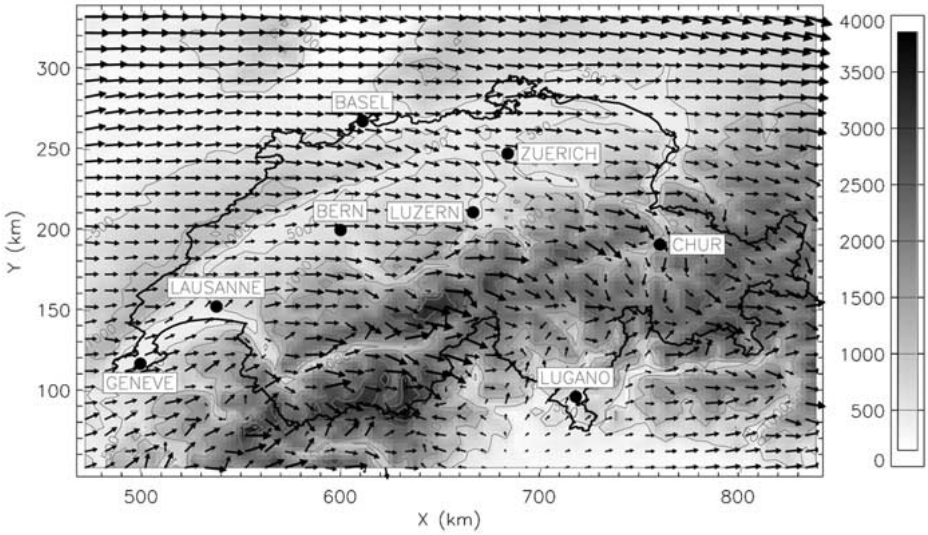

Figure 1. Afternoon (1700 CEST) wind speeds (max of 8 m s^{-1}) at the SAIMM simulation level of 50 m for July 29 over shaded topographic heights (m).

time) are shown in Figures 1 and 2. Airborne wind speed measurements over east of Zurich during the afternoon of July 30 were slightly higher than on July 29 (afternoon averages 3 and 4 m s^{-1} for July 29 and 30, respectively). Some ground stations, however, recorded lower speeds on July 30. The calculated wind direction is in quite good agreement with the measurements for both days. However, wind speeds on July 30 are overestimated by SAIMM in some parts of the model domain. For example, in the region where the aircraft measurements were carried out, wind speeds predicted by the model were about 3 and 5.5 m s^{-1} for July 29 and 30, respectively.

Afternoon (1700 CEST) ozone mixing ratios calculated by CAMx for the lowest model layer (0–50 m) are shown in Figures 3 and 4 for July 29 and 30, respectively. As seen in the figures, ozone levels are somewhat reduced in the urban areas and higher downwind of these regions. Concentrations in the northern part of the domain on July 29 are between 55–65 ppb and they are in the same range as the measurements shown in Dommen *et al*. (1999). The highest mixing ratios up to 85 ppb are predicted in the southern part of the domain. At some ground stations in the south however, higher ozone levels (> 100 ppb) were measured. This discrepancy is most likely due to the emission inventory which needs to be improved around the border regions. The wind blowing from south-west on July 30, leads to increased concentrations in the north-east part of the model domain (Figure 4). Airborne measurements showed concentrations up to 80–85 ppb northeast of Zuerich. On the other hand, the model predicts a maximum of 72 ppb farther downwind, due to the overestimated wind speed on July 30, which caused a faster transport of pollutants.

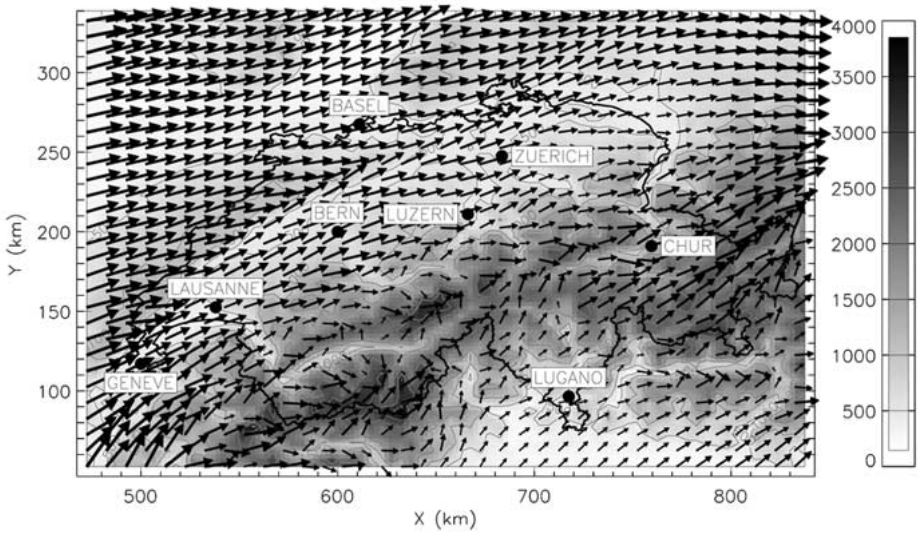

Figure 2. Afternoon (1700 CEST) wind speeds (max of 10 m s^{-1}) at the SAIMM simulation level of 50 m for July 30 over shaded topographic heights (m).

