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INSTITUTE FOR
TROPOSPHERIC RESEARCH
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GOTTFRIED WILHELM LEIBNIZ

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2002

10 years Institute for Tropospheric Research e.V. Leipzig

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10 years evolution of the troposphere over Eastern Germany and worldwide

INSTITUTE FOR TROPOSPHERIC RESEARCH
WISSENSCHAFTSGEMEINSCHAFT GOTTFRIED WILHELM LEIBNIZ

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Introduction & overview

Introduction and overview

In the outskirts of Leipzig, on the campus of the former Academy of Sciences, in close neighborhood of the Environmental Research Center, other research establishments and businesses you find the Institute for Tropospheric Research. It was founded in 1991 for the investigation of physical and chemical processes in the polluted troposphere (roughly the first 10 km of our atmosphere).

Fig. 1: *IfT main building.*

Meanwhile a well-defined and globally unique research profile emerged with a focus on aerosols, i.e. small airborne particles, and clouds. Despite their minute absolute amount, aerosols and clouds are essential parts of the atmosphere because they control the budgets of energy, water and trace substances of the Earth System. The research interest in these highly disperse systems is stimulated foremost by their potential change through human activities. These system changes feed back into the anthroposphere not only through regional and global climate change but also directly through health effects of inhaled haze and fog particles.

Despite strong connections between humans, aerosols, and clouds important physico-chemical processes of aerosol and cloud formation and the relationships with climate and health are poorly understood. This limitation is mainly due to analytical difficulties with the very small samples and with the complex behavior of tropospheric multiphase systems, in which individual processes seldom can be distinguished. In climate research this limitation is reflected in much larger uncertainties in predicted anthropogenic aerosol and cloud effects in comparison to numbers published by the Intergovernmental Panel on Climate Change for additional greenhouse gases.

Rapid advances in our understanding of tropospheric multiphase processes and an application of this process understanding to the prediction of the consequences of human impacts can only be expected from concerted approaches from several directions. Consequently, the Institute for Tropospheric Research conducts field studies in several polluted regions parallel to the development of analytical methods for aerosol and cloud research.

Fig. 2: Particle sampling systems during the INTERCOMP 2000 campaign at the research station Melpitz.

These tools are not only applied in field experiments but also in extensive laboratory investigations, which form a second major activity. A third and equally important approach consists of the formulation and application of numerical models that reach from process models to regional simulations of the formation, transformation and effects of tropospheric multiphase systems.

Field experiments

The atmosphere is an aerosol, i.e. a carrier gas mixture with suspended solid and liquid particles. Field experiments elucidate the atmospheric life cycle and related processes of aerosol and cloud particles. This task is vastly more difficult than comparable trace gas studies, in which only one number has to be known for each substance at each point in time and space. Particles sizes over more than six orders of magnitude occur in atmospheric aerosols and clouds, all of which play an important role in certain processes. All condensable substances of the Earth System can be found in the aerosol and a large number of them contributes to climate and biospheric effects. As a consequence of this multidimensional system essential aerosol and cloud properties are not well-established on a global scale yet.

The uncertainty and thus the studies of the Institute for Tropospheric Research start with particle sources. The combustion of fossil and contemporary fuels is one of the most prominent aerosol sources. However, these sources are still poorly characterised in terms of climate-relevant aerosol parameters. In collaboration with car manufacturers the institute establishes size dependent particle emission data of cars at test stands, in particular in the nanometer size range that was not covered by conventional emission studies. According to long-term measurements of the institute in a street canyon the car related emissions of particles and their precursor gases are subject to strong physical and chemical transformations even before they reach the sidewalk. These transformations will be investigated by a new mobile aerosol laboratory on a trailer behind a moving car in traffic.

Emission studies at cars are complemented by measurements at stationary combustion sources. Here the research focuses on particle properties that determine the absorption of solar radiation. Dedicated methods have been developed for the analysis of soot components, a major absorber of sun light. With aerosol measurements at welding stands finally the Institute for Tropospheric research characterises toxic industrial particle emissions. Health related aerosol studies will be expanded in the future in collaboration with the Environmental Research Center with coupled indoor and outdoor aerosol experiments and concurrent clinical investigations in the urban region of Leipzig.

Fig. 3: Impression from the SATURN experiment.

Even the largest highly polluted regions in the plumes of North America, Europe, the Indian subcontinent and Eastern Asia are insufficiently characterised in terms of aerosol burdens and ensuing climate effects. Thus, the institute focused the most recent field campaigns on the European and Indian plumes. These experiments were conducted within a large framework of international collaboration.

As baseline reference an austral area near Tasmania has been studied. By means of an intercontinental commercial aircraft, the results of

the regional experiments are connected through regular CARIBIC flights between Germany, the Indian Ocean and Southern Africa (cf. Fig. 4).
Process studies are conducted at suitable locations such as mountain observatories, tethered balloons and over cooling towers of large power plants.
These experiments are dedicated to particle nucleation, particle processing through clouds and the influence of anthropogenic aerosols on the optical properties of clouds.
The vertical aerosol distribution over Europe is observed with the worldwide first coordinated lidar network EARLINET (European Aerosol Research Lidar Network).

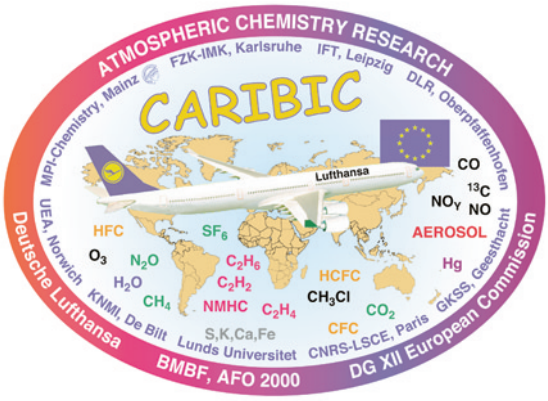

Fig. 4: *The logo of the international project CARIBIC (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container).*

Modeling

For the description of complex atmospheric processes, model systems of varying dimensions and complexity are developed, tested and applied to micro to mesoscale problems. The ultimate goal is to simulate the many interactions between aerosol particles, gases and clouds in a coupled three-dimensional meteorology-chemistry-transport model. With this model system as a toolbox scientific as well as legal tasks are addressed.
A long-term goal of the European Union states that no critical loads may be exceeded in the acidification and eutrophication of soils and water and for surface ozone. For the reduction of these critical loads, guidelines are requested for national limits in sulfur dioxide, nitrogen oxides, ammonia and volatile organics. Already today European guidelines define limits in particulate mass concentrations below 10 µm (PM10). It is expected that these guidelines will be extended shortly to smaller particles (PM2.5). To meet these guidelines significant regionally different efforts will have to be undertaken in order to reduce emissions.
Models are indispensable tools in the search for efficient and cost-effective means of meeting present and future limits of gaseous and particulate air pollution. As done in the past, the Institute for Tropospheric Research will give advice to the Saxonian government on preventative air pollution measures.
The knowledge of the present emissions is a prerequisite for the development and evaluation of strategies for emission strategies directed and an improvement of air quality. As a basis for the simulation of present and future scenarios the institute developed a dynamic emission inventory for Saxony. This inventory enables us to supply emission data for natural and anthropogenic air pollutants (SO_2, NO_x, CO, NH_3, non-methane hydrocarbons, dust, heavy metals, polychlorinated dibenzo-dioxines/-furanes. For extended investigations emissions of the greenhouse gases CO_2, CH_4 and N_2O can be supplied for numerical simulations of the transport and transformation of air pollutants over Saxony. The use of a geographical information system (GIS ArcInfo) allows a spatial resolution of emission data and a connection with political and or geographical structures including a digital road map and land use data.

Laboratory experiments

In atmospheric research, there is a continuous development of physico-chemical models for the description of the most relevant process. These

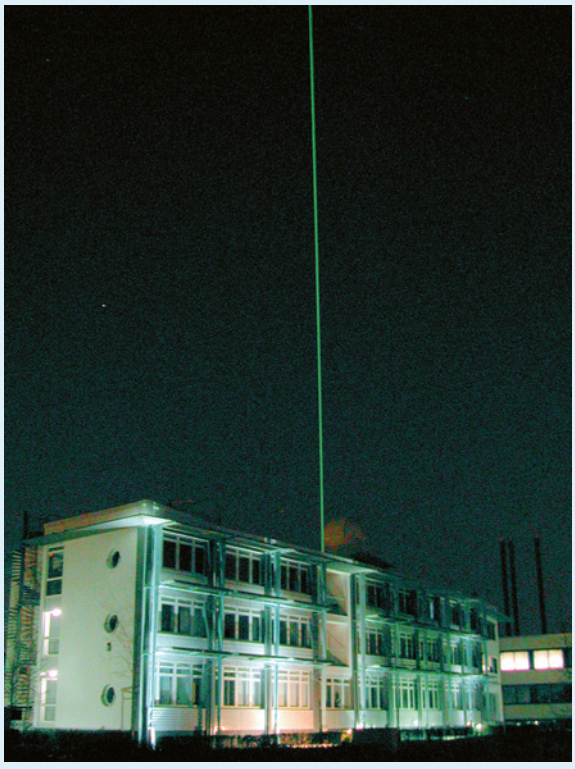

Fig. 5: *Multiwavelength Raman lidar measurement after sunset performed in the framework of the European Aerosol Research Lidar Network (EARLINET).*

Fig. 6: The IfT laminar flow tube reactor (IfT-LFT).

models are based on process parameters, which need to be determined in physical and chemical laboratory experiments.

In the physics section of the institute laboratory experiments cover the development of a large number of methods to characterize atmospheric particles and drops, in particular their size distribution and thermodynamic properties. Complex measuring and sampling systems are being designed for the characterization of cloud drops and interstitial particles.

Spectroscopic techniques such as the Differential Optical Absorption Spectroscopy have been developed for the analysis of trace gases and aerosol particles. The ongoing development of the multi-wavelength aerosol LIDAR (Light Detection and Ranging) technique includes the measurement of atmospheric state parameters such as temperature and relative humidity. A new Doppler LIDAR for vertical wind profiling will perform first test measurements in the autumn of 2003. Graphitic carbon is specified and quantified with a dedicated Raman spectrometer combined with multi-wavelength absorption measurements on aerosol samples.

Process-oriented laboratory studies are being carried out jointly by the physics and chemistry sections in two main areas. The first of these activities concerns a laminar flow tube reactor in which particle formation from (SO_2) and organic precursors (e.g., terpenes) is being investigated. In the second activity, the transition from a moist aerosol to a cloud will be simulated in a laminar flow channel.

The chemistry section conducts several process-oriented laboratory studies. Gas phase reactions of the radicals OH and NO_3 are being investigated in flow reactors. These reactions are important for ozone and particle formation caused by biogenic and anthropogenic emissions of volatile hydrocarbons. The chemical identity of atmospheric particles will be characterized in reaction chambers. In a single drop experiment, phase transfer parameters of trace gases and radicals are being determined for different chemical species and surfaces. Mechanisms of non-radical oxidations in the liquid phase are being studied with the stopped-flow technique and optical detectors. Experiments with radical reactions in the liquid phase form a core activity of the laboratory experiments because of their importance for processes in haze particles, fogs and clouds. For the understanding of the oxidation of organic trace gases in the tropospheric multi-phase system a large number of reactions with the OH and NO_3 radicals are being studied as well as reactions of halogenated oxidants. The latter species are of interest for the emission of reactive halogen compounds from sea salt particles.

Several laboratory experiments are dedicated to the chemical characterization of atmospheric organic aerosol components. Besides the conventional combustion techniques, mass spectroscopic and chromatographic techniques coupled directly to analysis by mass spectrometry or capillary electrophoresis with different sampling and segregation techniques are being developed. The close cooperation of the physics and chemistry section has lead to the development of a patented sampling method for narrow well-defined particle size ranges that is coupled directly to the mass spectroscopic analyses.

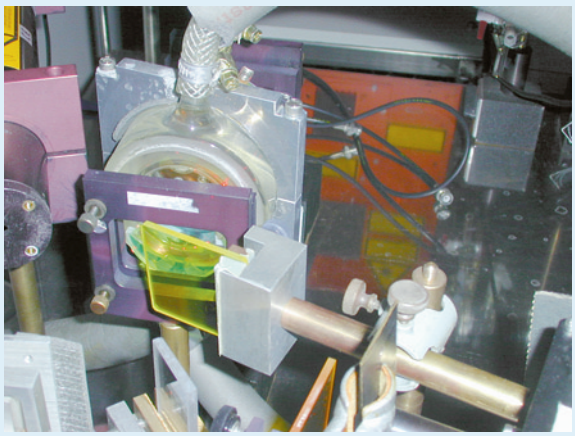

Fig. 7: A laser photolysis-long path laser absorption experiment for the study of nitrate radical kinetics in aqueous solution.

Detailed contributions *to current research projects*

SATURN: New particle formation in the lower troposphere

Holger Siebert, Frank Stratmann, Gerald Spindler, Ulla Wandinger, Birgit Wehner

From May 27 to June 14, 2002, the Institute for Tropospheric Research (IfT) carried out the field campaign “SATURN” to investigate new particle formation processes in the lower troposphere. The experiment took place in Melpitz which is the field research station of the IfT located about 40 km north-east of Leipzig. Members of all three departments of the IfT were involved in this project.

Motivation

The fundamental processes causing new particle formation, i.e., nucleation and subsequent growth into the size-range of a few nanometers are still not well understood. Most likely there is not only one mechanism that controls atmospheric nucleation processes. It is suggested by many authors that nucleation mechanisms may vary with altitude (Weber et al., 1999). Other researchers noted that favorable atmospheric conditions, such as turbulence due to breaking Kelvin-Helmholtz waves (Bigg, 1997) or boundary layer mixing processes (Easter and Peters, 1994; Nilsson and Kulmala, 1998), and atmospheric waves in general (Nilsson et al., 2000) can enhance nucleation rates by up to several orders of magnitude. However, most of the measurements reported in the literature are single-point measurements and therefore not suitable to gain insight into the above mechanisms.

In this work, which reports first results from the SATURN experiment, a new approach to gain further insight into possible processes controlling atmospheric nucleation processes is presented. Instead of performing only single-point measurements, vertical soundings of meteorological parameters such as wind, temperature, relative humidity, and particle number concentrations were carried out utilizing the balloon-borne measurement platform ACTOS (Airship-borne Cloud Turbulence Observation System). In addition, ground based measurements of meteorological, particle number size distributions, and gaseous precursor concentrations took place at the balloon site. Furthermore, at two additional locations (distance to the balloon site is about 40 km) particle number size distributions were measured. To gain information about the vertical structure and the development of the boundary layer, SODAR measurements were performed at the balloon site together with radiosondes, and a LIDAR was operated in about 50 km distance to the balloon site at IfT.

Meteorological situation

The SATURN experiment took place from May 27 to June 14, 2002. During this time period new particle formation events were observed on eight days. This paper focuses on June 3, 2002. During this day, the weather in central Europe was mainly influenced by a high pressure area leading from north Scandinavia over central Europe to the southeast of the Mediterranean Sea. This situation caused calm winds from southeasterly directions and sunny weather with only a few convective clouds at noon but no rain.

To gain information about structure and development of the planetary boundary layer (PBL), a multiwavelength aerosol Raman LIDAR was used at the Leipzig IfT-site. This instrument delivers the particle volume 180°-backscatter coefficient at 355, 532, and 1064 nm and the particle volume extinction coefficients at 355 and 532 nm as well as temperature and water-vapor mixing ratios.

Figure 1 shows the development of the PBL between 04:30 and 19:50 UTC on June 3, 2002, as measured with LIDAR at the IfT site in Leipzig. The time-height contour plot represents a measure of relative particle backscattering, which is shown, because measurements close to the LIDAR (i.e., close to the ground) are usually biased by the incomplete overlap between the laser beam and the receivers field of view. In the case of relative backscattering this overlap effect is canceled out. Thus, the observations cover the complete PBL. The elastic backscatter signal at 1064 nm is used for the presentation of Figure 1. This signal is most sensitive to changes in aerosol backscattering because of the low Rayleigh-backscattering contribution and is less influenced by transmission losses. Note that the LIDAR is sensitive to optically active particles only, i.e., backscattering is dominated by particles with radii > 50 nm.

Bluish-green colors in Figure 1 ($P_{rel} \approx 0$) indicate atmospheric regions with marginal changes in particle backscattering, whereas blue ($P_{rel} < 0$) and red colors ($P_{rel} > 0$) show strong negative and positive deviations from the mean, respectively. A

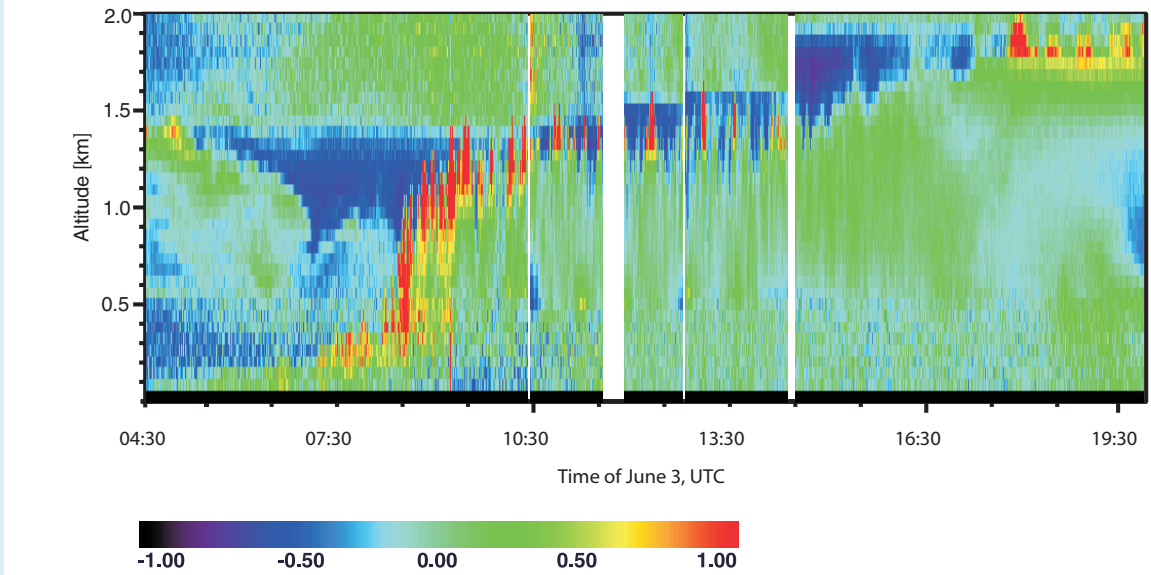

Fig. 1: Development of the planetary boundary layer in terms of the relative backscatter signal at 1064 nm observed with the IfT Raman LIDAR at Leipzig on June 3, 2002. The resolution is 60 m and 30 s.

rapid change between red and blue colors marks the atmospheric height regions of strongest dynamic activity, where particle-rich air is transported upwards and cleaner air moves downward. First updrafts developed below 400 m height between 07:00 and 08:00 UTC. A rapid increase of the mixing layer (ML) height from 300 to 1400 m associated with strong dynamic activity was observed between 08:00 and 09:30 UTC. From 09:30 - 15:00 UTC only slight variations of the PBL height around 1500 m occurred. In the late afternoon, a further increase of the PBL height to values around 1700 m, probably associated with a change in the airmass, was found.

Figure 2 shows a time-height contour plot of the backscattered signal intensity *S* measured with the SODAR (SOund Detecting And Ranging) on June 3, 2002 in Melpitz.

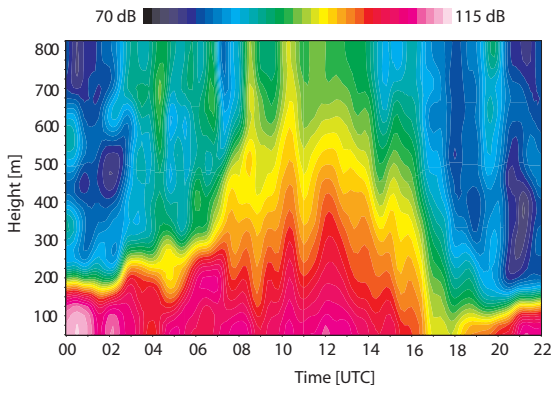

Fig. 2: Height-time contour plot of the backscattered signal intensity S as measured with SODAR in Melpitz on June 3, 2002. Sunrise is at 03:00 UTC, sunset is at 19:20 UTC, respectively.

One possibility to estimate the inversion height is the maximum of the vertical gradient of the signal *S*. In Figure 2, a local maximum of the gradient can be identified around 200 m between 00:00 and 03:00 UTC, after 03:00 UTC (sunrise) the gradient becomes smaller and the height of the maximum varies between 150 and 260 m. At about 07:00 UTC the nocturnal inversion layer breaks up and intensive vertical mixing begins. Within about one hour the ML rises from about 260 m to higher than ceiling of the SODAR. After sunset (19:20 UTC) again a strong vertical gradient of *S* with a maximum in about 150 m develops.

Figure 3 shows four vertical profiles of *T* (left) and r*H* (right) as measured with radiosondes. The first two profiles (launched at 05:30 and 06:30 UTC) show a significant inversion layer with positive temperature gradients below 260 m. The next two profiles (07:30 and 09:30 UTC) depict the situation after the inversion broke up, no inversion can be found over the entire profile which is in good agreement with the SODAR measurements. The humidity profiles show a strong decrease from about 70 to 50% above the inversion layer (first two profiles). A second jump from 50 to about 30% can be seen in 1000 m, which is likely an indication for the height of the nocturnal residual layer (RL). After the ground inversion disappeared, the humidity profiles show no significant structures up to the PBL height in 1200 (07:30 UTC) and 1400 m (09:30 UTC). The observations correlate well with the LIDAR data discussed above, however, the development of the PBL seems to be

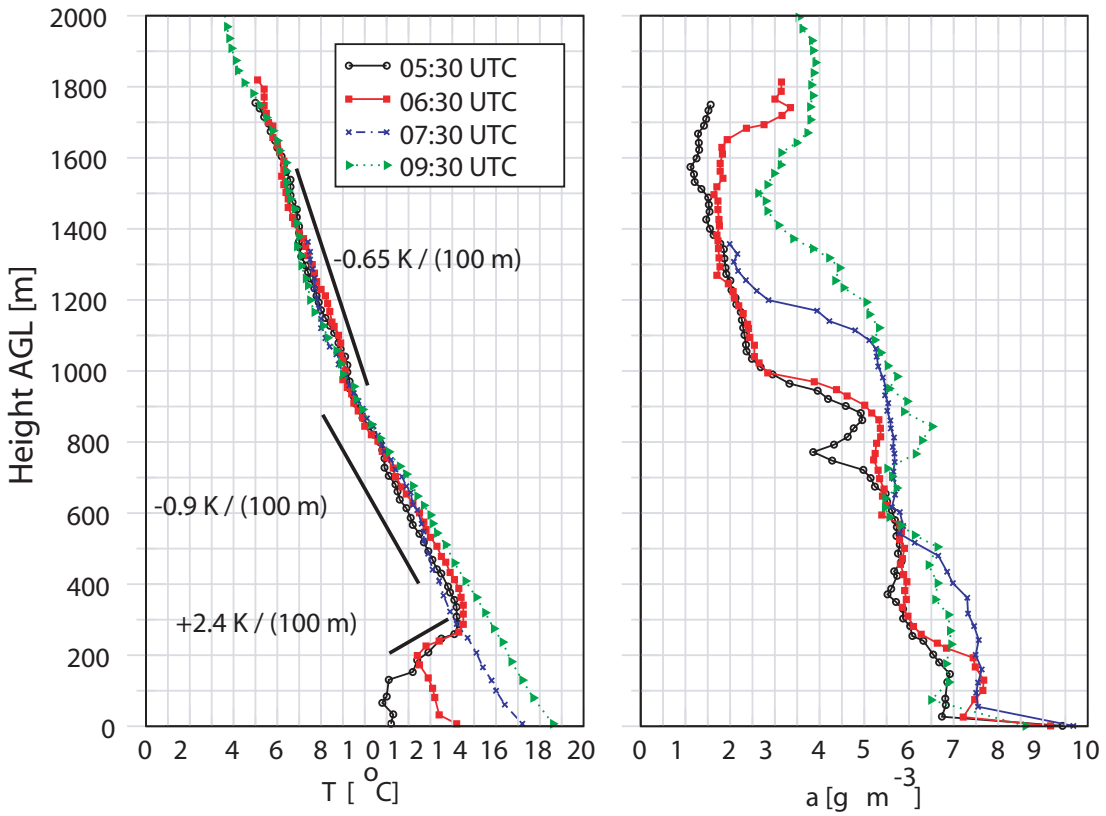

Fig. 3: *Vertical profiles of temperature T and absolute humidity as measured with rawinsondes at the Melpitz site. The depicted gradients are related to the sounding from 06:30 UTC.*

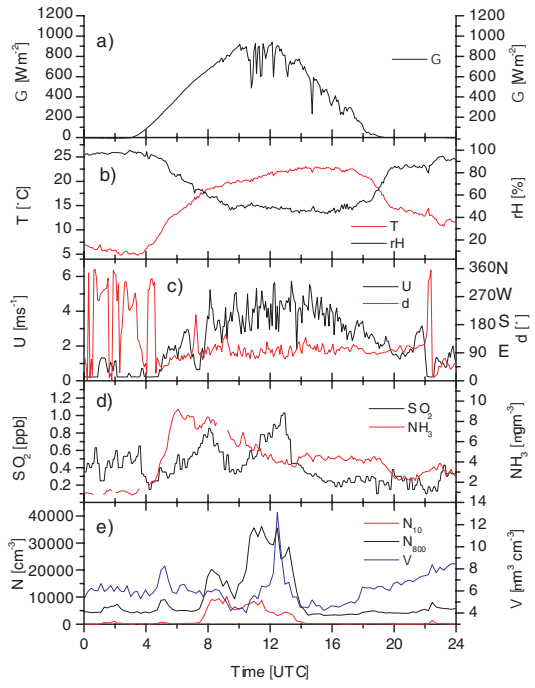

Fig. 4: *Diurnal variation of global radiation G, temperature T, relative humidity rH, wind speed U, wind direction d, SO_2, NH_3, particle number concentration (N10: 3 – 10 nm and N_{800}: 3 – 800 nm) and particle volume concentration V in Melpitz on June 3, 2002, at ground.*

time shifted for about 0.5 h between Melpitz and Leipzig which is obviously due to cirrus clouds over Leipzig.

Figure 4 shows results from the ground-based measurements of global radiation *G*, temperature *T*, relative humidity r*H*, wind speed *U*, wind direction *d*, SO_2, and NH_3 concentration, particle number concentration *N* (3 – 10 nm and 3 – 800 nm) and particle volume concentration *V* as function of time as determined in Melpitz on June 3, 2002.

From Figures 4 a-c, it can be seen, that June 3 featured an increase in global radiation (a radiation maximum near 1000 W m^{-2}) and temperature, and decreasing relative humidity during morning hours. Around 10:00 UTC the global radiation decreased and varies afterwards due to small cumulus clouds covering the sun intermittently. The wind speed was close to zero during the night and increased up to 4 m s^{-1} during the day forced by convection. The mean wind direction during the day was from east.

The SO_2 concentration (Figure 4 d) shows two peaks, the first one at 08:00 UTC, i.e., one hour after the break-up of the nocturnal inversion layer (up to 900 ppt) and a second one at 12:00 UTC (up to 1050 ppt). The ammonia concentration (Figure 4 d) increases fast in the early morning from 1 up to 9 µg m^{-3} which correlates well with the

global radiation and temperature. During daytime the NH_3 concentration is about 4-6 µg m^{-3}.
As can be seen in Figure 4 e, the number concentrations (N_{10}: 3 – 10 nm and N_{800}: 3 – 800 nm) show an significant increase at 07:30 UTC. N_{800} featured a second, higher maximum between 10:00 and 12:00 UTC, i.e., at the same time as the global radiation decreases due to the appearance of clouds. The volume concentration, as calculated from the number size distribution shows no correlation with the first morning maximum, it is even slightly decreasing between 08:00 and 10:00 UTC.

Balloon-borne profiles measured with ACTOS

Several vertical profiles and time series at a constant level were measured with ACTOS at June 3 in Melpitz. In this section, data concerning the static temperature *T,* the water vapor density *a*, the horizontal wind speed *U*, wind direction *d*, particle number concentrations N_I, N_{II}, and ΔN is presented. For the vertical profiles additionally the SO_2-concentration is given.

