6 CONCLUSION AND OUTLOOK

A process rate analysis tool has been implemented in the Aerosol-Chemistry-Transport Model RCG. This allows a broader inspection of how the model simulates air pollution concentrations. Moreover, the contribution of individual processes in accumulating or removing polluted mass can be integrated and a process related mass budget can be drawn. The reliability of the models simulation of the considered processes has been proven by a comprehensive model evaluation. The HoVerT campaign allowed to collect a unique data set of the aerosol chemical speciation at traffic, urban and rural sites. These observations were used for a thorough analysis of the aerosol part in the RCG model.

A comprehensive aerosol mass budget analysis has been performed on a yearly basis for a three-dimensional domain with strong emission gradients between the urban agglomerate of Berlin and the mostly rural surrounding area. Mass exchange rates analyses have shown a predominance of advective processes in dispersing mainly primary aerosols over the whole year from the city toward the surrounding areas, while secondarily built organic and inorganic aerosols exhibit seasonal characteristics.

Accumulation due to inflow of sulphate and of organic aerosol components depends on wind direction and on season.

Acidity of aerosol mass in Berlin has been determined analysing the contribution to secondary inorganic aerosol mass accumulation by aerosol building processes.

Primary PM10, including EC and OC, are produced in the city and dispersed via advection toward the surrounding region.

The local production to net transport ratio is 1.3 for primary aerosol components. Secondary PM constituents are net accumulated in Berlin via advection as well as via chemical production. Nitrate has been proven to play an important role in the accumulation process and behaves differently from sulphate and ammonium. The main nitrate precursors are nitrogen oxides which are produced by humans. Thus, a direct connection between Berlin and nitrate accumulation may exist.

Sulphate and secondary organic carbons accumulation in Berlin is due to advection from the South-East (ca. 40%), while all other components show a

preferred inflow from the west. The overall inorganic ions budget is in equilibrium giving neutral aerosols except of summer, when high ammonium production is responsible for alkaline PM in Berlin.

The comprehensive mass budget analysis can be used as a tool in validating the RCG-model showing clearly shortcomings in the parameterisation of local emissions such as wind-blown dust. We belief that the parameterisation presently implemented in the model is not adequate as it is developed for non-urban environments. Nevertheless, the measurements show a high percentage of "unknown particles" which to some extend are believed to be mineral dust from the surface. Thus the wind-blown dust approach in RCG has to be improved upon

EC and OC are treated as part of primary fine PM and behave like these. There is no specific treatment of hydrophobic to hydrophilic mutation in the black carbon part. A process parameterisation for EC and for primary OC should be added to the model. Furthermore, specific measurements could reveal the importance of the process in scavenging characteristics or in general in removal of EC. EC is overestimated in the city of Berlin and underestimated in the rural surrounding. This might explain at least theoretically this behaviour of being accumulated too much in the city, not being wash-out. Wet processes have to be described better in the model, but also in dedicated measurements.

There are no known chemical removal processes for sulphate. Wet deposition is the only process which effectively removes particulate sulphate form the atmosphere. Nevertheless, RCG simulated too steep gradients between SO_4 in the city and SO_4 outside the city. This might be due to the local emission sources present in the emission inventory. Reaction rates of sulphate production might be revisited in the light of this not-observed gradient or - what is even more feasible - the emission source strength might be checked for correctness.

For a corroboration of the findings about the importance of single processes, photochemically active trajectory analysis could give great benefit in understanding the fate of remotely freed aerosol precursors or components.

On the other hand, the importance of primary particles' transport has been stressed. A field campaign in and outside the city of wind induced particles in the air and their origin would be important in understanding the origin of mineral dusts in the air.

The implemented process and budget analyses tool will be used in future studies and can give insight how individual processes will change their importance when input-data will change for example in future emission scenarios or in future meteorological situations with higher temperatures and lower precipitations. Furthermore, sensitivity studies can corroborate the importance of individual processes.