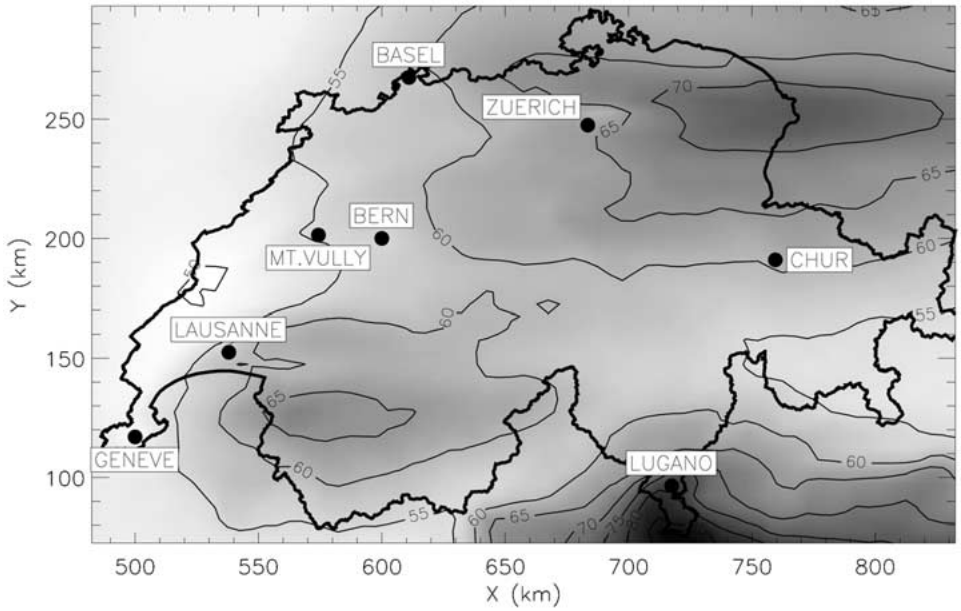

Figure 3. Afternoon (1700 CEST) ozone mixing ratios (ppb) at lowest CAMx simulation level (50 m) for July 29.

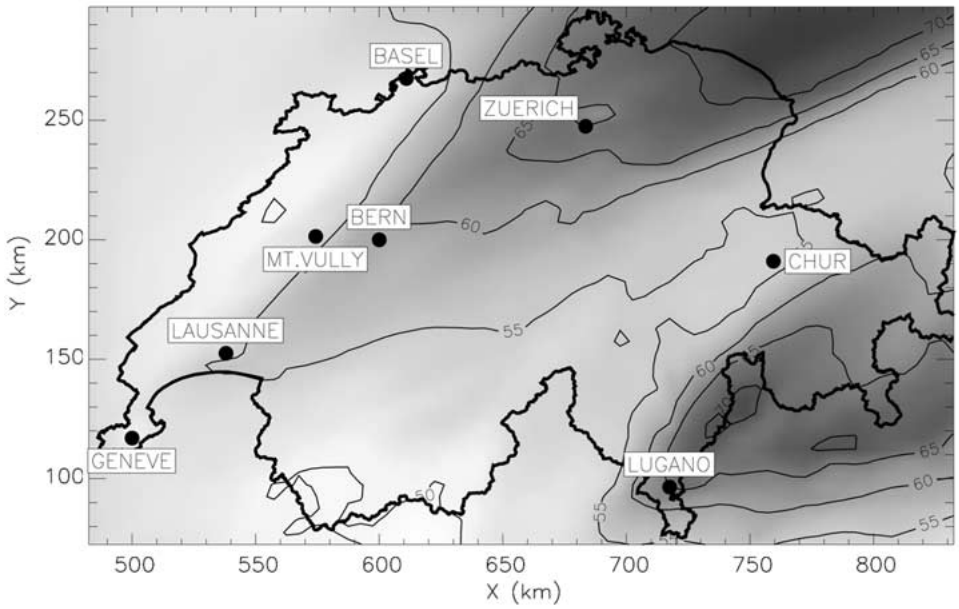

Figure 4. Afternoon (1700 CEST) ozone mixing ratios (ppb) at lowest CAMx simulation level (50 m) for July 30.

The flight path of the NCAR aircraft in the afternoon on July 29 is shown in Figure 5. The path for the second day was similar. The CAMx results from the levels matching the altitude of the aircraft were plotted along the flight path and compared with the measurements on July 29 and 30 (Figures 6 and 7, respectively). The model could reproduce the concentrations and temporal trends of NO_x and HCHO for the first day. On the other hand, ozone concentrations are in general 5–20 ppb higher and H_2O_2 concentrations about 0.5 ppb lower than the measurements. On July 30, formaldehyde and ozone levels were mostly underpredicted. Predicted H_2O_2 concentrations cannot be compared due to the missing measurement data.

The comparison of Ox (O_3 + NO_2) with the measurements is a better way to estimate the model performance than the comparison of ozone concentrations only, because it takes into account the variations in the ozone levels due to reactions with nitrogen oxides. This type of comparison is shown at four different sites in Figure 8. The measurements and predicted concentrations agree with each other reasonably well at rural stations such as Laegern and Chaumont. However, at urban and suburban stations such as Zuerich and Duebendorf, model results are higher than the ground-level measurements on July 29. Overestimation of Ox at night can be caused by underestimation of vertical mixing as well as of the deposition processes.