The first data set (Figure 5) was measured at 06:30 UTC during an descent from 830 m to GND. The temperature profile shows an inversion between 150 and 200 m with a mean gradient of about +2.8 K (100 m)$^{-1}$. Between 200 and 330 m the gradient is close to zero while for higher altitudes the profile shows an adiabatic lapse rate of -1 K (100 m)$^{-1}$. The water vapor density shows strong fluctuations within the ML with a slight decrease in the inversion from about 9.5 to 7 g m^{-3}. In the first 100 m above GND, *U* is close to 2 m s^{-1} while it has a local maximum around the inversion. In the RL, *U* varies between 3 and 4 m s^{-1} from easterly directions. The *N*-profiles show a clear cut between the mixing, inversion, and residual layer. In the ML N_{II} is nearly constant with height and with 250 particles cm^{-3} smaller than 10 nm. In the small region above the inversion (between 250 – 300 m) no small particles could be observed while higher up ΔN increases up to values of 800 cm^{-3} between 400 and 600 m. In this layer ΔN varies in the same range as in the time series shown in Figure 10. A similar behavior can be observed for the SO_2-concentration-profile which has also a

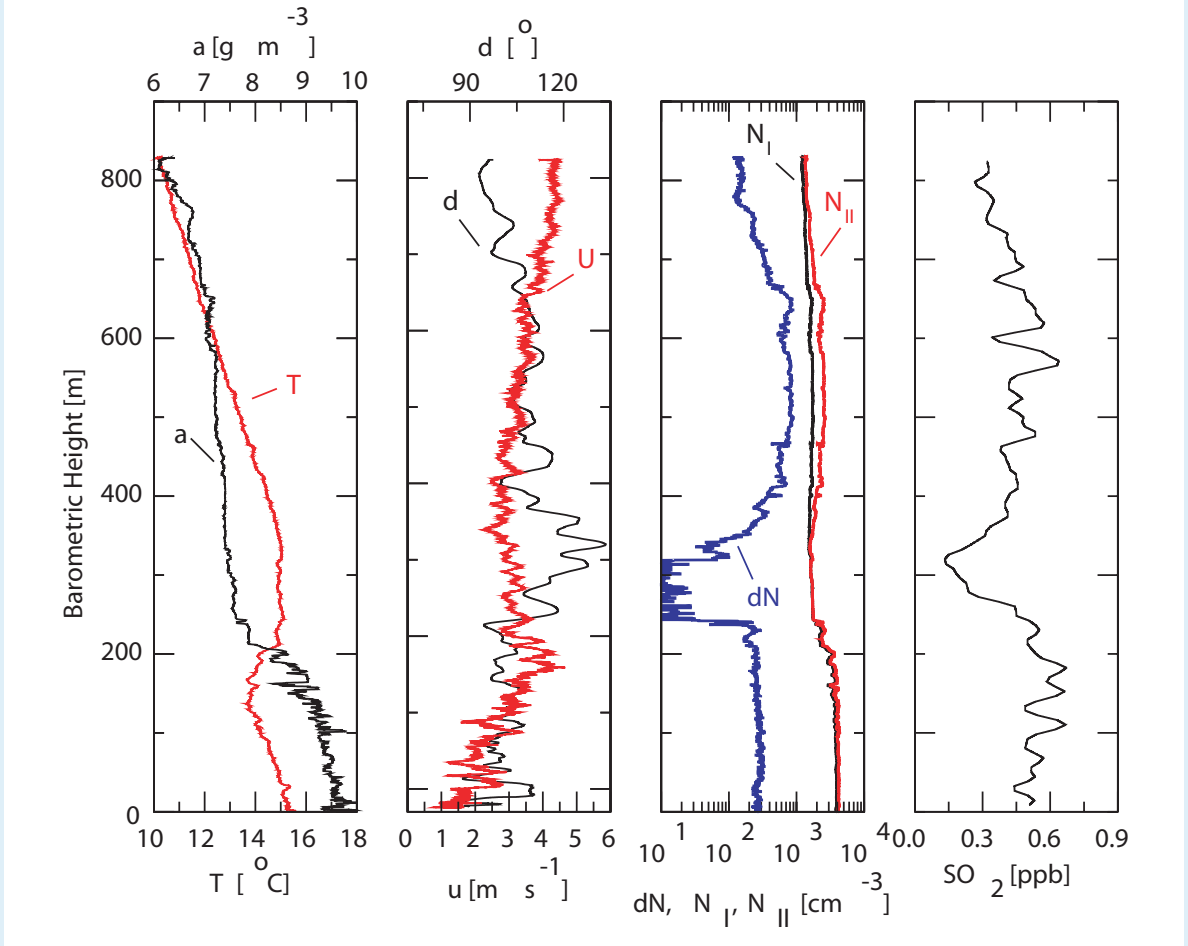

Fig. 5: *Vertical profile (descent from 830 m to ground, 06:31 - 06:53 UTC) of temperature T, absolute humidity a, horizontal wind speed U, wind direction d, particle number concentration N_I ($10 < D_p < 1500$ nm), N_{II} ($5 < D_p < 1500$ nm), ΔN ($5 < D_p < 10$ nm), and SO_2.*

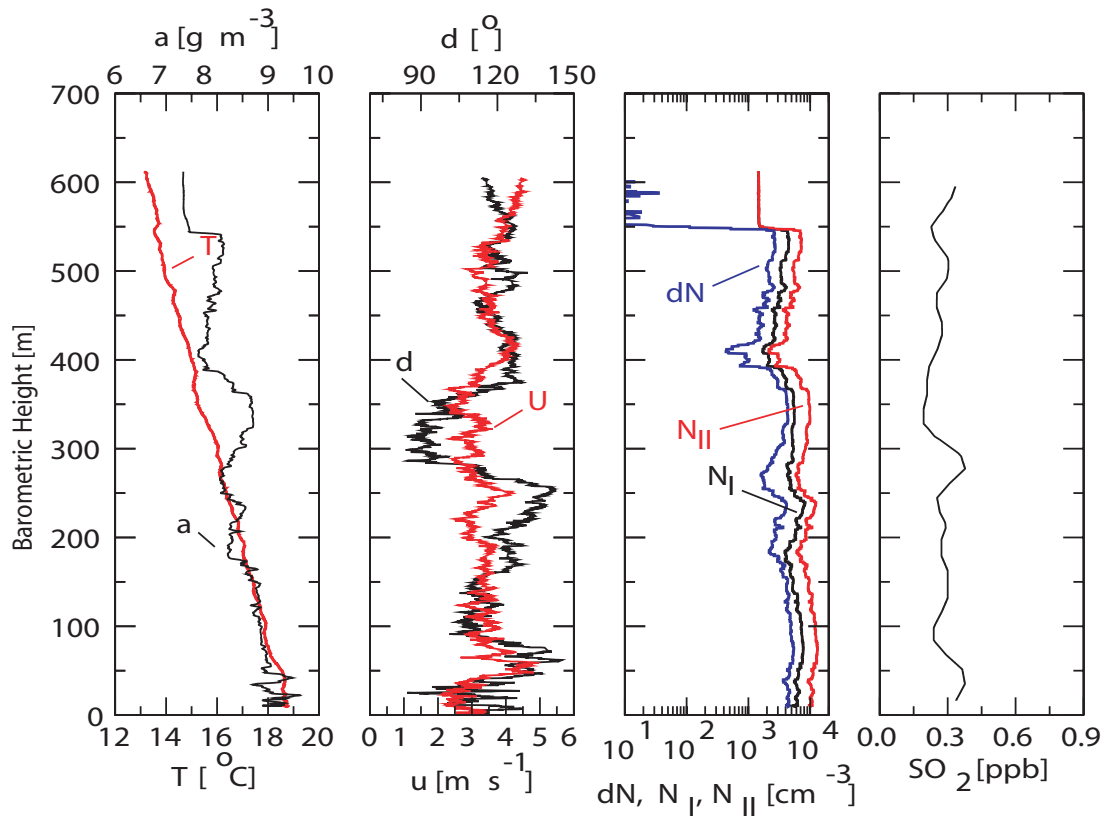

Fig. 6: *Vertical profile (ascent from ground to 610 m, 08:43 - 08:48 UTC) of temperature T, absolute humidity a, horizontal wind speed U, wind direction d, particle number concentration N_I ($10 < D_p < 1500$ nm), N_{II} ($5 < D_p < 1500$ nm), ΔN ($5 < D_p < 10$ nm), and SO_2.*

local minimum around 300 m.
The measured data suggests that at this stage, new particle formation has only been taking place in the RL and no particles are formed below the inversion. The small particles observed below the inversion are most likely due to traffic emissions as they were accompanied by high NO-concentrations.
Figure 6 shows vertical profiles measured about 2 h later than the measurements depicted in Figure 5. The profiles were measured during an ascent from GND to 610 m. The inversion layer in 200 m had disappeared and *T* shows a near-adiabatic lapse rate of -0.9 K (100 m)$^{-1}$ over the entire range. The *a*-profile exhibits some fluctuations over the complete profile but a sharp decrease in 550 m, i.e. in the same height where a small increase of *T* can be observed. In 350 m, *a* has a local maximum which corresponds well with a change of the wind direction from south-east to north-east and an increased *U*. The particle concentrations shows high values from the ground up to 550 m. In this range ΔN varies between 1000 and 5000 cm^{-3} and N_{II} between 2000 and 12000 cm^{-3}, respectively. Above 550 m ΔN decrease rapidly to zero and $N_{II} \approx N_I$ to about 1800 cm^{-3}. The SO_2-profile is nearly height independent with values around 0.3 ppb.
This all together indicates, that at this stage the ML has a height of about 550 m. After the vertical profiles were taken, the balloon was fixed again at 600 m. The corresponding time series (Figure 7), started about 10 min after the maximum height of the profiles shown in Figure 6 was reached.
The time series of *T* and *a* but also *U* and *d* show very similar structure, periods with high fluctuations are interrupted by periods with nearly no structure. The same intermittent behavior can be seen in the particle number concentrations. Most significant is ΔN which shows variations of several orders of magnitude with maximum values of 4000 to 5000 cm^{-3}. The total concentration N_{II} varies between 1300 cm^{-3}, which seems to be the background concentration (cf. also Figure 6), and maximum values of up to 12000 cm^{-3} between $t = 300$ and 340 s. This behavior can be explained with updrafts which temporarily penetrate into the capping inversion layer. Therefore, sometimes ACTOS is inside of such an updraft with highly increased turbulence and sometimes ACTOS is above the inversion where the turbulence is much weaker and N_p is much less than in the ML.
The second data set altogether indicates, that at this stage, the ML has a height of about 550 m and no new particle formation is taking place in the ML and RL. Particles are only transported by

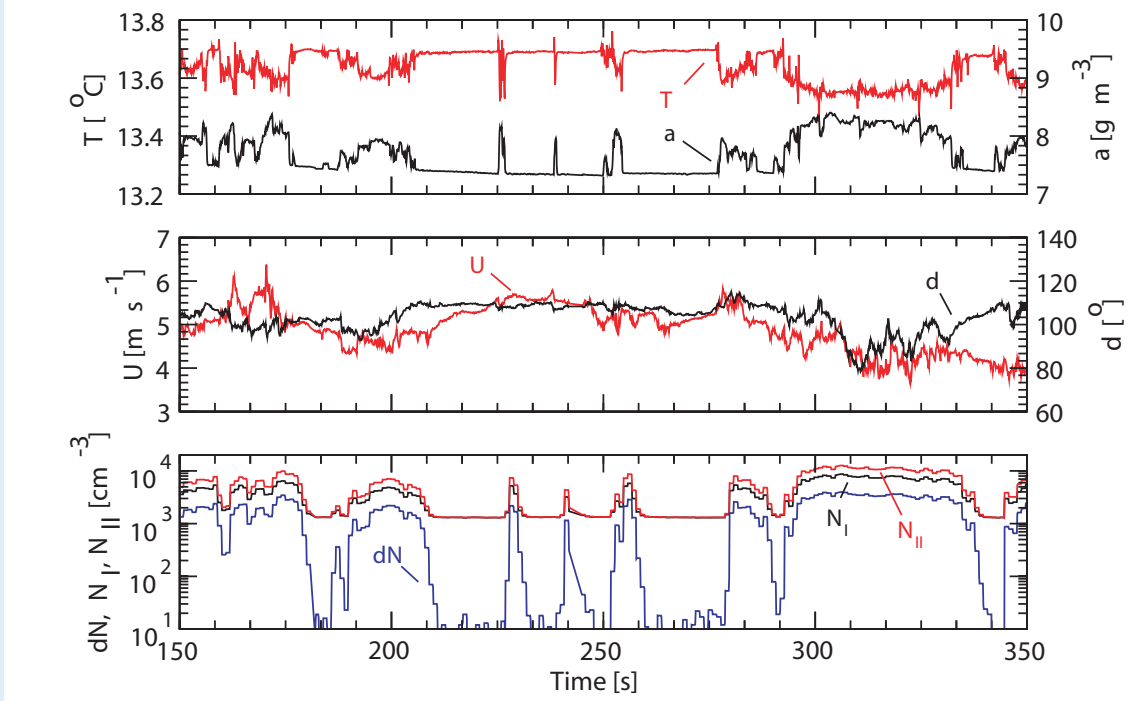

Fig. 7: Time series of temperature T, absolute humidity a, horizontal wind speed U, wind direction d, particle number concentration N_I ($10 < D_p < 1500$ nm), N_{II} ($5 < D_p < 1500$ nm), and ΔN ($5 < D_p < 10$ nm) at constant level in 600 m (08:59 - 09:05 UTC).

updrafts from the ML into the RL. This statement is supported by the size distribution measurements described below.

Comparison of size distributions at Melpitz, Collm, and Panitzsch sites

Figure 8 shows the diurnal variation of the normalized number size distributions for the three ground based measurement sites on June 3, 2002.

Each number size distribution as measured by the TDMPS-systems was divided by the total number concentration from 3 to 800 nm to emphasize the location of the mean maximum diameter independent of the total number concentration. From 00:00 to 07:00 UTC the size distributions were dominated by a stable maximum in concentration between 50 and 80 nm at all three sites. Mainly in Melpitz, particles in the 10 – 50 nm size range showed some variation most likely due to traffic emissions in the morning. Approximately at 07:30 UTC a rapid increase in number concentration of particles with diameters between 3 and 30 nm is recognizable in particular in Melpitz and at the Collm site. The size distributions have a bimodal structure (local maxima around 5 and 20 nm). At the Panitzsch site, the size distributions have only one local maximum around 20 nm and no around 5 nm. At the Melpitz and Collm sites this behavior could be observed nearly at the same

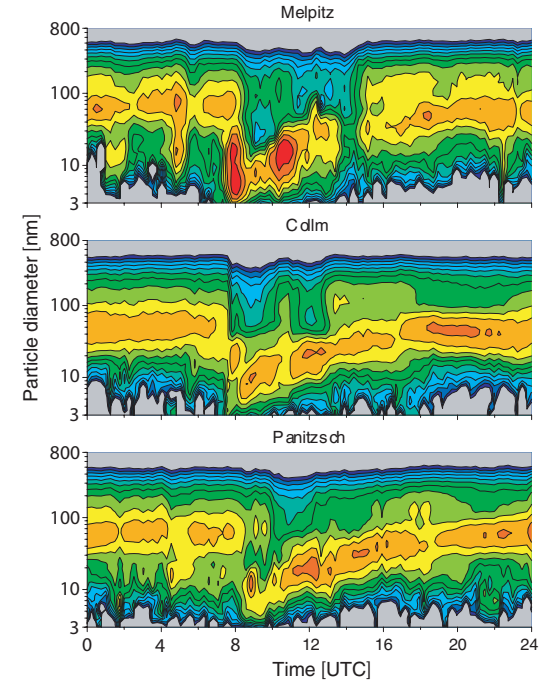

Fig. 8: Diurnal variation of number size distributions normalized by the total number concentration in Melpitz, Collm, and Panitzsch on June 3, 2002. Red colors indicates relatively high concentrations, whereas blue indicates relatively low concentrations.

time whereas in Panitzsch it becomes obvious about half an hour later. The occurrence of these maxima in the ultrafine size range (< 20 nm) was followed by a subsequent growth of the maximum diameter up to 50 – 80 nm at 18:00 UTC at all three sites

These results, together with the balloon-borne measurements, indicate that the local maximum at 20 nm is due to particles that were formed and grew in the RL and were mixed down during the break-up of the inversion. The local maxima at 5 nm could be explained by assuming new particle formation processes induced by the mixing processes taking place while the inversion breaks up. However, this is a hypothesis which cannot be proven with the data gained during the SATURN experiment.

Summary and conclusions

First results of the SATURN experiment from June 3, 2002 are presented. Detailed investigations of the vertical distribution of several meteorological parameters and particle number concentrations were performed with the balloon-borne system ACTOS together with continuous and ground-based measurements at the Melpitz site. Additionally ground-based measurements of the particle size distribution were carried out at two further sites to analyze the spatial and temporal correlation of new-particle formation events. The data were collected during a typical radiation day mainly influenced by a high-pressure area, only a few convective clouds occurred around noon.

Two different scenarios were observed during June 3, 2002: Before the break-up of the inversion, new particle formation took place in the RL and no particles were formed below the inversion. During the break-up of the inversion, particles which had been formed in the RL are mixed down. At the same time, new-particle formation processes, probably induced by the mixing processes, took place. This latter hypothesis is consistent with the findings of the SATURN experiment described here. However, further experimental proofs appear desirable.

Outlook

The data gained during the SATURN experiment will be further analyzed to gain information regarding e.g., the different scenarios under which new particle formation took place and vertical turbulent transport of aerosol particles.

To verify the hypothesis made above, it will be necessary to perform additional experiments regarding the vertical distribution of particle number concentrations under different meteorological conditions. It is planned to make long-time observations in a constant height in the residual layer including size resolved measurements.

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Cooperation
- Bundeswehr (WTD-71, Eckernförde)
- enviscope GmbH (Frankfurt/Main)

Variability of physical properties of the urban aerosol within one city

Thomas M.Tuch, Birgit Wehner, Ulrich Franck (UFZ; Center for Environmental Research), Alfred Wiedensohler

Introduction

Elevated concentrations of ambient particulate matter have been associated with increases in all-cause mortality, mortality from respiratory and cardiovascular diseases, hospital admissions, and exacerbation of respiratory symptoms in chronically ill patients (Dockery et al.,1994, Pope et al.,1999). Standards to reduce particle mass below aerodynamic diameters of 10 or 2.5 µm (PM_{10}, $PM_{2.5}$) have been established in many countries. These particle mass-based metrics usually neglect, however, ultrafine particles (<0.1 µm in diameter) because of their comparatively small mass concentration. On the other hand, these particles are efficiently deposited in the gas exchange region of the lungs and might pose a health risk as indicated by recent toxicological studies (Oberdörster et al., 1992, 1995, 2000) and epidemiological data (Pekkanen et al., 1997, Peters et al., 1997, Wichmann et al., 2000).
$PM_{2.5}$ mass measurements at a single urban background station dominated by the mass concentration of particles larger than 0.1 µm in diameter are well accepted as representative for the entire respective city. Particle number concentrations dominated by particles smaller than 0.1 µm in diameter are believed to be more variable due to their faster dynamics that cause a higher dependence on the particle sources and their variability.
Obtaining representative aerosol particle data for a certain geographical region requires long-term measurements, which can eliminate anomalies caused by variations in meteorological conditions. Such data are suitable for epidemiological studies associating average exposure with health effects in the urban environment. Furthermore they represent valuable input parameters for climate models.
Particle number size distributions in the size range from 3 to 800 nm have therefore been measured at the IfT (urban background) since February 1997 (Wehner et al., 2000, 2002). Measurements also included meteorological and trace gases. The resulting data set is complex, and therefore, detection of relationships among the measured parameters requires an in-depth analysis that includes statistical methods such as correlation analysis and principal component analysis. Here, this analysis was done to characterize urban background aerosol by determining seasonal, weekly, and diurnal variation and to find relations between the measured parameters.

Many epidemiological studies investigate correlations between short-term (daily and sub-daily) variations of aerosol parameters with short-term health effects. Exposure assessment of these studies is typically based on a single measurement site in a city. This strategy requires that short-term exposure to the aerosol at different places within a city is highly correlated to assure that variations of the exposure determined at the measurement site is typical for the whole study area. Buzorius et al. (1999) have shown that total particle number concentrations at several sites in the city of Helsinki are highly correlated. It has not yet been shown that smaller size classes of the urban aerosol, which are currently investigated in epidemiological studies, are correlated as well. In February 2002 we have established a second continuous measurement site in cooperation with the UFZ (Centre for Environmental Research) that can be used to determine the correlation of smaller particle size ranges within the city. This site is located in a street canyon representing a worst case scenario compared to urban background aerosol. This paper presents first results of the correlation studies between these two sites.

Methods

Site description. Leipzig is located in the Saxon-Thuringian basin. It has about 500000 inhabitants. The nearest higher natural elevations are located about 80 km southeast of the city.
The core measurement site is located at of the Institute for Tropospheric Research (IfT) in a mixed semi-industrial area about 5 km from the center of the city. There are no major industrial particle sources within 1 km of the site. The IfT is surrounded by several major roads, approximately ~100 m away. The inlets for aerosol (commercial low-flow PM10) and trace gas measurements are mounted on the roof of the IfT building about 16 m above the ground to minimize the influence of local emission sources such as passing cars. This core site operates continuously since February 1997. In February 2002 a second continuous measurement site has been established in a street canyon approximately 1.5 km south-west of the IfT. This site is located in an apartment in a typical residential area close to the intersection of two busy streets. Ambient air is sampled from 5 m above the ground using an identical inlet (see Figure 1).

Fig. 1: Map of Leipzig with measurement sites at the IfT and in a street canyon (Eisenbahnstrasse).

Instrumentation. Particle size distributions at both sites are measured using identical TDMPS systems (twin differential mobility particle sizer, Birmili et al, 1999) consisting of two Hauke-type DMAs (differential mobility analyzer). The first DMA is an Ultrafine DMA (UDMA) that selects particles from 3-22 nm in diameter at an aerosol/sheath-air flow rate of 2/20 l/min, and the second DMA selects particles from 22-800 nm at a aerosol/sheath-air flow rate of 0.5/5 l/min. The RH of the sheath air is stabilized at below 5%. Particles are counted downstream of the DMA using a condensation particle counter (CPC), model TSI 3010 (TSI Inc., St. Paul, MN), and downstream of the UDMA using an Ultrafine CPC, model TSI 3025 (TSI Inc., St. Paul, MN). Custom software using the actual transfer functions of the DMAs is used for the inversion of the raw mobility distributions (Stratmann and Wiedensohler, 1996). The TDMPS system measures a complete particle size distribution (0.003-0.8 µm) every 15 minutes. Trace gas data and metrological data are measured at the IfT with a time resolution of 1 and 10 minutes respectively (for instrumentation see Table 1). In addition NO

Parameter	Instrument / Manufacturer	Time resolution
O_3	Ozone-Analyser, model 8810 Monitor Labs Inc., Englewood (CO), USA	1 minute
NO, NO_2	NO/ NO2-Analyser, model 8841 Monitor Labs Inc., Englewood (CO), USA	1 minute
SO_2	SO2-Analyser, model AF 21 M Ansyco Inc., Karlsruhe, D	1 minute
CO	CO-Analyser, model CO 11 M Ansyco Inc., Karlsruhe, D	1 minute
Temperature	Platinum thermometer Pt 100	10 minutes
Relative Humidity	Hair hygrometer	
Atmospheric pressure	Aneroid barometer	
Wind speed	Cup anemometer	
Wind direction	Wind vane	
Precipitation	Ombrometer HP Adolf-Thies GmbH Klima- Mess- und Regeltechnik Göttingen, D	
Global Radiation	Pyranometer CM 11 Kipp & Zonen, Delft, NL	10 minutes

Tab. 1: Trace gas measurements and meteorological data measured at the IfT site.

and NO_2 are measured at the Eisenbahnstrasse with a time resolution of one minute using an identical instrument.

Data analysis IfT-data. Statistical time series analysis was applied to the complete data set (1997-2001). First, auto-correlation functions were calculated to reveal periodic behaviour of single variables (e.g., particle number concentration in a certain size range) and to identify typical time scales. Then, cross-correlation functions were used to determine the correlation coefficient between two time series that were shifted by an increasing time lag. The methods are described in textbooks, such as Einax et al. (1997).
Most of the measured parameters were directly analysed without further manipulation. One exception was particle concentrations in the different size ranges, resulting from integration of number size distributions over a certain size range by the measurement instrument (e.g., DMA). The number concentration between 3 and 10 nm in diameter is labelled *N3* and so on (see Table 2). All of the data (particle, trace gas, and meteorological) were segregated into the two different seasons (summer and winter) and different days of the week to find longer-term differences.

Parameter	Symbol
Particle number concentration	
3 – 10 nm	*N3*
10 – 20 nm	*N10*
20 – 30 nm	*N20*
30 – 50 nm	*N30*
50 – 100 nm	*N50*
100 – 800 nm	*N100*
Trace gas concentration	
O_3	*O3*
NO	*NO*
NO_2	*NO2*
Meteorological parameter	
Atmospheric pressure	*p*
Global radiation	*Q*
Wind speed	*u*

Tab. 2: *Parameters for statistical analysis.*

Street canyon data. Synchronized 15 minute data measured from March 1st 2002 to June 30th 2002 at both sites have been aggregated in an SPSS file (SPPS base 11.5, SPSS Inc., Chicago, IL). Based on this data set, two-sided Pearson correlation coefficients have been calculated for all 40 size channels (bins) (N=7867). In addition subsets for weekdays (Monday through Thursday, N=4495) and Sundays (N=981) as well as for days with particle formation events (N=1748) compared to days without events (N=6119) have been calculated. Fridays and Saturdays have been excluded from the analysis due to different traffic patterns compared to weekdays and Sundays.

Results

(a) Auto-correlation. Figure 2 shows auto-correlation functions calculated from measurements of the number size distributions limited to weekdays during summer and winter in 1997-2001. Results of weekends are not presented here, because they do not show this cyclic behavior of weekdays caused by a more irregularly traffic pattern on weekends.

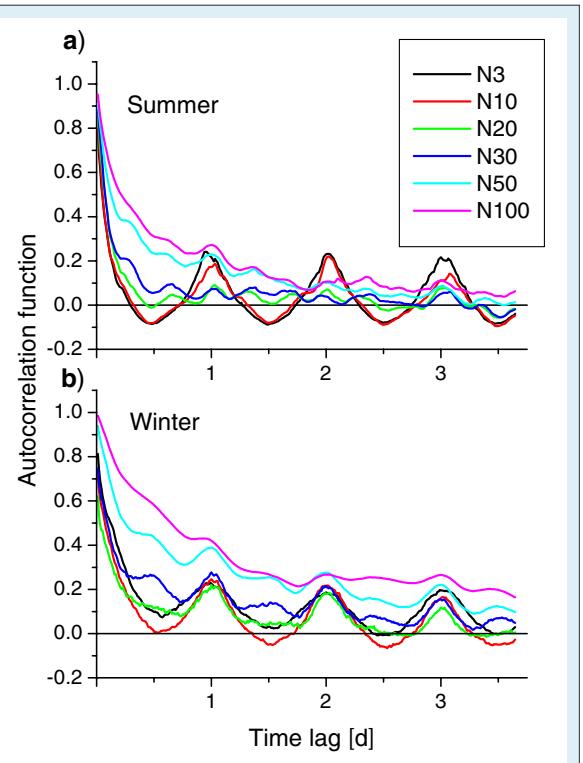

Fig. 2: *Auto-correlation functions of different size classes calculated for weekdays in (a) summer and (b) winter at the IfT.*

In summer, particles less than 20 nm (*N3* and *N10*) showed a clear periodic behavior with a period of one day (peaks at multiples of 1 day). The relative peaks at time lags of 1/2 day, 1 1/2 day, etc., correspond to a negative correlation for these time lags. With increasing particle size, the peaks became less pronounced compared to these two smaller size classes (*N3* and *N10*). Thus, only small particles (D_p < 20 nm) showed a typical diurnal cycle.
In winter, particles between 3 and 50 nm (*N3* - *N30*) in particular also clearly showed a cyclic behavior with a period of about 1 day. These size classes are strongly influenced by car traffic, and peaks in traffic emissions are cyclic. The correlation coefficients for winter decreased slower

with time lag than those for summer, indicating that the winter data were more interdependent due to slower variations caused by less vertical dilution.

(b) Cross-correlation. The time series of particle concentrations (*N3* and *N100*) were correlated with all measured meteorological parameters and trace gases. However, only the key results showing clear correlations are presented here. Figure 3 shows these cross-correlation functions between the time series of global radiation and particle concentration data for weekdays in summer.

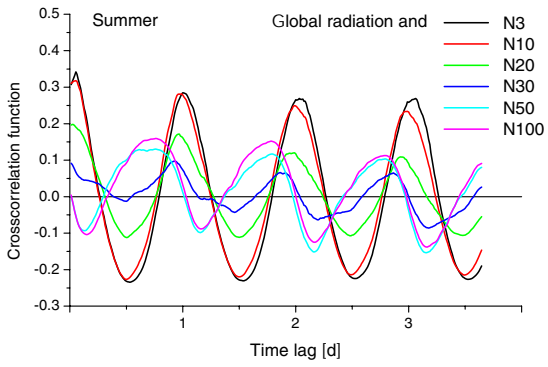

Fig. 3: *Cross-correlation functions of different particle size classes and global radiation for weekdays in summer at the IfT.*

Particles less than 20 nm (*N3* and *N10*) correlated well with the global radiation at a time lag of less than 1 hour, which is recognizable as the maximum (> 0.3) in the correlation curve. The global radiation as well as these small particles peaked around noon or one to two hours later within a cycle of 24 hours, reflected by relative peaks in the correlation coefficient at time lags of 1 day. The time series of particle concentration was shifted with respect to the global radiation, indicating that the global radiation peaked before the small-particle concentration peaked. This result confirms the hypothesis that small particles (D_p < 20 nm) are formed as a consequence of photochemical processes, which strongly depend on the intensity of global radiation (e.g., O'Dowd et al., 1999; Birmili and Wiedensohler, 2000).

For particles between 20 and 50 nm (*N20* and *N30*), the correlation coefficient was lower than that for particles less than 20 nm (*N3* and *N10*), and the peak for *N30* was shifted slightly to time lags of less than 1 day. This lower coefficient and slight shift indicate that the dependence between particle concentration and global radiation decreases with increasing particle diameter. A similar shift was also seen for particles larger than 50 nm (*N50* and *N100*); however, the maximum correlation coefficient was slightly higher for these two largest size classes compared with *N30*. The phase shift between the two time series (i.e., between global radiation and *N50* or *N100*, respectively) was 16 - 18 hours. However, it is implausible that the radiation would influence large particles to such a long time lag; the higher particle concentration should rather be a result of the morning rush hour, other combustion processes, or boundary layer development and the corresponding dilution. At a time lag of zero, the correlation coefficient was also almost zero between the two largest size classes (*N50* and *N100*) and the global radiation, indicating no direct relationship.

The NO concentration measured close to roads is known to be an indicator of car traffic emissions. Particle size classes influenced by car traffic should therefore correlate well with the NO concentration, and thus show no large time lag. Figure 4 shows the corresponding cross-correlation for winter. Particles larger than 30 nm (*N30* - *N100*) showed correlation coefficients between 0.35 and 0.6 with the NO concentration at a time lag of zero. Only particles less than 10 nm (*N3*) were nearly uncorrelated for a time lag of zero. After a lag of 1 day, *N10* and *N20* show the best correlation with NO (r_{xy} = 0.17 for *N10* and *N20*; r_{xy} = 0.13 for *N30* and *N50*), indicating their cyclic behaviour connected with car traffic. The weakest correlation was for particles greater than 100 nm, and the next weakest was the fraction of these particles less than 10 nm. These results correspond with the observed diurnal variation, where the particles between 10 and 100 nm seem to be the most strongly influenced by car traffic and somewhat by domestic heating.

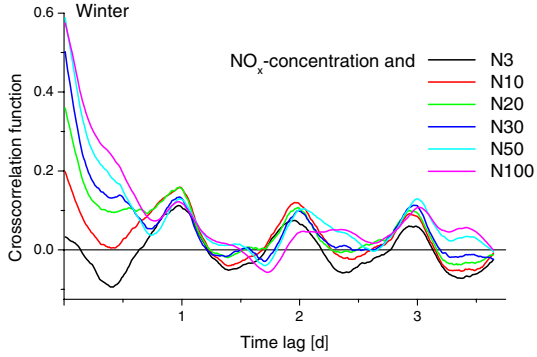

Fig. 4: *Cross-correlation functions of different particle size classes and NO concentration on weekdays in winter.*

(c) Diurnal variation of the particle size distribution at two sites in Leipzig. Results from the 5 year measurements at the IfT shown above indicate that traffic plays a major role for the urban aerosol. The following paragraph focuses on results comparing the IfT site with the new street canyon site at the Eisenbahnstrasse. The average diurnal variation of the aerosol at both sites by weekdays (a) and Sundays (b) is shown in Figure 5. The average total number concentration at the

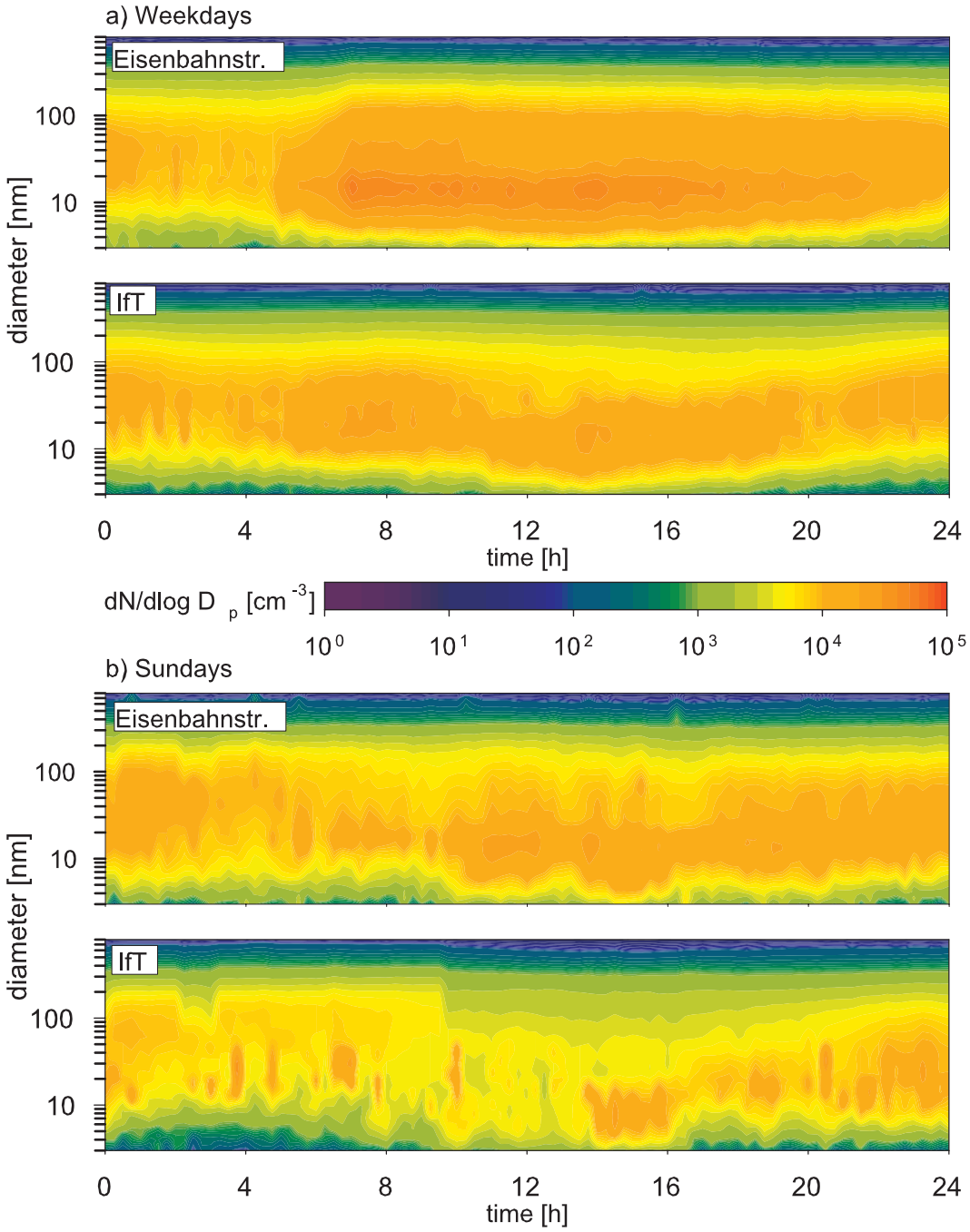

Fig. 5: *Average diurnal variation of aerosol particle number concentrations at both sites in Leipzig.*

Eisenbahnstrasse was 19000 cm^{-3} on weekdays compared to 10000 cm^{-3} at the IfT (ratio: 1.9:1). Average concentrations on Sundays where 11000 cm^{-3} and 7000 cm^{-3} respectively (ratio 1.6:1).

