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Effects of some model parameters on the indicator values for ozone production sensitivity

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Abstract

It is very important for ozone control strategies to know whether ozone production is sensitive to NO_x or VOC emissions to avoid unwished effects due to the nonlinearity of the photochemistry. Indicator species derived from measurements can be useful to assess the sensitivity. However, indicator values can vary under certain circumstances. In an earlier model study, we showed that the thresholds of indicators are affected by the wind fields and emissions. In this work, we studied the variability of indicator thresholds further by model simulations. A 3-day episode in July 1993 in Switzerland was modelled using the meteorological model SAIMM and the photochemical model CAMx. The studied indicators are NO_y, $HCHO/NO_y$, O_3/NO_z and H_2O_2/HNO_3. The effects of some factors such as the definition of sensitivity, boundary concentrations and degree of emission reductions are discussed.

1 Introduction

The relation between ozone and its main precursors NO_x ($NO + NO_2$) and VOC (volatile organic compounds) is very important for environmental policy because of the adverse impacts of ozone on human health and on crops and forests (NRC [1]). Some researchers proposed the use of observable species as indicators for ozone production sensitivity to changes in the precursor emissions. Milford et al. [2] proposed NO_y as indicator and Sillman [3] expanded the concept of indicator species to include several others such as O_3/NO_z, H_2O_2/HNO_3, $HCHO/NO_y$. Indicators have been used over different areas by various numerical models (e.g.

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Vogel et al. [4], Lu and Chang [5]). Our earlier study suggested that the proposed indicator values are not universally applicable and the threshold values of indicators may vary with the location and environmental conditions (Andreani-Aksoyoglu et al. [6]). In this paper, we continued investigating the variability of indicator values by checking for the effects of some parameters such as boundary concentrations and the degree of emission reductions.

2 Models

2.1 Meteorological model

The meteorological data such as wind fields, humidity, pressure, temperature and vertical exchange coefficients were calculated using the SAI Mesoscale Model, SAIMM (SAI [7]). Basic meteorological quantities used by the SAIMM are time and space dependent soundings and surface measurements of pressure, temperature, relative humidity, wind velocity and direction. The model uses geometric terrain-following coordinates. The horizontal resolution for this study was 5 km x 5 km in a 470 km x 385 km grid. There were 19 layers. The bottom of the lowest level is the topographic surface and the top of the upper level is at constant height of 9000 masl. Hence the thickness of a given vertical layer generally varies within the domain and is smaller for higher altitudes. As input for this model, the data from the meteorological network ANETZ and output of the Swiss Model (SM) of the Swiss Meteorological Institute were used. The simulated meteorological quantities were nudged towards the experimental data to obtain a better agreement.

2.2 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 [8]). It has the possibility of choosing CBM-IV or SAPRC 97 chemical mechanisms. The model domain covered an area which was 370 km in the west-east and 285 km in the north-south direction i.e. it was embedded in the larger SAIMM domain. The horizontal resolution was 5 km x 5 km, and there were 8 vertical layers with varying heights from 50 m to 3000 m above ground.

2.3 LOTOS Model

The European grid model LOTOS, (Builtjes [9]) was used to provide the CAMx model with the boundary concentrations. The LOTOS – long term ozone simulation – is a 3-D Eulerian grid model of intermediate complexity. It is focused to calculate photooxidant formation, and is recently extended with an aerosol module. The model is intended for hour-by-hour calculations over extended periods of several years.

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LOTOS covers all of Europe in grids of, for this calculation, 0.5 x 1.0 latlong. The vertical extension is 3.5 km, with 4 vertical layers including a parametrised surface layer. The chemical scheme is CBM-IV. The emission data are based on CORINAIR, and for the remaining countries on the LOTOS-emission database. Per source category time and temperature dependencies are given. Boundary conditions follow from the global model TM-3.

The meteorological input is diagnostic, using the experience and interpolation methods of the Free Univ. Berlin.