The model results of the two days were also compared with measurements carried out at a rural station on Mt. Vully (653 msl) in the western part of Switzerland. Figure 9 shows the comparison of calculated concentrations of gaseous species

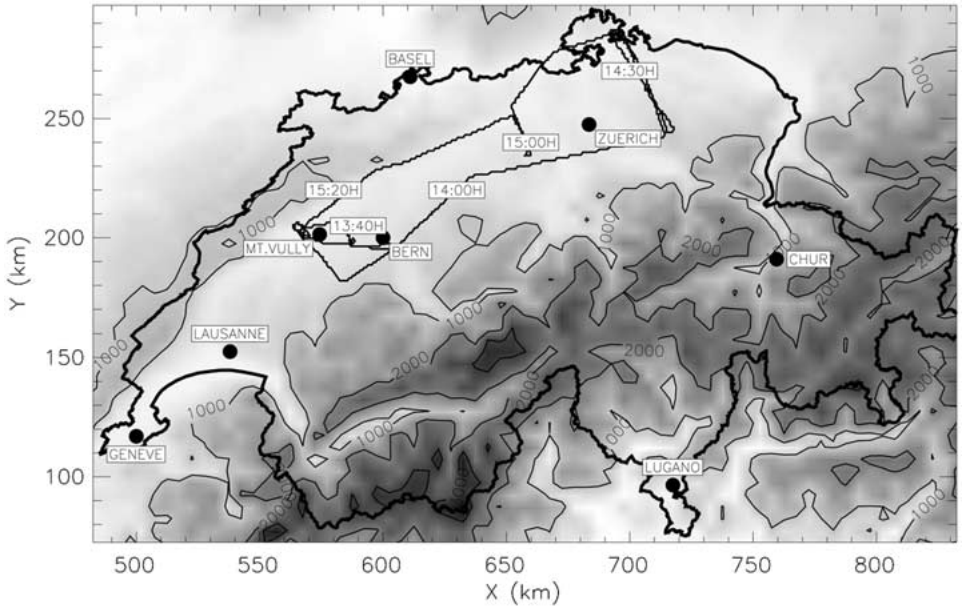

Figure 5. Afternoon (1330–1540 UTC) NCAR flight path for July 29 with shaded topographic heights (m).

such as O_3, H_2O_2, NO_2, HNO_3, PAN, NH_3 and particulate NO_3^-, NH_4^+, SO_4^{2-} and OC (secondary organic carbon) with the measured ones, if available. The predicted afternoon concentrations of ozone and NO_2 are close to the measurements with 10 and 20% differences, respectively. Ozone levels at night are overestimated. Measured H_2O_2 data are higher than predicted because measurements include not only hydrogen peroxide but partly also organic peroxides, while model results refer only to hydrogen peroxide. The agreement between model results and measurements for PAN in the afternoon is within 20% for July 29, but measurements are nearly twice higher on July 30. The calculated concentrations of HNO_3 as well as of particulate NO_3^- agree quite well with the measurements, while sulfate is underestimated. The secondary aerosol concentrations were calculated for the particle size smaller than 2.5 μm. The predicted particle nitrate concentrations at Mt.Vully show the same diurnal behaviour as the measurements, high at night and in the morning and low in the afternoon. Particle NH_4^+ on the other hand, is influenced by the NH_3 concentrations which are underestimated by the model for both days. This is believed to be due to the high uncertainty in the NH_3 emission data. There were only a few measurements of SO_4^{2-} with time resolution 3 to 8 hours during the two days studied (Figure 9). Model values are in general lower than the measurements. They agree with each other better in the afternoon of July 29. For the secondary OC, only the model results are shown, because there were no OC measurements.

The results obtained by the CAMx model seem to be better compared to the previously used models UAM and UAM-V (Andreani-Aksoyoglu and Keller, 1998).