The diurnal pattern is visibly different at both sites on Weekdays and on Sundays. On Weekdays a peak of particles in the size range from 10 to 150 nm, centered at about 13 nm, was observed at both sites starting at about 5:00 in the morning. This peak coincides with the onset of the morning rush hour and elevated NO concentrations (see Figure 6). The pattern suggests that this morning peak is caused by direct traffic emissions. At the Eisenbahnstrasse elevated concentrations prevail until 22:00 indicating that this site is primarily influenced by direct traffic emissions throughout the day. At the IfT the traffic peak fades at about 10:00. It is replaced by the typical pattern of particle formation associated with global radiation. Such events start at the smallest observable diameters (less than 5 nm) and grow to larger size ranges. An exemplary diurnal variation with a traffic peak and

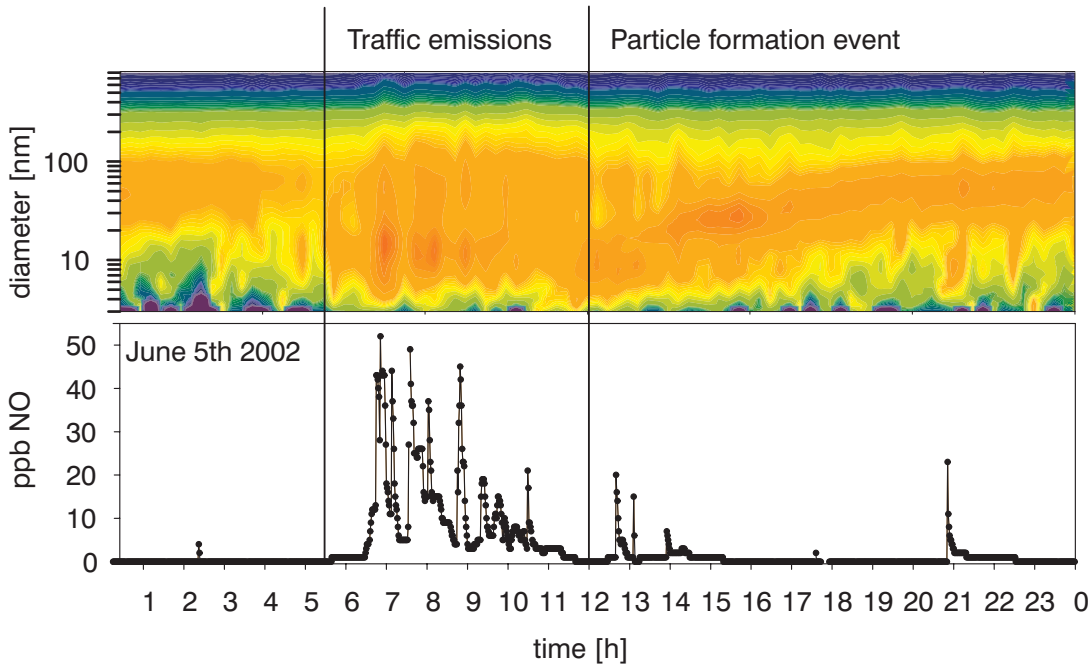

Fig. 6: *Exemplary diurnal variation of the aerosol at Eisenbahnstrasse (June 5, 2002) and NO concentration. Aerosol originating from traffic is associated with elevated NO levels where new particle formation events are not.*

a particle formation even at the Eisenbahnstrasse shown in Figure 6. It demonstrates that such peaks are typically not associated with elevated levels of NO.

On Sundays no distinct onset of a rush hour was observed in the Eisenbahnstrasse. Average diurnal variations at both locations indicate that two distinct sources contribute to the ultra-fine aerosol at both measurement sites: Traffic and new particle formation events associated with global radiation. This observation is consistent with the auto-correlation analysis of the long-term measurements at the IfT core site.

(d) Bin-wise correlation between the IfT and the Eisenbahnstrasse. Two-sided Pearson correlation coefficients of individual size bins of the number size distribution can be used to investigate how well particle number concentrations measured at one site reflect number concentrations at a second measurements site. This information is of special interest in exposure studies associating health effects with particulate air pollution, since such studies typically use exposure measurements from a single measurement site to asses effects for a larger area.

Figure 7 presents bin-wise correlation coefficients between the IfT and the Eisenbahnstrasse for synchronized 15 minute measurements at both sites. Note that all correlation coefficients presented are significant at $p<0.05$. The blue curve in both Figures 7a) and b) represents the average correlation coefficients for the whole study period. Correlation coefficients of sub-micron particles were highly size-dependent. They ranged from 0.3 to 0.65 for particles smaller than 10 nm in diameter. The weakest correlation (<0.2) was found in the size range from 15 to 40 nm. Average correlation coefficients for particles larger than 50 nm increased up to 0.8 indicating that measurements of these particles are well comparable at both sites. Note that correlation coefficients for both particle ranges smaller than 4 nm and larger than 500 nm are reduced to weak counting statistics of the TDMPS systems in these size ranges.

To investigate influences of the major sources (e.g., traffic and particle formation events associated with solar radiation) correlation coefficients for two subsets of the data set have been calculated.

Figure 7a) elucidates the influence of traffic on the correlation coefficients between the two sites. It compares workdays (red curve: high traffic densities, heavy duty diesel traffic allowed) with Sundays (green curve: lower traffic densities, heavy duty diesel traffic restricted). It is noticeable that correlation coefficients below 10 nm are virtually not influenced by changes of the traffic emissions. Correlation Coefficients in the size range from 10 to 40 nm are smaller on Sundays compared to weekdays. This suggests that secondary particles from traffic are not well mixed within the atmosphere: Minor traffic peaks at the Eisenbahnstrasse are therefore not observed at the

IfT (see Figure 6) resulting in reduced correlation coefficients. Changes in traffic densities induce, on the other hand, pronounced changes of the correlation coefficients in the size range from 40 nm to 300 nm. Particles in this size range are originate from direct traffic emissions. Furthermore, large particles measured in a city may originate from mesoscale transport to the measurement site. Whereas both stations measure highly correlated to almost identical concentrations in this size range on Sundays, correlation coefficients are reduced on weekdays. This suggests that whereas the street canyon site is highly traffic influenced on weekdays (reduced correlation coefficients due to local source) both sites are more influenced by the background aerosol on Sundays (increased correlation coefficient due to remote sources).

Figure 7 b) shows the influence of particle formation events associated with solar radiation on the size dependent correlation coefficients between the two measurement sites. Since sufficient amounts of precursor gases for particle formation events are typically available in larger areas and differences in solar radiation are relatively small such events

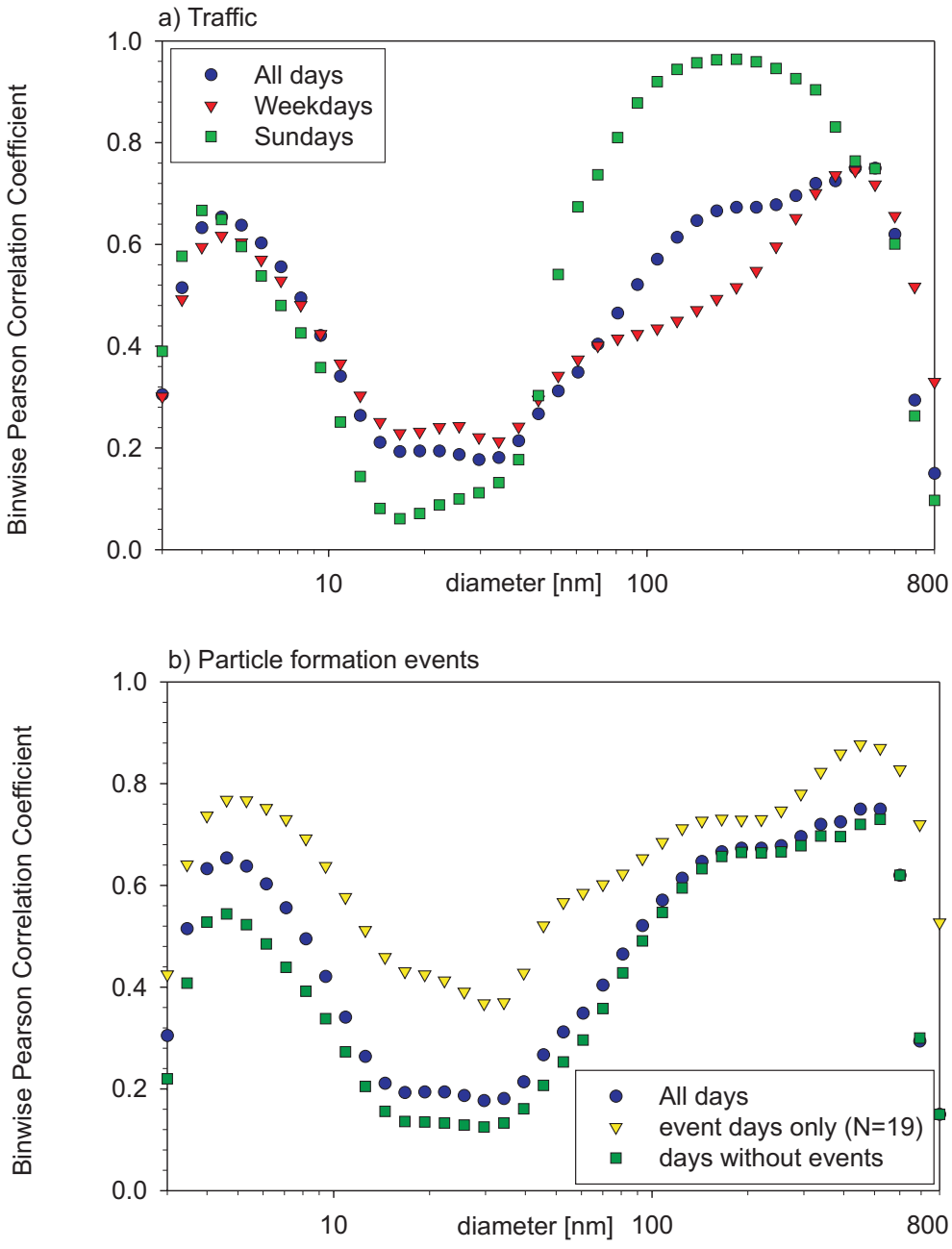

Fig. 7: *Average size dependent Pearson correlation coefficients between measurements at the IfT (blue circles) and influence of traffic (a) and particle formation events (b) on the correlation coefficients.*

are typically non-local. 19 event days have been visually identified from diurnal variation plots. Correlation coefficients have been calculated both for event days (yellow) and non-event days (green). Particle formation events start with particles smaller than 5 nm in diameter. Since the source of these particles is non-local, such events have been observed in parallel at both measurement sites. This cause an increased correlation coefficient for particles smaller than 5 nm in diameter on event days. Subsequent to the particle formation event particles grow to size ranges larger than 5 nm. Due to this particle growth correlation coefficients for these size ranges are higher on event days.

Conclusions

Auto- and Cross-correlation analysis confirmed the typical diurnal cycle of particles less than 100 nm with a peak in the number concentration during the morning rush hour. Furthermore, time series of ultrafine particles (< 20 nm) were well correlated with those of global radiation, particularly during summer. High correlations between the number concentration of particles larger than 20 nm and the NO concentration were found in winter, indicating that the occurrence of these particles is directly related to emissions from combustion processes.
Our parallel measurements have shown that correlation coefficients between two sites only 1.5 km apart are highly size-dependent. Furthermore these correlation coefficients are even modified by changes of typical major sources of particles in this size range. The weakness of the correlation for particle size ranges smaller than 100 nm along with the variability of the correlation coefficients induced by changes of emissions and meteorological conditions poses a major challenge to epidemiological studies investigating short-term health effects associated with size classes of ultrafine particles. Our data suggest that the typical exposure assessment strategy using a single urban background stations may not be appropriate for such studies.

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Cooperation
- Centre for Environmental Research (UFZ)

On the impact of turbulence length-scales, derived from large-eddy simulations on predicted mesoscale fields using the "Lokal-Modell"

Jürgen Helmert and Olaf Hellmuth

Motivation

Turbulent transport processes within the planetary boundary layer (PBL) influence phase transfer and redistribution of water, chemical species, aerosols, and cloud development. Due to their widespread occurrence, persistence, and radiative properties, clouds are of fundamental importance for the global energy budget. They belong to the most important factors regulating the current climate system and future climate change (Ramanathan et al., 1989). However, turbulent transports of heat, moisture, momentum as well as evaporation and condensation are not resolved in mesoscale and large scale models (weather forecast models, climate models).
Therefore, these models require suitable parameterizations for turbulent transport processes. A parameterization approach, often used in mesoscale and large scale models is the down-gradient approximation for the eddy diffusivity coefficient (e.g., Mellor and Yamada, 1974). This turbulence parameterization is based on a prognostic equation for the turbulent kinetic energy (TKE) and a formulation for the lengths scales of turbulent mixing processes. However, the turbulent mixing lengths are related to various characteristic turbulence length-scales of turbulent processes, whose approximation is a serious problem in turbulence parameterizations. Although various diagnostic equations for the turbulence length-scales have been proposed, there is no single best solution. In order to augment the database of measured boundary layer turbulence data, for more than three decades numerical simulations of atmospheric boundary layer flows have been used.
One of the most successful approach for atmospheric boundary layer applications is large-eddy simulation (LES). Consequently, since more than a decade LES has been increasingly used to develop and evaluate parameterization schemes in mesoscale, large scale and climate models (Ayotte et al., 1996). LES explicitly resolves the energy-containing turbulent motions that are related to turbulent transport of momentum, heat and moisture. The residual, less energy-containing small-scale motions are modelled by subgrid-scale (SGS) parameterizations. Because only a small portion of turbulent processes has to be treated by the SGS parameterizations, LES results are much less sensitive to deficiencies in turbulence parameterizations.
By varying a number of setup parameters such as surface layer heating, mean thermodynamic structure, and geostrophic wind profile, LES allows to simulate the three-dimensional turbulent structure of the atmospheric boundary layer for a wide range of atmospheric conditions in a controlled fashion and to determine the characteristic turbulence length-scales of energy-containing turbulent motions.
Therefore, the LES approach has been extensively used in this study, to generate a comprehensive dataset of varying boundary layer conditions with different atmospheric stability. This dataset allows the determination of a generalized approximation for the turbulence length-scale of energy-containing turbulent processes, which can be implemented in mixing length formulations of turbulence parameterizations used in mesoscale and large scale models.

Large-eddy simulation database

In order to generate the boundary layer dataset, the LES model of the Max-Planck-Institut für Meteorologie Hamburg has been used (Chlond, 1992). This model contains a water cycle with cloud formation and takes into account radiation processes. The simulations were performed on model domain of size 6.4 x 6.4 x 4.4 km^3, using a grid resolution of 100 x 100 x 40 m^3.
As an example, Figure 1a shows the final three-dimensional structure of a convective boundary layer after 9 hours of integration time. It exhibits isosurfaces of vertical velocity, which identify strong buoyancy-induced motions. These are narrow updraft regions with high positive vertical velocity fluctuation elongated in mean wind direction (small arrows). These updrafts originate as small-scale thermals near the surface and grow in diameter with increasing distance from the ground, merging with their neighbours, and forming larger-scale regions of upward-moving warmer fluid in the mixed layer at about z=0.5 km, where the greatest horizontal length-scale is assumed. Near the PBL top at z=1.6 km, the updraft diameter is decreasing. These upward moving structures are accompanied by large compensating regions of negative vertical velocity fluctuation with small amplitude (downdrafts).
In the present study, the characteristic turbulence length-scale of the energy-containing turbulent updraft-processes is defined as integral of vertical

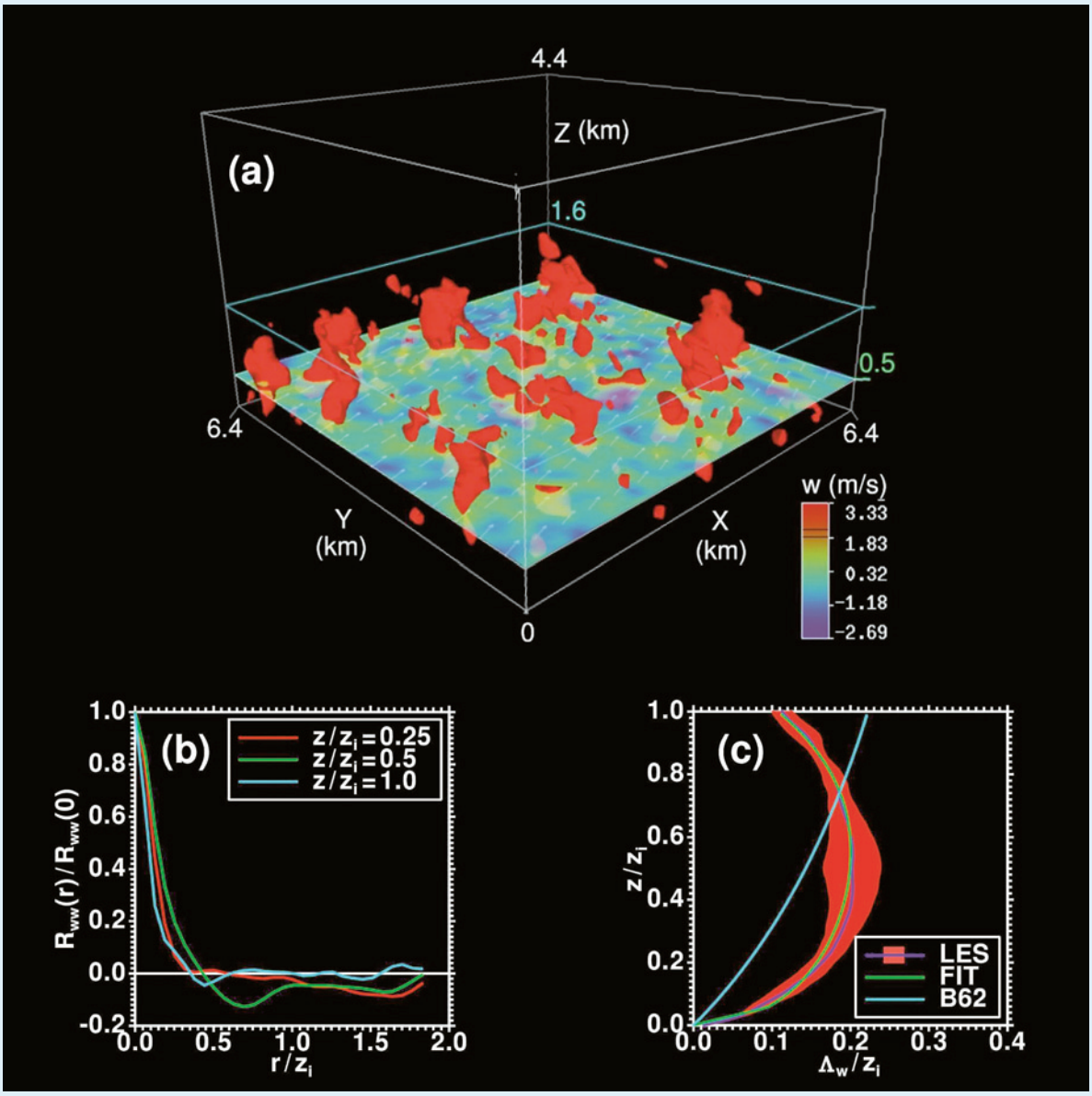

Fig. 1: *(a) LES domain with three-dimensional iso-surfaces and horizontal cross-section of vertical velocity and horizontal wind; (b) normalized one-dimensional autocorrelation functions of vertical velocity at different heights in the convective boundary-layer; (c) vertical profile of normalized vertical velocity integral length scale.*

velocity autocorrelation function (Figure 1b). The Fourier-Transformation approach has been used to derive the horizontally averaged autocorrelation function at each level of the LES domain. In order to increase the statistical significance of the resulting integral length-scale, the LES output has been averaged over a large number of time steps. Figure 1c shows the mean value (magenta solid line) and standard deviation (red-shaded area) of integral length-scale under conditions of well-developed turbulence for one LES dataset.

Using a non-linear least squares method after Levenberg-Marquardt a look-up-table of turbulence length-scale profiles has been derived from the LES database (e.g., Figure 1c, green solid line). Input parameters are normalized PBL height, friction and convective velocity. For comparison, the cyan solid line in Figure 1c shows the turbulence length-scale approximation proposed by Blackadar (1962), representative for neutral atmospheric stratification and widely used as turbulence length-scale of energy-containing motions in mixing length formulations of down-gradient turbulence parameterizations approaches. However, it was shown in Ayotte et al. (1996), that even in buoyancy-driven convective boundary layers the down-gradient approach associated with a turbulence length-scale of neutral boundary layer has strong deficiencies.

Examination of mesoscale response to turbulence parameterization: The LITFASS study

To examine the sensitivity of a mesoscale model against the turbulence length-scale approximation, the LES based look-up-table of turbulence length-scales has been implemented into the present turbulence scheme of the "Lokal-Modell" (LM) of the German Weather Service (Doms and Schättler, 1999). In cooperation with the R&D department of the German Weather Service, the June 18, 1998 from the LITFASS field-experiment has been chosen as example case for the sensitivity study. It was one of the "Golden Days" of the experiment, where during daytime, convective conditions as well as clouds in different altitudes and precipitation events were observed.

The examination is based on LM V2.16. Two 24 h-simulations, denoted by RUN I and RUN II were performed. RUN I used the turbulence model suggested by Mellor and Yamada (1974) associated with a length-scale approximation after Blackadar (1962). In RUN II, for unstable stratification the former length-scale approximation has been replaced with the LES-based look-up-table of turbulence length-scales whereas for stable stratification the treatment of turbulent processes is identical between RUN I and RUN II.

The LM simulations were performed on a model domain, consisting of 171 x 171 grid points that covers central Europe. Figure 2 shows results of RUN I after 12 h integration time. It exhibits clouds at different altitudes with cellular structure over continental areas, indicating the occurrence of buoyancy driven boundary layers. Due to a low-pressure system located in the Northwest of the model domain, these clouds are moving with the mean flow from Northwest to Southeast.

The model simulations have been compared with analysis data of 2m-temperature, specific humidity, 10m-wind, as well as low, medium and high cloud coverage at several times (6, 12, 18 UTC). The analysis data are based on an operational, integrated modelling and a 4D assimilation approach.

A statistical examination in terms of correlation coefficient and centered pattern RMS difference after Taylor (2001), shows that the agreement of near surface values of temperature, wind speed and moisture between analysis data and LM simulations is higher than those of cloud coverage.

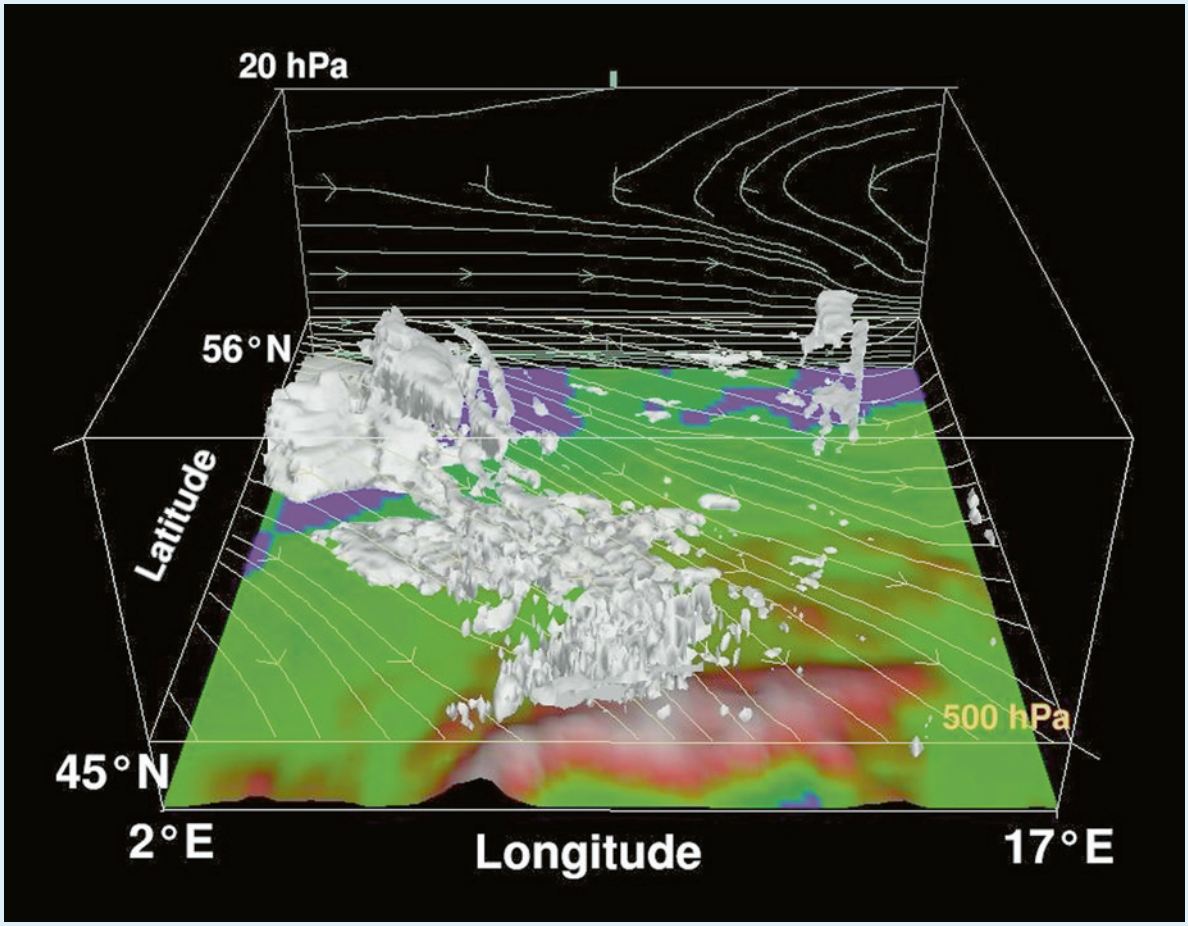

Fig. 2: Three-dimensional view from south into the domain of the "Lokal-Modell" at 18 June 1998 12:00 UTC. The white shaded area is the cloud cover, the vector fields depict the vertical wind field and the horizontal wind field at 500 hPa.

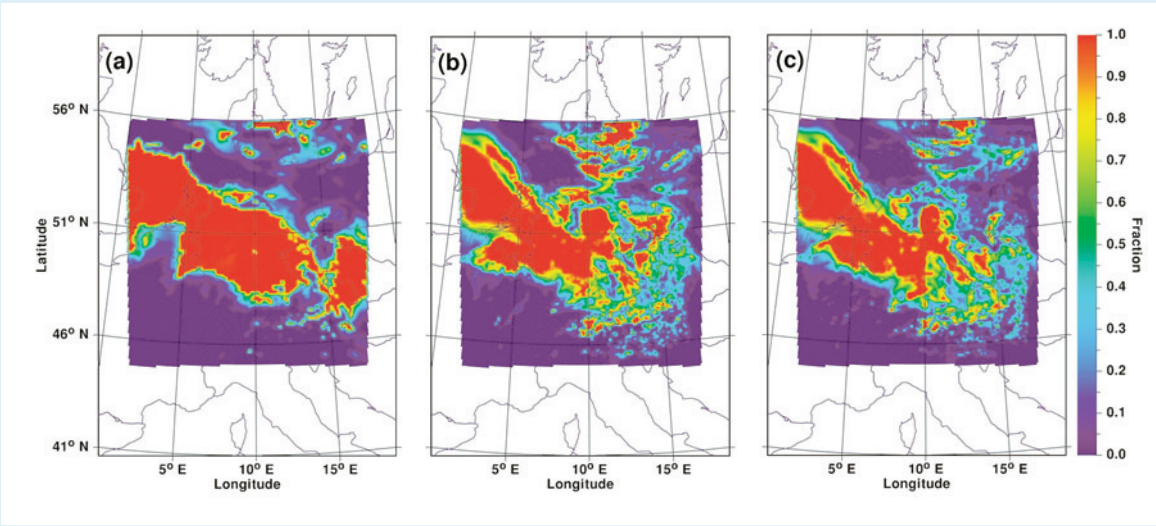

Fig. 3: *Fraction of low-cloud coverage at 12:00 UTC determined from interpolated analysis data (a), RUN I (b) and RUN II (c).*

However, it was found that even the prediction of low-cloud formation benefits from the new LES-based turbulence mixing lengths approximation, indicated by higher correlation coefficient and smaller centered pattern RMS difference between RUN II and analysis, than between RUN I and analysis.

Figure 3 shows the fraction of low-cloud coverage at 12 UTC determined from model analysis data (a), RUN I (b) and RUN II (c). The general structure of the low-cloud field from analysis data is well captured in both simulations, showing a large area covered by low clouds, which is elongated from the Northwest to the Southeast of the LM domain. The large area of low-cloud coverage located at the southeastern boundary of the domain was not predicted in both runs. However, the forecast of RUN II predicts fewer clouds than RUN I in the North and the Northeast of the LM domain, which agrees quite well with the analysis data.

A closer examination of the spatial differences in the low-cloud fields between analysis data and the forecasts of RUN I and II is provided by the two-dimensional crosscorrelation performed in a centered 128 x 128 gridpoints subsection of the LM domain. Figure 4 shows the crosscorrelation of low-cloud coverage at 12 UTC between analysis data and forecasts of RUN I and II. The elongation of the cloud field is clearly reflected by a stretching of the corresponding crosscorrelation in the direction of elongation. Thus, the cloud cover is more correlated in the direction of elongation than perpendicular to it. A closer examination shows that the area of high correlation coefficient R>0.8 is not centered but shifted towards positive lags. This is related to the occurrence of increased cloud coverage with

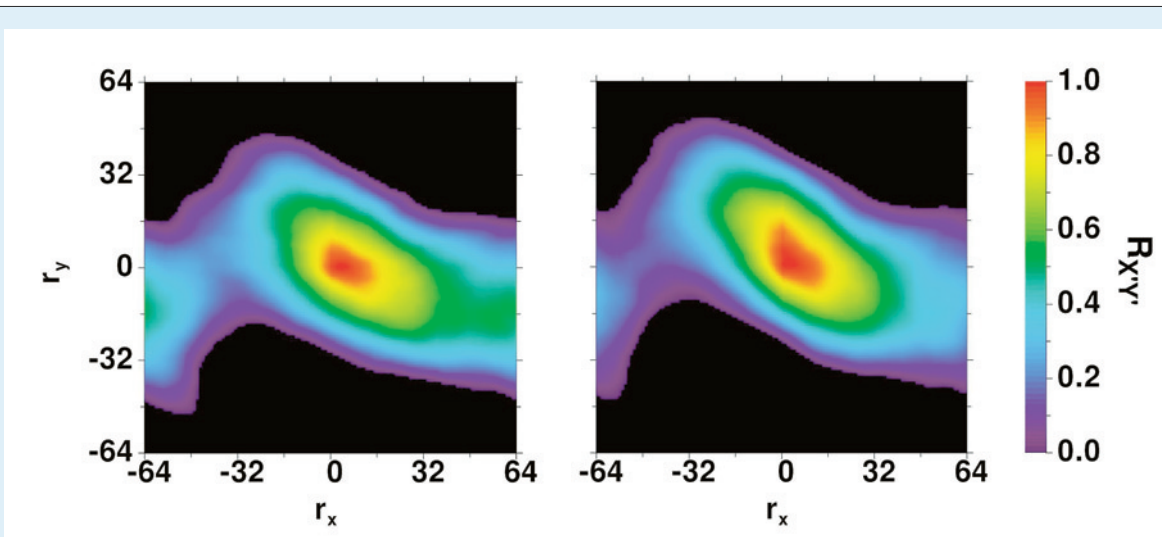

Fig. 4: *Two-dimensional crosscorrelation of low-cloud coverage between analyis data and RUN I (left) and analysis data and RUN II (right).*

low clouds in the Northeast of the domain in RUN I and RUN II compared to the analysis. However, the crosscorrelation between analysis data and results of RUN II exhibits a larger area of high crosscorrelation coefficient than the crosscorrelation between analysis data and results of RUN I. This result and the greater correlation coefficient for lags in north direction emphasizes the above mentioned findings that the low-cloud forecast at 12 UTC of RUN II is closer to the analysis data than that of RUN I for the 18 June 1998.

