Preliminary simulation of a summer smog situation in Switzerland using the Urban Airshed Model (UAM)

S. Andreani-Aksoyoglu, W.R. Graber, J. Keller
Laboratory for Environmental Research and Systems Analysis, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

Abstract

The photochemical smog situation in Switzerland on July 29, 1993 was simulated using the 3- dimensional grid model UAM (Urban Airshed Model). Results were compared with the field measurements. The calculations showed that ozone formation in Switzerland is mainly NO_x limited. However, some areas seem also to be sensitive to VOC emissions. Calculation of dry deposition of the pollutants indicated that most of the O_3 formed over the Swiss Plateau is deposited in the Alpine regions which have high forest coverage. The impact of biogenic VOC on O_3 concentrations was predicted to be small (<5%). Their effect on PAN and H_2O_2 concentrations however, is found to be significant.

1 Introduction

During the last few years, considerable effort has been devoted to the development and application of air quality computer models in order to understand the complicated photochemical processes and to plan efficient emission control strategies [1]. It is essential for control strategies in Switzerland to clarify carefully the photochemical processes, because about half of the area is covered by Alpine mountains without anthropogenic emissions. As described by Graber et al. [2], the thermally driven, daily changing wind system gives rise to an exchange of air masses between the densely populated Swiss Plateau and the Alps. Trajectory modeling studies have already been carried out in Switzerland, mainly in the Swiss Plateau region [2-3]. However, no three dimensional modeling of the entire country has been done so far. The aim of this study is to simulate a photochemical summer smog situation in Switzerland, using the three-dimensional Urban Airshed Model [4] covering both urban and rural domains. The sensitivity analyses, emission reduction

scenarios and the comparison of UAM with a multi-parcel Lagrangian model for the same case study have been discussed in detail elsewhere [5]. In this paper, the emphasize is given to the concentration distribution and dry deposition of the pollutants.

2 Methodology

The Carbon Bond Mechanism (CBM-IV) implemented in UAM [6] was modified by addition of O_3 and OH radical reactions of α-pinene. The modeled region is 350 km in the east-west and 220 km in the north-south direction with a grid size of 5 x 5 km^2 and two vertical layers. The initial and boundary concentrations as well as the meteorological data were taken from the POLLUMET field measurements [7] and literature [8-9]. The simulations for July 29 were initialized by a pre-run for July 28.

The five land-use classes used were urban, mixed forest, water, agricultural and rocky land [4]. Land-use data have been obtained from Geostat [10]. Deposition velocities are calculated in the model using initially given surface resistances of the species [4].

The generation of wind fields is described elsewhere [2]. In general, the west wind dominates, however, at lower levels, the wind direction is influenced by the valleys. A provisional emission inventory with anthropogenic NO_x and VOC emission fields as described by Graber et al. [2] was used. Hourly biogenic VOC emissions from the forests were calculated for each cell of the modeling region for that particular day. The method used to calculate the natural emissions from the forests is described elsewhere [11]. The influence of biogenic VOC on the pollutant concentrations was studied by eliminating them from the reaction mechanism. Emission reduction scenarios were calculated by reducing in one case NO_x, in another case anthropogenic VOC emissions by 80%. In the third case, both emissions were reduced by the same amount.

During the field experiments, various measurements were carried out. Airplane measurements covered a large part of the Swiss Plateau. The flights were operated by the National Center for Atmospheric Research in Boulder (NCAR), and by the MetAir company. Data from these airplane measurements were used for comparison with the calculations for the lower layer. The trace-gas flux measurements [7] were carried out by Eddy correlation technique.

3 Results and Discussion

The highest ozone concentrations (80-85 ppb) were predicted between 16:00 and 17:00 CEST (Central European Summer Time) downwind of Zürich (Fig. 1). Peak ozone concentrations are about 70 ppb at Zürich, 60 ppb at Genève, 70 ppb north-east of Genève, and 55 ppb at Bern. Increased ozone concentrations in the Alpine valleys are evident. In the region of the Alps, ozone

concentrations are not higher than 50 ppb, whereas in the south of the Alps levels are between 50 and 60 ppb. High ozone concentrations in the south-east part of the model domain are due to the high emissions transported by the wind from northern Italy and the Lugano area. Increased concentrations at north-east of Genève represent the Genève plume. PAN concentrations at the time of peak ozone were between 0.5 and 2.5 ppb over the Swiss Plateau, the highest being downwind of Zürich. H_2O_2 concentrations varied between 1 and 3 ppb mainly over the Swiss Plateau and in northern Switzerland.