3 Simulations

In this study, the CAMx model was applied to simulate the air quality in Switzerland during the period of July 28-30, 1993. The simulation of the first day was used to initialize the model. The results discussed in this paper refer to July 29. The anthropogenic emissions were compiled from various data sources and include emissions from traffic, industrial processes, and residential areas (BUWAL [10]). The biogenic emissions were included in the emission inventory as well (Keller et al. [11], Andreani-Aksoyoglu and Keller [12]). In this study, the CBM-IV chemical mechanism was used. The initial and boundary concentrations of chemical species are either compiled from the measurements and previous simulations (referred to as Swiss boundaries, SB) or provided by the European LOTOS model (referred to as LOTOS boundaries, LB). The concentrations of some selected species at four lateral boundaries in the lowest layer are shown in Figure 1 for both Swiss and LOTOS boundaries. The main difference between the two sets of data is in the NO_x concentrations which are higher in the Swiss boundaries.

The 8 simulations performed in this study are described in Table 1. The first 5 runs were carried out using the Swiss boundary concentrations. In the other 3 simulations, the boundary concentrations were provided by the European LOTOS model. In each group there is one base case with 100% emissions. Additional runs were performed with reduced anthropogenic NO_x and VOC emissions (65 %) separately to investigate the sensitivity of ozone production to NO_x and VOC. In the first group (with the Swiss boundaries) two additional simulations were carried out to check for the effect of the degree of emission reduction on the indicator values. In this way the comparison of runs 1-3 and runs 6-8 serves to investigate the effect of boundary concentrations whereas comparison of runs 2-3 and runs 4-5 can show how the amount of emission reduction affects the results.

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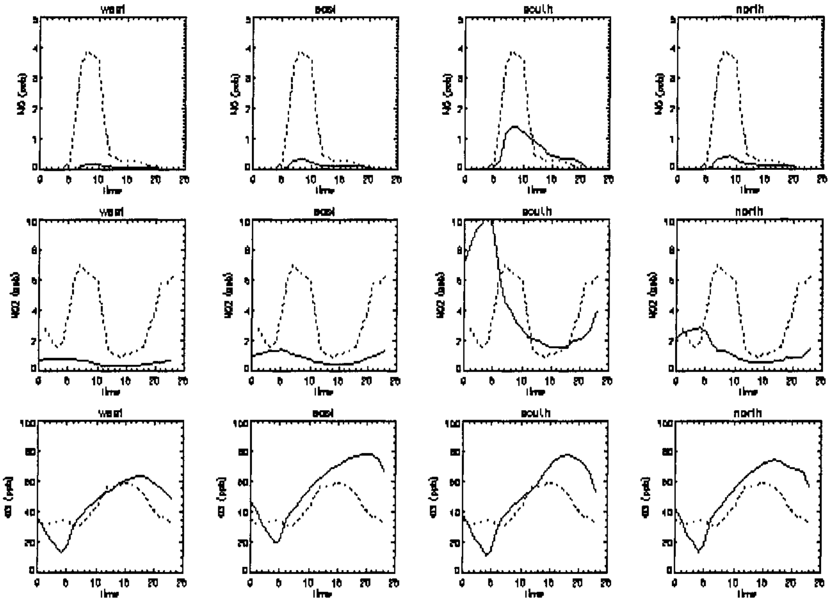

Figure 1: The concentrations of NO, NO_2 and O_3 in the Swiss (...) and LOTOS (__) boundary datasets for the four lateral borders of the model domain.