The time evolution of correlation coefficient and centered pattern RMS difference between RUN I and RUN II in terms of cloud coverage, liquid water content (LWC), and water vapor mixing ratio is shown in Figure 5. Obviously, the largest differences between both simulations, indicated by low correlation coefficients and large centered pattern RMS differences occur between 6 and 18 UTC. The cloud coverage shows a minimum correlation coefficient between level 35 and 32 at about 6 UTC. The vertical range of minimum correlation coefficient is increasing with time in height, reaching from the surface to about height level 25 (1.7 km) at 11 UTC. After 12 UTC the minimum of correlation coefficient vanishes in the range of top PBL while after 18 UTC low values of correlation coefficient occurs in the PBL and at high altitudes (level 12).

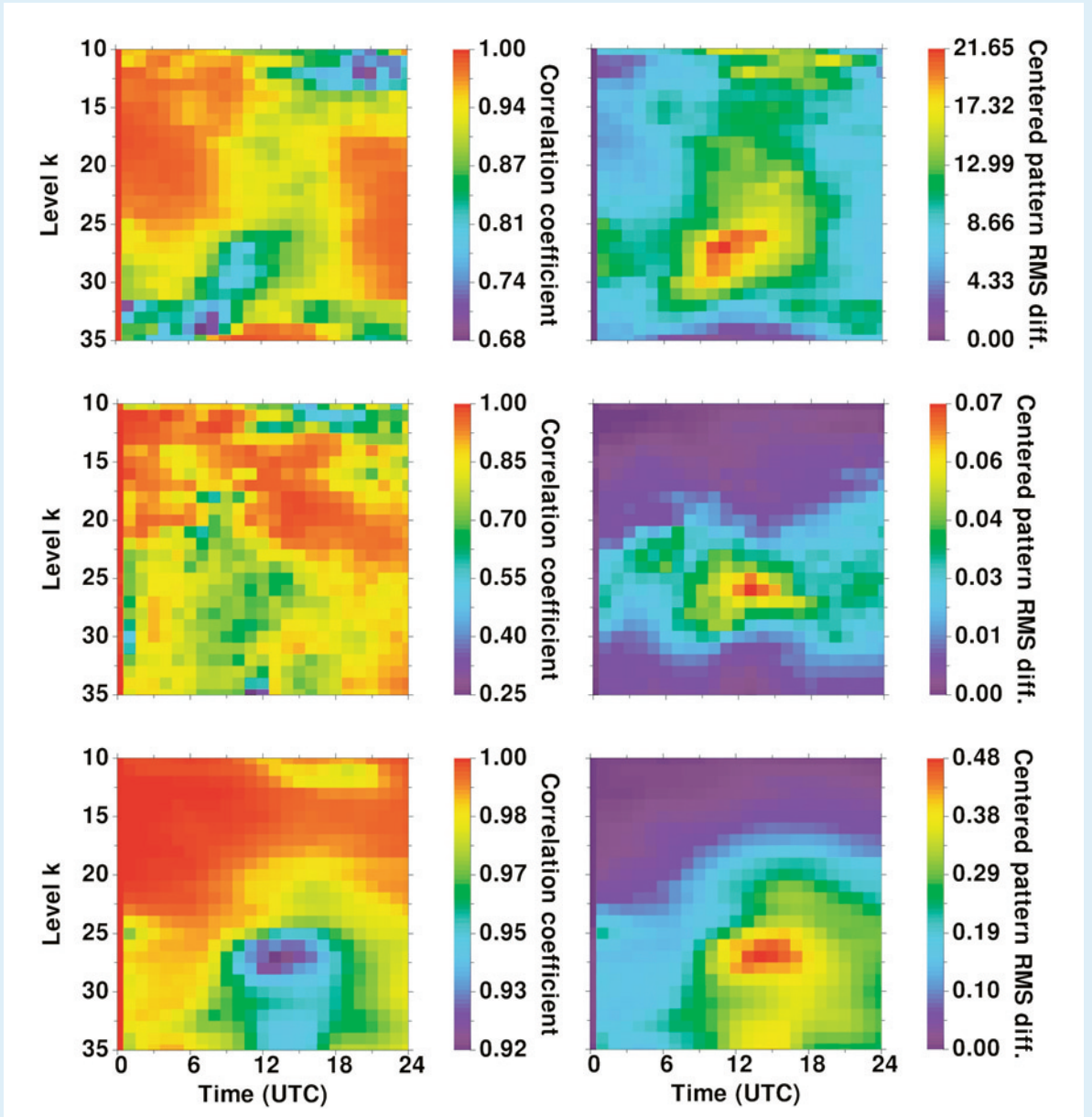

Fig. 5: Time evolution of the correlation coefficient (left) and the centered pattern RMS difference (right) between RUN I and RUN II for the model cloudiness (top), the cloud water content (center) and the water vapor (bottom) in dependence on altitude.

The corresponding centered pattern RMS difference of cloud coverage shows a pronounced maximum at 11 UTC at level 28 developing with time to higher altitudes. It is located near level 25 at 15 UTC. In agreement with a vanishing minimum of correlation, the centered pattern RMS difference of cloud coverage decreases after 18 UTC. This indicates that the difference between RUN I and RUN II decreases in the night.

The time evolution of the correlation coefficient for the LWC is similar to that of the cloud coverage. However, the minimum of the correlation coefficient exhibits a larger vertical extent up to about level 17. This vertical extent is decreasing after 12 UTC to about level 27 at 18 UTC.

The centered pattern RMS difference of the LWC shows significantly smaller values than those of the cloud coverage. In agreement with the cloud coverage it exhibits a pronounced maximum developing after 11 UTC at level 26 that remains nearly constant in altitude and vanishes after 18 UTC.

Obviously, the forecast of 3D water vapor is quite similar between RUN I and RUN II indicated by the large correlation coefficient. The time evolution for this correlation coefficient shows a pronounced minimum from 11 to 16 UTC at about level 28 (1040 m), which is related to the top of the PBL. Accordingly, the centered pattern RMS difference exhibits a maximum at the same level and in the same time interval.

It should be noted that large differences between RUN I and RUN II, indicated by low values of the correlation coefficient and high values of centered pattern RMS difference do not only occur at PBL model levels. As shown in Figure 5, after about 12 UTC large differences between both simulations occur at high altitudes. The reason for these differences at high altitudes is not quite clear. However, the time shift of about 6 hours between occurrence of largest differences near the boundary layer and those at high altitudes could be related to differences in turbulence parameterization in RUN I and RUN II. Although turbulent processes have their origin in the boundary layer, they have obviously impact on the forecast of 3D cloud

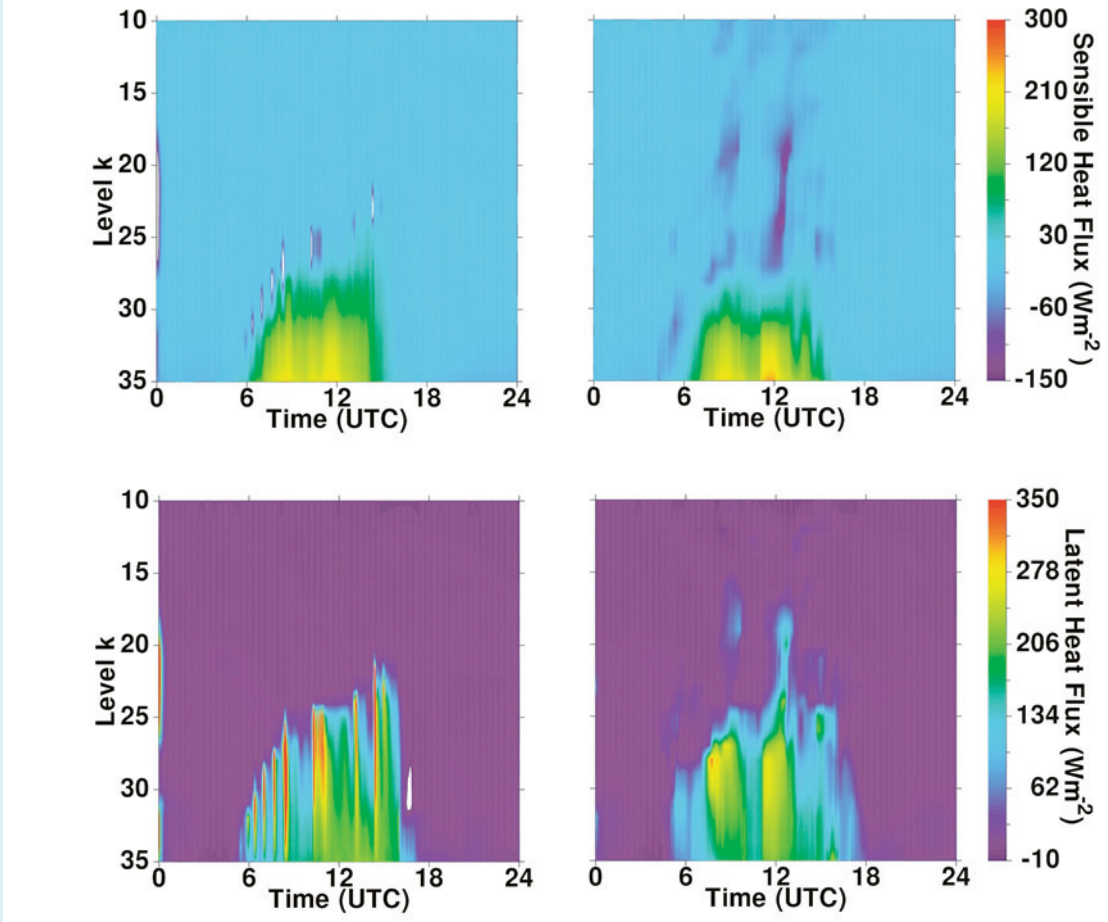

Fig. 6: *Time evolution of sensible (top) and latent (bottom) heat flux of RUN I (left) and RUN II (right) at grid point (129,109) of the LM domain related to the measurement site of Meteorological Observatory Lindenberg.*

coverage and LWC. Therefore, it is possible that the observed differences move to higher altitudes with increasing integration time. However, details of this mechanism are not quite understood and require further investigations.

Figure 6 shows the diurnal cycle of the sensible and latent heat flux at the measurement site of Lindenberg (LM grid point 129, 109). The sensible and latent heat fluxes strongly increase after 6 UTC. The vertical profiles show the typical behaviour for the convective boundary layer and are quite similar to profiles obtained from LES.

and gridscale precipitation (RAIN CON, RAIN GSP) and for the total amount of precipitation (TOT PREC) is very similar, showing a decrease of the correlation coefficient in the morning and an increase in the afternoon. However, the correlation coefficient of precipitation rate (PRR) shows a quite different behaviour. The time evolution of correlation coefficient related to precipitation rate of grid-scale rain (PRR GSP) shows a sharp drop off in the early morning, followed by a strong increase with a maximum at noon and a decrease in the afternoon while those of precipitation rate

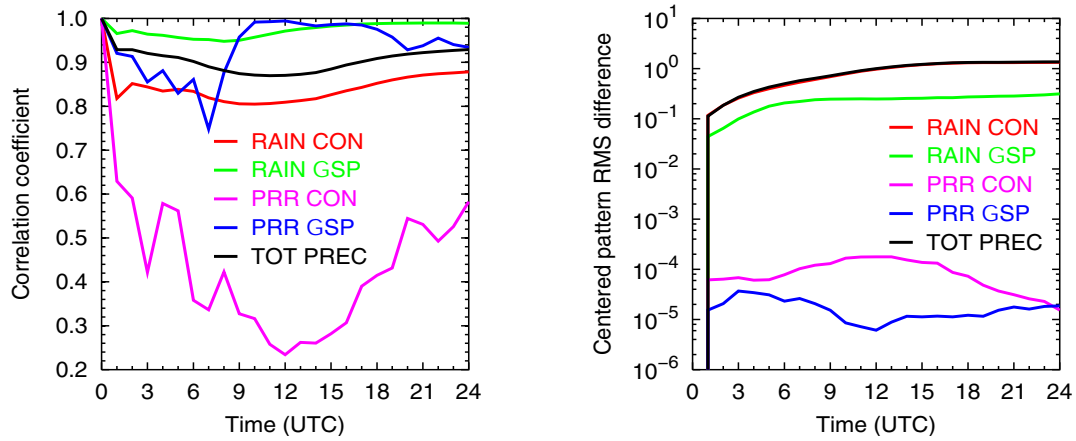

Fig. 7: *Time evolution of correlation coefficient (left) and centered pattern RMS difference (right) between RUN I and RUN II for amount of rain from convective (RAIN CON) and from grid-scale (RAIN GSP) precipitation, precipitation rate of convective (PRR CON) and of grid-scale (PRR GSP) rain and total amount of precipitation (TOT PREC).*

The largest difference between RUN I and RUN II was found in the magnitude of minimum sensible heat flux and in the vertical extent of the entrainment zone located at the top of the convective boundary layer, which is much smaller in RUN I. Whereas RUN I indicates a sharp drop of the PBL top after 15 UTC, RUN II predicts a smooth transition to lower PBL top values.

Although the time evolution of the predicted latent heat flux is similar to that of the sensible heat flux, differences between RUN I and RUN II are much more pronounced than for the sensible heat flux. RUN I indicates a periodic structure in the morning hours from 6 to about 9 UTC with rather high maximum values of about 350 W/m^2 not visible in RUN II. The large differences in time evolution of latent heat flux between RUN I and RUN II after 15 UTC are similar to that of the sensible heat flux, indicating a different behaviour of the decreasing PBL in both simulations.

Figure 7 shows the impact of turbulence parameterization on the predicted precipitation fields. The time evolution of the correlation coefficient for the amount of rain from convective

of convective rain (PRR CON) exhibits a strong decrease to a minimum value of about 0.2 at 12 UTC followed by an increase to about 0.6 at 24 UTC. Apparently, the correlation coefficients related to the convective parts of precipitation (RAIN CON, PRR CON) are lower than for the grid-scale part (RAIN GSP, PRR GSP). This emphasizes, that the different turbulence parameterization approaches have implications for the prediction significance of the precipitation variables even in unstable conditions.

The time evolution and values of corresponding centered pattern RMS difference agree among variables related to the amount of rain (RAIN CON, RAIN GSP) and total precipitation showing an increase in the early morning and nearly constant values after 6 UTC. However, centered pattern RMS differences of variables related to precipitation rate (PRR CON, PRR GSP) differ in order of magnitude from other precipitation variables. Whereas PRR CON indicates a maximum of centered pattern RMS difference at 12 UTC, PRR GSP exhibits a minimum at noon.

Conclusion and outlook

LES is a very useful tool to provide turbulence data with high resolution in space and time. Thus, it can fill the gap of field measurements for a wide range of atmospheric conditions. In the present case, LES of atmospheric boundary layers with various atmospheric stability states have been performed. The resulting database of synthetic atmospheric boundary layers has been used to determine the turbulence length-scale of energy-containing motions, representative for turbulent transport processes of momentum, heat and moisture. Based on a generalized turbulence length-scale approximation in terms of PBL scaling properties, a non-linear least squares method has been used to derive a look-up-table of turbulence length-scales. This look-up table has been implemented in the mixing length formulation of the turbulence parameterization of LM V2.16, which is based on the Mellor-Yamada turbulence scheme. The sensitivity of LM simulations on the mixing length formulation has been examined for an example case of the LITFASS field-experiment.

Although a comprehensive LM validation study is beyond the scope of the present investigation, the examination clearly depicts the dependence of several cloud and rain properties on mixing length formulation.

A comparison with analysis data has shown that particularly the significance of low-cloud coverage prediction was advantageously affected by the LES-derived mixing length formulation.

Acknowledgements

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Cooperation
- Max-Planck-Institut für Meteorologie, Hamburg
- Deutscher Wetterdienst (DWD), Abteilung Forschung und Entwicklung
- Deutscher Wetterdienst (DWD), Meteorologisches Observatorium Lindenberg

Long-term simulation of ozone and PM using the model system LM-MUSCAT within the CityDelta project

Olaf Hellmuth, Ralf Wolke, Oswald Knoth, Eberhard Renner, Wolfram Schröder, Birgit Heinrich, Stefan Benz

Motivation

In the framework of the CAFE program (**C**lean **A**ir **f**or **E**urope) of the European Community the response of predicted urban air quality to changes in urban emissions is investigated. The model inter-comparison CityDelta, initiated by the Joint Research Center (JRC), focusses on health-relevant matrices of exposure (e.g., long-term concentrations) to fine particles and ozone in urban areas. The range of response resulting from the model-intercomparison study will be used in the cost-effectiveness analysis of CAFE with the aim to balance Europe-wide emission controls against local measures. As the IfT contribution to CityDelta long-term simulations of ozone and PM for the city area of Berlin will be performed for the base case szenario 1999 and seven emission scenarios for 2010 with modified NO_x, VOC and PM emissions. For the calculation, the IfT meteorology-chemistry-aerosol-transport forecast system will be used. In the present paper, the model system and simulation approach will be described as well as the project progress reported.

General remarks

Atmospheric chemistry-transport models are useful tools for the understanding of pollutant dynamics in the atmosphere. Such air quality models are numerically expensive in terms of computing time. This is due to the fact that their resulting systems of ordinary differential equations (ODE) are non-linear, highly coupled and extremely stiff. In chemical terms, a stiff system occurs when the lifetime of some species are many orders of magnitude smaller than the lifetime of other species. Because explicit ODE solvers require numerous short time steps in order to maintain stability, most current techniques solve stiff ODEs implicitly or by implicit-explicit schemes (Verwer et al., 1998).
For air quality studies, the chemistry-transport-model MUSCAT (**MU**ilti**S**cale **C**hemistry **A**erosol **T**ransport) has been utilized (Wolke and Knoth, 2000; Knoth and Wolke, 1998b). The code is parallelized and tested on several computer systems. Presently, MUSCAT has an online-coupling to the parallel, non-hydrostatic meteorological code LM (Doms and Schättler, 1999; Schättler and Doms, 1998) which is the operational regional forecast model of the German Weather Service. Both parallel codes work on their own predefined fraction of the available processors and have their own separate time step control (Figure 1). The coupling scheme simultaneously provides time-averaged wind fields and time-interpolated values of other meteorological fields (vertical exchange coefficient, temperature, humidity, density). Coupling between meteorology and chemistry-transport takes place at each horizontal advection time step only.
The code is parallelized by distributing these blocks among the available processors. This may lead to load imbalances, since each block has its own time step control defined by the implicit time integrator. Therefore, dynamic load balancing is proposed and investigated.

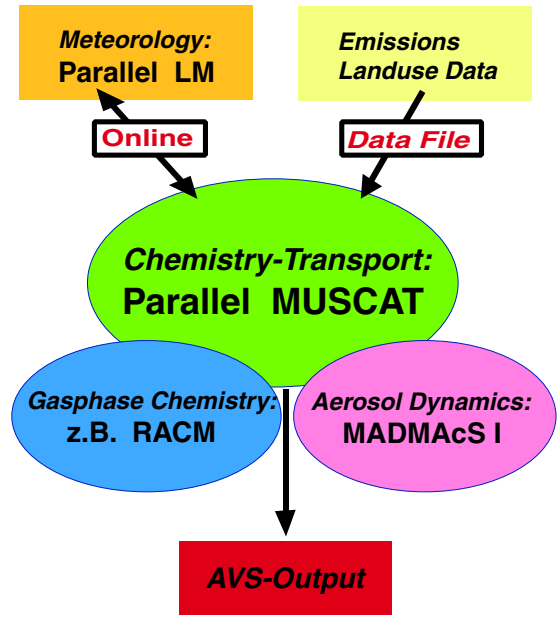

Fig. 1: *The coupled model system LM-MUSCAT.*

The chemistry-transport code MUSCAT

Air quality models base on mass balances described by systems of time-dependent, three-dimensional advection-diffusion-reaction equations.

(a) Multiblock grid. In MUSCAT a static grid nesting technique (Wolke and Knoth, 2000; Knoth and Wolke, 1998b) is implemented. The horizontal grid is subdivided into so-called “blocks”. Different resolutions can be used for individual subdomains

in the multiblock approach (Figure 2). This allows fine resolution for the description of the dispersion in urban regions and around large point sources. This structure originates from dividing an equidistant horizontal grid (usually the meteorological grid) into rectangular blocks of different size. By means of doubling or halving the refinement level, each block can be coarsened or refined separately. This is done on condition that the refinements of neighbouring blocks differ by one level at the most. The maximum size of the already refined or coarsened blocks is limited by a given maximum number of columns. The vertical grid is the same as in the meteorological model.

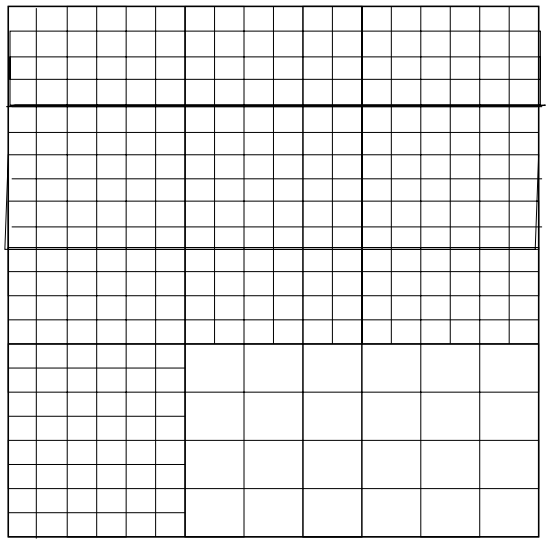

Fig. 2: *Multiblock grid.*

The spatial discretization is performed by a finite-volume scheme on a staggered grid. Such schemes are known to be mass conservative because of the direct discretization of the integral form of the conservation laws. For the approximation of the surface integrals, point values of the mixing ratio and its first derivative are needed on the cell surfaces. To approximate the mixing ratio at the surface we implemented both a first order upwind and a biased upwind third order procedure with additional limiting (Hundsdorfer et al., 1995). This scheme has to be applied to non-equidistant stencils which occur at the interface of blocks with different resolutions (Knoth and Wolke, 1998b). Figure 3 illustrates the choice of the grid values involved in the interpolation formula for the two different upstream directions. In this case the grid cell has five cell wall interfaces.

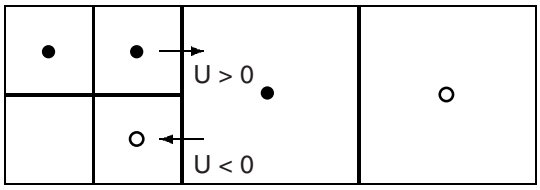

Fig. 3: *Stencil for advection operator at the interface between different resolutions. ● for U > 0, O for U < 0.*

(b) Time integration. For the integration in time of the spatially discretized equation an IMEX scheme ("implicit-explicit scheme") has been applied (Wolke and Knoth, 2000; Verwer et al., 1998). This scheme uses explicit second order Runge-Kutta methods for the integration of the horizontal advection and an implicit method for the rest. The fluxes resulting from advection are defined as a linear combination of the fluxes from the current and previous stages of the Runge-Kutta method. These horizontal fluxes are treated as "artificial" sources within the implicit integration. Thus, a change of the solution values as in conventional operator splitting is avoided. Within the implicit integration, the stiff chemistry and all vertical transport processes (turbulent diffusion, advection, deposition) are integrated in a coupled manner by the second order BDF method ("backward differential formula"). We apply a modification of the code LSODE (Hindmarsh, 1983) with a special linear system solver and a restriction of the BDF order to two. The non-linear corrector iteration is performed by a Newton method in which the sparse linear systems are solved by linear Gauss-Seidel iterations. The time step control is the same as in the original LSODE code. The error control can lead to several implicit time steps per one explicit step. Furthermore, different implicit step sizes may be generated in different blocks. The "large" explicit time step is chosen as a fraction of the CFL number h_{cfl} ("Courant-Friedrichs-Levy number"). This value has to be determined for each Runge-Kutta method individually in order to guarantee stability and positivity. Higher order accuracy and stability conditions for this class of IMEX schemes are investigated in Knoth and Wolke (1998a).

(c) Gas phase chemistry. The chemical reaction systems are given in ASCII data files in a notation easy to handle. For reading and interpreting these chemical data a pre-processor has been developed. The output file contains all data structures required for the computation of the chemical term as well as the corresponding Jacobian. Changes within the chemical mechanism or the replacement of the whole chemistry can be performed in

a simple and comprehensive way. Several gas phase mechanisms (e.g., RACM, RADM2, CBM IV) are used successfully in 3D case studies. Time resolved anthropogenic emissions are included in the model via point, area and line sources. It is distinguished between several emitting groups. Biogenic emissions are parameterized in terms of land use type, temperature and radiation.

(d) Aerosol dynamics. For simulation of aerosol-dynamical processes the model MADMAcS I (Wilck and Stratmann, 1997) was included in MUSCAT. The particle size distribution and the aerosol-dynamic processes are described using the modal technique. The mass fractions of all particles within one mode are assumed to be identical. The particle size distribution changes owing to various mechanisms, which are divided into external processes like particle transport by convection and diffusion, deposition and sedimentation as well as internal processes like condensation and coagulation.

(e) Parallelization. The parallelization is based on the distribution of blocks among the processors. Inter-processor communication is realized by means of MPI ("Message-Passing Interface"). The exchange of boundary data is organized as follows. Since the implicit integration does not treat horizontal processes, it can be processed in each column separately, using its own time step control. An exchange of data over block boundaries is necessary only once during each Runge-Kutta sub-step. Each block needs the concentration values in one or two cell rows of its neighbours, according to the order of the advection scheme. The implementation of the boundary exchange is not straightforward because of the different resolutions of the blocks. The possibilities of one cell being assigned to two neighbouring cells or of two cells receiving the same value must be taken into account. We apply the technique of "extended arrays": the blocks use additional boundary stripes on which incoming data of neighbouring blocks can be stored. Hence, each processor only needs memory for the data of blocks that are assigned to it.

Online-coupling to the parallel meteorological model LM

In Wolke and Knoth (2000) an online-coupling technique between the chemistry-transport code MUSCAT and the mesoscale non-hydrostatic meteorological model METRAS (Schlünzen, 1990) is proposed. The meteorological and the chemistry-transport algorithms have their own separate time step control. The coupling procedure is adapted to the applied IMEX schemes in the chemistry-transport code. Each stage of the IMEX scheme requires a new calculation of the horizontal fluxes and an implicit integration cycle. The new mass-conservative coupling provides constant time-averaged horizontal wind fields for the computation of the horizontal fluxes and time-interpolated meteorological fields (vertical exchange coefficient and wind speed, temperature, humidity) during the implicit integration part. In this way, average temporal changes of the meteorological values are considered, even though the meteorological solver generally generates these variations with a more accurate time resolution.

All meteorological fields are given with respect to the equidistant horizontal meteorological grid. They have to be averaged or interpolated from the base grid into the block-structured chemistry-transport grid with different resolutions. The velocity field is supplied by its normal components on the faces of each grid cell, and their corresponding contravariant mass flux components fulfill a discrete version of the continuity equation in each grid cell. This property has to be preserved for refined and coarsened cells. Because only the mass flux components are needed for the advective transport of a scalar, the necessary interpolation is carried out for these values. The interpolation is done recursively starting from the meteorological level.

The same approach is applied to the coupling with the meteorological driver LM. Since LM solves a compressible version of the model equations, an additional adjustment of the meteorological data is necessary. The velocity components are projected such that a discrete version of the continuity equation is satisfied. The main task of this projection is the solution of an elliptic equation by a preconditioned conjugate gradient method. This is also done in parallel on the LM

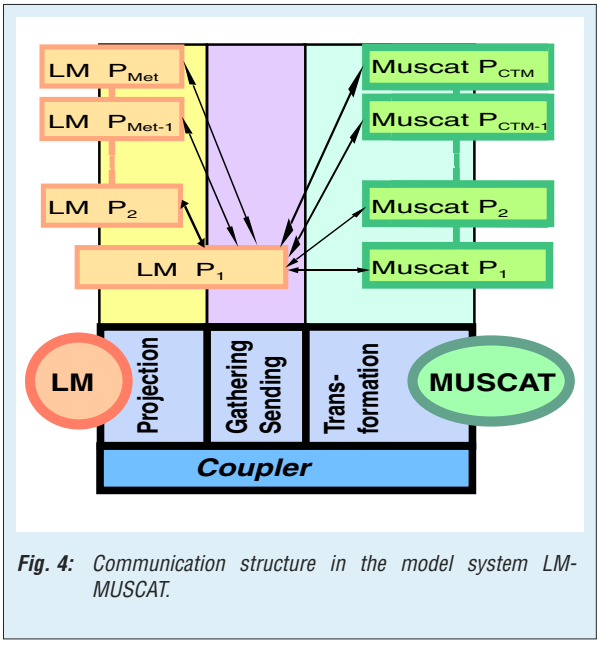

Fig. 4: Communication structure in the model system LM-MUSCAT.

processors. The projected wind fields and the other meteorological data are gathered by one of the LM processors. This processor communicates directly with each of the MUSCAT processors, see Figure 4.

Parallel performance and load balancing

To discuss parallel performance and run time behavior of the model system a typical meteorological "summer smog" situation has been set up. The model area covers approximately 640 km x 640 km with a multiscale grid with resolutions of about 4 km-8 km-16 km, see Figure 5. In the vertical direction the model domain is divided into 19 non-equidistant layers between the surface and a height of approximately 3000 m. The used chemical mechanism RACM (Stockwell et al., 1997) considers 73 species and 237 reactions. Time-dependent emission data from both point and area sources are taken into account.

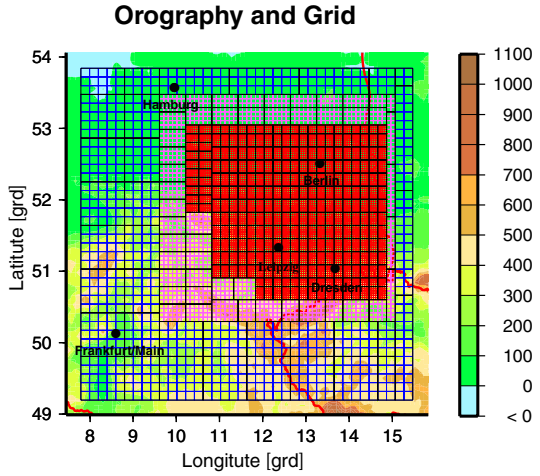

Fig. 5: Orography and multiblock grid for reference simulation on parallel performance.

All tests are run on a Cray T3E with different numbers of processors. In all cases, 20% of the available processors are used for the meteorological code LM, the others for MUSCAT. The equidistant meteorological grid is decomposed in several subdomains the number of which corresponds to the number of LM processors. The MUSCAT runs are performed with two different prescribed error tolerances *Tol* of the BDF integrator.

Dynamical load balancing. Consider a static partition where the blocks are distributed between the processors only once at the beginning of the programs run time. Here, we use the number of horizontal cells (i.e., of columns) as measure of the work load of the respective block. Therefore, the total number of horizontal cells of each processor is to be balanced. This is achieved by the grid-partitioning tool ParMETIS (Karypis et al., 1998). It optimizes both the balance of columns and the "edge cut", i.e., it takes care of short inter-processor border lines.
In order to improve the load balance, techniques for redistribution of blocks have been implemented. A blocks work load is estimated using the numbers of Jacobian and function evaluations applied during a past time period. According to the work loads of the blocks, ParMETIS searches for a better distribution, besides minimizing the movements of blocks. The communication required for the exchange of block data can be done by means of similar strategies as for the boundary exchange.

Performance and scalability. In the three cases, only the work load of the MUSCAT processors is changed by the step control and by the dynamic load balancing procedure. The run time behavior of the LM processors differs only in their idle times (see also Figure 7). The parallel performance of the model runs is presented in Figure 6. In the Figure with CPU times, a logarithmic scale is used for the "number of processor" axis. Higher accuracy requirements increase the work load needed for

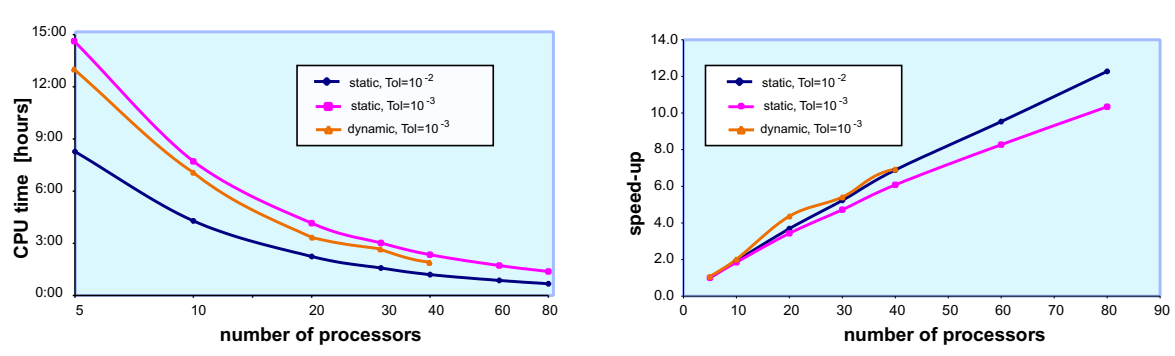

Fig. 6: CPU times and speed-up on the Cray T3E.

the implicit integration in MUSCAT significantly. By using dynamic load balancing the CPU times can be reduced by a factor of about 0.8-0.9 in comparison to the static grid decomposition. The speed-up is defined with respect to the runs with 5 processors. For $Tol=10^{-3}$ we take the dynamic case as reference case. An optimal speed up is reached if the CPU curves show a linear behaviour with slope one. The measured speed-up values show that the code is scalable up to 80 processors.
The computational behavior for the 40 processor run is presented in more detail in Figure 7. In all cases the coupling effort is negligible. As expected, the idle times of the LM processors grow if the work load of the MUSCAT processors caused by tighter tolerances is increased. Therefore, the number of LM processors can be reduced especially in the runs with higher accuracy. However, the basic strategy consists of avoiding idle times on the MUSCAT processors side. The costs for the projection of the wind fields are comparable with that of the LM computations. But it seems that this work load can be reduced by more suitable termination criteria of the cg-method ("conjungate gradient method").