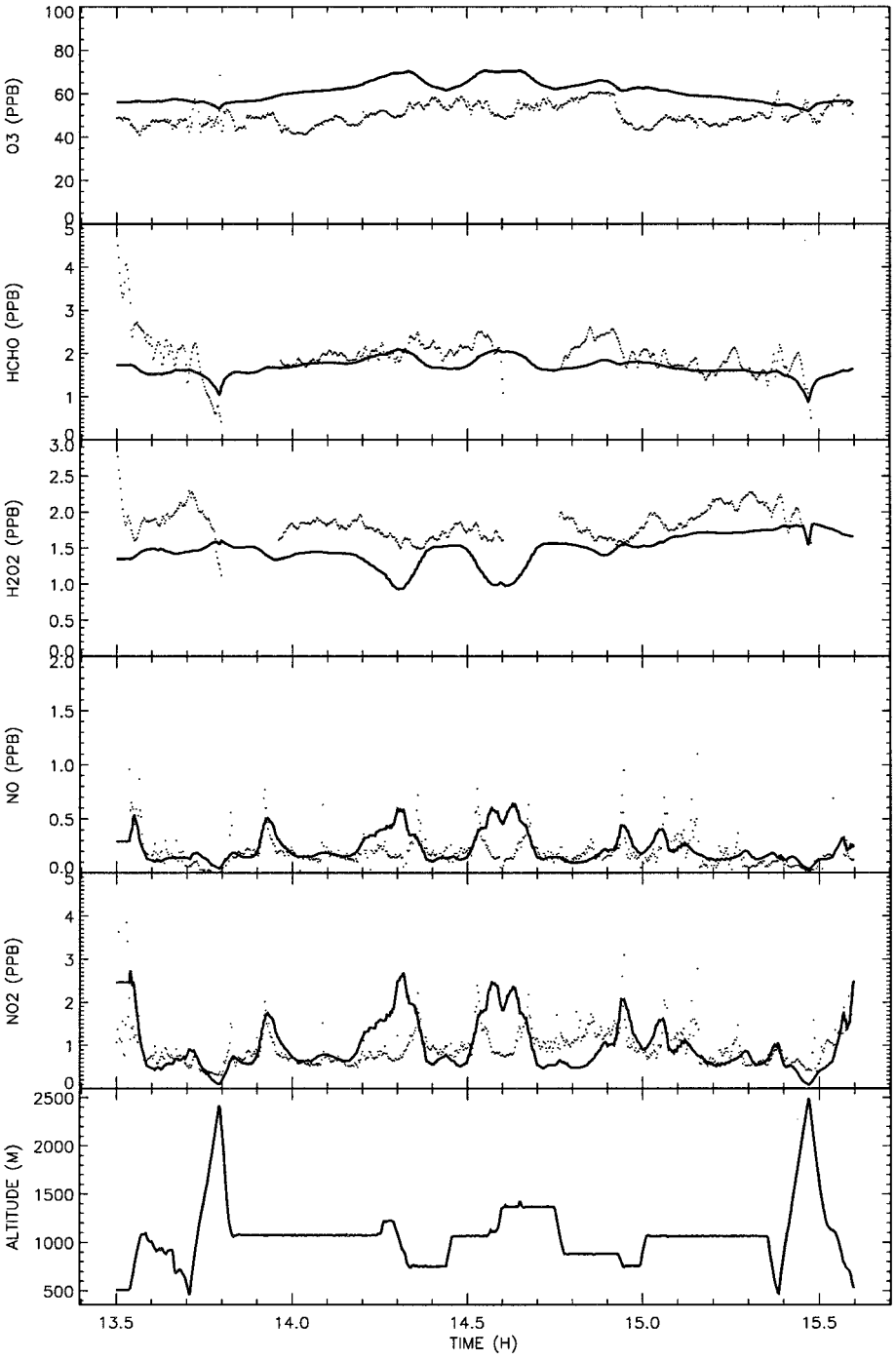

Figure 6. Aircraft (dots) and lowest level (50 m) CAMx model results (solid lines) as a function of decimal hours (UTC) along July 29 flight path (altitude shown) of Figure 5.