Table 1: Model Runs

Run number	boundary type	NOx emissions (%)	VOC emissions (%)
1	Swiss	100	100
2	Swiss	100	65
3	Swiss	65	100
4	Swiss	100	50
5	Swiss	50	100
6	LOTOS	100	100
7	LOTOS	100	65
8	LOTOS	65	100

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4 Results

There was a light westerly wind on July 29, 1993. Meteorological parameters calculated by the meteorological model SAIMM, were in general reasonable for July 29, although the model performance for another day was not satisfying (Andreani-Aksoyoglu et al. [13]). Therefore, only the results of July 29 were used in this paper. Figure 2 shows the predicted wind fields for July 29. The predicted wind velocities were about 3-4 m s^{-1} in the afternoon over the Swiss Plateau and both wind directions and velocities agreed with ground measurements. A comparison of photochemical model results and airborne measurements of some species carried out on board the NCAR aircraft during the afternoon over the Swiss Plateau is shown in Figure 3. NO_x, NO_y and HCHO concentrations could be reproduced by the model quite well. The predicted H_2O_2 concentrations are about 0.5 ppb lower than the measurements in general. Although ozone concentrations match the measurements well at some locations, they are in general overestimated by the model. The performance of the meteorological and the photochemical models for the period studied was discussed in detail elsewhere (Andreani-Aksoyoglu et al. [13]). The main emphasis here will be given to the variability of indicator values with the parameters mentioned in the previous section.

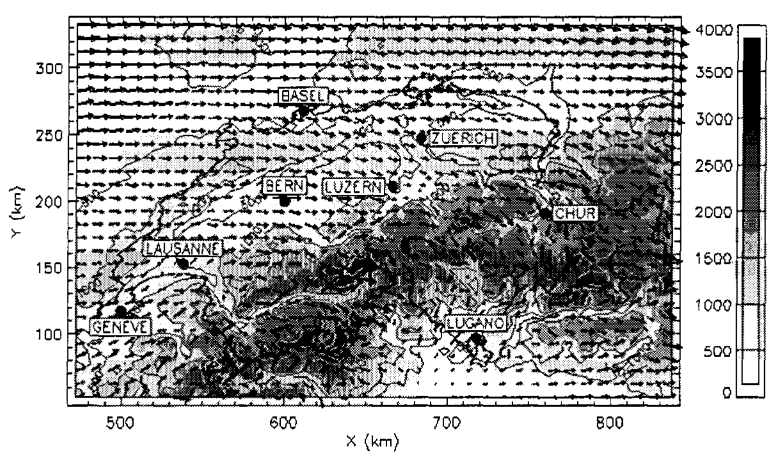

Figure 2: Afternoon wind fields simulated by SAIMM for July 29, 1993 over the topography (shaded).

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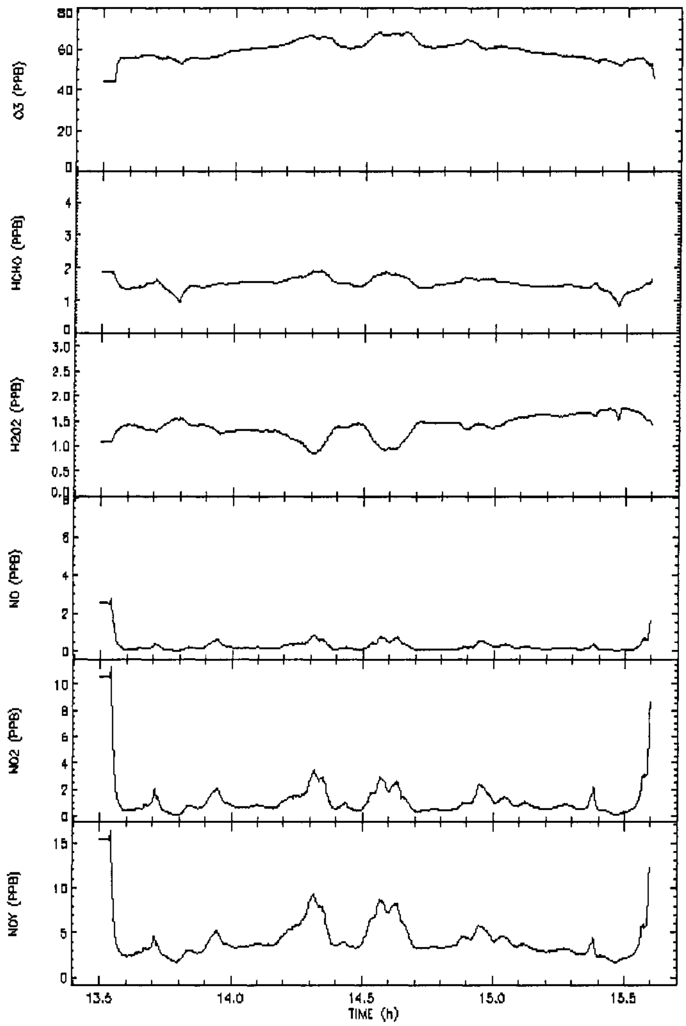