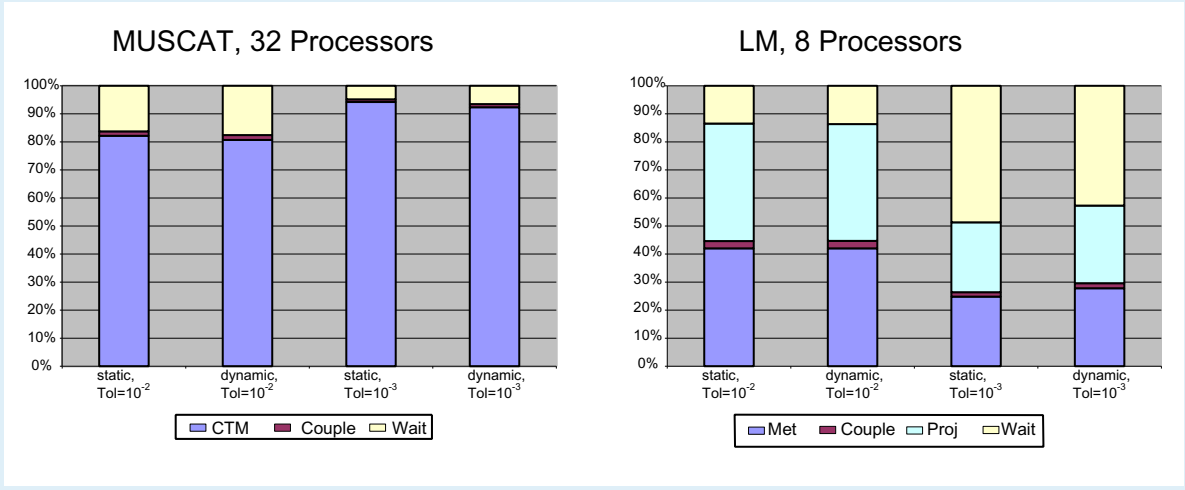

Fig. 7: *Analysis of the required CPU time for runs with 40 processors.*

Simulation for CityDelta

In its first phase, CityDelta is focussed on the following questions:
(1) What is the influence of local versus regional emission reductions on health-relevant matrices for fine particle and ozone in urban air ?
(2) What are the differences between predictions from regional models and those obtained with finer resolved models ?
(3) What is the agreement between different scale dispersion models on the level of response to emission changes ?
To answer these questions, long-term simulations for the city areas of London, Paris, Marseille, Milan, Copenhagen, Berlin, Prague and Katowice will be performed by alltogether 19 modelling groups from several European countries. IfT is focussing on the city area of Berlin.
For long-term simulations it is of crucial importance to compromise between the degree of model sophistication and project requirements. In the present study, the model will be integrated over the whole year 1999 (reference year). Primary project intention is to determine so-called Deltas, i.e., differences between different models and their sensitivity against changes in emissions and grid resolution. Although model validation is of lower ranking in the CityDelta framework it is important for the assessment of the reliability of simulation results. Therefore, accompanying validation studies will be performed. CityDelta can best be described as "screening study".

To accomodate the project requirements, especially with regard to long-term integration, some model simplifications have been realized:
(a) The MADMAcS aerosol model has been substituted by a more simplified aerosol model similar to that of the EMEP Eulerian model MADE50 (Multi-level Acid Deposition model for Europe with 50 km resolution)(e.g., Berge, 1993, 1997; Jakobsen et al., 1995, 1996, 1997; Jonson and Berge, 1995). In this approach, the formation of ammonium sulphat and ammonium nitrat aerosols is calculated from equilibrium reactions (mass-based approach to calculate PM2.5 and PM10).
(b) To ensure comparability with other model results, in the framework of the multi-block grid structure an uniform grid resolution is used.
(c) For consistency reasons, it is important that all models use similar boundary conditions. The

boundary conditions for the chemical species have been provided by the Regional EMEP model. With regard to required CPU time and planned time schedule the option to provide boundary conditions from a preceding large scale LM-MUSCAT simulation has been dispensed (this issue will be pursued in parallel). The boundary conditions for chemistry include 24 gas- and 5 aerosol-phase species on 20 EMEP vertical levels with a time resolution of 3 hours.

(d) For emissions, the Berlin Emission Inventory provided by the Federal Environmental Agency (UBA) and TrUmF Troposphärische Umweltforschung, FU Berlin has been taken (Stern, 2002). This emission inventory includes emissions from the administration of the German states Berlin, Brandenburg, Sachsen-Anhalt and Sachsen, which have been harmonized with respect to structure, coordinate system, species and source groups. It contains species such as NO_x, NMVOC, SO_2, CO, PM10, PM2.5, benzene. Source group categories of CORINAIR have been used, i.e., snap code 01-10. It should be noted that some CORINAIR source groups are not available, for instance agriculture. Data are given in a geographical co-ordinate system with area sources resolved by 0.03125° longitude and 0.015625° latitude. For Sachsen-Anhalt there are no data west of 11° longitude, for Sachsen there are no data south of 51° latitude. There are also no finely resolved data for Poland and Mecklenburg-Vorpommern. These gaps are filled with CORINAIR large scale data with grid resolution 0.5° longitude and 0.25° latitude. Emissions are given as annual averages for the reference year 1999 for area and point sources. There is no information about NH_3 and biogenic VOC emissions. For point sources volumetric flow rate, stack height, exit temperature, stack area, monthly operation hours and emission mass flux are given. Monthly, daily and hourly time variations are also included in the emission inventory (Stern, 2002).

Figure 8 shows the flow diagram for the LM-MUSCAT CityDelta modelling. Via a project contract with DWD 6-hour GME analysis data are available for initialization and large-scale forcing of LM. GME data from several databases have been "merged" to obtain an unique dataset with meteorological boundary conditions for the whole year 1999. Using the pre-processor GME2LM, the GME analysis data have been interpolated to the

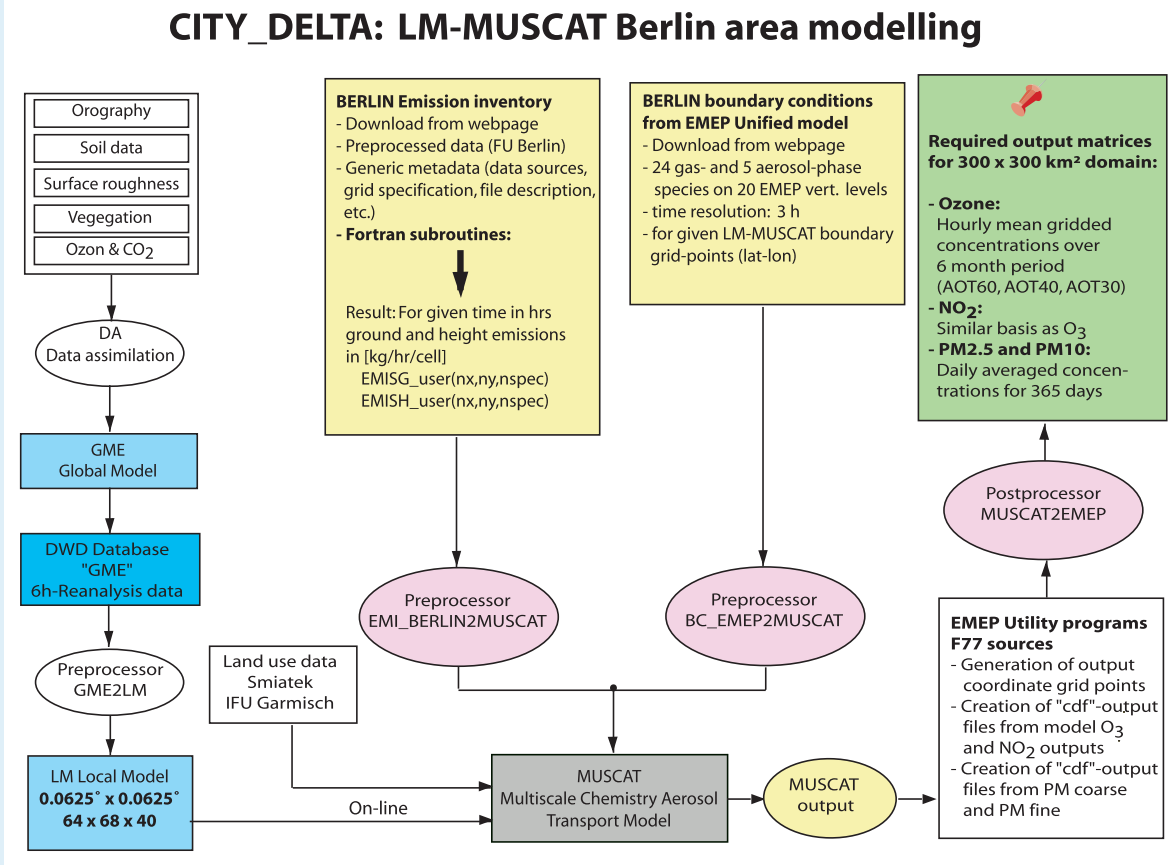

Fig. 8: CityDelta modelling flow diagram.

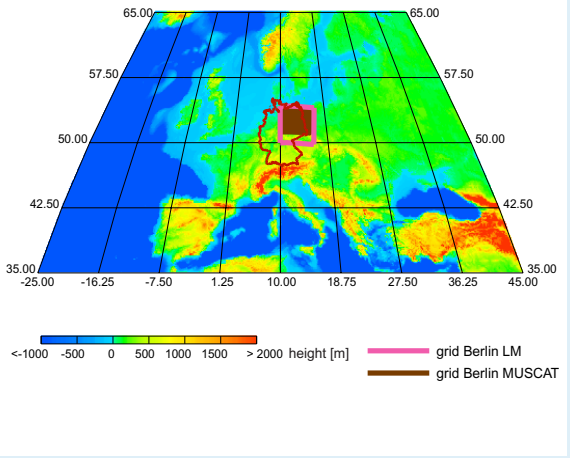

Fig. 9: Location of LM and MUSCAT grid.

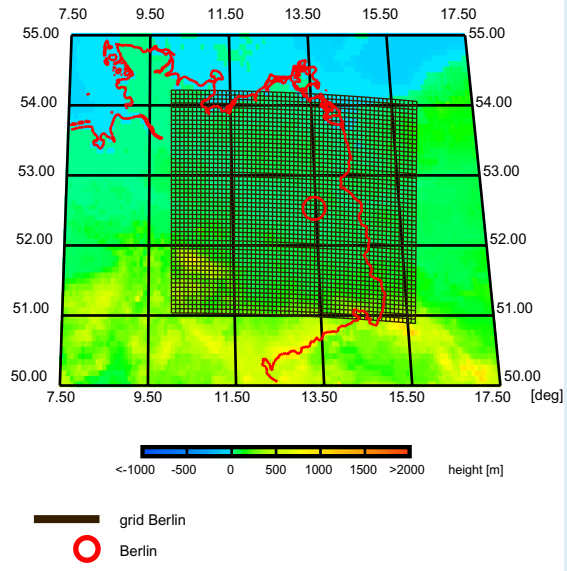

Fig. 10: Evaluation domain.

LM model domain (64 x 68 x 40 grid cells) with horizontal grid resolution of 0.0625° longitude and 0.0625° latitude (approx. 7.5 km). Several pre-processors for input of emission data and chemical boundary conditions as well as for data post-processing have been developed. The grid resolution of MUSCAT is identical to that of LM, but the MUSCAT grid has been imbedded into the LM grid (Figure 9). Thus, the influence of the nudging function in the LM boundary zone is limited to regions outside of the evaluation domain. The propagation of boundary effects into the region of interest is avoided (Figure 10).

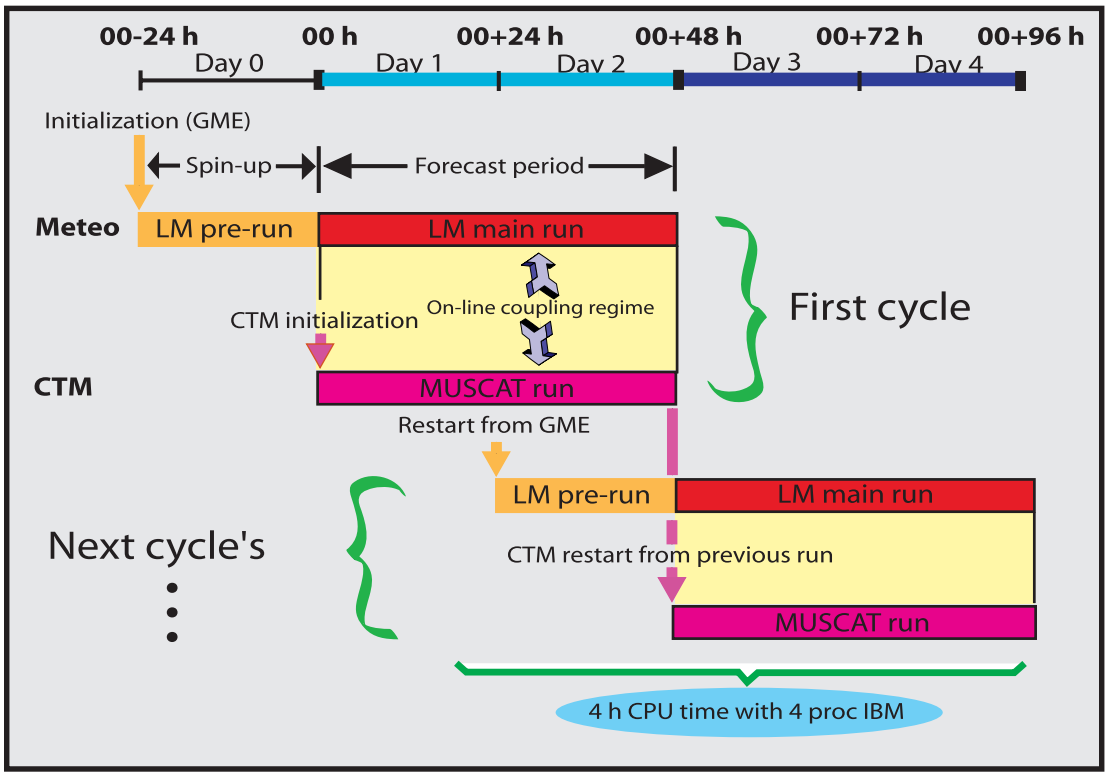

Fig. 11: CityDelta simulation cycles.

To keep the long-term evolution of meteorological fields as close as possible nearby observations, the integration period has been subdivided into overlapping short-term forecast cycles. Each of these cycles consists of an one-day LM pre-run for spin-up of the meteorology followed a two-day coupled run of meteorology and meteorology (Figure 11). The LM pre-run is initialized with GME data. For initializing MUSCAT final concentration fields of the previous cycle are taken. Thus, the CTM is actually restarted rather than initialized. While model initialization is an update of complete 3D fields, the external forcing via nudging is limitited to the boundary grid cells of the model domain, only. The designed forecast cycle has been compared with other cycles and found to be suitable with respect to consistency of meteorological fields between two cycles.

Figures 12 and 13 show, for example, the annual emissions of NO_x and PM10 for area source. Next, preliminary results from the pilot simulation May-June 1999 will be shown. Modelled wind and concentration fields reveal a rational and comprehensible overall evolution of meteorological fields as well as chemical concentrations and particulate matter in space and time. The development of meteorological fields corresponds

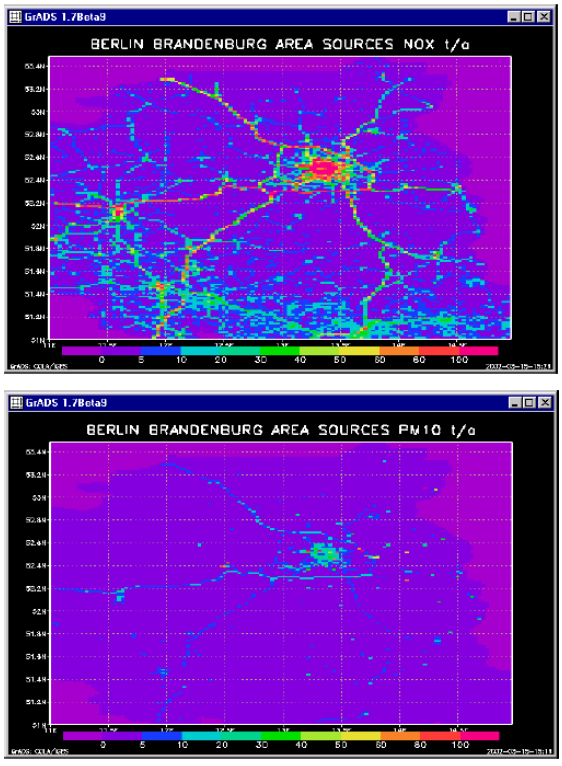

Fig. 12: *Annual NO_x and PM10 area emissions in tons/year for 1999 (Stern, 2002).*

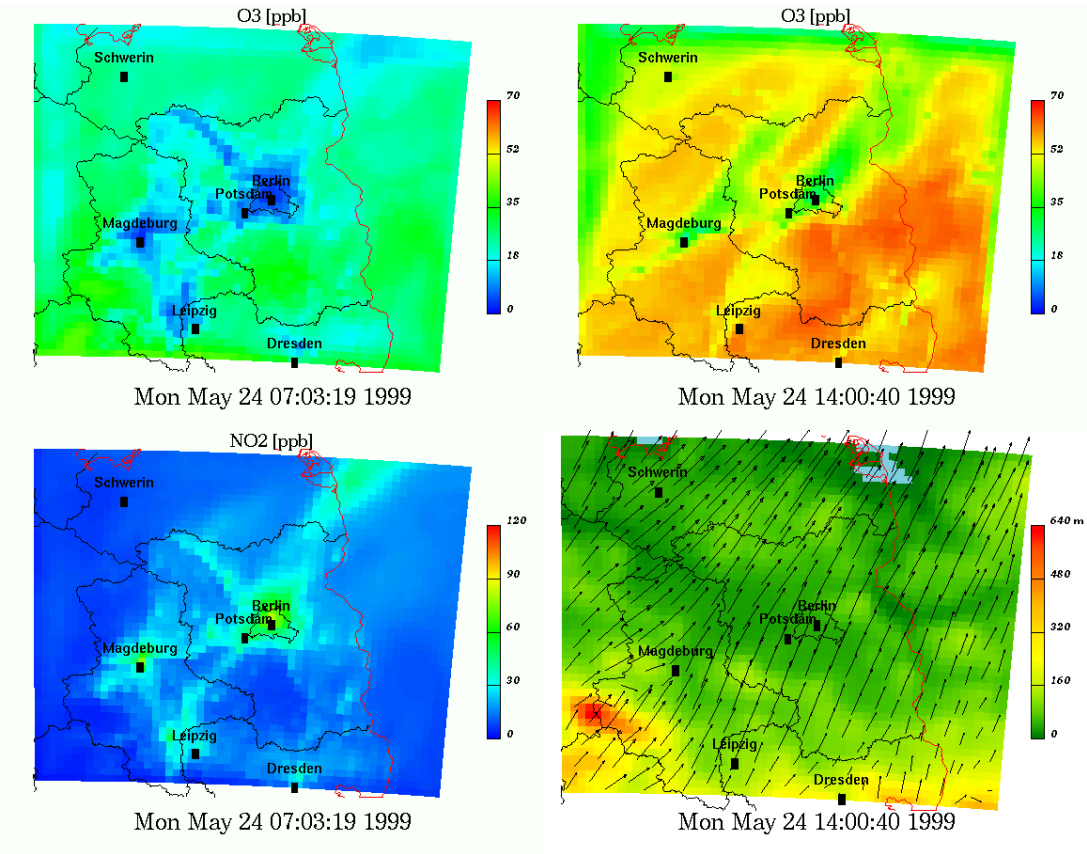

Fig. 13: *Ozone, NO_2 concentration fields and wind fields.*

well with the synoptic weather situation. Also small scale convergences in the wind feld variable in space and time obey common expectations of mesoscale modelling.

Simulated wind, ozone and NO_2 concentrations during an ozone episode shown in Figure 13 as an example. In the last ten days of May 1999, large scale advection of ozone from southwest and southeast led to a clear enhancement of the background ozone concentration in the region of interest except for Berlin, where ozone remained at comparably low levels. Beginning in the late afternoon, accelerating in the evening and during the night ozone systematically decreases. This evolution was pronounced along the motorways. Modelled data are compared with meteorological and air pollution measurements from the sites shown in Figure 14. As seen from Figure 15 for

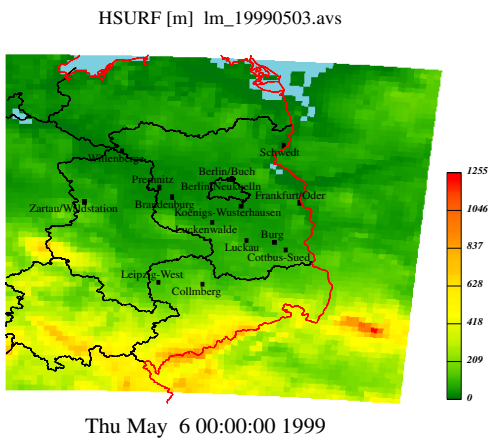

Fig. 14: *Observation network.*

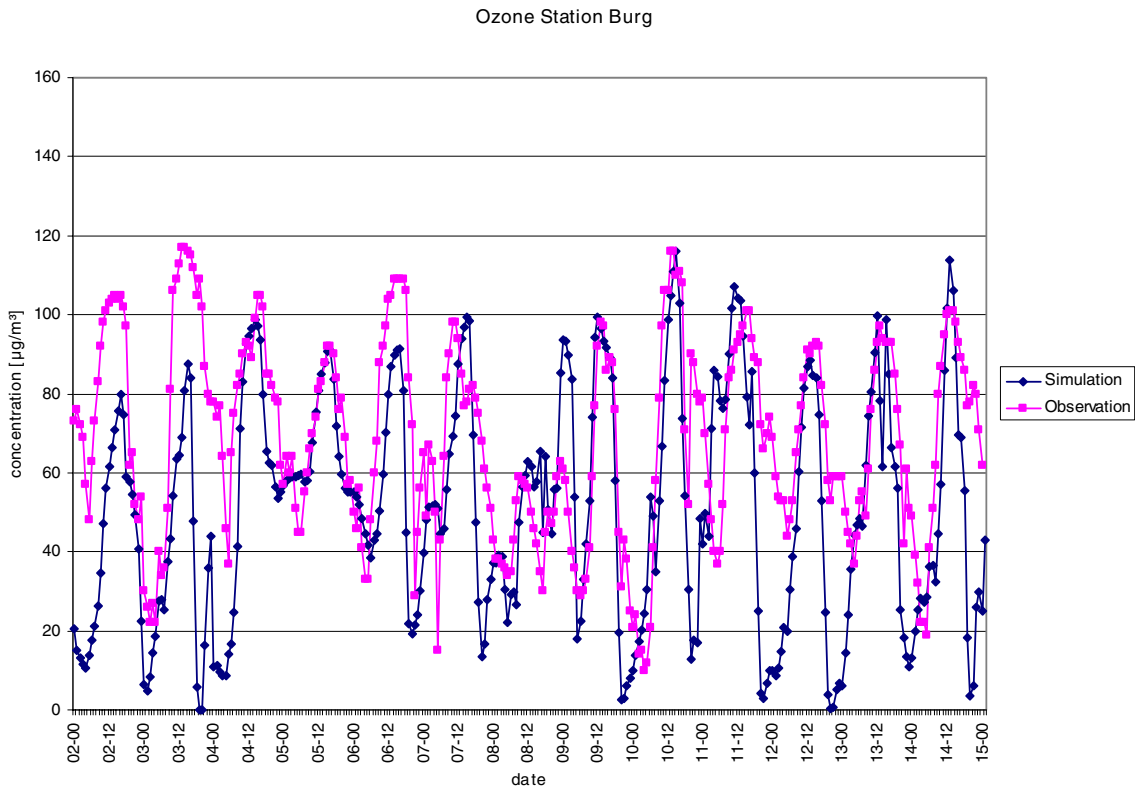

Fig. 15: *Modelled and observed ozone concentration, May 2-15 1999, Burg (rural) (abscissa: e.g., 03-12, that is May 3, 12 UTC).*

the rural station Burg, the agreement between observed and predicted time series of hourly ozone concentrations is encouraging. A closer look reveals that ozone has been underestimated, especially at night, and NO_x is clearly overpredicted. One reason could be that the vertical exchange in LM is too low at night, leading to higher NO_x in the lowest layer than observed. Another important issue regards the NO_x emissions. The reported NO_x emissions in the Berlin Emission Inventory are considerably higher than those reported in the Sachsen emission inventory generated by IfT. A reference simulation with NO_x emissions reduced by approximately 30 percent results in a better agreement of observed and modelled ozone concentration, i.e., leading to an increase of ozone concentration. Nevertheless, even if in this case the ozone maxima agree better with observations, the resulting ozone minima are too high. Further investigations are necessary to understand these discrepancies. Screening of

PM simulations shows also a coherent evolution. For validation there are no PM2.5 observations available. PM10 has been measured at three stations, only.

Conclusions and outlook

The model system LM-MUSCAT is a powerful instrument for environmental studies. The parallel efficiency is encouraging. The results from the CityDelta pilot study are encouraging too. The availability of a fully 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.

Acknowledgements

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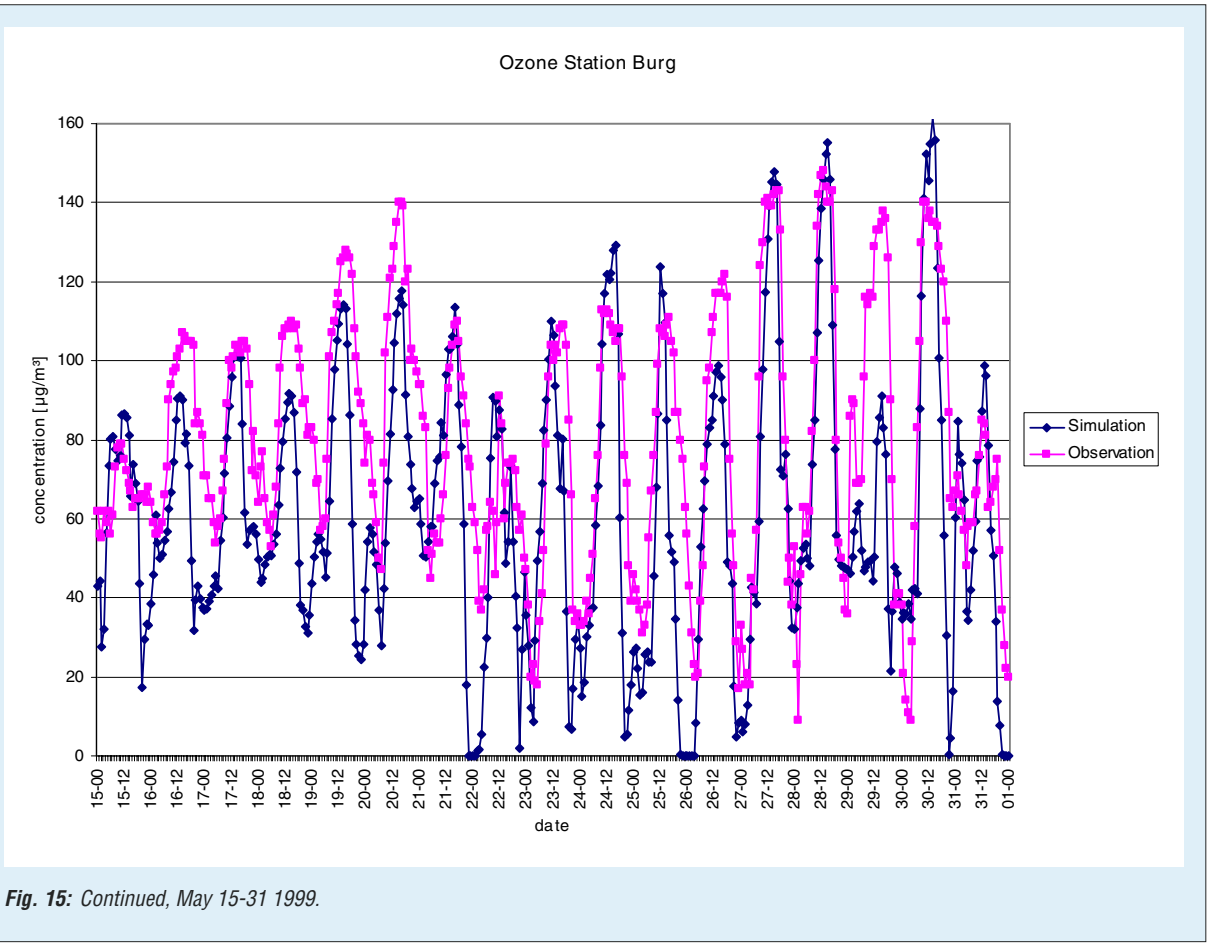

Fig. 15: *Continued, May 15-31 1999.*

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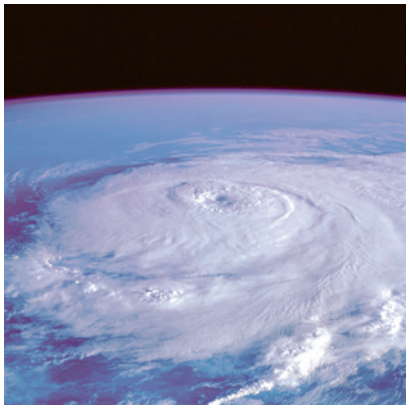
Cooperation
- John von Neumann Institute for Computing Jülich
- Zentrum für Hochleistungsrechnen, TU Dresden
- Deutscher Wetterdienst Offenbach
- Joint Research Center



Short contributions *to current research projects*

A detailed tropospheric aqueous phase chemical mechanism

Zoltan Majdik and Hartmut Herrmann

The development of the detailed and extended chemical mechanism describing tropospheric aqueous phase chemistry **CAPRAM** 2.4 (**MODAC** mechanism) (**C**hemical **A**queous **P**hase **Ra**dical **M**echanism, **Mo**del **D**evelopment for Tropospheric **A**erosol and Cloud **C**hemistry Herrmann et al., 2000, Ervens et al., 2002, http://www.tropos.de/CHEMIE/multimod/CAPRAM/CAPRAM24.pdf) proceeded in three main directions:

- Application of the model to a size-segregated system in order to investigate the influence of size and liquid water distribution on the mass transport and on the multiphase chemistry in cloud droplets for three different standard CAPRAM scenarios.
- Extension of CAPRAM 2.4 (MODAC mechanism) to a more realistic organic chemistry considering at least C_3 and C_4 compounds.
- Development of the CAPRAM halogen module for the simulation of halogen activation for aerosols and clouds.

phase mechanism **RACM** (**R**egional **A**tmospheric **C**hemistry **M**echanism) (Stockwell et al., 1997). Uptake processes of soluble species are included in the mechanism following the approach by Schwartz (1986) considering gas phase diffusion, mass accommodation coefficients, Henry solubility and chemical reaction within the aqueous phase. The calculations were performed considering different numbers of size bins for the particle size range 1 µm < $r_{droplet}$ < 64 µm assuming a lognormal number-size distribution. In the present study three different CAPRAM standard scenarios (urban, remote and marine) are considered which differ only in the initial concentrations and in the emission/deposition fluxes.

The results show that size segregation has a great effect on phase transfer processes and, as a consequence, on diurnal concentration profiles. An interesting size effect can be observed during the night in the case of Fe(III)-Fe(II) especially in the remote and marine cases as shown in Figure 1.

