Mass Budget Simulations of Ozone in the City Plume of BERLIN for an Episode of the BERLIOZ Experiment

A Guest contribution to subproject GLOREAM

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Summary

Mass budget simulations have been carried out with the mesoscale Chemistry-Transport-Model KAMM/DRAIS for July 20th, 1998, an episode during the BERLIOZ experiment that took place in Berlin and its surroundings. On this day an ozone plume has been developed north-north-west of the City of Berlin. This city plume has been analysed using the mass budget methodology. The calculations have been performed for three regions. The first region comprises the city of Berlin and the other two are located north of it. Above each region four layers are considered. The lowest one represents the surface layer. The next two layers reach to the top of the boundary layer, followed by the last layer extending to the top of the domain. In the surface layer above the city, chemical processes decrease the mean ozone concentration during the whole day. Nevertheless, an increase of the ozone concentration occurs during daytime, because higher ozone concentration are mixed down from the layer above. Independent of the region, chemical processes are mainly responsible for the daytime changes of the ozone concentration in the mixing layer above the surface layer. As expected, the chemical ozone formation rate is largest in region 2, which is located directly downwind of the city. The maximum production rate in the lower part of the boundary layer is 7 ppb/h and it occurs in the late morning (11 UTC). Farther to the north, in region 3, the model underestimates the formation rate. This result reflects the underestimation of the ozone concentration in the plume at larger downwind distances.

Introduction and Objectives

Mass budget analyses describe the temporal change of the mass M[1] of an air pollutant due to different production (P) and loss (L) processes: advective (ADV) and turbulent (TRB) transports, emission (E), deposition (D), and chemical transformations (CH) (Fig 1). It has been shown by other investigations (e.g. Memmesheimer et al., 1997), that mass budget considerations allow determining the relevance of the processes in dependence on time and space. Because of their non-local character, they provide more general results. On July 20th, 1998, an episode of the BERLIOZ Experiment, an ozone plume developed north-north-west of the City of Berlin. For this episode simulations have been carried out with the model system KAMM/DRAIS. The purpose was to analyse the relevance of the different ozone mass budget components along the plume. Three different areas have been considered (Fig. 2). The first one includes the City of Berlin, the other two adjoin in the direction of the plume. In order to demonstrate the vertical variations of the relevant processes, four different layers have been considered above each area (Tab. 1).

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[1] Instead of mass, volume averaged concentrations in terms of mixing ratios will be used throughout the text

Table 1: Vertical layers for mass budget calculations

	Vertical extension (m above ground)	Characterisation
Layer 1	2000 - 5000	Free troposphere
Layer 2	1200 - 2000	Upper mixing layer
Layer 3	75 - 1200	Lower mixing layer
Layer 4	Ground - 75	Surface layer

Performance of the simulation with the model system KAMM/DRAIS

The KAMM/DRAIS model system (Nester and Fiedler, 1998) consists of the non-hydrostatic meteorological mesoscale model KAMM and the dispersion model DRAIS that takes into account the deposition and chemical transformation of all 41 RADM2 species (Stockwell et al., 1990). The model possesses a module, which allows calculating the individual contributions of emissions, advective and turbulent fluxes, deposition and chemical transformation to the mass budget of each species being considered (Panitz et al.,1997).

The model domain has an extent of 200 km * 200 km and encloses the City of Berlin being located nearly in the centre of the domain. The horizontal grid resolution is 2 km. The emission data have been provided by IER, Stuttgart. Land-use and topography data are taken from the data base delivered by IFU, Garmisch-Partenkirchen. The external forcing for the KAMM model and the initial- and boundary conditions for the species concentrations in the DRAIS model have been derived from the results of the European scale model EURAD (Ebel et al., 1997), using a procedure developed by Nester et al. (1995) to couple the KAMM/DRAIS system to the EURAD model.

In the early afternoon of July 20th, 1998, the ozone concentration distribution near ground level shows a pronounced plume downwind of the City of Berlin. The maximum ozone concentration in this plume is 68 ppb. It occurs about 70 km north-north-west of the city centre. Meteorological and chemical measurements have been carried out onboard of an aircraft, flying through the plume in different levels. The comparison of the measured and modelled wind direction and speed shows an acceptable agreement. This is true also for the ozone concentration, except for the periods when the aircraft flew in the area of Menz, about 70 km north of Berlin (Fig. 2). Here, the model underestimates the measured concentrations by about 10 ppb.

