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Modelling of Atmospheric Chemistry Transport Processes

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Regional modelling of atmospheric trace gases and particulate matter is of major importance for air pollution studies as well as climate considerations. In this context, the chemistry transport model system LM-MUSCAT is applied for several air quality studies and the investigation of local climate effects. Two selected applications from different objects of research are presented in the paper. The first study enables a detailed quantification of the contributions of cooling tower emissions to particle concentration levels in specific Saxonian urban areas. In the second project, the influence of Saharan dust emissions on radiative forcing and, hence, on the meteorology is quantified. These regional effects are also relevant for climate change studies. Regional scale models are well suited for simulation of individual dust storm events, or for comparisons with *in-situ* observations made during field experiments. Improvements in the parameterisation of dust processes which are obtained by such regional model investigations can help to improve parameterisations in global scale models. Both applications require a detailed process description as well as a high spatial resolution. Such simulations are very expensive in terms of computing time and demand the use of powerful parallel computers.

1 Introduction

The physical and chemical processes in the atmosphere are very complex. The systems of differential equations resulting from atmospheric chemistry transport models are nonlinear, highly coupled and extremely stiff. Therefore, CPU costs are very high for advanced air pollution models containing a large number of species. Furthermore, the physical and chemical processes that determine the distribution of air pollutants occur simultaneously, coupled and in a wide range of scales. Here, the lack of adequate resolution limits the ability to accurately model individual processes and their interactions. For example, when plumes are injected into coarse grid cells in regional models with a uniform grid, the emitted material is diluted immediately within the cell and the details of the near field chemistry are lost. Multiscale models can provide finer resolution in certain key regions.

The modelling department of the IfT has developed the state-of-the-art multiscale model system LM-MUSCAT[19,20]. It is qualified for process studies as well as the operational forecast of pollutants in local and regional areas. The model system consists of two online coupled codes. The operational forecast model LM[4] (Local Model) of the German Weather Service is a non-hydrostatic and compressible meteorological model and solves the governing equations on the basis of a terrain-following grid. The model includes the dynamic kernel for the atmosphere as well as the necessary parameterisation schemes for various meteorological processes, boundary conditions and surface exchange relations. It describes the atmospheric flow and phenomena between the meso-β and micro-α scale (i.e. grid resolutions from 50 km to 50 m), in particular near-surface properties, convection, clouds, precipitation, orographical and thermal wind systems. The model is capable of self-nesting and four-dimensional data-assimilation.

Driven by the meteorological model, the chemistry transport model MUSCAT[18,7] (Multi-Scale Atmospheric Transport Model) treats the atmospheric transport as well as chemical transformations for several gas phase species and particle populations. The transport processes include advection, turbulent diffusion, sedimentation, dry and wet deposition. Due to the online coupling between LM and MUSCAT, the calculations exploit the actual properties of the atmosphere. The implicit-explicit time integration scheme of MUSCAT operates independently from the meteorological model, thus allowing for autonomous time steps and different horizontal grid resolutions in selected regions of the model domain. For this purpose, the required LM fields (e. g., wind, temperature, humidity, exchange coefficients) are interpolated temporarily and spatially. The chemical part of MUSCAT contains the gas phase mechanism of RACM[15] considering 76 reactive gas species with 239 reactions. Secondary particle formation and appropriate interactions with the gas phase are also included. The radiation activity and biogenic emissions are calculated autonomously, whereas information on the cloud cover, temperature, and other meteorological parameters is taken from LM. The anthropogenic emissions in Saxony are accounted for by several spatial registers of point, line, and area sources (including plume rise). Annual averages of emissions are disaggregated in time (monthly, weekly, daily). EMEP emission data were used in the outer region.