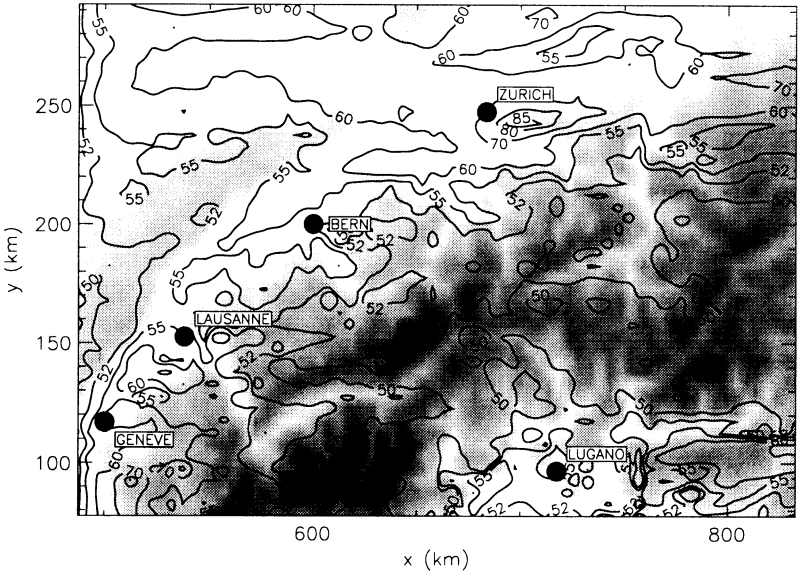

Figure 1 : Average O_3 concentrations (ppb) (contour lines) between 16:00 and 17:00 CEST, plotted over the Swiss topography (darker regions have higher altitudes)

The ratio of VOC/NO_x concentrations is an indicator of either NO_x (high ratio) or VOC (low ratio) limiting ozone formation [12]. In general, this ratio is high in Switzerland. A few areas with low VOC/NO_x ratios (20-30) are in the western part of the Zürich area, in the north-west of Switzerland, and in the north-east, south-west and south-east corners of the model domain. Reduction scenarios calculated by reducing the emissions indicated that ozone formation in Switzerland is mainly NO_x limited while a few locations show sensitivity to VOC controls as well. VOC controls seem to be effective at NO_z ($NO_z = NO_y - NO_x$, where $NO_y = NO_x + PAN + HNO_3$ + other nitrates, and $NO_x = NO+NO_2$) concentrations above about 2 ppb in the base case, whereas NO_x controls are predicted to be effective also below this threshold [5].

One simulation was carried out without isoprene and α-pinene reactions. It is known that in general, biogenic VOC enhance the formation of peroxy radicals and ozone. Ozone concentrations decreased only by 2-3 ppb (up to 5%) as a result of elimination of biogenic VOC. The effect of biogenic VOC is evident in the areas with high NO_x and relatively low VOC. The rest of the modeling area was not affected by the absence of these compounds. The effect of biogenic VOC seems to be more significant for the formation of other pollutants. When biogenic VOC were excluded from the calculations, formaldehyde concentrations decreased by 10-20 %, H_2O_2 by 5-10 %, and PAN concentrations by 10-20 %.

Agreement between the predicted and measured ozone and NO_2 concentrations are quite good especially in the afternoon (Fig. 2). In the morning, ozone concentrations are overestimated by 5-7 ppb. H_2O_2 concentrations are underestimated. These comparisons show that the afternoon data agree well with the predictions. However, the model overestimates ozone concentrations by about 10-15 % in the morning. The results indicate the necessity of using more layers to simulate the air quality more accurately.