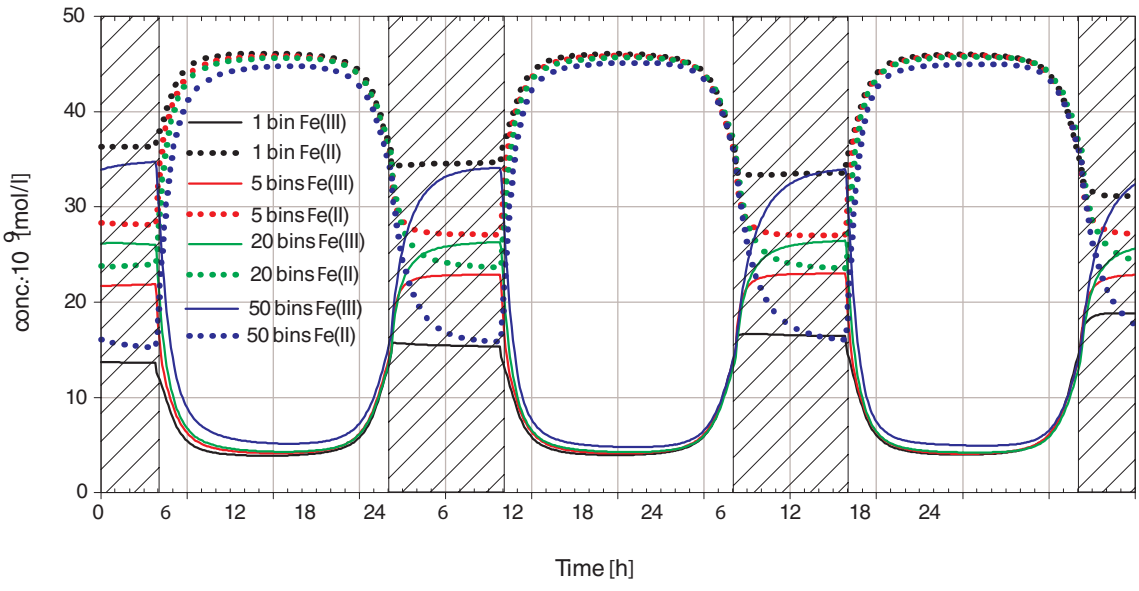


Fig. 1: *Decreasing [Fe(II)] and increasing [Fe(III)] with a finer droplet spectrum.*

Model description and simulation results

Size-resolved system

The detailed and extended chemical mechanism describing tropospheric aqueous phase chemistry (147 species and 438 reactions) CAPRAM 2.4 (MODAC mechanism) was used coupled to the gas

Increasing the number of size bins (1, 5, 20, 50) causes higher Fe(III) concentration, [Fe(III)]. At the same time [Fe(II)] decreases. Table 1 contains results obtained for three scenarios, considering a monodisperse distribution (r = 10 µm) and a size dependent distribution with 50 different size bins (r = 1 µm – 64 µm). The numbers in Table 1 represent maximum concentrations and are

expressed in cm^{-3} for gas phase species and $mol \cdot l^{-1}$ for aqueous phase species. For most species, the biggest differences between the two distributions are found for the marine scenario, due to the stronger contribution of phase transfer processes compared to the other scenarios.

Glyoxal and methyl glyoxal are ring cleavage products from the aromatic oxidation in the gas phase. Their oxidation in the aqueous phase ultimately leads to oxalic and pyruvic acid, respectively (Figure 2). These pathways may significantly contribute to the sources of oxalate and pyruvate identified both in polluted and unpolluted environments. Despite the implementation of such processes, at the current state of multiphase mechanism development source processes of dicarboxylic acids are still missing.

gas phase	urban 1 size bins	50 size bins	remote 1 size bins	50 size bins	marine 1 size bins	50 size bins
O_3	$3.5 \cdot 10^{12}$	$3.75 \cdot 10^{12}$	$5.8 \cdot 10^{11}$	$7 \cdot 10^{11}$	$6.4 \cdot 10^{11}$	$5.75 \cdot 10^{11}$
NO_2	$6 \cdot 10^{10}$	$5.2 \cdot 10^{10}$	$8 \cdot 10^{9}$	$6 \cdot 10^{9}$	$1 \cdot 10^{9}$	$1 \cdot 10^{9}$
NO	$1.35 \cdot 10^{10}$	$1 \cdot 10^{10}$	$6 \cdot 10^{10}$	$4 \cdot 10^{10}$	$7 \cdot 10^{8}$	$8.4 \cdot 10^{8}$
NO_3	$4 \cdot 10^{7}$	$6 \cdot 10^{7}$	$4.5 \cdot 10^{6}$	$7.5 \cdot 10^{6}$	$1.3 \cdot 10^{6}$	$1 \cdot 10^{4}$
OH	$1.75 \cdot 10^{6}$	$2 \cdot 10^{6}$	$3.25 \cdot 10^{6}$	$5 \cdot 10^{6}$	$2.75 \cdot 10^{6}$	$1.25 \cdot 10^{6}$
HONO	$1.8 \cdot 10^{9}$	$2.65 \cdot 10^{9}$	$4.7 \cdot 10^{8}$	$6 \cdot 10^{8}$	$7 \cdot 10^{7}$	$1 \cdot 10^{4}$
N_2O_5	$5 \cdot 10^{7}$	$2 \cdot 10^{8}$	$1.75 \cdot 10^{6}$	$4.5 \cdot 10^{6}$	$7.5 \cdot 10^{5}$	$5 \cdot 10^{0}$
HO_2NO_2	$2.25 \cdot 10^{9}$	$2.5 \cdot 10^{9}$	$1 \cdot 10^{7}$	$3 \cdot 10^{7}$	$1 \cdot 10^{6}$	$1 \cdot 10^{1}$
aqueous phase						
OH	$1 \cdot 10^{-13}$	$6 \cdot 10^{-14}$	$2 \cdot 10^{-13}$	$1.5 \cdot 10^{-13}$	$5 \cdot 10^{-13}$	$1.3 \cdot 10^{-12}$
NO_2	$7 \cdot 10^{-11}$	$6 \cdot 10^{-11}$	$8 \cdot 10^{-12}$	$7 \cdot 10^{-12}$	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$
Fe(II)	$1.5 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$4.5 \cdot 10^{-7}$	$4.5 \cdot 10^{-7}$	$4.5 \cdot 10^{-8}$	$4.5 \cdot 10^{-8}$
Fe(III)	$5 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	$2 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$1.5 \cdot 10^{-8}$	$3.5 \cdot 10^{-8}$
Cu^{2+}	$2.5 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$5 \cdot 10^{-8}$	$5 \cdot 10^{-8}$	$1 \cdot 10^{-9}$	$1 \cdot 10^{-9}$
Cu^{+}	$2 \cdot 10^{-9}$	$1.6 \cdot 10^{-9}$	$1 \cdot 10^{-9}$	$1 \cdot 10^{-9}$	$1 \cdot 10^{-10}$	$1 \cdot 10^{-11}$
HO_2	$6.2 \cdot 10^{-11}$	$4 \cdot 10^{-11}$	$2 \cdot 10^{-10}$	$2 \cdot 10^{-10}$	$2.9 \cdot 10^{-10}$	$3 \cdot 10^{-11}$
O_2^-	$1.4 \cdot 10^{-8}$	$1 \cdot 10^{-8}$	$7.8 \cdot 10^{-10}$	$6.4 \cdot 10^{10}$	$4.5 \cdot 10^{-9}$	$1.8 \cdot 10^{-10}$
O_3	$2 \cdot 10^{-9}$	$2.15 \cdot 10^{-9}$	$3.4 \cdot 10^{-9}$	$4 \cdot 10^{-9}$	$3.5 \cdot 10^{-10}$	$3 \cdot 10^{-10}$
HONO	$1.5 \cdot 10^{-9}$	$2.5 \cdot 10^{-9}$	$2 \cdot 10^{-9}$	$2.3 \cdot 10^{-9}$	$1 \cdot 10^{-10}$	$5 \cdot 10^{-14}$
NO_2^-	$1 \cdot 10^{-10}$	$1.1 \cdot 10^{-10}$	$7.5 \cdot 10^{-7}$	$7.5 \cdot 10^{-7}$	$7.5 \cdot 10^{-8}$	$5 \cdot 10^{-11}$
HO_2NO_2	$2.5 \cdot 10^{-6}$	$2.5 \cdot 10^{-6}$	$1.075 \cdot 10^{-9}$	$2.5 \cdot 10^{-9}$	$5 \cdot 10^{-10}$	$1 \cdot 10^{-15}$
Oxalate	$6 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	$3.75 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$4.75 \cdot 10^{-7}$	$4 \cdot 10^{-8}$
HSO_3^-	$7.5 \cdot 10^{-9}$	$1.1 \cdot 10^{-8}$	$1 \cdot 10^{-7}$	$1 \cdot 10^{-7}$	$1 \cdot 10^{-8}$	$5 \cdot 10^{-10}$
SO_3^{2-}	$1 \cdot 10^{-13}$	$2 \cdot 10^{-13}$	$8 \cdot 10^{-9}$	$8 \cdot 10^{-9}$	$1 \cdot 10^{-9}$	$5 \cdot 10^{-11}$

Tab. 1: *Overview of the size effect of key species.*

Organic extension

After evaporation of cloud water, a residual aerosol particle contains sulfate, nitrate, organics and trace metals. The mass of soluble organic carbon in such particulate samples contributes up to 10% of the total aerosol mass but mostly fractions of 1% - 2% are found. For the organic chemistry extension of CAPRAM, especially formation processes of low volatile di- and ketocarboxylic acids were considered giving insight about the composition of the organic content in aerosol particles. Chemical source and sink processes of mono- and difunctional C_3 and C_4 compounds are considered. For the simulations the organic extension was coupled to the CAPRAM 2.4 reduced mechanism which is suitable for larger scale simulations.

Halogen module

The tropospheric particle phase plays an important role as a source of halogen-containing species. The effect of halogen atoms on the ozone budget was emphasized in both field and modeling studies. It has been demonstrated in model studies before that sea salt aerosol particles act as a source for halogen species such as Cl_2, Br_2, BrCl, HOCl and HOBr, and subsequently Cl, Br, ClO and BrO. In the sea salt particles chemical processes are

mainly controlled by effects of quite low pH values and high ionic strengths. It is known from model studies (Herrmann et al., 2000, Ervens et al., accepted) that not only tropospheric gas phase but also particle phase chemistry may be strongly influenced by radical chemistry. In this study an urban and a marine scenario are considered. The simulations were carried out with CAPRAM 2.4 (MODAC mechanism) coupled to the newly developed CAPRAM halogen module. The mechanism presented here includes direct uptake or release processes of these radicals for the first time. In contrast to other mechanisms explaining halogen activation, the main focus was to elucidate better the role of radical processes in the particle phase and check if not only marine aerosols but also clouds may in principle represent sources of active halogens in the troposphere.

It has been shown that, at the current stage of mechanism development, in the case of marine and urban clouds direct phase transfer of halogen atoms, formed in the particle phase by radical conversions via NO_3, SO_4^- and OH, represents an important source of halogen atoms in the gas phase. If the particle phase consists of sea salt the multiphase reaction system including the formation of Cl_2 and Br_2 in the particle phase and photolytic processes in the gas phase ar responsible for halogen atom formation in the gas phase.

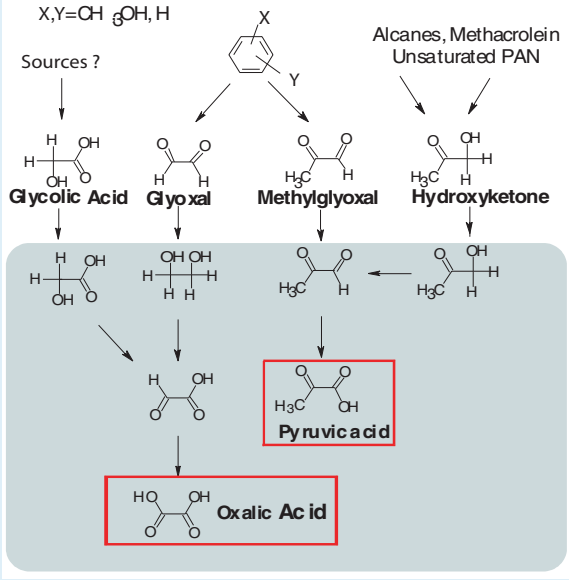


Fig. 2: Formation pathways of pyruvate and oxalate in the multiphase system.

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Interaction of free radicals with organics in the tropospheric aqueous phase

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Introduction

Free radicals such as OH, NO_3 and halogen contain radicals, play an important role in the chemistry of aqueous tropospheric particles (Zellner and Herrmann, 1995). The OH radical is the most important oxidant in both the tropospheric gas and aqueous phase i.e. in clouds droplets, fog, rain and aqueous particles. Among the various organic compounds emitted are organic solvents which are used in a large number of industrial processes (i.e. cleaning and degreasing processes, polymer disposal agents and adhesives). Due to their volatility, in many of these processes they are emitted either directly or indirectly into the atmosphere. A number of organic compounds employed as solvents at the present time has been shown to have adverse health effects (i.e., carcinogenic, mutagenic and reprotoxic properties). The rate constants of the OH radical reactions with such organic compounds have been investigated in aqueous solution, under different conditions.
Nitrate radicals in aqueous solution may undergo reactions with aromatic compounds which have been shown to proceed faster than the corresponding gas-phase reactions by orders of magnitude. From phenol, a mixture of *ortho*-and *para*-nitrophenol is formed. The corresponding reaction in gas phase has been shown by us to give essentially the *ortho* isomer. The *ortho*/*para* ratio in the nitration of phenol could be a useful tool for distinguishing among these mechanisms.
Tropospheric particles may release halogen atoms into the gas phase. Model calculations suggest that halogen-atom and related species could play a role not just in the gas phase but also in cloud water and, possibly, in aerosol particles. Therefore, Reactions of Cl atom with organic compounds were studied at different temperature and ionic strength by HeCd-laser absorption.

Kinetic studies

A laser-photolysis long path laser absorption (LP-LPLA) set-up (Figure 1) was used for kinetic studies of OH radical reactions. Due to the small extinction coefficient of OH and the overlapping of the stronger absorption of organic peroxyl radicals formed in correspondence to the OH decay, kinetic measurements were carried out by competition kinetics using thiocyanate as reference substance.
In contrast to most existing studies the reaction rates were investigated as a function of temperature. Considering available temperature-dependent values both from literature studies and from the present study, it was shown that correlation exist between the activation energy and the bond dissociation energy (Table 1 and Figure 2).

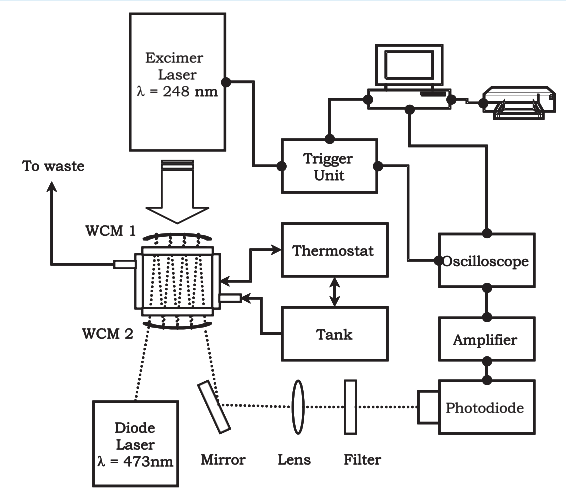

Fig. 1: *Laser flash photolysis set-up (LFP-LPLA).*

Compound	k_{298K} / $M^{-1}s^{-1}$	Ea / kJ/mol	BDE / kJ mol^{-1}
Acetone	$(2.1 \pm 0.6) \cdot 10^8$	(18±11)	411
2-Butanone	$(1.5 \pm 0.7) \cdot 10^9$	(23±10)	411
Acetonylacetone	$(7.6 \pm 1.1) \cdot 10^8$	(16±8)	400
Isobutyraldehyde	$(2.9 \pm 1.0) \cdot 10^9$	(6±3)	313
Ethyl Formate	$(3.2 \pm 0.8) \cdot 10^8$	(10±4)	336

Tab. 1: *Observed rate constants for the reaction of OH with organic compounds and BDEs of the weakest C-H bond.*

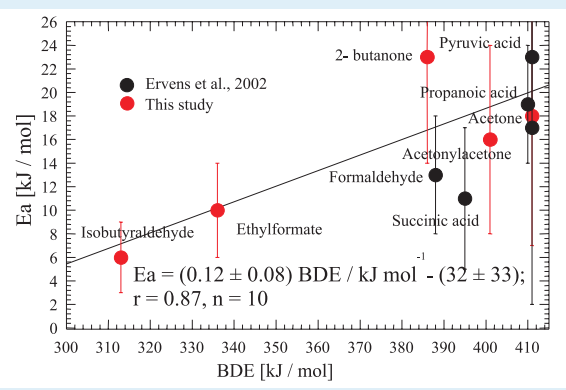

Fig. 2: *Bell-Evans-Polanyi plot for H-atom abstraction reactions of OH in aqueous solution.*

Chlorine atoms were generated by excimer-laser-photolysis at 248 nm of aqueous solutions containing chloroacetone as a precursor. For the kinetic studies a laser-photolysis-longpath-laser-absorbance apparatus (LP-LPA) was used. The reaction cell could be thermostated, allowing temperature- dependent measurements in the range from 278 K to 318 K. The Cl atom absorption was measured directly using the 325 nm output of an He-Cd laser. The time-resolved Cl atom absorption profiles were monitored by a photodiode and then transferred to a digital oscilloscope.

Compound	k_{2nd} (298K) / M^{-1} s^{-1}	E_A / kJ mol^{-1}	BDE / kJ mol^{-1}
2-Propanol	(3.2 ± 0.7) 10^9	23 ± 8	381 (1)
1-Propanol	(2.2 ± 0.4) 10^9	19 ± 7	385 (2)
Ethanol	(2.2 ± 0.3) 10^9	19 ± 7	389 (1)
Methanol	(1.0 ± 0.1) 10^9	34 ± 6	401 (2)
Hydrated Formaldehyde	(1.4 ± 0.3) 10^9	26 ± 6	388 (2)
Acetic Acid	(1.0 ± 0.2) 10^8	41 ± 11	410 (3)
Propionic Acid	(1.2 ± 0.3) 10^9	44 ± 10	410 (2)*
Isobutyric Acid	(1.7 ± 0.3) 10^9	26 ± 4	397 (2)*
Formic Acid	(2.8 ± 0.3) 10^9	20 ± 6	387 (2)

(1): Lide and Frederikse, 1994; (2): Benson, 1976; (3): Singleton et al., 1989.

Tab. 2: *Observed rate constants at 298K and activation energies for the reaction with Cl atom in aqueous solution; BDE of the weakest C-H bond. * values approximated to the calculated BDEs of, respectively, acetic acid and Isobutanol (C2).*

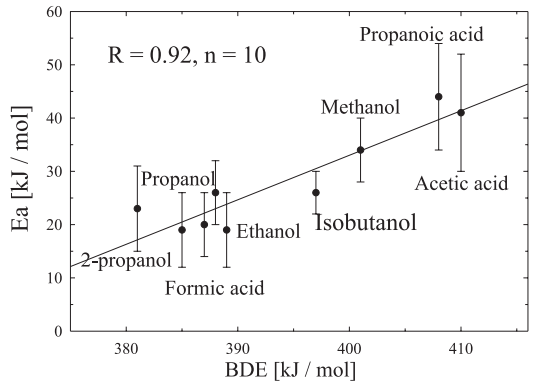

Fig. 3: *Evans-Polanyi plot for H-abstraction reactions of Cl atom in aqueous phase.*

According to the principle of Evans and Polanyi there is a correlation between the activation energy and the enthalpy for reactions that follow the same chemical mechanism (Evans and Polanyi, 1938): $E_a = A + B \cdot \Delta H_r$. To test whether the reactions of $Cl_{(aq)}$ with organic compounds do follow the same reaction mechanism (H-atom-abstraction), the activation energies for the reaction of $Cl_{(aq)}$ with different organic compounds were measured and plotted against the bond dissociation energies (Table 2 and Figure 3).

The kinetic data obtained from this study will be applied as input parameters for a tropospheric multiphase model **CAPRAM** 2.4 (**MODAC** mechanism) (**C**hemical **A**queous **P**hase **Ra**dical **M**echanism, **Mo**del **D**evelopment for Tropospheric **A**erosol and Cloud **C**hemistry) clarifying the influence of cloud chemistry in the tropospheric multiphase system.

Product and uptake studies

The flash photolysis of nitrate anions at λ = 248 nm and of peroxydisulphate anions at λ = 351 nm was used to study the oxidation process of phenol by OH / NO_2 and NO_3 / NO_2 in aqueous solution under different experimental conditions.

An isocratic HPLC procedure was developed and used to analyze mixtures of phenols and nitrophenols in laboratory studies since the relative abundance of the isomeric nitrophenols may be significant for the identification of the mechanism of oxidation process involved.

Two different mononitrophenol (isomer *ortho* - and *para* -) and a dihydroxy derivative (catechol) were identified as the main reaction products by means of HPLC-DAD-ED technique and their yields of formation were directly compared with the initial radical concentrations of OH, NO_2 and NO_3, respectively (Figure 4) (Barzaghi and Herrmann, 2002).

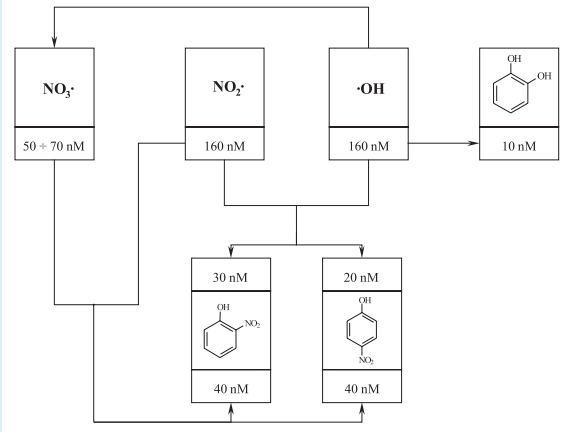

Fig. 4: *Summarising scheme comparing the initial maximal radical yields and the product yield with the possible reaction pathways for the phenol oxidation.*

Furthermore, the uptake of the nitrate radical on a liquid surface was investigated using the novel Single-drop-experiment (Figure 5). A scavenger, namely Alizarin Red S, was added in order to

make the process observable. An estimated lower limit for the mass accommodation coefficient α was derived by means of a computer model which allows corrections for the influence of the gas phase diffusion (Schütze and Herrmann, 2001).

The "Pendant Drop" method which is used for the determination of the surface tension from the shape of a drop was adapted to the system. This allows the characterization of organic films on the drop. The investigation of uptake processes on such organically coated surfaces is planed for the future.

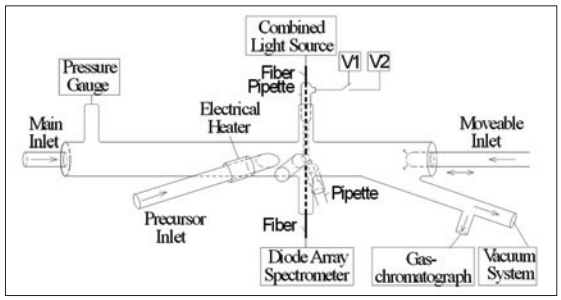

Fig. 5: *Schematic diagram of the flow tube reactor.*

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On collision processes in a mixed-phase cloud

Karoline Diehl, Rebekka Posselt, Martin Simmel, Sabine Wurzler

Introduction

At subzero temperatures, tropospheric clouds are in general mixed-phase clouds, i.e., they consist of super-cooled liquid drops and ice particles. Cold precipitation from these clouds occurs as graupel, hail, or snow: ice particles grow by water vapor diffusion, by riming, or by collision with other ice particles. Warm precipitation results from drop collisions and melted ice particles. Thus, the ice phase is an important requirement for precipitation. An efficient ice forming process is the contact mode where the freezing of a super-cooled drop is initiated by the collision with a potential ice nucleus (Pruppacher and Klett, 1997). Therefore, collisions between aerosol particles, drops, and ice particles are of great importance in mixed-phase clouds. The efficiencies of two of these processes are studied: collisions between drops and aerosol particles and collisions of different kinds of ice particles.

Methods

In the contact mode, the freezing temperature is independent of the drop size (Pruppacher and Klett, 1997). For atmospheric ice nuclei such as soot, mineral and biological particles median freezing temperatures between –4.5° C and –12° C were found (e.g., Pitter and Pruppacher, 1973; Gorbunov et al., 2001; Diehl et al., 2002). However, contact freezing is restricted by the efficiency with which the drops collide with the ice nuclei. This is strongly dependent on drop and particle sizes. Using a two dimensional field of drop and aerosol particle sizes together with the efficiencies according to Kerkweg et al. (2003) allows the combination of different collision partners. For the present case of collision between drops and particles, drop radii ranged from 0.1 to 3000 µm, particle radii from 0.01 to 30 µm.

Higher efficiencies are expected for ice particle collisions than for drop collisions. Reasons are the special shapes of ice crystals with larger and irregular surfaces together with their fall pattern showing secondary motions (Pruppacher and Klett, 1997). Model calculations were undertaken to study the time development of an ice particle size distribution owing to the collision of plates compared to the collision of columns. The collision efficiencies and terminal velocities for the ice particles were calculated according to Posselt et al. (2003). A form factor of the collision products was considered. The calculations started with an exponential size distribution of the mass density of the ice particles.

Results

The collision efficiency between drops and particles is given in Figure 1. One can note that high efficiencies can be expected for collisions of large drops with large particles or for collisions of small drops with small particles. Thus, drop freezing via contact freezing can be supposed for large drops by large biological particles (i.e., pollen and leaf litter) and large mineral particles, for small drops by soot particles, small biological particles (i.e., bacteria), and small mineral particles.

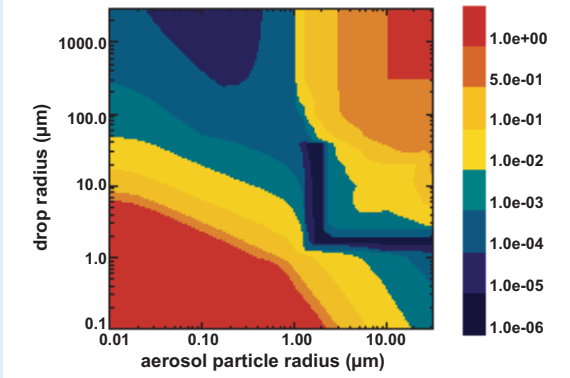

Fig. 1: Collision efficiencies for drops and aerosol particles.

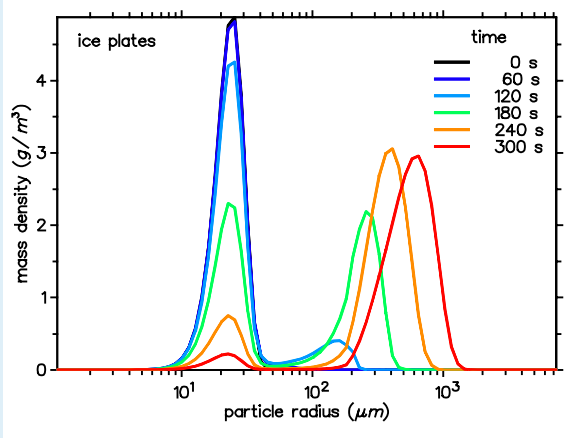

Fig. 2: Time development of the mass density distribution in (g/m³) for colliding ice plates.

Figure 2 shows the time development of the mass density distribution per bin for ice plates, Figure 3 for ice columns. One can notice that due to collision processes in both cases the single modal distributions developed into bimodal distributions towards larger ice particle sizes. However, for ice plates the time development resulted in larger sizes and more ice mass in the precipitation sized ice particle range than for ice columns. These significant differences indicate the importance of the ice particle shape for collision processes.

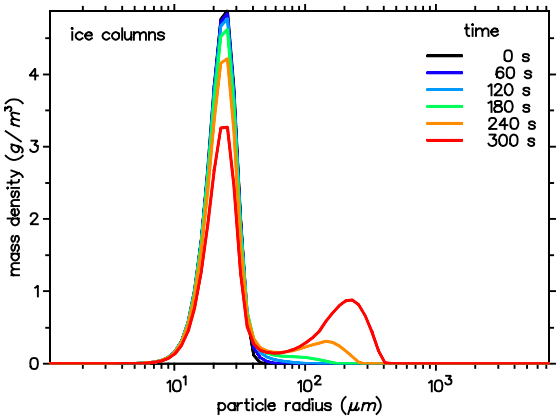

Fig. 3: *Time development of the mass density distribution in (g/m³) for colliding ice columns.*

Conclusions and outlook

Collision processes in mixed-phase clouds determine not only ice formation via contact freezing but also the growth of ice particles by collision and, therefore, the formation of precipitation.

(1) Contact freezing is restricted by the size of the involved potential ice nuclei. Small particles affect the freezing of small drops, large particles the freezing of large drops.

(2) The shape of ice particles influences significantly their growth by collision: ice plates grow much faster than ice columns.

For future work it is planned to incorporate the presented approaches into cloud models with detailed microphysics including the ice phase to describe drop freezing via contact freezing and the growth of ice particles by collision.

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Warm microphysics in an orographic cloud

Martin Simmel, Karoline Diehl, Sabine Wurzler

Introduction

In warm orographic clouds at the tops of low mountains, most drops do not fall out as precipitation but evaporate releasing cloud-processed aerosol to the atmosphere. Compared to the aerosol particles the cloud droplets formed on, it shows modified size distributions and chemical compositions resulting in changed radiation and condensation properties (Feingold et al., 1998; Wurzler et al., 2000). The cloud - aerosol interactions in such a cloud were simulated with two cloud models describing the gas and particle processes before, during, and after cloud lifetime.

Methods

Two adiabatic air parcel models with a sectional description of the cloud microphysics and the interactions between aerosol particles and drops are employed. One of the cloud models treats the cloud microphysics with one-dimensional distributions implying that drops of the same size have the same composition. The other air parcel model includes a two-component treatment of the cloud microphysics where drops of the same size can have different contents of aerosol particles and, therefore, different gas scavenging behaviours. The warm microphysical processes considered in both models are: growth/shrinking and impaction of aerosol particles as well as nucleation, growth/evaporation and collision-coalescence of drops. The collision process is described by the Linear Discrete Method (Simmel et al., 2002) using an improved collision-coalescence kernel (Kerkweg et al., 2003). Furthermore, a state of the art iterative condensation scheme was developed.
The initial conditions for the cloud simulations and reference values were taken from the field campaign FEBUKO (see e.g., http://projects.tropos.de/afo2000g3). The models were initialised with data measured at the valley station: temperature, pressure, dry aerosol particle size distribution and composition. The relative humidity was derived from the cloud base height. The integration time, vertical velocities, and adiabatic cooling rates were deduced from the tracer experiments and the observed wind speeds. Sensitivity studies for 300, 600, 900, and 1800 seconds were carried out.

Results and discussion

The model results are compared to experimental data from the mountain station. From Table 1 it can be seen that the results of the two models are similar. The model results of temperature and liquid water content (LWC) are the same for all cases because they are determined only by the thermodynamical conditions and are not sensitive to the aerosol particle distributions. The model calculated LWC is about 20% higher than the measured one. Taking into account the rather simple model assumptions (missing entrainment, no deposition etc.), this is a very good result. Higher updraft velocities (i.e., shorter integration times) lead to higher supersaturations (S_{max}), more activated particles, and, therefore, to smaller drop sizes. Effective radii (r_{eff}) from the model simulations

Integration time (s)	Measured	Modeled (1D/2D) 300	600	900	1800
Temperature (°C)	7.2	6.9	6.9	6.9	6.9
LWC (g/m³)	0.26	0.31	0.31	0.31	0.31
r_{eff} (µm)	5.22	2.94/2.90	3.53/3.45	3.97/4.07	4.81/4.75
N_{drop} (1/cm³)	632	3100/3125	1896/1916	1269/1153	705/794
N_{ap} (1/cm³)	5131	5659/5134	6239/6255	6791/6953	7167/7128
N_{tot} (1/cm³)	6291	8232/8259	8135/8171	8060/8106	7872/7922
S_{max} (%)	no data	0.30/0.33	0.19/0.20	0.15/0.16	0.10/0.10

Tab. 1: *Comparison of model results to field experimental data from the FEBUKO project at mount Schmücke in October 2001. r_{eff} is the effective radius, LWC the liquid water content, N_{drop}, N_{ap}, and N_{tot} the number of drops, aerosol particles, and their sum, respectively, and S_{max} the maximum supersaturation. Model results are from simulations with different integration times of 300, 600, 900, and 1800 seconds, i.e., vertical velocities of about 1.2, 0.6, 0.4, and 0.2 ms^{-1}.*

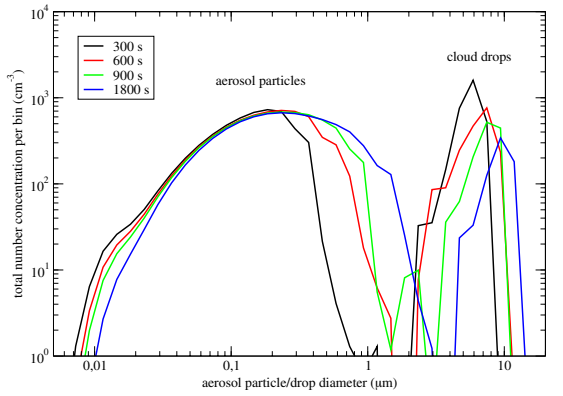

Fig. 1: Total number concentration of aerosol particles and cloud drops per size bin at Mount Schmücke for the 1D model with various (300 s, 600 s, 900 s, and 1800 s) integration times.