Both models work widely independent and have their own separate time step control. Coupling between meteorology and chemistry transport takes place at each horizontal advection time step only. Inter-processor communication is realised by means of MPI. The parallelisation is performed by domain decomposition techniques, but LM and MUSCAT use different horizontal grid structures. In MUSCAT, finer and coarser resolutions can be used for individual sub-domains in the multiblock approach[7]. This structure originates from dividing the meteorological grid into rectangular blocks of different sizes. The code is parallelised by distributing these blocks on the available processors. In the first version, both parallel codes work on their own predefined fraction of the available processors. This fixed *a priori* partitioning of the processors leads to imbalances between LM and MUSCAT. Therefore an alternative coupling scheme is implemented which guarantees the load balance between the both codes[9].

2 Formation and Dispersion of Secondary Aerosols by Single Cooling Towers Modelled for an East-German Region

Motivation. Fine and ultra-fine particles are suspected to cause damages in human health and natural environment. Several methods to reduce emissions by traffic, industry, agriculture and other sources have been successfully realised. Nowadays, the focus of environmental sciences and politics is directed towards understanding the physico-chemical formation and growth processes of secondary particles rather than the primary production[2]. Modifying factors in this context are the meteorological conditions, and the variable effective heights of the dominant emission sources. Therefore, long-term real weather simulations were performed in a mesoscale industrial region of Saxony. To investigate, to which extent the plume exhaust of the natural draft cooling towers in Boxberg and Lippendorf contributes to the formation of secondary aerosols. In spite of the relatively low emissions of primary particulate matter (PPM), interferences of the air quality are expected from the formation of sulfate and nitrate aerosols by the SO2 and NOx gas emissions and reactions with ammonia.

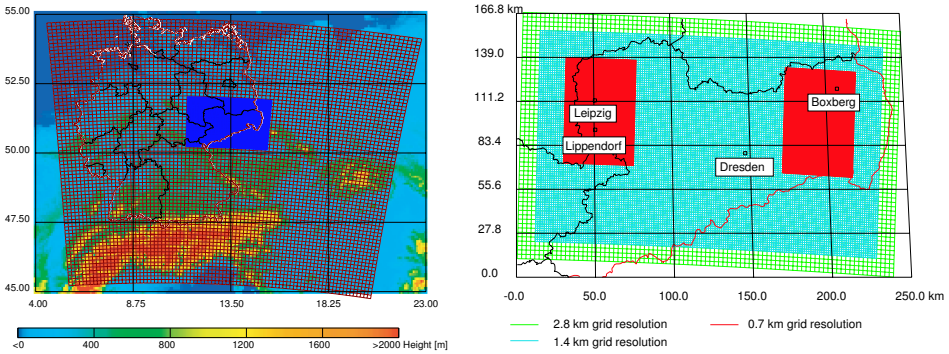

Figure 1. Model regions of Central Europe (left side) and Saxony (right side) with structure of nesting and grid refinement, respectively.

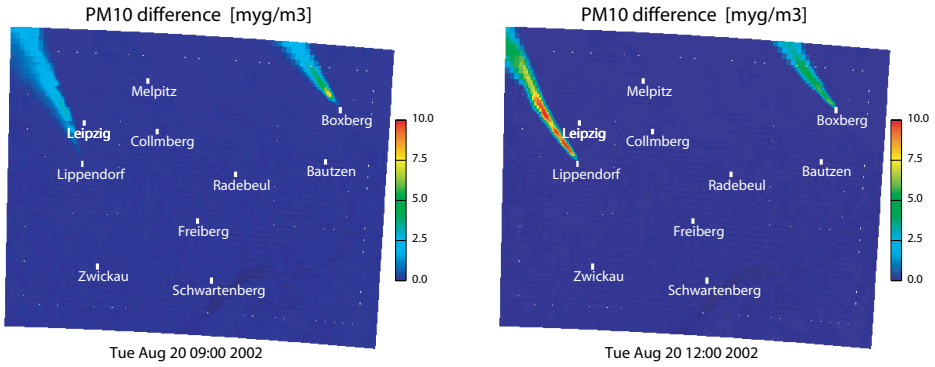

Figure 2. PM_10 concentration in Saxony caused by the emissions in Boxberg and Lippendorf at August 20-th 2002, 9:00 LT (left side) and 12:00 LT (right side).