Mass budget simulations

As illustrated in Fig. 1, the mass budget module in the KAMM/DRAIS model calculates the contributions of the different processes to the change of the mean concentration in a predefined volume (Fig. 1).

In order to analyse the relevance of the processes that determine the development of the ozone concentration in the city plume, the ozone budget has been calculated in four different layers (Tab. 1) above three different areas (Fig. 2).
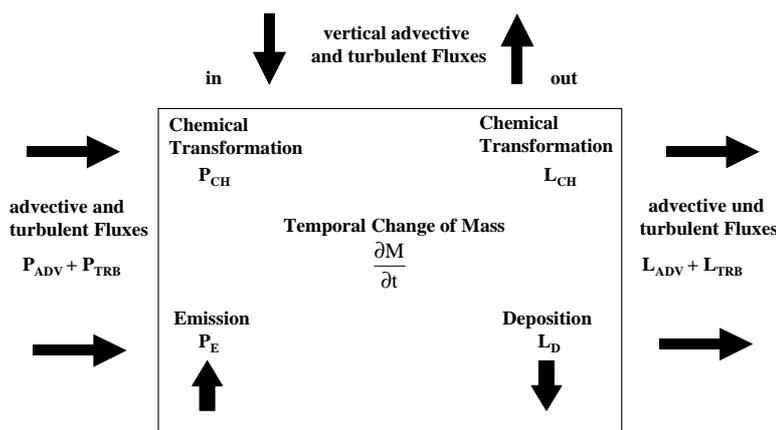
Figure 1. Illustration of mass budget calculation
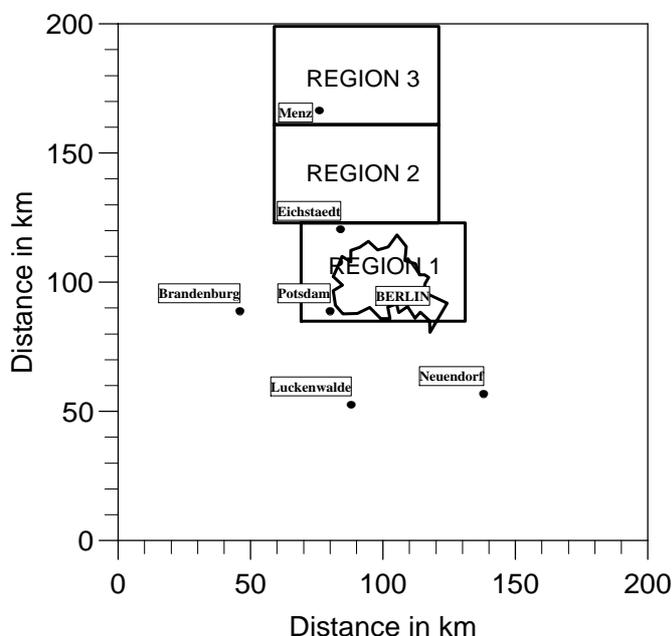
Figure 2. Arrangement of the regions

Although all mass budget components have been determined, only the changes of the ozone concentration due to chemical transformations are presented. The determination of this quantity was one of the aims of the experiment. Fig. 3 shows the hourly change of the chemical ozone formation rate in dependence on the regions and the three lowest layers 2-4 (Tab 1). During the whole day, a loss of ozone due to chemical reactions is found in the surface layer above the city. However, caused by vertical mixing from above, the ozone concentration increases in this layer during the day. In contrast to region 1, chemical production of ozone occurs in the two other regions, where the diurnal variation of the ozone formation behaves quite similar. The highest formation rate (7 ppb/h) appears in the second region, which is closest to the city area. During daytime, ozone is produced chemically throughout the lower boundary layer (layer 3) above all regions. As expected, the production rate is most pronounced in region 2. It is lowest during the afternoon in region 3. The peak formation rates of ozone are similar in the surface layer 4 and the lower mixing layer 3 above region 2. Generally, they are remarkably reduced in the upper mixing layer (layer 2). Again