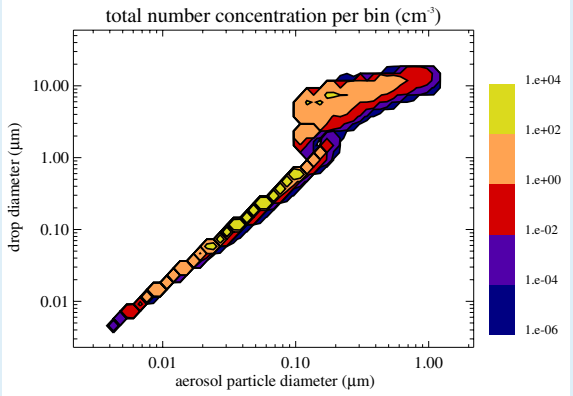

Fig. 2: Same as Fig. 1, but for 600 s integration time, obtained with the 2D model.

are much too low and drop numbers (N_{drop}) are too high for realistic integration times (300 s, 600 s, 900 s). When choosing an unrealistically long integration time (1800 s), the correspondence between model and experiment becomes quite good. Total particle numbers (N_{tot}) are too high in the models, which is apparently one of the reasons that the calculated effective radii remain smaller than those observed in the experiment. Figures 1 and 2 show the total number concentrations calculated with the one-component model for all integration times and with the two-component model for an integration time of 600 seconds, respectively.

Conclusions and outlook

The present results show that the cloud models describe the thermodynamics rather well but for the microphysical processes some work still has to be done. Changes in the dynamics (time dependent cooling rate according to the slope of the hill and the height dependence of the wind speed), and the drop activation scheme (e.g., non-ideal solutions) will be investigated next. Possible influences of deposition due to the coniferous forest at the site and of entrainment will be considered, too. Furthermore, additional comparisons with experimental data will be carried out and the cloud evaporation will be studied. Here the data from the redescending air flow over the mountain will be of importance as these actually describe the cloud processing of aerosols. In the cloud processing situation, we expect major differences between the results of the two model types and that the two-component microphysics gives more realistic results.

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Tracking of cloud interfaces by the volume-of-fluid method

Detlef Hinneburg and Oswald Knoth

Introduction

Most numerical models describing the atmospheric processes are based on Eulerian grids. Due to the numerical diffusion induced by fixed grids, the spatial distributions of simulated variables adopt rather smoothed forms. This unfavorable peculiarity disintegrates especially the cloud/environment interface because of the jumps in temperature and vapour/condensate content. Ignoring the addressed disadvantage leads to permanent dilution and loss of cloud water and cloud volume culminating in a rapid collapse of the cloud.
Consequently, atmospheric models with cloud modules should find a remedy like, for example, the application of the volume-of-fluid (VOF) method (e.g., Margolin et al., 1997). This method was successfully applied to multi-phase systems in Eulerian grids with the gain of a numerically undisturbed coexistence of the phases during the transport through the grid. Emphasis is placed on the scale-adapted and separate treatment of the different phases and their specific properties rather than an expensive reproduction of the interface in the sub-scale.

Volume-of-fluid (VOF) method

The VOF method matches in a straightforward way with usual grid models and may be considered as an additional tool managing exclusively the cloud relevant processes of those grid cells which cover the cloud/environment interface (i.e., 2-phase cells). All other cells, variables and processes are subject to the standard model procedures. The method operates with some additional prognostic variables introduced only in the concerned cells: the partial cloud volume and separate variables of water vapour, condensed water and temperature each for the cloud and environment fractions.
The primary process is the advection of cloud volumes across the cell boundaries with regard to their spatial arrangement. A geometric shaping and localization procedure for the volume fractions is therefore required. VOF as a fast and efficient method, generates a reliable structure of the cloud without any sub-scale information beyond the volume fractions. The transport of the associated variables is performed secondarily via the shifted volume fractions.
The realized VOF method applies only to cells with fractional cloud volume and proceeds in each time interval according to the following steps:

(1) Selection of the VOF cells. All 2-phase cells are marked in order to be involved in the VOF scheme. Only this limited set of cells determines the extent of the allocated fields for the extra variables.

(2) Localizing/shaping of the cloud fractions. The partial cloud volume of a cell is shaped as a parallelepiped adjacent to that cell face, where the centroid vector of neighbouring cloud material is directed with its largest component. The extent perpendicular to this cell face is defined by the local cloud volume. Fractional cloud volumes are thus controlled to some degree by the direct neighbours.

(3) Advection of the cloud volume fractions. The balanced transport fluxes on the cell faces are interpreted as cross-border volume shifts. Depending on position, size and shape, the cloud volumes lie either outside or partially/completely inside the shifted volumes. The collected cloud portions can be calculated simply due to their parallelepiped shapes. The balance of fluxes or volumes, resp., is devided into two equations: for the cloud and the complementary non-cloud volumes. Using these shifted volumes, the upwind advection of the cloud and non-cloud variables is treated separately. For the 3-dimensional model the operator split techniques is used, where steps 1 to 3 have to be repeated for each direction.

(4) Cloud processes. After the advection has been finished, the cloud processes are applied separately to the cloud and non-cloud phase in each grid cell.

(5) Update of the global variables. The phase-split variables of the VOF cells weighted by the corresponding volume fractions are averaged to update the standard model variables of temperature, vapour and condensate content, which then determine the other model processes (e.g., buoyancy).

Application in cloud modeling

The used atmospheric model is non-hydrostatic, inelastic, implicit, designed in the z-coordinate system, and qualified for applications from the microscale to the global scale. The 2-dimensional

simulations analyzed here are constrained to the condensation-evaporation process and the appropriate buoyant effects, excluding diffusion.
The test example (Grabowsky and Clark, 1991) describes the undisturbed dynamics of a rising warm and moist parcel in a stable atmosphere ($d\theta/dz$ = 3.7 K/km) driven by the buoyancy and retarded by the weight of condensed water. The parcel starts as a saturated bubble (radius 80 m) surrounded by a shell (thickness 80 m) with linear decrease to the environmental humidity (20%). In the domain of 640 m x 800 m a uniform spatial resolution of 20 m, 10 m, or 5 m was chosen. Each simulation was executed both with and without applying the VOF method.

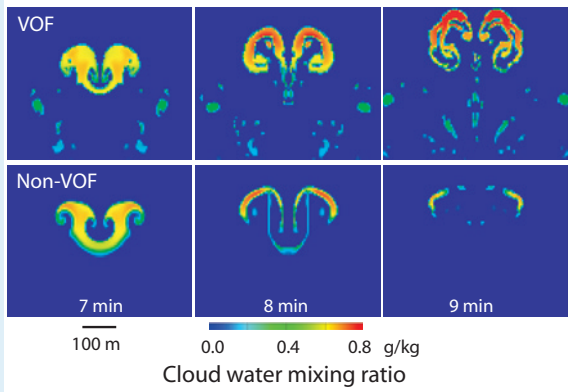

Fig. 1: *Cloud water content after 7, 8, and 9 min for spatial resolution of 5 m; with and without the VOF method.*

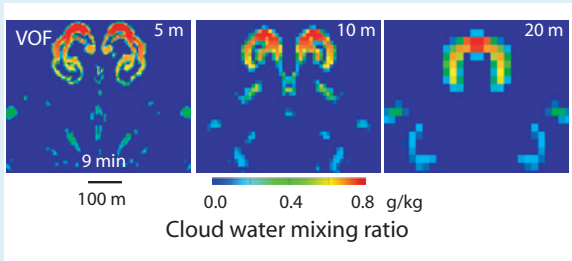

Fig. 2: *Cloud water content after 9 min for spatial resolutions of 5 m, 10 m, and 20 m; with the VOF method.*

Figures 1 and 2 demonstrate the different cloud development for the two methods under various spatial resolutions (a selected region is shown). Caused by the buoyant motions, the bubble is subject to deformations in an increasing degree.
The ***conventional method*** (Figure 1, bottom) is unable to maintain the cloud filaments, in spite of the high spatial resolution and actual condensation conditions. Moreover, the cloud vanishes the earlier the more the resolution is reduced.
In contrast, the ***VOF method*** (Figure 1, top) allows the cloud to pass through an unbroken evolution. The cloud development turns out to be almost independent of the spatial resolution (Figure 2); for very low resolution at least the regional total of the cloud water content is preserved.

Resulting improvements

The improvements in cloud modeling due to the application of the VOF method can be summarized as follows:

- Cloud water once produced is no longer subject to arbitrary averaging and dilution but is transported like a parcel with well-defined properties and boundaries.
- The VOF method accepts and sustains the distinctive properties of the cloud and its direct environment.
- Outside the region of origin, clouds and their fragments attain a substantially higher life-time.
- Reliable simulation results are produced even for very low spatial resolution.

Furthermore, no significant impact on run-time and storage of the model is observed.
In a future step, the shaping procedure for cloud fractions will be given the license of tilted positioned interfaces instead of the restriction to face-parallel orientation (see point 2 in the second section).

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Model-to-satellite approach for cloud verification using the "Lokal-Modell" (LM)

Olaf Hellmuth, Barbara Heide, Gerd Vogel (Deutscher Wetterdienst)

Motivation

The treatment and prediction of clouds (cloud cover, liquid and ice water content, cloud thickness, emissivity etc.) is of crucial importance for mesoscale and large scale atmospheric modelling, e.g., for numerical weather prediction, air pollution modelling and climate simulation. Although some progress has been made in the last decade, there are still large observational gaps, especially with respect to in-situ cloud measurements. Remote sensing data from operative weather satellites which are available over the whole globe are very useful for the verification of cloud prediction. Here, results of a verification study on LM cloud prediction using Meteosat-7 infrared (IR) and water vapour (WV) data are presented.

Method

On the basis of predicted 3D temperature, humidity, cloud cover, liquid water content (LWC), ice content and pressure fields as well as 2D surface albedo, surface temperature and surface pressure fields, infrared brightness temperatures in the Meteosat-IR/WV channel are computed using a radiation transfer code of Morcrette (1991). Sensor response functions have been updated for Meteosat-7. LM simulations are performed for three different cloud parameterizations:

(a) Warm-rain scheme (GSCP1);
(b) One-category ice scheme for mixed-phase precipitating clouds including cloud snow (GSCP2);
(c) Two-category ice scheme for mixed- and ice-phase stratiform clouds including cloud snow and cloud ice (GSCP3) (Doms and Schättler, 1999).

The synthetic infrared brightness temperatures are compared with observed Meteosat-7 IR/WV data using correlation coefficient *r* and centered pattern RMS difference *e* (Taylor, 2001).

Results

Due to the availability of LWC profiles inferred from microwave radiometer (from Deutscher Wetterdienst (DWD)) the 6 May 2001 has been chosen for LM setup. Figure 1 shows exemplary simulated brightness temperatures in the IR and WV channels on the base of LM forecast fields, Figure 2 the corresponding Meteosat-7 observations at 12 UTC. Even if the observed Europe-wide cloud pattern is more or less captured by the LM simulation, the differences between predicted and observed data are larger than those within the three LM runs. The figures show, that either predicted cloud coverage and/or cloud water content (liquid, ice) is clearly underestimated. Also, the predicted upper-troposheric humidity is to low. Nevertheless, to some degree the convective clusters with heavy thunderstorms over Austria, Slovenia and Croatia are simulated at least by GSCP3. The predicted synoptic structures are well-correlated and the spatial distribution agrees with the observed structures in both channels. Considering the non-linearity of the underlying physical processes, the cloud patterns are well reproduced by the simulations.

To get some information about the gain in cloud predictability, the model runs have been compared with a reference simulation. In present case, the warm-rain scheme (GSCP1) is used to be the reference forecast. The reference forecast has been verified using the satellite data by determination of *r* and *e*. This has been done also for GSCP2 and GSCP3 versus observations. Then, relative changes in *r* and *e* with respect to the reference run are calculated (Table 1). An enhancement of predictability is indicated by an increase in *r* and decrease in *e*.

The correlation between observed and predicted brightness temperatures is quite poor, i.e., lower than r=0.5. The centered pattern RMS difference is comparably large, i.e., ca. 14 K for IR and ca. 4 K for WV. The agreement between observed and predicted infrared brightness temperatures is better in the WV than in the IR channel. The difference between the warm-rain scheme (GSCP1) and the one-category ice scheme (GSCP2) is very low, seen by a small decrease in *r* and *e* in both the IR and WV channel. Much more increase in cloud predictability can be observed in the case of the two-category ice scheme. Here, an increase in *r* of about 20% in the IR channel, and a decrease in *e* of about 1% in the WV channel can be detected.

Figure 3 shows time series of the brightness temperature at the measurement site Lindenberg (microwave radiometer) and its neighbourhood. At the measurement site the time evolution of IR brightness temperature corresponds well with the GSCP3 run. GSCP1 and GSCP2 show an irregular evolution with strong zigzag and sharp temperature drop around noon caused by predicted convective

Reference scheme [1]	Verification scheme [2]	Band	$r_{obs,1}$ obs-[1]	$r_{obs,2}$ obs-[2]	$10^2(r_{obs,2}-r_{obs,1})/r_{obs,1}$ [%]	$e_{obs,1}$ obs-[1] [K]	$e_{obs,2}$ obs-[2] [K]	$10^2(e_{obs,2}-e_{obs,1})/e_{obs,1}$ [%]
GSCP1	GSCP2	IR	0.277	0.270	-2.3	14.525	14.520	-0.03
		WV	0.442	0.424	-4.136	4.357	4.293	-1.462
GSCP1	**GSCP3**	IR	0.277	0.332	**19.9**	14.525	13.083	**-9.928**
		WV	0.442	0.491	**11.036**	4.357	3.675	**-15.645**

Tab. 1: *Correlation coefficients (r) and centered pattern RMS differences (e) between satellite-based (obs) and modelled IR/WV brightness temperatures (6 May 2001, 12 UTC).*

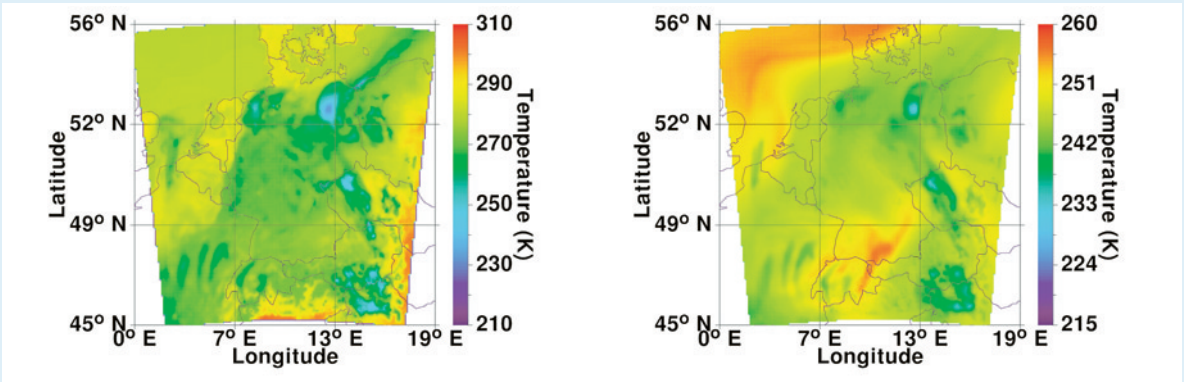

Fig. 1: *Synthetic brightness temperatures from LM run, 6 May 2001, 12 UTC: (a) IR-GSCP3; (b) WV-GSCP3.*

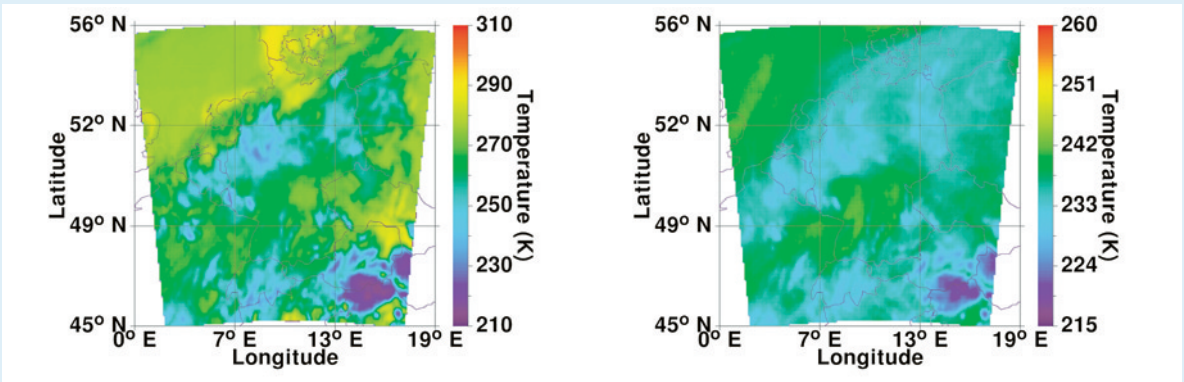

Fig. 2: *Meteosat-7 brightness temperature, 6 May 2001, 12 UTC: (a) IR channel, (b) WV channel.*

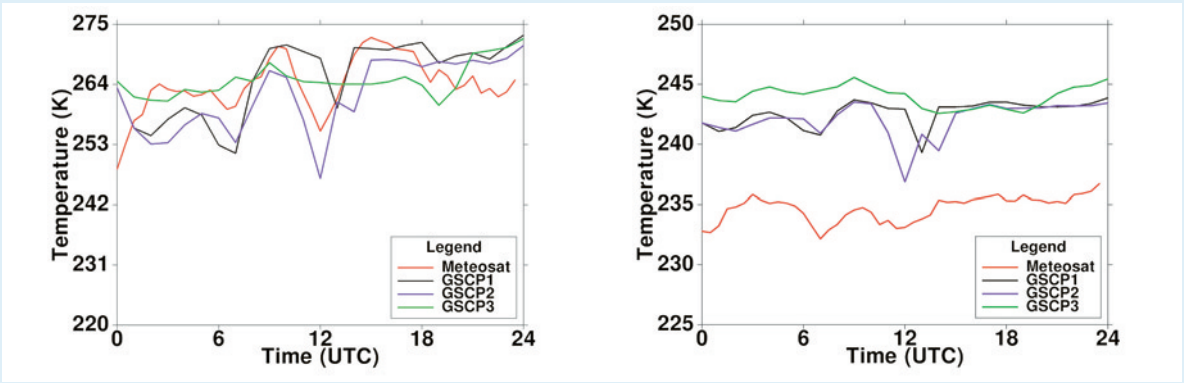

Fig. 3: *Time evolution of modelled and observed IR/WV brightness temperatures at Meteorological Observatory Lindenberg, 6 May 2001: (a) IR brightness temperature averaged over 3x3 LM grid cells around the measurement site; (b) as (a) but for WV brightness temperature.*

clouds passing the grid cell (seen from predicted LWC profiles, but not observed). The same effect can be observed in the WV spectral band. The general offset between observed and modelled WV brightness temperatures indicates a deficit in the predicted upper-troposheric humidity compared to the observation. Apart from this, the observed temperature evolution is best reproduced by GSCP3 run. The overall behaviour is slightly modified when considering the average over the 3x3 adjacent grid cells. The smoothing effect is slightly more pronounced in the WV than in the IR channel. It also depends on the parameterization scheme. This clearly reflects the disjunctive information content of both channels. As the interpretation of the different signals might be ambiguous, the two-channel signal must be reduced to a more complex indicator. Further investigation is needed on this topic.

Conclusion and outlook

The detected differences between observed and predicted cloud fields in the mesoscale clearly depicts the great challenge and imperative to improve the physical parameterization of numerical weather prediction models, especially with respect to cloud dynamics, microphysics and exchange processes. It should be noted, that mesoscale models like LM already contain sophisticated cloud parameterization compared to large scale and climate models.

Meteosat radiometer data provides very useful information for the verification of LM cloud prediction. Synthetic brightness temperature represents a complex signal and integral measure of the cloud predictability, but to some degree also of other thermodynamic forecast values. On the basis of the described verification approach, predicted cloud fields can be inspected hourly. Thus, important conclusions regarding the refinement of cloud parameterization, especially the treatment of microphysics can be drawn. With the given approach, a potent tool for the accompanying examination of cloud evolution for long term air pollution simulations (cloud scavenging, photolysis) is available at IfT.

To avoid ambiguous interpretation of two-channel signals, determination of semi-transparent cirrus cloud temperature and emissivity from modelled and observed infrared radiances is in progress. Apart from this, the sensitivity of modelled brightness temperature against input parameters as well as the benefit of LWC profiles from microwave radiometer data will be elucidated. To draw statistically significant conclusions, a long-term cloud evolution study is necessary.

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Cooperation
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Saharan dust over a Central European EARLINET-AERONET site: Combined observations with Raman lidar and Sun photometer

Detlef Müller, Ina Mattis, Ulla Wandinger, Dietrich Althausen, Albert Ansmann

Introduction

A strong Saharan dust outbreak was observed across Europe in October, 2001 (Mattis et al., 2002; Kaminski and Weller, 2002). Detailed characterizations of the dust plumes were made at the lidar stations which participate within the European Aerosol Research Lidar Network (EARLINET) (Bösenberg et al., 2001). Simultaneous observations with the institute's Raman lidar and Sun photometer, which is operated by the Aerosol Robotic Network (AERONET) (Holben et al., 1998) allowed for monitoring and rigorous quality assessment of the derived aerosol properties. Deficiencies of both instruments, e.g., column-averaged information from measurements by Sun photometer and restriction to minimum measurement height for lidar, could be compensated for with the combined observations. For the first time it became possible to derive extinction profiles with the IfT lidar under daylight conditions. The effect of non-spherical shape of the observed particles in part led to false aerosol properties derived on the basis of the Sun photometer data. This result will have impact on the retrieval algorithm used by the AERONET team within NASA.

Instruments

The Raman lidar is described in detail in Mattis (2002). The instrument emits laser pulses at 355, 532, and 1064 nm. From the detected signals follow particle backscatter coefficients at the three emitted wavelengths, particle extinction coefficients and particle backscatter-to-extinction (lidar) ratios at 355 and 532 nm, particle depolarization ratio at 532 nm, and profiles of water-vapor and temperature. The pure rotational Raman lines, which are used for the retrieval of temperature profiles for the first time were used for the retrieval of extinction profiles at 532 nm under daylight conditions. The automatically-operated Sun photometer is used for observations of Sun and aureole radiances in 8 channels between 340 and 1020 nm. Measurements of sky radiance are made at 440, 670, 870, and 1020 nm. From the signals follow columnar spectral optical depths, particle phase functions, and column-averaged microphysical particle parameters (Dubovik and King, 2000).

Observations

Figure 1 shows the particle backscatter coefficient at 532 nm for several measurement periods between 1706 UTC on October 12 and 1936 UTC on October 15, 2001. According to backward trajectory analysis the particles originated from the Sahara region north of 10° N and west of 25° E. The particles showed high depolarization ratios up to 25% at 532 nm. The dust plume reached top heights of 5 km. In the beginning of this event there was a clear separation between aerosols in the boundary layer and in the dust plume above. The dust plume penetrated into the planetary boundary layer on October 14, 2001.

Figure 2 shows the time series of optical depth at 532 nm on the basis of the Sun-photometer and the lidar observations from 0716 UTC on October 13, 2001, until 1807 UTC on October 14, 2001. In the case of the lidar observations we present total columnar optical depth, the contribution by the dust layer above 1000-m height, and the layer below 1000-m height. Total optical depth increased from 0.25 to 0.6 during the dust event. According to the lidar observations the dust layer above 1000-m height contributed 70%-90% to total optical depth. There is excellent agreement of optical depth derived from the extinction profiles to optical depth derived from Sun photometer for the timeframe from 1151-1327 UTC on October 14, 2001. At that time absolute cloud-free conditions prevailed.

Figure 2 also shows the comparison in terms of column-averaged Ångström exponents. For that purpose extinction information from lidar observations at 355 and 532 nm, and from Sun photometer observation at 380 and 670 nm, respectively, was used. We note a constant decrease of the Ångström exponents during the event with lowest values around 0 on October 14, 2001. The mixing of Sahara dust into the planetary boundary layer can be seen from almost equal values for the Ångström exponent above and below 1000-m height at noontime on October 14, 2001.

Figure 3 shows the comparison of the lidar ratio at 532 nm derived from lidar to the column-averaged value from Sun photometer for the observational period from 1151-1327 UTC on October 14, 2001. The lidar ratio is defined as the ratio of the volume extinction (absorption plus scattering) coefficient to the scattering coefficient at 180°. A column averaged lidar ratio of 62±10 sr follows from the lidar data. The column-averaged lidar ratio from Sun photometer is approximately 29±15 sr,

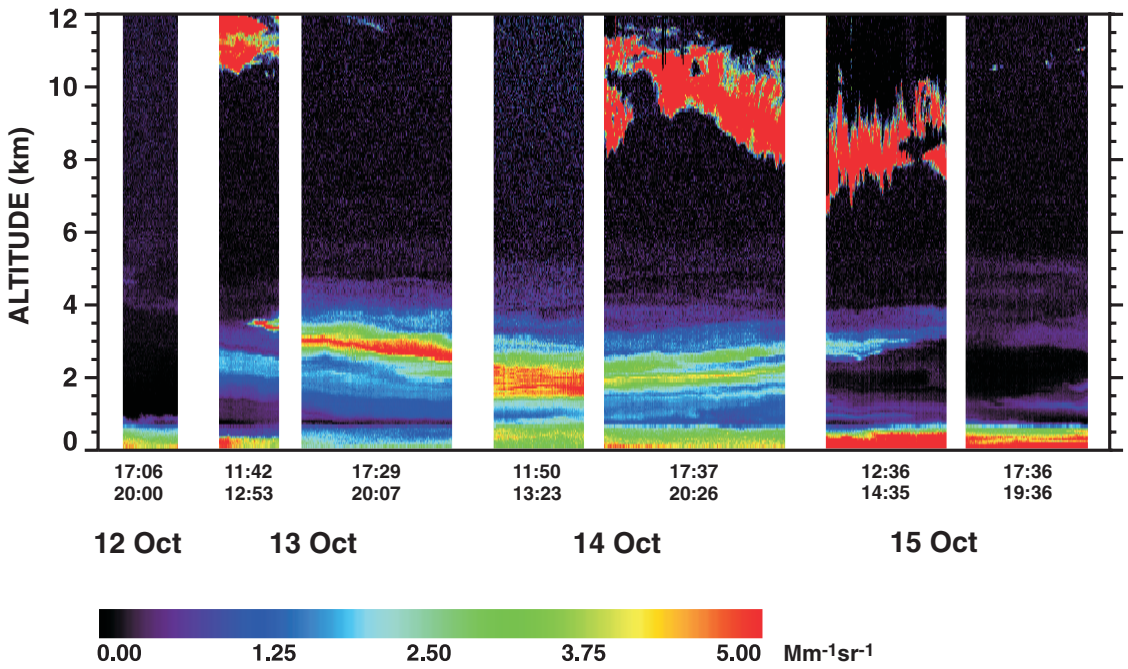

Fig. 1: *Particle backscatter coefficient at 532 nm.*

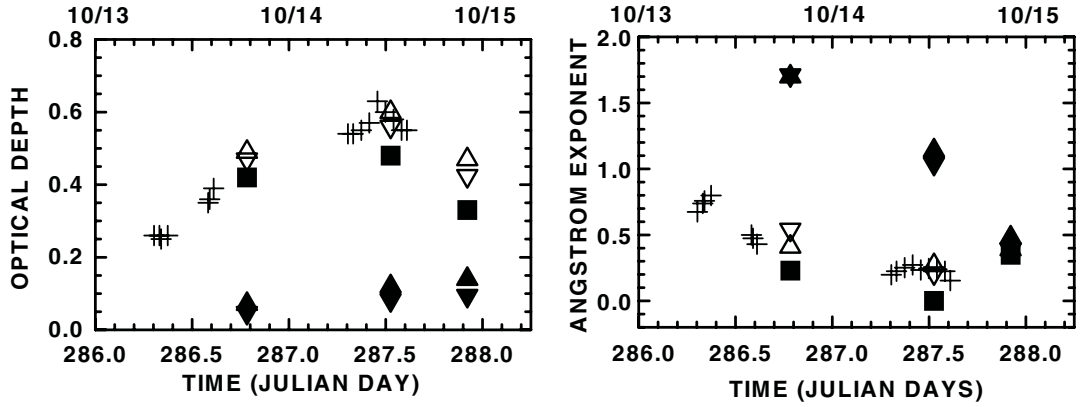

Fig. 2: *(Left) Particle optical depth from Sun photometer (cross), and from lidar (open triangle). Also shown is the contribution above 1000-m height (filled square), and below 1000-m height (filled triangle). Upward and downward pointing symbols follow from error analysis and denote maximum and minimum reasonable value. (Right) Ångström exponent. Meaning of symbols same as on left side.*

if spherical shape of the particles is assumed. The use of spheroidal particle shapes, i.e., oblate and prolate geometries, results in a lidar ratio of 35±18 sr. This substantial deviation of a factor of two compared with the mean value from lidar most-likely is caused by an erroneous particle model, which is used for the calculation of the particle lidar ratio and the microphysical properties of dust.

As outlined by Mattis et al. (2002), and Müller et al. (2002) the observed lidar ratios were unexpectedly large. Two effects may be responsible. During the transport from the source regions to the lidar site a large amount of coarse-mode particles above 1 μm in diameter may have been removed by sedimentation. Theroretical studies have shown that lidar ratios increase if particle size decreases. The most important reason however is believed to be the nonspherical shape of the scatterers, which causes the reduction of the particle backscatter coefficient. Respective effects have been described in theoretical work by, e.g., Mishchenko et al. (1997), and Kalashnikova and Sokolik (2002). An analysis of the present example showed that there is an increase of the particle lidar ratio by a factor of 1.8-4.4 due to the nonspherical particle shape.

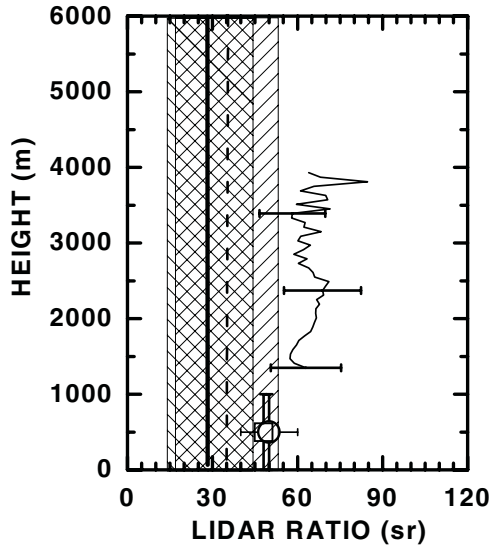

Fig. 3: *The profile of particle lidar ratio above 1000-m height. Below 1000-m height mean values are given (circle, square). Column-mean values from Sun photometer are given on the basis of spherical particle shape (solid line) and spheroidal particle shape (dashed line). Uncertainties in the retrieval are denoted with areas shaded to the left and to the right, respectively. Error bars denote standard deviation.*

Outlook

Combined observations of multiwavelength lidar and Sun photometer seem to be a promising tool for vertically resolved characterization of mineral dust outbreaks. The particle models used for the retrieval of microphysical particle parameters are highly sensitive to optical effects caused by particle shape, which can be formulated in terms of the depolarization ratio. The substantial deviation of the particle lidar ratio derived from the Sun photometer observations shows that more work is needed to develop appropriate inversion schemes for the retrieval of microphysical particle parameters.