Figure 3: Model results (solid line) and aircraft measurements (dotted line) of some species on July 29, 1993 over the Swiss Plateau.

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The NO_x and VOC sensitivity regimes were defined using the method described by Andreani-Aksoyoglu et al. [6]. The three different sensitivity regimes derived from the simulations 1-3 are shown in Figure 4. Reduction in peak ozone concentrations due to NO_x emission reduction by 35% is plotted against the reduction in peak ozone concentrations due to VOC emission reduction by the same amount. After evaluating various slopes between 1 and 100, a slope of 10 was chosen for this study to separate different chemical regimes. The points refer to the grid cells in the domain. This figure shows that most of the grid cells are in the transition range, i.e. ozone formation is sensitive to both NO_x and VOC emissions. There are many grid cells in the NO_x sensitive regime and a few cells in the VOC sensitive regime. If we apply the definition of Sillman [3], all the grid cells belong to the transition range because the differences in ozone concentrations in the NO_x and VOC reduced cases are within 5 ppb (Figure 5). The difference between the figures 4 and 5 shows the effect of definition of sensitivity. The following calculations were carried out using our definition with the boundary slope of 10.

The values for indicators such as NO_y, O_3/NO_z, $HCHO/NO_y$, H_2O_2/HNO_3 were calculated for each sensitivity regime. To avoid outliers, the values were confined to a range between the 2nd and the 98th percentiles. For a successful indicator, the range of values of the NO_x and VOC sensitive regimes should not overlap. Figure 6 shows the ranges of the indicators in the NO_x and VOC sensitive regimes for three different cases. These cases refer to simulations 2-3 (35% SB), 4-5 (50% SB) and 7-8 (35% LB). On this figure one can see the effects of both the boundary concentrations and degree of emission reductions on the indicator values. When we compare the two cases with 35% and 50% emission reductions using the same boundary concentrations (Swiss boundaries), we see little difference for each indicator. This means that reducing the emissions by 35% or 50% does not change the distribution of the grid cells to different chemical regimes significantly in this study. On the other hand, we see differences when we compare the two cases with different boundaries using the same emission reductions (35%). In case of NO_y and $HCHO/NO_y$, the two regimes are better separated from each other with the LOTOS boundaries. For the other two indicators, the two regimes are always overlapped but the ranges are wider in the case of LOTOS boundaries. On the other hand, the effect of boundaries on peak ozone concentrations seems to be less important: when the Swiss boundaries were replaced by the LOTOS boundaries, peak ozone concentrations increased by 3-4 ppb around the borders of the model domain. A slight decrease (1-2 ppb) was predicted over the Swiss Plateau.

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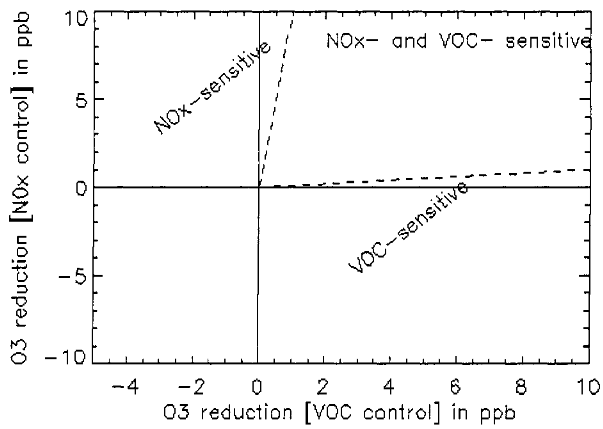

Figure 4: Relation between ozone reductions due to NOx controls and ozone reductions due to VOC controls using the boundary slope of 10.