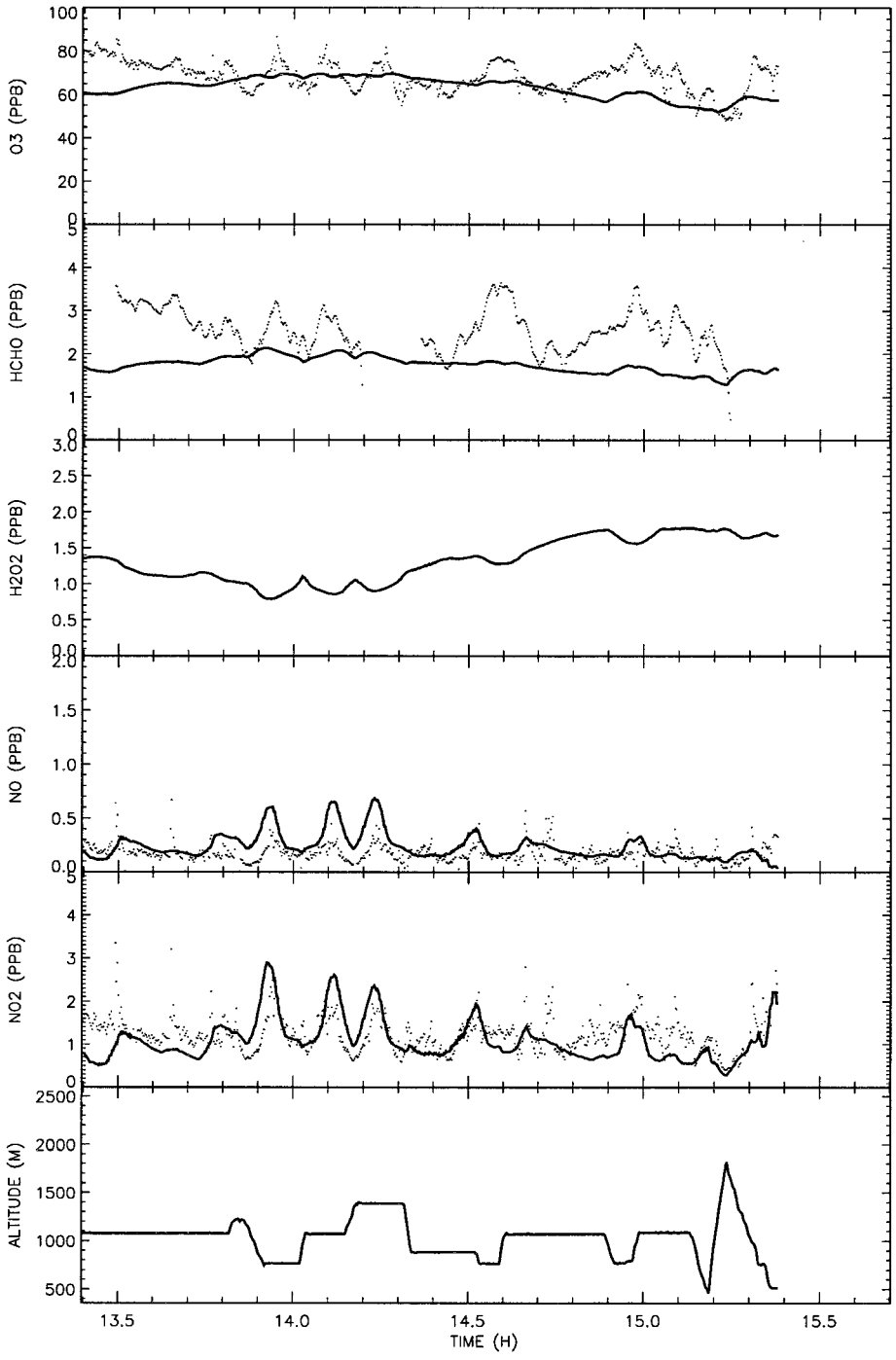

Figure 7. Aircraft (dots) and lowest level (50 m) CAMx model results (solid lines) as a function of decimal hours (UTC) along July 30 flight path (altitude shown).

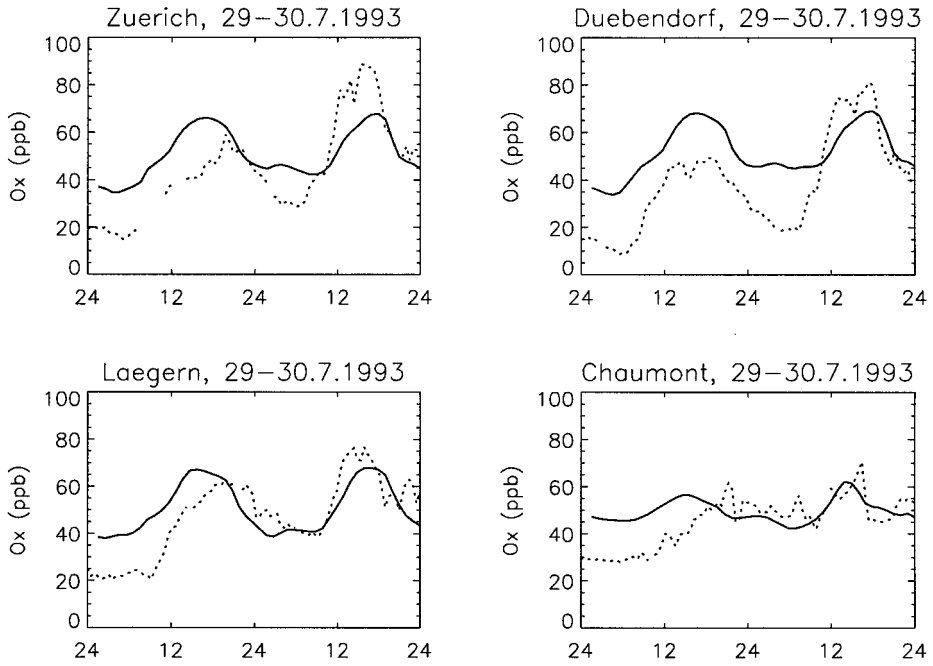

Figure 8. Observations (dots) and lowest level (50 m) CAMx model results (solid lines) of Ox (O_3 + NO_2) for July 29–30 as function of time (CEST) at four ground-level stations.

A comparison of different model results with measurements at Mt. Vully is shown in Figure 10 for HNO_3 and O_3. The same initial and boundary conditions were used in each case. Clearly, UAM shows the weakest performance for both species. Because of the different vertical structure in UAM which was improved in UAM-V, the large difference compared to the other two models is not unexpected. Although the results of UAM-V and CAMx for O_3 are very similar, they show significant differences in the case of HNO_3. CAMx reproduces the measured HNO_3 concentrations much better mainly due to the inclusion of aerosol chemistry.

Airborne measurements of atmospheric particles and trace gases over the Swiss Plateau during the Pollumet period showed a high positive correlation between the accumulation mode particle number concentration (with diameter $d > 0.17$ μm) and the secondary gaseous pollutants such as O_3 and NO_z as well as NO_y and HCHO (Hering *et al.*, 1998). The correlations with the primary pollutants were less pronounced and negative with H_2O_2. Since HCHO was measured in photochemically aged air, it was probably mostly secondarily produced. Hering *et al.* (1998) concluded that under conditions of high pollution and strong photochemical activity the aerosol particles mainly grow by means of secondary processes (heterogeneous nucleation). Similar correlations were also found in the present model results. The correlations of predicted O_3 and HCHO concentrations with