The calculated amount of ozone deposited during the simulation period is shown in Fig. 3. Dry deposition is predicted to be higher in the forested and agricultural areas with relatively high ozone concentrations. However, high ozone deposition is remarkable in the Alpine regions although the concentrations are relatively low. These results indicate that some of the ozone produced over the Swiss Plateau is transported and then deposited in the Alpine regions which have high forest coverage. The predicted values were compared with some measurements at Siselen which is at north-east of Bern. The measured deposition fluxes of NO_2 and O_3 were shown together with the calculated ones in Fig.4. Although the agreement between the measurements and calculations for the maximum values seems to be good, the morning flux for ozone is overestimated. This is partly due to the overestimated ozone concentrations in the morning. The sudden jumps at 7:00 and 21:00, are likely to be due to the different deposition velocities calculated for day and night in the model. The calculated ozone deposition at Siselen is 32 mg m^{-2} d^{-1} while measured values are around 20 mg m^{-2} d^{-1}. Deposition of PAN varies between 0.2 and 1 mg m^{-2} d^{-1}, and the highest deposition is predicted to be in northern Switzerland, around Zürich. Although the deposition of nitrogen compounds represents a nutrient transfer from atmosphere to biosphere, excess nitrogen might contribute to the forest decline. NO_2 deposition flux was predicted to be between 1 and 4 mg m^{-2} d^{-1}, concentrated around Zürich where NO_x concentrations are high. The deposition flux of HNO_3 was calculated to be 4-8 mg m^{-2} d^{-1}, mainly in the Alpine regions.

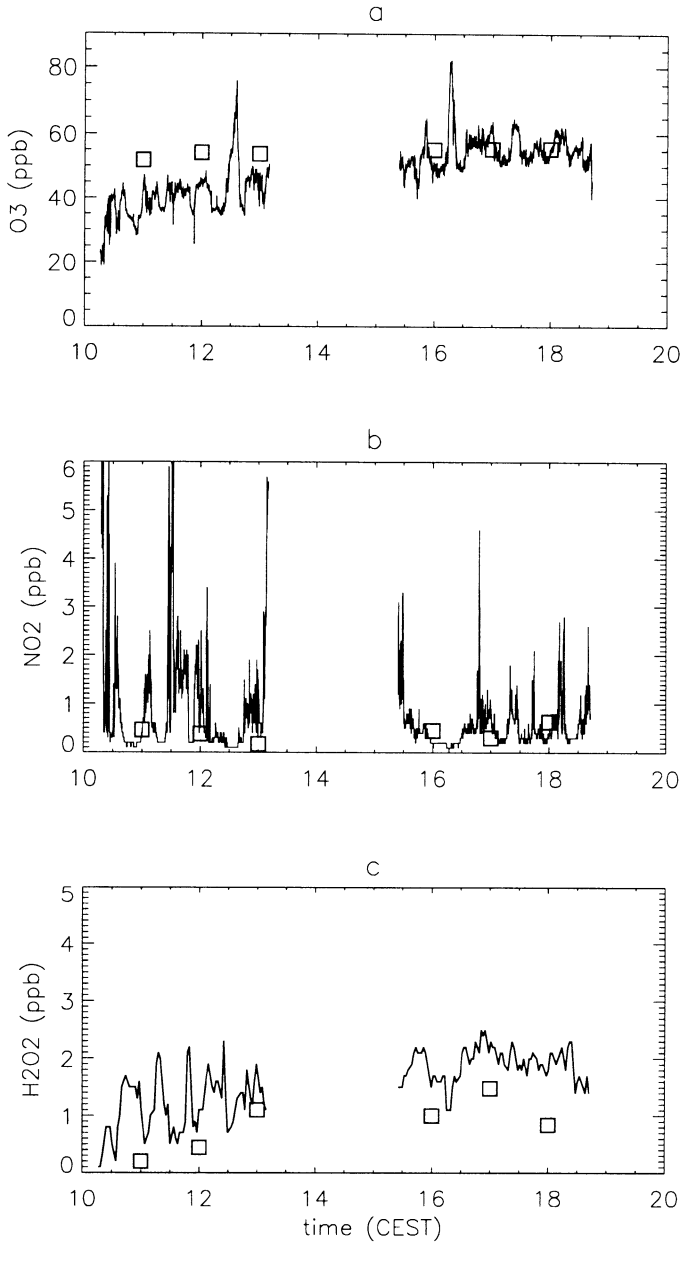

Figure 2 : Comparison of calculations with the airplane (MetAir) measurements. The solid line shows the airplane measurements; predictions are given as squares.