Model setup. The study mainly aims at problems of the secondary formation and growth of anorganic particles with sizes below 10 μm (PM10). The dominant contribution to mass accretion is provided by the heterogeneous condensation of gaseous compounds on preexisting aerosols[1], involving pollutants such as ammonia and sulfuric or nitric acid (generated by complex pathways from the precursor species SO2 and NO2). The model system LM-MUSCAT was applied in a nested hierarchy with the superior control by the global reanalysis data of GME (Fig. 1). The innermost region of interest covers an area of 240 km x 156 km (Saxony) with variable resolution between 2.8 km and 0.7 km, where the finest grid was arranged around the dominant emission sites Boxberg and Lippendorf. The outer region extends over about 1200 km x 1000 km (Central Europe) with a constant resolution of 8 km in the main part. The model simulation in this domain generates the initial and boundary concentrations for the inner region.

Two typical summer/winter periods in 2002 each of 36 days were chosen for the simulations. The plume rise of the cooling towers in dependence on the meteorological conditions was accounted for by utilizing the adequate model of Schatzmann and Policastro[12]. One virtual neutral tracer per cooling tower was additionally emitted for identification purposes. Considering the meteorological and emission/immission conditions within the reference periods, two 8-day intervals were selected, for which the simulations on Saxony were repeated with the cooling tower emissions switched off. Thus, the difference between the switch-on/off simulations allows extracting the direct and indirect influence of the corresponding emissions on the immission situation[3]. The outstanding SO2 production rates by the sites Boxberg (1.2 t/h) and Lippendorf (2.1 t/h) as compared to the extremely small dust emissions (0.04 t/h, 0.06 t/h, resp.) actually require the examination of the secondary formation of particulate matter. In this respect, the assumed partitioning of the cooling tower SO2 emission data (88% SO2, 12% SO4 ions) is of great significance for the results.

Simulation results. The results are summarised as follows (an example is shown in Fig. 2):

1. Significant direct or indirect influences of the cooling tower gas emissions on the particle immission situation can be noticed only within limited zones of very narrow (highly concentrated) plumes and mainly for several hours about noon in summer.
2. The intrinsic particle formation caused by the considered emission sites frequently reaches peak values of about 10 μg/m^3 PM10 (maximal 20 μg/m^3), consisting predominantly of ammonium sulfate. Primary particles and ammonium nitrate formation contribute to only 10%.
3. The small probability for maximum plume intensities at a fixed location on the surface lowers the temporal averaged particle exposition down to 0.1 μg/m3 PM10.

3 Regional Modelling of Saharan Dust Aerosol

Motivation. Soil derived mineral dust contributes significantly to the global aerosol load and is suspected to impact the climate system by changing the energy balance of solar and thermal radiation[6]. The worldwide largest sources of dust are located in North Africa; about 50-70 percent of global dust emissions are estimated to originate from the Saharan desert. Radiative forcing by soil dust aerosol is complex, since dust not only scatters but also partly absorbs incoming solar radiation, and also absorbs and emits outgoing longwave radiation ('greenhouse effect'). Any changes in atmospheric dust loads result in changes of the radiation balance and surface temperatures. The magnitude and even the sign of the dust forcing depends on the optical properties of the dust, on the vertical distribution of the dust, on the presence of clouds, and on the albedo of the underlying surface[8]. Dust can also influence the climate indirectly by altering cloud optical properties[10]. The research project SAMUM[13] (2005-2007)) has the goal of quantification of radiative properties and effects of Saharan dust aerosols by means a field experiment in Morocco.