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the strongest ozone formation (nearly 2 ppb/h) appears in region 2. In the free troposphere (layer 1, not shown) above all regions, the change of the ozone concentration due to chemical processes is much lower than in the boundary layer.
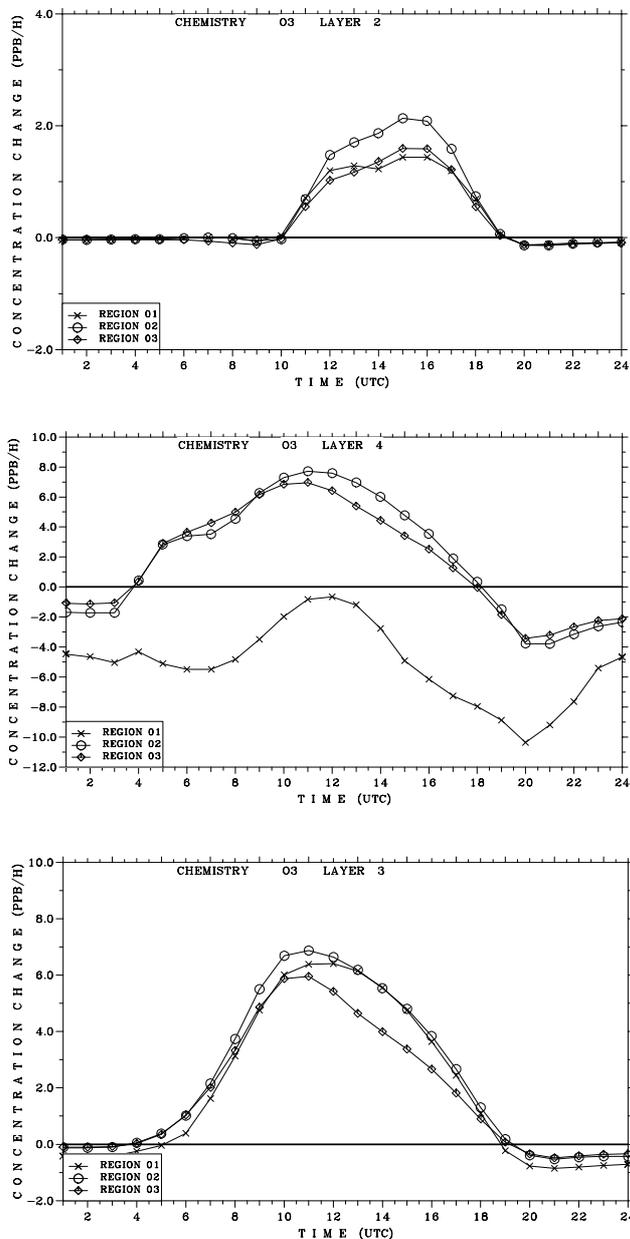
Figure 3. Diurnal cycle of ozone formation rate due to chemical reaction in three layers over three regions

From aircraft measurements it has been derived that the formation of ozone in the urban plume achieves a value of about (6.5 ± 1.0) ppb/h in the afternoon (Corsmeier et al., 2001). The aircraft flew in the lower mixing layer 3 in different levels and crossed the regions 2 and 3 several times. Averaging the modelled ozone production rates over corresponding periods leads to a value of 5.1 ppb/h for regions 2. Because layer 3 above region 2 represents a larger volume of air than the aircraft flight, this is still an acceptable agreement. For region 3 the model calculation gives an averaged ozone formation rate of 3.5 ppb/h, which is obviously too low. A corresponding comparison for the city provides the values (4.5 ±1.0) ppb/h and 5.0 ppb/h for the measured and simulated ozone production rates, respectively

Conclusions

The mass budget calculations for ozone show that the relevance of the different processes depends on the selected region and on the considered vertical layer. In the surface layer, deposition and vertical diffusion dominate the change of the mean ozone concentration. Over the city, the chemical processes decrease the mean ozone concentration during the whole day. Nevertheless, an increase of the ozone concentration occurs during daytime, because air from above, carrying a higher ozone load, is mixed downward. In the mixing layer above the surface layer, chemical processes are mainly responsible for the change of the ozone concentration during daytime, independent of the considered region. In the late afternoon and the early morning, advection dominates the change of the ozone concentration in all layers, because the contributions of the other processes are reduced, but also because of changes in the wind field. The largest increase of the mean ozone concentration due to chemical reactions occurs in region 2, which is located on the downwind side of the city. The maximum formation rate of ozone (7 ppb/h) occurs during the late morning (11 UTC) in the lower mixing layer above region 2.

The comparison of the simulated ozone formation rates with those derived from aircraft measurements show an acceptable agreement over the city and the adjacent downwind region. In region 3 the model underestimates the formation rate. This result reflects the underestimation of the ozone concentration in the plume at farther downwind distances.

Acknowledgement

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