a) O_3 b) NO_2 c) H_2O_2

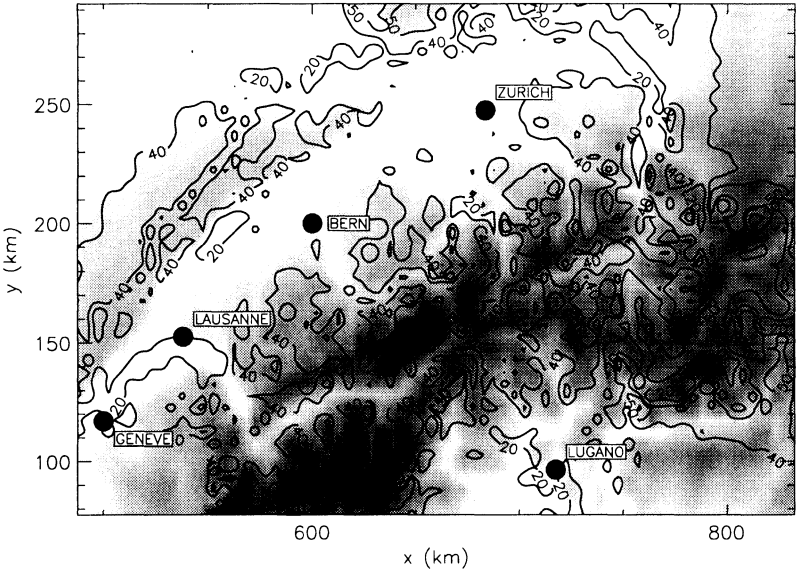

Figure 3 : Dry deposition of O_3 (mg m^{-2} d^{-1}) (contour lines) on July 29, 1993, plotted over the Swiss topography (darker regions have higher altitudes)

4 Summary and Conclusions

A photochemical smog situation in Switzerland on July 29, 1993 was simulated using the 3-dimensional grid model, UAM (Urban Airshed Model). The highest ozone concentrations (80-85 ppb) were predicted downwind of Zürich. UAM results were found to be in a good agreement with airplane measurements over the Swiss Plateau. Ozone concentrations were overestimated by 5-7 % in the morning which caused also overestimated ozone deposition. The calculated maximum dry deposition fluxes for NO_2 and O_3 are in a good agreement with the measurements. The highest ozone deposition is predicted to be in the Alpine regions with high forest coverage. Results of this study indicate that increasing the number of layers in the simulations might improve the predictions. Emission reduction scenarios indicated that ozone formation in Switzerland is mainly NO_x limited whereas a few locations seem to be sensitive to VOC controls as well. VOC controls are predicted to be effective at NO_z concentrations in the base case above 2 ppb, while NO_x controls were effective also below this level.

The influence of biogenic VOC on ozone concentrations was found to be relatively small. When biogenic VOC were eliminated, a decrease of up to 5 % in ozone concentrations was found at locations with low VOC/NO_x ratio. However, they had a stronger effect on H_2O_2 and PAN concentrations.