Model setup. The model computes dust emissions in non-vegetated areas depending on surface wind friction velocities, surface roughness, soil particle size distribution, soil moisture, and snow cover[16]. Soil particle size distributions are derived from global soil texture data. Enclosed topographic depressions are treated as preferential 'hot-spot' source regions, and the soils in these areas are assumed to consist of uniformly textured particles

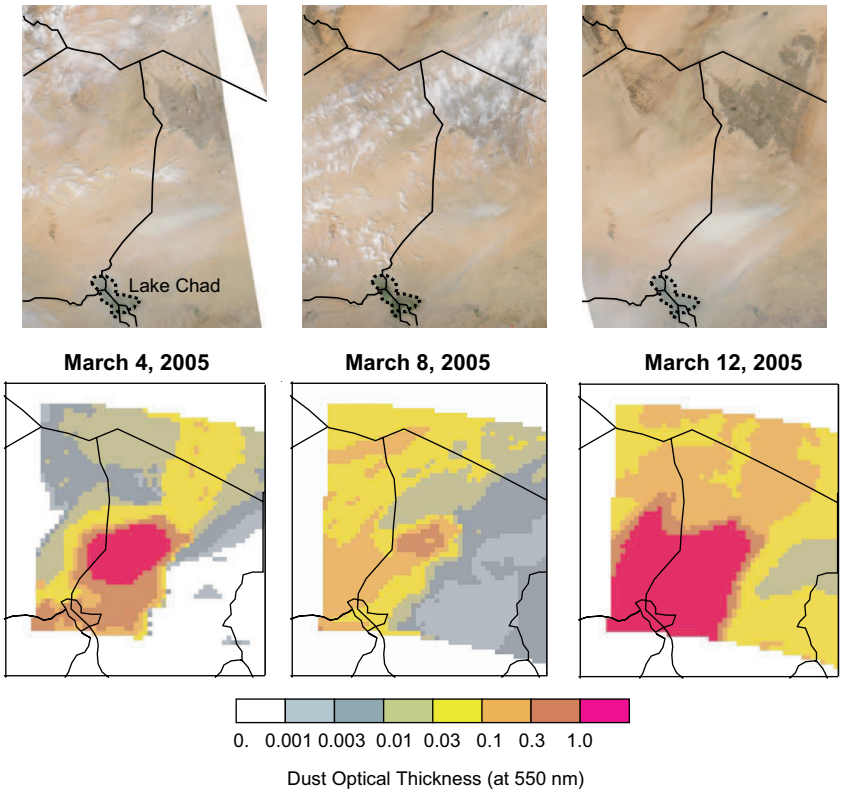

Figure 3. Dust episodes during the BODEX field study: True-colour retrievals by the NASA MODIS instrument on the Aqua satellite for the days March 4, March 8 and March 12, 2005 for the Lake Chad region (Chad) (top panels, source: NASA) and dust optical thickness for the same days simulated with the regional dust model at 7 km horizontal resolution (bottom panels).

which disaggregate into micrometer-sized particles during saltation events. The threshold friction velocities which must be reached for initiation of dust emission are computed depending on soil particle size distribution following the approach by Marticorena and Bergametti (1995), but assuming constant surface roughness within each model gridcell. Size distributions of the mobilised dust depends on surface properties as well as the surface wind speed. This dust emission scheme is coupled online into LM-MUSCAT. Emission fluxes are computed with surface winds and soil moisture fields from the meteorological model LM, dust is transported as passive tracer in 5 independent size bins covering the size range from 0.1 to 25 microns with the MUSCAT tracer scheme, and the aerosol deposition parameterisation in LM-MUSCAT is adapted for dust aerosol properties. The development of the regional Saharan dust model started late 2004, and tests of the model are carried out for previously observed Saharan dust episodes.

Simulation results. So far, the performance of this regional dust model has been tested for two well documented events of Saharan dust transport to Europe in August and October 2001, and a dust storm that occurred in the Bodele depression (Chad) in March 2005 during