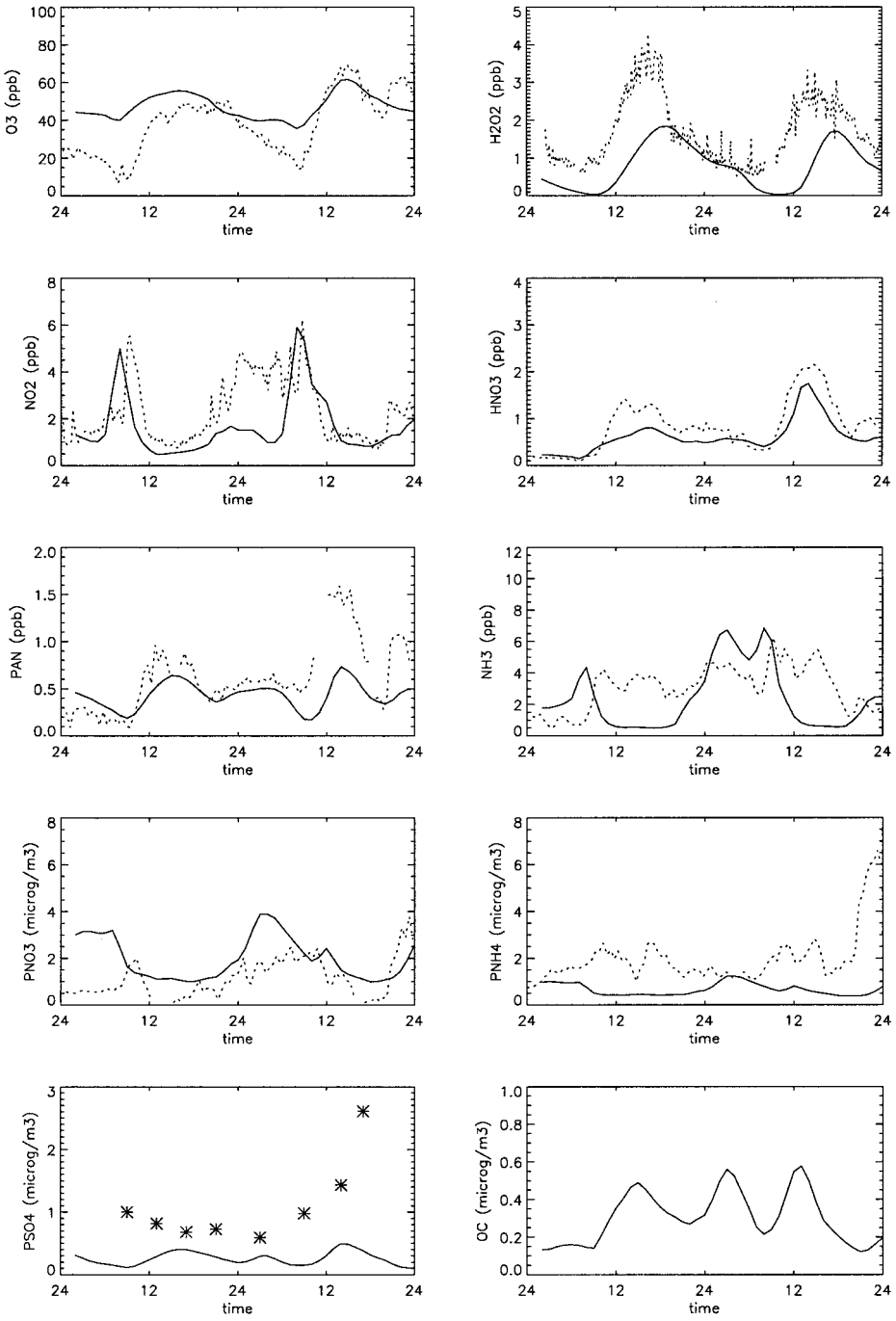

Figure 9. Observations (dots) and lowest level (50 m) CAMx model results (solid lines) for July 29–30 at Mt.Vully as a function of time (CEST).

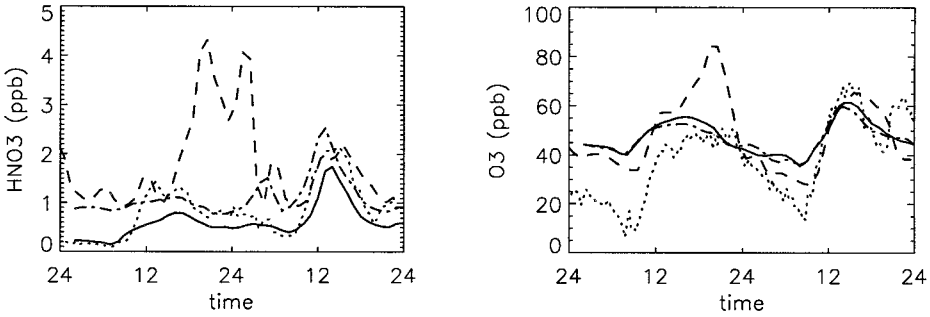

Figure 10. Observations (dots) and lowest level model results for CAMx (solid), UAM (dashes), UAM-V (dash-dots) for July 29–30 at Mt Vully as a function of time (CEST).