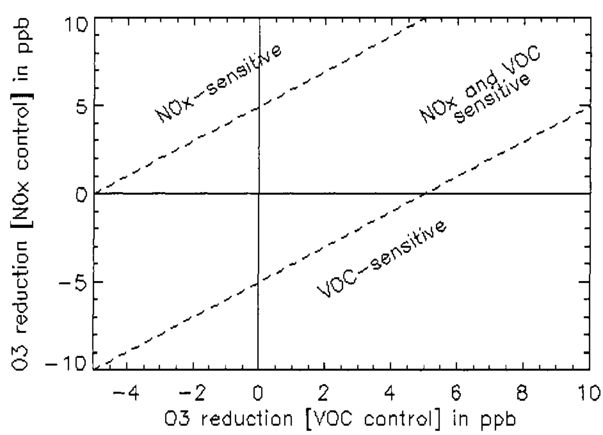

Figure 5: Relation between ozone reductions due to NOx controls and ozone reductions due to VOC controls using Sillman's definition.

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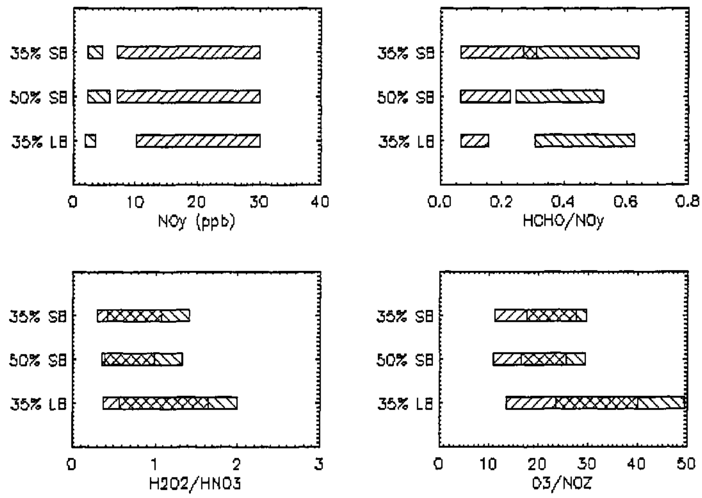

Figure 6: Variation of indicator values with the degree of emission reductions and boundary concentrations. SB: Swiss boundaries, LB: LOTOS boundaries. 35 % and 50% are the emission reductions.
VOC sensitive regime
NO_x sensitive regime

5 Conclusion

In this study, a 3-day episode in Switzerland was simulated by the SAIMM (meteorological) and CAMx (photochemical) models. The values for some selected indicators were evaluated for one day and variations by some model parameters were investigated. It was shown that the grid cells can be attributed to different chemical regimes depending on the definition of sensitivity used. When Sillman's definition was used, all the grid cells were predicted to be in the transition range. Our definition attributed most of the grid cells to the transition range, but some of the cells were also in the NO_x and VOC sensitive regimes. Changing the emission reduction from 35% to 50% did not change the values of indicators. These results show that the chemical regimes were not affected by this change in the degree of emission reduction. On the other hand, replacing the Swiss boundary concentrations by the boundaries calculated by the European LOTOS model led to significant differences in the indicator values, although the effect on the peak ozone concentrations was less pronounced. As suggested earlier in Andreani-Aksoyoglu et al. [6], this study showed that the indicator values may vary with some parameters and therefore, they are not universally applicable.

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