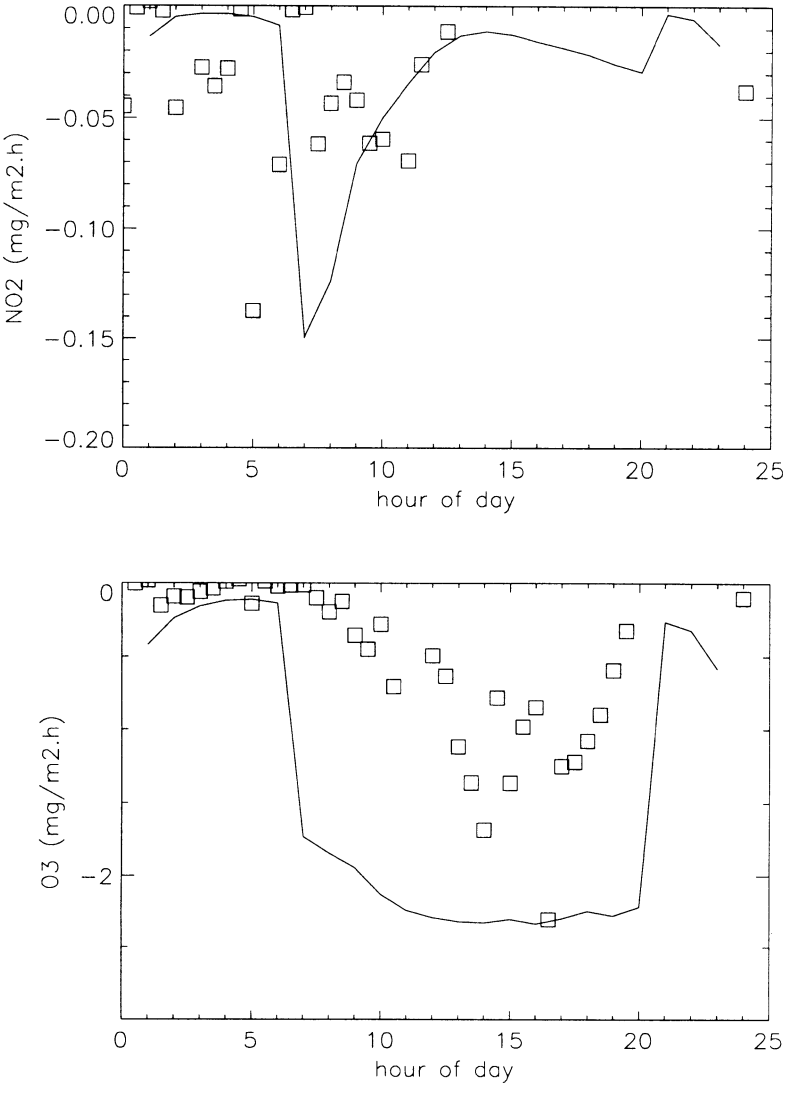

Figure 4 : Dry deposition fluxes of NO_2 and O_3 (mg m^{-2} h^{-1}) at Siselen on July 29, 1993. Solid line shows the model predictions; measurements are given as squares. Negative values are due to downward fluxes.

Acknowledgements

We would like to thank the Swiss Federal Institute for Forest, Snow and Landscape Research (WSL), the Swiss Meteorological Institute (SMA), MetAir and NCAR for providing us with data. We appreciate also the helps of W. Eugster.

References

[1] Sillman S. & Samson P.J. Nitrogen oxides, regional transport, and ozone air quality: results of a regional-scale model for the midwestern United States. *Water, Air and Soil Pollution,* 1993, **67**, 117-132

[2] Graber W.,K., Andreani-Aksoyoglu S., Keller J.E. & Roesselet C.M. A multi-parcel Lagrangian model for the quantification of the influence of the Alpine air mass exchange on the production of photo-oxidants. *Atmos. Environ.*, 1995, (in press)

[3] Roesselet C.M. & Kerr J.A. *Photochemical modelling of photo-oxidant levels over the Swiss plateau and emission reduction scenarios.* PSI Report Nr. 93-03, Paul Scherrer Institute, 1993

[4] Morris R.E. & Myers T.C. *User's guide for the Urban Airshed Model Volume 1: User's manual for UAM (CB-IV).* EPA-450/4-90-007A,USEPA, 1990

[5] Andreani-Aksoyoglu S., Graber W.K., & Keller J. Three dimensional modeling of photochemical air pollution in Switzerland and comparison with a multi-parcel Lagrangian model. (Submitted to Atm. Environ.), 1995

[6] Gery M.W. & Whitten G.Z. A photochemical kinetics mechanism for urban and regional scale computer modeling. *J.Geophys. Res,* 1989, **94**, 12925-12956

[7] Pollumet Data base, 1993

[8] Ehhalt D.H. Concentrations and distributions of atmospheric trace gases. *Ber. Bunsenges. Phys. Chem.*, 1992, **96**, No.3, 229-240

[9] Isaksen I.S. & Hov O. Calculation of trends in the tropospheric concentration of O_3, OH, CO, CH_4 and NO_x. *Tellus,* 1987, **39B**, 271-285

[10] GEOSTAT Bundesamt für Statistik, Bern, 1992

[11] Andreani-Aksoyoglu S. & Keller J. Estimates of monoterpene and isoprene emissions from the forests in Switzerland. *J. Atmos. Chem,* 1995, **20** (in press)

[12] Finlayson-Pitts B.J., Pitts J.N. Jr. *Atmospheric Chemistry*, John Wiley & Sons Inc. USA, 1986