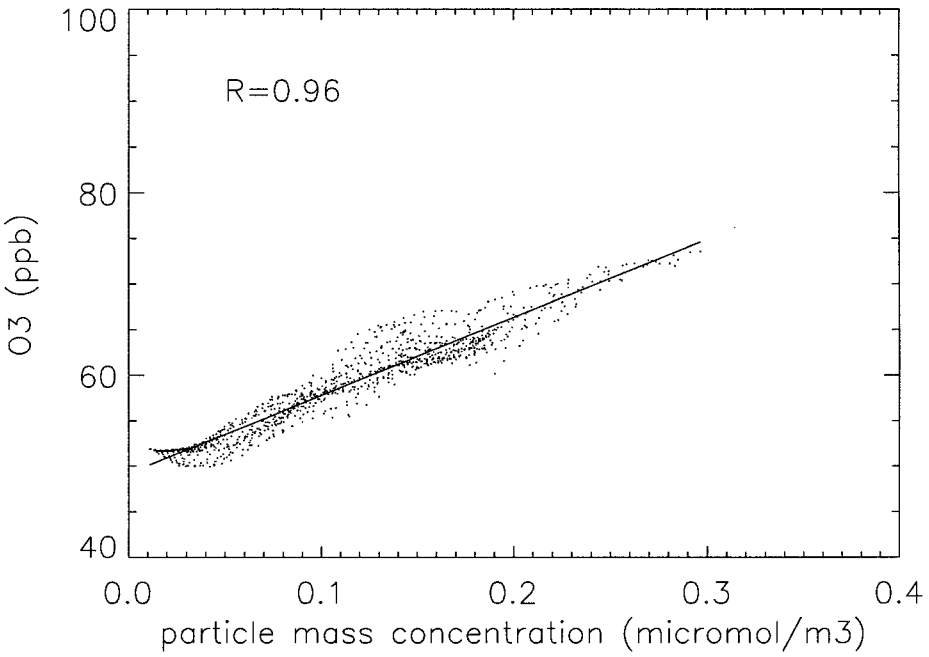

Figure 11. Correlation between lowest level (50 m) CAMx O_3 mixing ratios (ppb) and total secondary particle mass concentration (PNO_3, PSO_4, PNH_4, OC) (μ mole m^{-3}) at 1700 CEST on July 29.

the predicted total secondary particle mass concentration are shown in Figures 11 and 12, respectively. The values refer to afternoon of July 29 over the Swiss Plateau. High positive correlations of particle mass especially with O_3 and also with HCHO, and negative correlation with H_2O_2 (not shown) are in agreement with the observations indicating that a considerable fraction of the aerosol mass might be photochemically produced in parallel to the ozone and formaldehyde formation.

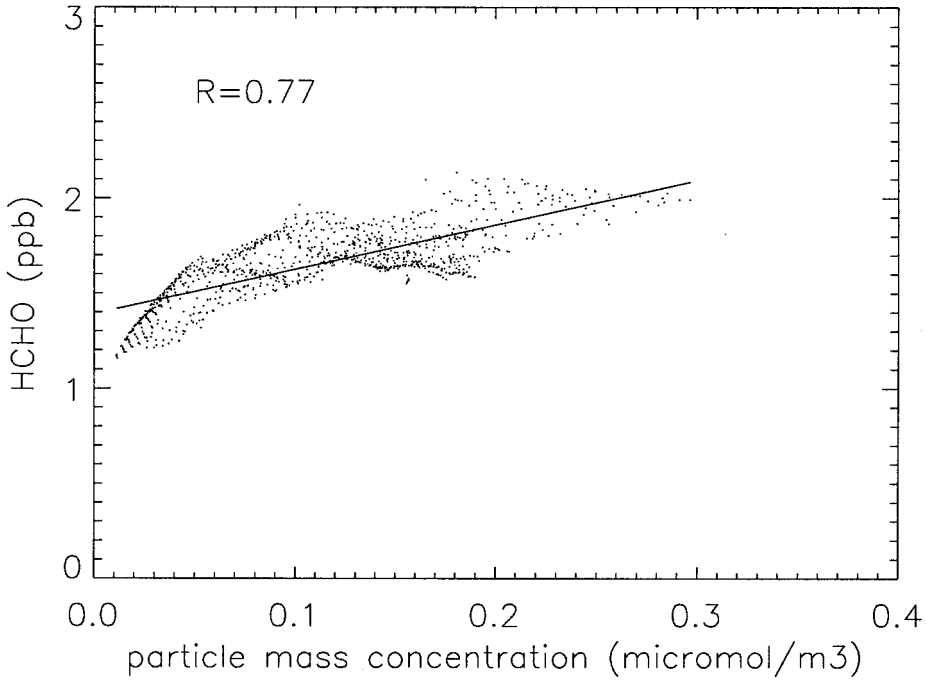

Figure 12. Correlation between lowest level (50 m) CAMx HCHO mixing ratios (ppb) and total secondary particle mass concentration (PNO_3, PSO_4, PNH_4, OC) (μmole m^{-3}) at 1700 CEST on July 29.

Another available data set concerning aerosols are the monthly and annual averages of measurements taken at 4 sites between April 1, 1998 –March 31, 1999 (Hüglin, 2000). These data are shown in Table I, together with the calculated daily averages for July 29 and 30 (see Figure 13 for the diurnal variation of predicted particle concentrations at the same stations as in Table 1). With these data, the model cannot really be validated. The table is shown to give an idea about the first, preliminary results of this study, and the data are not meant to be compared because of the following reasons. First of all, measurements and calculations do not refer to the same year. In addition, measurements are annual and monthly averages and calculations are the daily averages only for July 29 and 30. One can see the similar trend in the predicted values, lower in rural station Chaumont, higher in urban sites, as in the measurements. However, there is an obvious discrepancy between measured and calculated values of sulfate and nitrate. We believe that too low sulfate concentrations are due to underestimated SO_2 levels (about half of the measured concentrations at these stations). This is most likely caused by the underestimation of SO_2 emissions. Low sulfate levels lead to more ammonium nitrate formation which explains overestimated nitrate concentrations.

