1 INTRODUCTION

Understanding comprehensively the atmospheric aerosol concentration and growth and its interactions with advective accumulation and deposition processes is paramount to take adequate reduction strategies. In the last years much effort has been taken to understand the single processes such as the building of secondary inorganic and organic aerosol components and its simulation in smog chambers as well as in chemical transport models (CTM) (Matta et al., 2002, Nenes et al., 1998, Schell et al., 2001, Grosjean and Seinfeld, 1989 and references therein, Pandis et al., 1992, Odum et al., 1996). Also long range processes of primary coarse particles such as Saharan Dust (Vautard et al., 2005) or the lifetime of fine and ultrafine particles such as elemental and organic carbons has been studied extensively (Bukowiecki et al., 2003).

Gathering all this knowledge and assembling it is a difficult task and has been done in a comprehensive, but theoretical way by Seinfeld and Pandis (1998), for instance. Nevertheless, an adequate deterministic description with reasonable results for all processes is still a challenging task (Tilmes et al., 2002, Römer et al., 2003, van Loon et al., 2004)

In order to describe the Aerosol concentration in urban environments and in relation to rural surroundings long term measurement campaigns are still seldom done. The German AFO-2000 HoVerT campaign (Atmosphärenforschung 2000 “Untersuchung von vertikalen und horizontalen Transportprozessen und deren Einfluss auf die bodennahe Belastung durch Ozon und Staub” – Atmospheric research 2000 “Investigations on vertical and horizontal transport processes and their influence on surface Ozone and aerosol concentrations”) aimed at increasing the observational data basis for chemically and size specified aerosols in a Central European region with strong anthropogenic influence, the Berlin Brandenburg area. Dedicated one year lasting (from September 2001 to September 2002) measurements in a network of about ten traffic, urban, peri-urban and rural sites provided valuable information to assess the urban/ regional contrast in concentrations and sources of different aerosol components and for model evaluation. The HoVerT data base is enhanced by routine observations from the regional Berlin and Berlin-Brandenburg air quality networks and from specific measurements at traffic influenced sites in Berlin, supported by the Department of Urban development (Senatsverwaltung fuer Stadtentwicklung) of Berlin.

The Aerosol Chemistry Transport Model REM_CALGRID (RCG), developed at Free University of Berlin with the support of German Environmental Agency (Umweltbundesamt – UBA) (Stern, 1994, Stern, 2003, Stern et al., 2003) can be viewed as an air quality model of intermediate complexity. It covers the lower troposphere with nesting options; it uses in general a carbon bond mechanism (CBM-4) – chemistry (Gery et al., 1989) and includes an aerosol scheme distinguishing between a fine (aerodynamic diameter < 2.5µm) and gross (< 10µm) mode and between different chemical fractions (primary particulate matter, elemental carbons, primary organic carbons, secondary organic aerosols, inorganic ions). In the past, RCG has been mainly used for simulations of emission abatement scenarios (Stern, 2003) and for ozone forecasts (Flemming et al., 2001, Tilmes et al., 2002). Continental (European) scale photo-oxidant simulations for a summer season were evaluated on several occasions (e.g. Römer et al., 2003) and continental scale simulations of secondary inorganic aerosol are described in Hass et al. (2003).

Aerosol Chemistry Transport Models are built in order to use in a feasible way all the knowledge of intensive observation measurements in nature and in smog-chambers. They also give the unique opportunity to analyse the contributions of single processes to the total aerosol mass. Thereby, contributions of different physical and chemical processes to the changes in species concentrations can be tracked and give a processes related mass budget. This mass related budget analysis of the interactions of different contributions to the final modelled fields allows a better insight in the importance of processes to the final result. Moreover, grid models are able to represent better the spatial distribution of pollutants concentrations than point measurements. While punctual comparisons of station measurements to station based simulations are necessary to obtain a model evaluation, a mass budget analysis can give hints to better understand shortcomings or strengths of modelled interactive processes and their reciprocal relationships. Thus, a mass budget analysis approach can also serve as a model evaluation tool.

Mass budget considerations have become a frequently used tool to estimate the contribution of the single processes to the total concentration fields in many different air quality models (Jeffries and Tonnessen, 1994, Jeffries, 1995, 1996, Jang et al., 1995a, 1995b). Mass budget analyses with Eulerian grid chemistry transport models have been carried out by Memmesheimer et al. (1997) with the FLuMOB-Data-Set (FLuMOB-Project, Stark et al., 1995) and Panitz et al. (1999) in different German regions for Ozone production and loss in the lower troposphere determining the importance of single precursors to the accumulation near the ground. The simulation periods never exceeded a couple of months, while the chosen areas comprised different sizes.

The mass budget study described in this thesis has been conducted within the HoVerT aerosol measurement project. The Aerosol Chemistry Transport Model RCG has been used to simulate the same period on two different scales – one Europe-wide with a resolution of approximately 25 km and a nested domain around Berlin with a resolution of 4 km. Observational results have been used to evaluate and to improve the model (Beekmann et al., 2007).

This monograph is intended to present a mass budget model-experiment for the whole HoVerT campaign period, i.e. a one year integration of mass changes due to individual processes is analysed. The RCG-model has been used and every individual simulated process has been investigated. The considered area comprises the urbanised area of Berlin with an extension of approx. 1600 km^2. One of the most prominent characteristics of the chosen area is the net difference between the emission-intensive urbanised Berlin area and the mostly rural surrounding domain with very few industries and households and only few traffic lines. The main land use type in the surroundings is forest and agriculture. RCG is able to simulate elemental carbon as well as primary and secondary organic carbons. This makes it possible to analyse the accumulation and loss terms for a variety of species over a long term period in a particular area characterised by strong emission gradients between urban and surrounding regions.

The following sections will describe the major finding of the HoVerT-campaign, the chemistry transport model RCG and its evaluation by the sampled aerosol measurements, the mass budget methodology used and the most prominent processes contributing to the accumulation and to the loss of mass in the chosen control volume over Berlin. These processes are emissions, advection and diffusion, chemistry and deposition. Finally, the most important results will be summarised and outlooks for further works will be given.