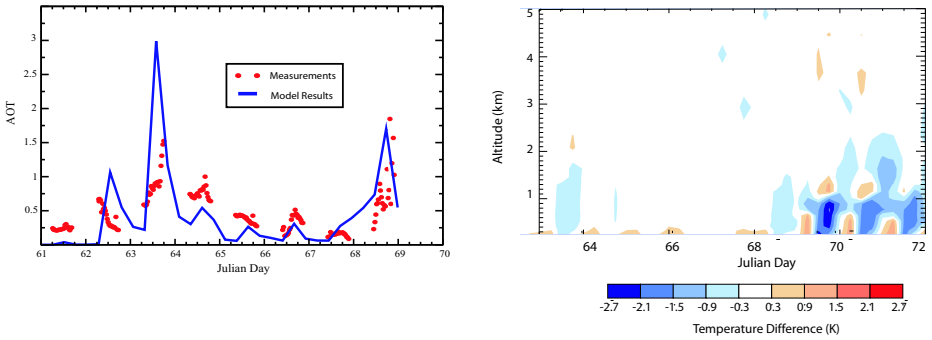

Figure 4. Modelled dust optical thickness at BODEX field site (at 16.5N, 18.3E) compared to sunphotometer measurements (left) and vertical distribution of simulated differences in air temperature compared to a simulation without dust forcing (right) for March 1 to March 12, 2005.

the British BOdele Dust EXperiment (BODEX). We present first results for this latest dust event, for which *in-situ* measurements of meteorological parameters like wind speed and surface temperatures as well as measurements of dust optical properties are available. The Bodele depression, which lies between the Tibesti Mountains, Lake Chad, and the Djouf region in Mali is the most active dust source in the Sahara desert, where the presence of dust is detected throughout the year. For simulation of this dust episode, dust production and transport was computed for an area of ca. 200000 km^2 using a horizontal model resolution of 7 km. Deposition of the large dust particles is mostly by dry deposition, the stronger gravitational settling of larger particles leads to changes in the size distribution of atmospheric dust. Dust optical thicknesses are computed from the size-resolved dust concentrations. Within LM-MUSCAT, changes in radiative fluxes caused by dust are computed online, using dust aerosol optical properties derived from sunphotometer measurements (Todd et al., submitted). The radiative forcing by dust aerosol is allowed to impact on the meteorology and feeds back on dust emissions and atmospheric distributions.

With an updated parameterisation for the dust emission model, the BODEX dust case was simulated twice, once including the dust feedback on climate, and once with dust as passive tracer. Size-resolved dust emission fluxes for the BODEX location and time period of the field study are computed with the regional dust model. Although satellite retrievals of the regional extent of the dust plumes are difficult to obtain over highly reflective desert surfaces, a comparison of true-color retrievals by the MODIS instrument onboard the NASA Aqua satellite with model results for the days March 4, 8, and 12, 2005 shows good agreement in the occurrence of modeled dust plumes with the remote sensing images (Fig. 3). The variation of modeled dust aerosol optical thickness during the dust episode compared to measurements from the sunphotometer instrument (Fig. 4, left) also shows good agreement. Only on March 4 (Julian day 63), the modeled optical thicknesses briefly exceed the measurements by about a factor of two. The difference in modeled air temperatures for the field site between the LM-MUSCAT model run in which dust was included as radiatively interactive tracer and temperatures computed with the LM without including dust aerosol for the same period shows the effects of the presence of dust aerosol on the atmospheric temperatures (Fig. 4, right). Atmospheric cooling occurs during daytime when

the atmospheric dust scatters or absorbs a part of the incoming sunlight, which results in a reduction in solar radiation fluxes below ca. 1 km height. This cooling is only evident for days when the dust optical thickness exceeds the value of 1, with a maximum cooling reaching about 3 degrees. During night the presence of dust caused a slight warming by about 0.5 K compared to the simulation without dust, because the large dust particles absorb a small part of the outgoing terrestrial radiation. Surface temperature measurements show a drop in maximum day temperatures by about 10K from March 10 to March 11, which is well matched by the modeled surface temperatures.

4 Concluding Remarks

The availability of a full coupled meteorology chemistry aerosol model with well defined interfaces to external emission inventories and regional scale forcing data for chemistry and aerosols, e.g., *EMEP*, is an important step forward in regular model applications on behalf of the European Community or other entities. The model system LM-MUSCAT is also a powerful instrument for process studies[14] and the interpretation of field experiments[5]. In this context, the nested mesoscale model LM-MUSCAT could fill the gap between local measurements and the rather coarse resolution of large scale models. The model performance and parallel efficiency is encouraging[20].

Acknowledgments

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