TABLE I

Measured (annual and July) (Hüglin, 2000) and CAMx lowest level (50 m) daily average PM2.5 concentrations (μg m^{-3}) for particulate ammonium, nitrate, sulfate and organic carbon at four locations

	NH_4^+				NO_3^-				SO_4^{2-}				OC			
	measured		calculated		measured		calculated		measured		calculated		measured		calculated	
Location	annual	July	29.7	30.7	annual	July	29.7	30.7	annual	July	29.7	30.7	annual	July	29.7	30.7
Basel	1.9	1.3	0.9	0.7	2.5	0.2	2.6	2.0	3.8	3.2	0.4	0.3	2.9	1.0	0.4	0.4
Bern	1.4	1.2	0.8	0.7	2.3	0.7	2.5	2.2	2.7	2.6	0.4	0.3	4.6	2.6	0.4	0.4
Chaumont	0.9	1.2	0.5	0.6	0.5	0.1	1.5	1.7	2.4	3.0	0.2	0.2	1.2	1.5	0.2	0.2
Zurich	1.9	1.4	2.0	1.1	2.7	0.6	5.9	3.2	3.3	3.2	0.7	0.6	3.1	1.8	0.8	0.8

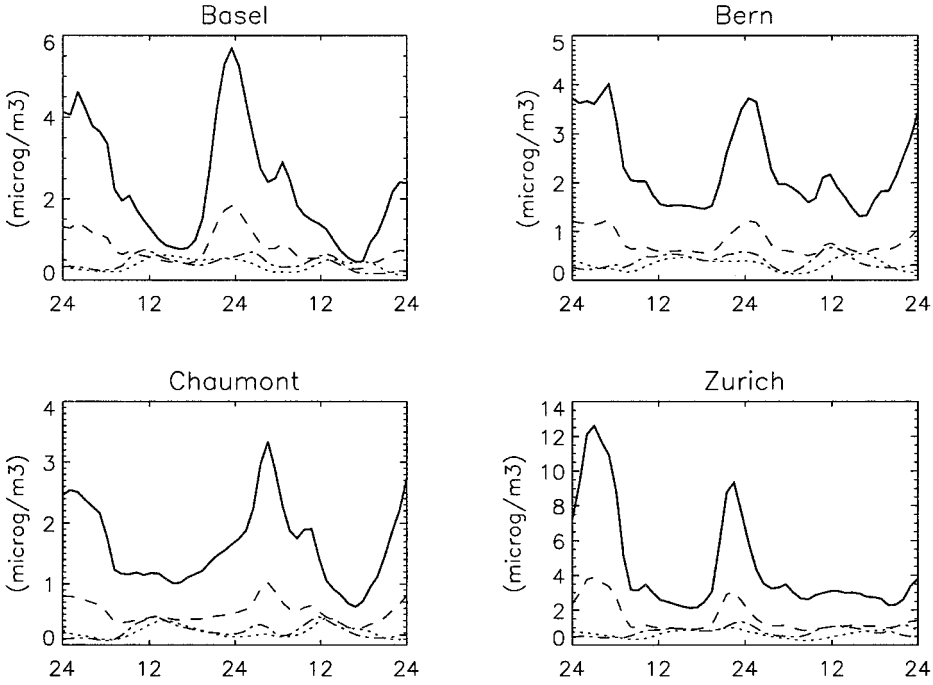

Figure 13. Lowest level (50 m) CAMx aerosol concentrations (μg m^{-3}) as a function of time (CEST) on July 29–30 at four sites for: PNO_3 (solid), PSO_4 (dots), PNH_4 (dashes), OC (dash-dots).

5. Conclusion

A photochemical air pollution episode in Switzerland during July 28–30, 1993 was simulated by the three-dimensional Eulerian model CAMx, including for the first time both gas-phase and aerosol chemistry. The meteorological input data were

generated by the SAI Mesoscale Model (SAIMM). Evaluation of model results together with the measurement data from the POLLUMET field experiment showed that CAMx predicted the pollutant concentrations for July 29 better than the UAM and UAM-V models used previously. However, the wind fields produced by the SAIMM model overestimated the wind speeds for July 30, causing a fast transport of pollutants in some parts of the model domain.

Inclusion of aerosol chemistry led to an improvement of model results especially for HNO_3. The positive correlations found from airborne measurements between the total particle concentration and secondary gaseous pollutants were reproduced also by the model indicating that a considerable fraction of the aerosols might be photochemically produced. In general, the evaluation of the calculated particle species was difficult due to very few measurement data. However, these first, preliminary calculations for the particles performed in this study, gave reasonable results which can be improved and evaluated better when more experimental data is available in the future.

The results obtained from this first application of CAMx model in Switzerland lead to the following conclusion. Although model performance looks better than the previously used UAM and UAM-V models, boundary conditions and emission inventory still need to be improved. Another outcome is the need of a meteorological model which can generate more suitable meteorological data for the complex terrain. In the future, a non-hydrostatic meteorological model will be used. The aerosol chemistry module which is still under development, looks promising. More measurements on aerosol species are needed for model evaluation.

Acknowledgements

We would like to thank the Swiss Meteorological Institute for providing the meteorological data for the SAIMM calculations. We also thank G. Kok (NCAR), A. Neftel (FAL) and A. Hering (ETH) for the measurements. C. Hueglin (EMPA) is gratefully acknowledged for providing the measured PM2.5 data. We would also like to thank C. Emery and G. Yarwood from ENVIRON for their helps to apply the aerosol routines. Finally, we are grateful to U. Baltensperger for his valuable comments on the manuscript.

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