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Size-segregated characterization of PM_{10}, $PM_{2.5}$, PM_1 at the IfT research station Melpitz downwind of Leipzig

Gerald Spindler, Erika Brüggemann, Konrad Müller, Hartmut Herrmann

Up to the end of the 80-ies the region around Leipzig was one of the most polluted areas in Central Europe. The collection and characterization of environmental particulate matter has been in the interest of atmospheric sciences, human health and environmental policy for several years now. Nevertheless, the present knowledge of primary formation, source identification and the effects of particulate matter is unsufficiently (e.g. Georgii and Warneck, 1999, Pope and Dockery, 1999, Grassian, 2002).

At the IfT-research station Melpitz (downwind of the conurbation Leipzig-Halle-Merseburg, Figure 1) the concentration and chemical compostion of particles was investigated from the year 1992 on together with dry deposition of gaseous species and wet deposition of pollutants (Spindler et al. 2001). $PM_{2.5}$ and PM_1 sampling started in 1995 and 1999, respectively (Spindler et al., 1999). Long term series of measurements and investigations are the observational basis for the distinction of real atmospheric changes and short weather-related events in the atmosphere (Heintzenberg et al., 1998). The physical properties of particles - diameter and humidity growth - are of relevance for their lifetime, cloud processing, and transportation effects (Swietlicki et al., 2000).

The low flow sampler (LF) is the Partisol 2000 Air Sampler (Rupprecht and Patashnik Co. Inc., USA, Figure 1). For the weekly filter samples PM_{10}, $PM_{2.5}$ and PM_1 Teflon filters with 47 mm diameter (Millipore, Eschborn, Germany, Type 4700, 3 µm pore size) were used. The weekly sampling volume is 84 m^3. The cut-offs are realized with virtual impactors. The particle mass concentration was determined by weighing under constant conditions (50% relative humidity, 20°C). After a conditioning time of at least 24 hours the particle mass was determined gravimetrically (Mettler AT 261 Delta Range balance, Mettler Toledo GmbH, Germany). Water soluble ions were determined from half of each filter. Standard ion chromatography with columns from Dionex, USA and Metrohm, Switzerland was used for that purpose.

The particle mass concentration and the mass of water soluble ions are available as weekly means for three particle size fractions (PM_1: fine particles,

Fig. 1: *Overview to west at the Melpitz site. In the forground (right hand side) the Partisol 2000 air sampler with different impactors.*

$PM_{2.5}$-PM_1: medium size particles and PM_{10}-$PM_{2.5}$: coarse particles) from the LF sampler. Figure 2 shows the mass distribution in the particle fractions for the three years 1999 to 2001. The yearly course of the mass concentration distribution between the size fractions demonstrates the absolute maximum for the largest particles during summer time. During summers more coarse mode particles exist, because surfaces, especially those covered with short vegetation, dry faster (Klemm et al., 2002), the absolute precipitation time is lower, and precipitation events more intense than in other seasons. Coarse particles are caused from re-emission by wind and agricultural activities in the surroundings, transported to the site over short distances. The size fraction ($PM_{2.5}$-PM_1) shows the smallest particle mass concentration over the whole year, that means most of $PM_{2.5}$ is PM_1. Figure 3 shows the mean contribution of the water soluble ions to the mass of the three particle fractions for the investigated years. Sulphate and ammonium have the highest contribution to the fine particles (22 and 13%), as a result of a long range transport from natural and anthropogenic sources and particle modification by gas to particle conversion. Nitrate has the highest mass fraction in the fine (16%) and medium particles (21%). This gives a hint that combustion processes are an important source. The lowest content of water soluble ions (SO_4^{2-}, NH_4^+ and NO_3^-) in the coarse particles (PM_{10}-$PM_{2.5}$, 17%) in comparison with the medium fraction ($PM_{2.5}$-PM_1, 45,0%) and the fine particles (PM_1, 51%) indicates re-emitted crustal material. Most of the total particle mass was found in the fine particle fraction PM_1 (56%). The smallest particle mass concentration was found for the medium ($PM_{2.5}$-PM_1) particle size fraction (14%) and the fraction for the coarse particles (PM_{10}-$PM_{2.5}$) has a mass contribution of 30% This result is in line with the course of particle size mass ratios in Figure 2.

All discussed results of this study are from filter measurements. Therefore in summer losses of ammonium nitrate and ammonium sulphate by evaporation caused at high temperatures and low relative humidity can reduce the mass of fine and medium particles, in particular.

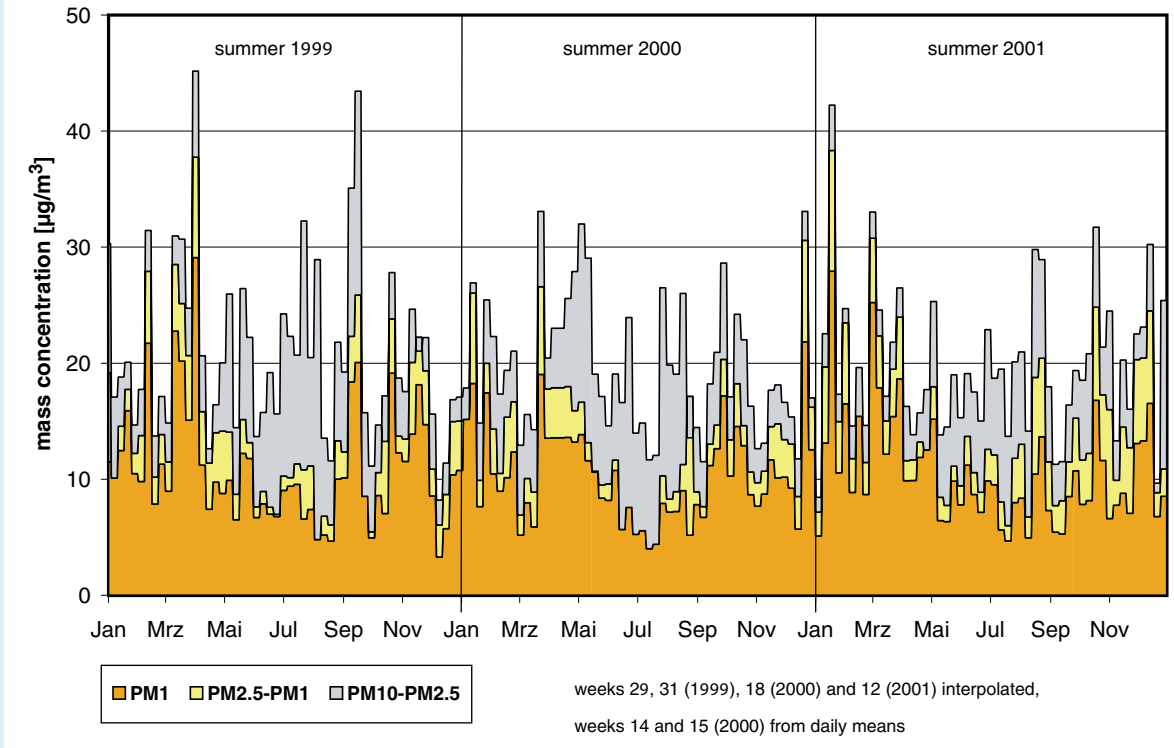

Fig. 2: *Mass distribution in three particle fractions for the years 1999, 2000 and 2001 (weekly means, from the LF sampler).*

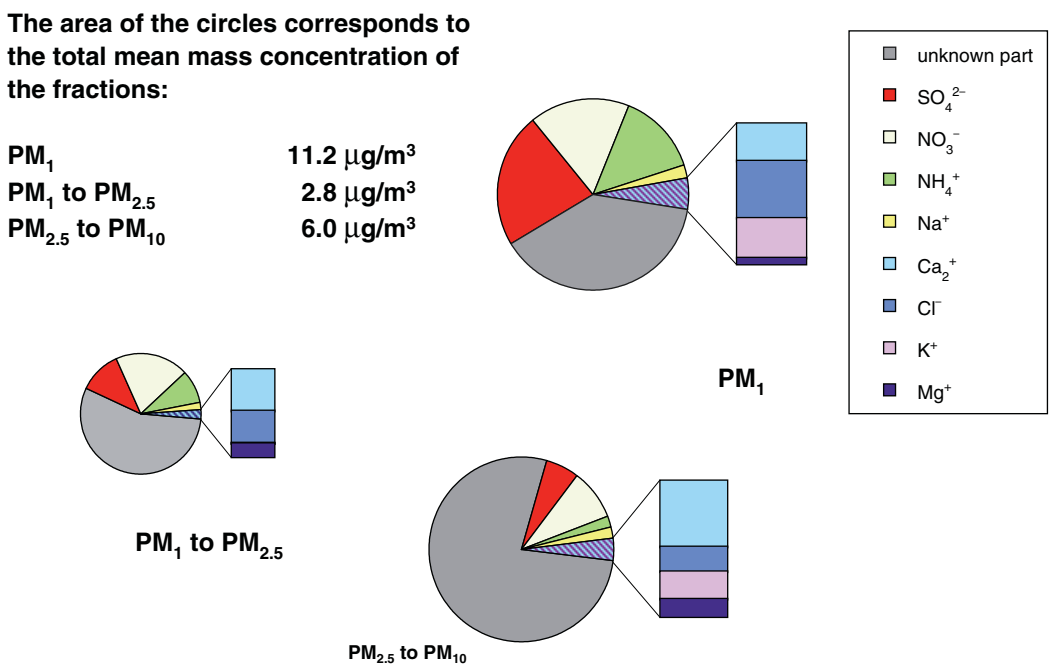

Fig. 3: *Mean mass contribution to the size fractions PM_1 (fine particles), PM_1 to $PM_{2.5}$ (medium particles) and $PM_{2.5}$ to PM_{10} (coarse particles) with major water soluble ions.*

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BEWA 2000 – Carbonyl compounds and particles in a coniferous forest atmosphere
(Subproject of AFO 2000)

Erika Brüggemann, Thomas Gnauk, Hartmut Herrmann, Diana Hofmann, Konrad Müller, Antje Plewka, Cornelia Sedello, Gerald Spindler

Emissions of biogenic volatile organic compounds (BVOC) are estimated to exceed anthropogenic VOC emissions approximately by one order of magnitude on the global scale. BVOC include both hydrocarbons like ethene, isoprene or monoterpenes and oxygenated compounds like alcohols, carbonyl compounds or carboxylic acids. In contrast to deciduous trees emitting mainly isoprene coniferous trees release predominantly C_{10}-monoterpenes, among them alicyclic (myrcene, ocymene), monocyclic (limonene) or bicyclic (α-pinene, β-pinene) monoterpenes being found often at the same time. Monoterpenes are extremely reactive against ozone or radicals like OH and NO_3. Their oxidation results in polar compounds showing substantially reduced volatility compared to their precursors. Such low volatility compounds are suspected to condense on existing particles modifying their surface or even to form new particles by homogeneous nucleation (Witter et al., 2001).

The BEWA project is a joint research project to investigate the influence of biogenic emissions in Central Europe on tropospheric chemistry on three levels:

1 - laboratory experiments,
2 - field experiments and
3 - modelling studies.

The IfT is contributing to BEWA 2000 in two projects of level 1 and 2:

1 – Particle Modification and Formation from BVOC Emissions of Coniferous Forests in Germany,
2 – Determination of Carbonyl Compounds in and above the Canopy of a Norway Spruce Forest.

To understand the complex atmospheric multiphase chemistry of particles formed from or modified by biogenic emissions it is necessary to perform both field measurements as well as laboratory expements under controlled conditions. To this end, in the Teflon reaction chamber of the IfT chemistry department the formation of SOA (Secondary Organic Aerosol) from the reaction of α-pinene and ozone is investigated.

The aim of the field experiments is the investigation of airborne particles in (12 m height) and above (24 m height) the canopy of a Norway spruce forest by size-segregated particle sampling with BERNER impactors, and high volume filter sampling, and analysing the chemical composition of the particle fractions. For this purpose main particle components (inorganic ions, carbon as sum parameters of organic carbon (OC) and elemental carbon (EC)) have been determined. Additionally

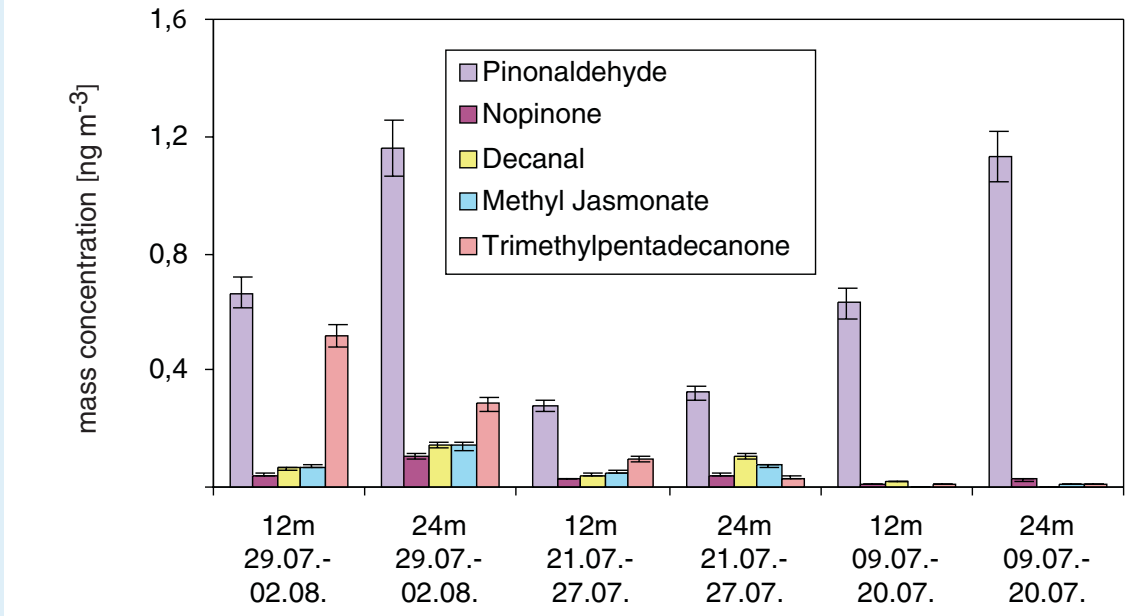

Fig. 1: *Particulate matter (PM) concentrations of typical BVOC measured in 2001 campaign on $PM_{2.5}$ filter samples from two heights.*

high-volume samples with high particle mass were used for identification and quantification of organic compounds in airborne particles (Figure 1). Differences of the chemical composition in and above the canopy should hint at primary emission or photochemical formation of BVOC as well as the formation of secondary organic particles. The results will contribute to the further development of multiphase models including particle modification and formation processes.

The total organic carbon (TOC) content of PM_{10} during the BEWA 2001 campaign varied between 0.6 and 2.6 µg m^{-3}. Between both heights in the group analyses no significant differences could be observed but differences were found in the analyses of filters when searching for organic species of biogenic origin. The concentration of pinonaldehyde (main product of the reaction of α-pinene with ozone or OH) and nopinone (main product of the reaction of β-pinene with ozone or OH) were found to be higher above the canopy indicating secondary photochemical formation followed by phase transfer into the particles.

After the successful development of the new deployed sampling unit for carbonyl compounds with the 2.4-Dinitro phenyl hydrazine (DNPH) method, two of these samplers have been deployed during the summer campaign at the Waldstein. At a height of 12 m (within the tree tops) and at 24 m (above the tree tops) 90-minutes-samples were taken simultaneously over the whole period of the experiment. These measurements follow the direct emission investigations from branches.

In 2002 the tube elution method was improved for small air volume analysis from relaxed eddy accumulation (REA) samples (Oncley et al., 1993). For the REA analysis three hourly samples were taken according to the direction of vertical winds (upwinds, downwinds and dead band). The elution of non-derivatized DNPH by 3 ml H_2O was followed by elution of carbonyl-DNPH by 3 ml acetonitrile and the evaporation of excess acetonitrile at 45 °C by a purified N_2 stream to 500 µl sample volume.

During the 2002 field campaign the LABVIEW-controlled REA system (Figure 2) was tested successfully. Hourly measurements were carried out. Both flow directions were observed for the main components. The further development of the controlling software is in progress. A site-dependent parametrization of the REA system will be added by measuring fluxes of CO_2 and water vapor using the eddy covariance method.

From the concentration gradient between both heights primary emissions were observed for

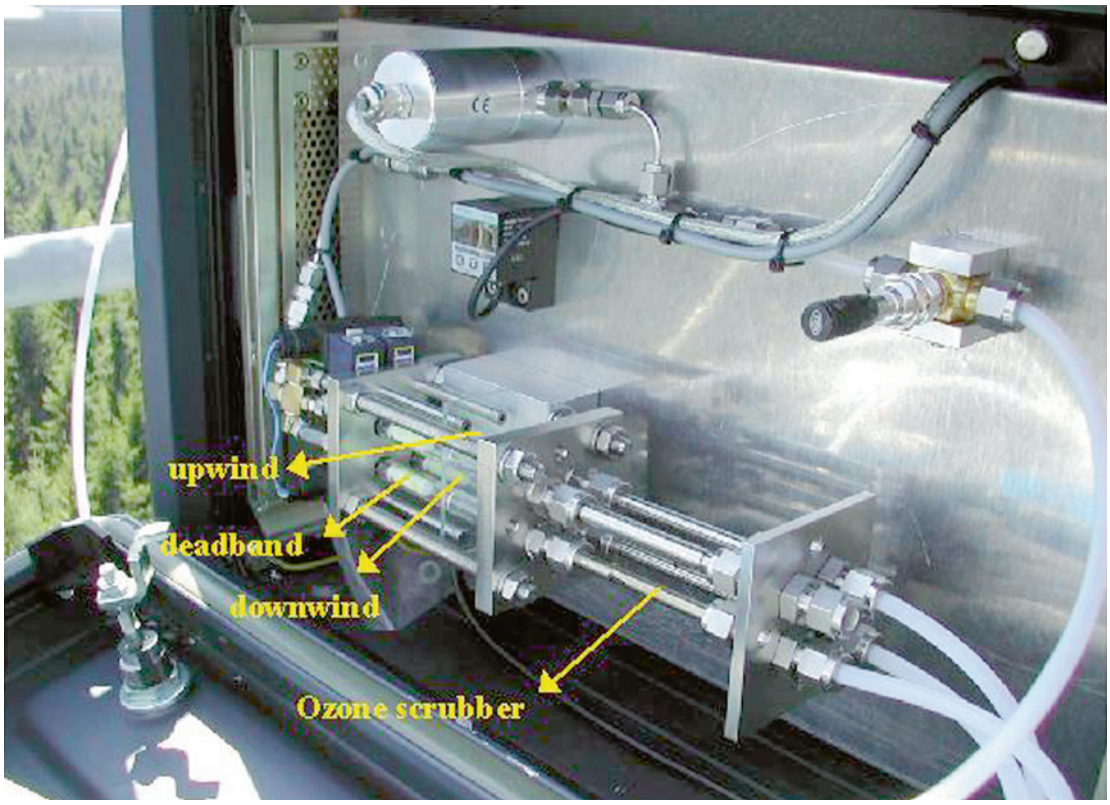

Fig. 2: *Sampling unit of the new developed relaxed eddy accumulation system (mounted on the back side of the computer) for carbonyl compounds.*

acetaldehyde (Figure 3), propionaldehyde and methyl ethyl ketone during the morning hours. Highest gas phase concentrations were observed for acetone, mainly above the canopy. Besides the long range transport from anthropogenically influenced sites the secondary formation from biogenic hydrocarbon emissions is a possible source. From anthropogenically influenced sites a mix of formaldehyde, acetone and other carbonyl compounds will typically be emitted but the formaldehyde concentrations detected are low, so a primary emission or a secondary formation of acetone from biogenic sources in the near surrounding is expected to explain the measured acetone concentration.

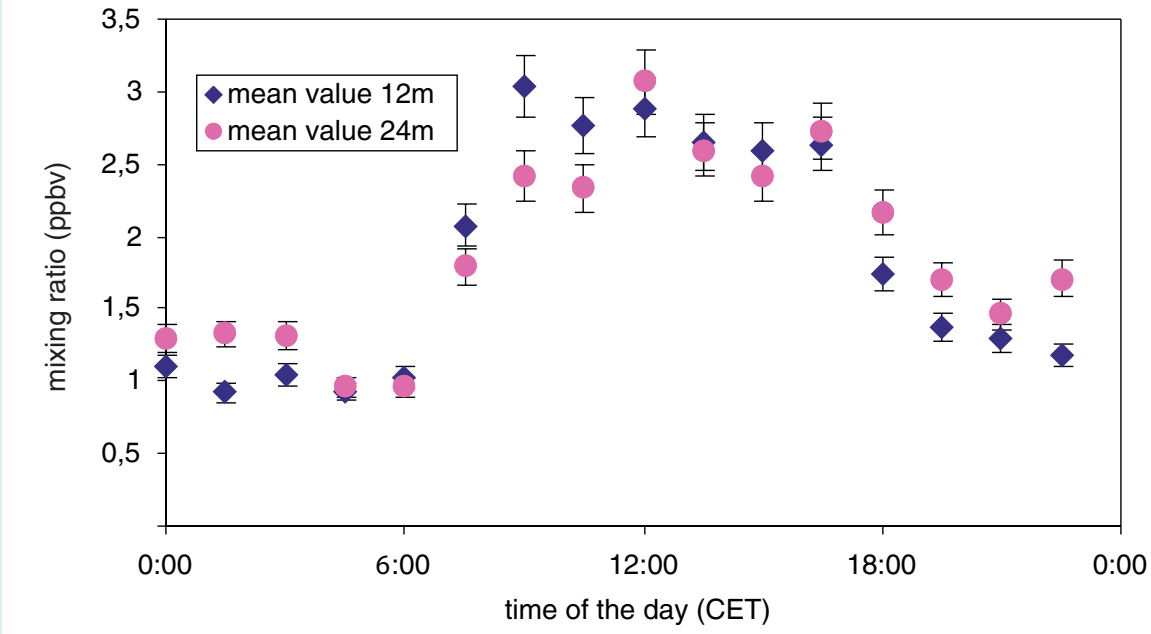

Fig. 3: *Mean daily variation of acetaldehyde at both heights during the "golden days period" (2001-07-22 to 2001-08-03).*

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- University of Freiburg
- University of Innsbruck

Atmospheric oxidation of α-pinene: Characterization of chemical and physical properties of particles in chamber experiments

Yoshiteru Iinuma, Olaf Böge, Thomas Gnauk, Konrad Müller, and Hartmut Herrmann

Introduction

Various organic species are emitted to the atmosphere from vegetation. Biogenic hydrocarbons, especially monoterpenes, play an important role in the formation of secondary organic aerosols (SOA) initiated by their atmospheric oxidation. It has been estimated that they contribute a significant mass to the total aerosol mass in the troposphere, ranging from 30 to 270 Tgy^{-1} (Andreae and Crutzen, 1997).
The identification and quantification of reaction products from the oxidation process of terpenes in the gas phase has been receiving great attention over the past two decades. More recently, the particle formation during the oxidation process of terpenes in the atmosphere has also been investigated (Pandis et al., 1991; Zhang et al., 1992; Hoffmann et al., 1997). Although this process is believed to be a major source of SOA in the troposphere only a limited number of experiments has been carried out to understand the quantitative and chemical nature of the produced aerosol (Griffin et al., 1999; Winterhalter et al., 2001). Therefore understanding the nature of these species and their formation and conversion pathways in both gas and particle phases is an essential step towards in assessing the role of these species in aerosol chemistry and physics.

Experimental

Reactions were run in a 9 m^3 Teflon reaction chamber. Figure 1 shows the schematic diagram of the experimental setup. The initial concentration of α-pinene was 100 ppbV. Ozone was produced by the photolysis of O_2 by means of a Hg lamp and the initial concentration of ozone was set to 70±5 ppbV. The concentration of ozone was determined by UV absorption at λ=254 nm. Relative humidity and temperature were kept at 45±5% and 20±3 ˚C, respectively. Sulphuric acid or ammonium sulphate seed particles were produced by a nebuliser and then dried with a diffusion drier prior to the entering the chamber. A bipolar charge neutraliser was used to neutralise the dried seed particle. The size distributions of particles in the reaction chamber as a function of time were measured by a differential mobility particle sizer (DMPS) and ultrafine differential mobility particle sizer (UDMPS). The wall and coagulation loss was characterised using the DMPS by introducing seed particles and ozone only to the chamber facility. The seed particles and ozone are injected to the reaction chamber and mixed with a fan prior to the reaction with α-pinene. After defining the initial concentrations of ozone and seed particles, α-pinene was injected and mixed for five minutes with a fan. The samples were collected on a PTFE

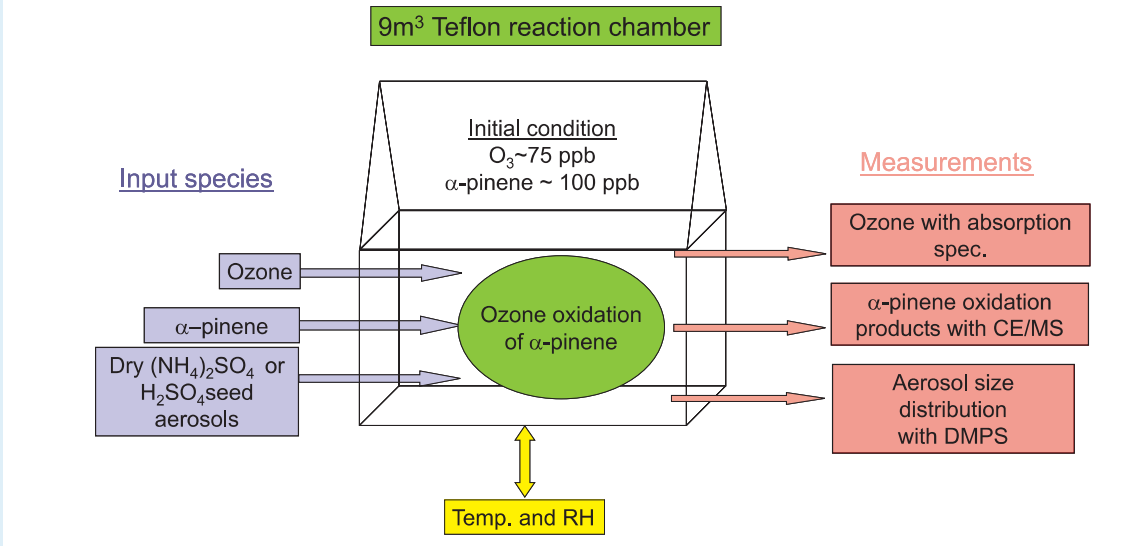

Fig. 1: *Schematic diagram of chamber setup.*

filter (47 mm diameter, 1 µm pore size) and a quartz fiber filter (47mm diameter). Teflon filters were extracted in adequate amount of Milli-Q water for CE-MS analysis. A buffer system containing 20 mM ammonium acetate/10% methanol at pH 9.1 (adjusted by NH_4OH) was used. A fused silica capillary with internal diameter of 75 µm and total length of 63 cm was used for CE separation. Total organic mass was determined using a thermographic method. The thermographic method oxidises organic matter on filters at 650 ˚C to produce carbon dioxide. The produced carbon dioxide is measured by an IR detector for quantification.

Results and discussion

A major objective of the aerosol chamber experiments is to determine the secondary aerosol formation potential of organic compounds and the identification and quantification of produced secondary aerosol species. Figures 2 and 3 show examples of increased particle mass (ΔM) and size distribution change as a function of time. Increase in particle number, mass and mode diameter were observed shortly after α-pinene was injected. In the presence of seed particles, newly formed ultrafine particles or condensation of organic vapour on the existing particles form organic layer, hence increases in particle size and mass. Partitioning of produced organic compounds or water vapour into the organic layer causes further growth in size and mass over the course of an experiment. The resulting particles contain a mixture of organic compounds, inorganic compounds and water. ΔM reached at the maximum about 2.5 hours after α-pinene injection. A significant growth in a size distribution was observed for the experiments with seed particle, ozone and α-pinene. ΔM is calculated from the DMPS size distributions assuming that the inorganic core of particle is unchanged, hence ΔM is the final particle mass minus the initial particle mass (Cocker III et al., 2001). The comparison of calculated ΔM and TOC from thermography (TOC_{th}) shows reasonable agreement (Figure 2). Figure 4 shows a typical electropherogram. Only filter samples from experiments with ammonium sulphate seed particles are shown at this time. CE-MS analysis showed that major water soluble ionic products in the particle phase were pinic acid, 10-hydroxy-pinonic acid, norpinic acid, pinonic acid relatively small concentration of norpinonic acid and possibly pinalic acid. CE-MS provides very fast and sensitive analysis for the α-pinene oxidation products.

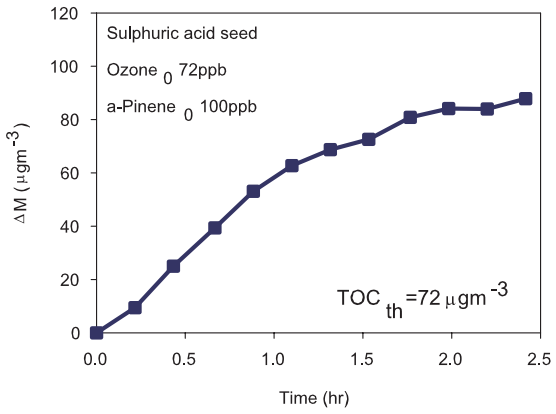

Fig. 2: ΔM as a function of time.

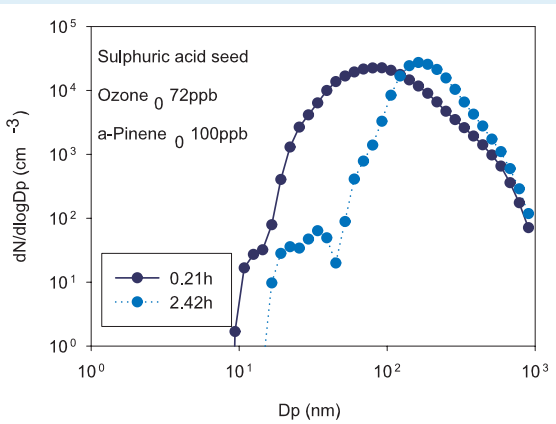

Fig. 3: dN/dlogDp as a function of Dp.

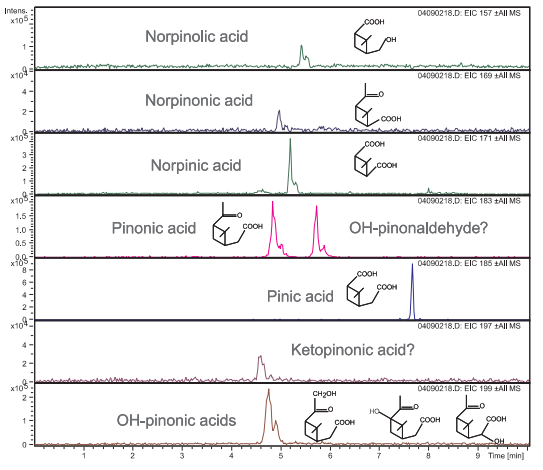

Fig. 4: A typical electropherogram of α-pinene oxidation products.

Summary

In this study, some first results from chamber experiments for the characterisation of α-pinene ozone oxidation products in the particle phase are presented. Evolution of particles (ΔM and dN/dlogDp) as a function of time are shown for sulphuric acid seeded experiments. Further

experiments are planed in order to characterise aerosol formation and evolution from α-pinene oxidation.
Oxidation products were analysed by CE-MS and it is found that pinic acid, 10-hydroxy-pinonic acid, norpinic acid and pinonic acid are major ionic species in the particle phase. CE-MS provides a fast and sensitive analysis for the identification of ionic species from α-pinene oxidation products.

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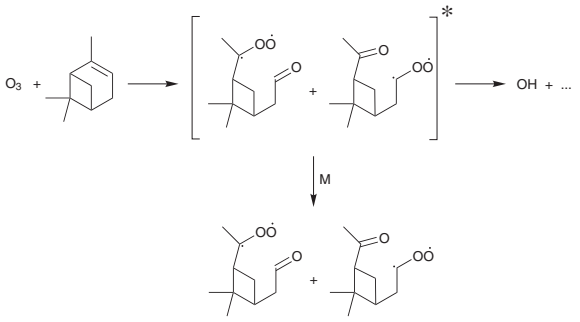
Particle formation in the system O_3/α-pinene/(SO_2)

Torsten Berndt, Olaf Böge, Frank Stratmann

Terpenes are released into the atmosphere from vegetation with an estimated global emission rate in the order of 10^{14} g yr^{-1} (Günther et al., 1992). α-Pinene (35%), β-pinene (23%) and limonene (23%) are the most important compounds in this process accounting for ca. 80% of the emitted terpenes. Their atmospheric degradation process in the gas phase is initiated by the attack of OH and NO_3 radicals or O_3. In the last years interest focused on the contribution of terpene oxidation products on the formation and growth of particles, especially in the case of the ozonolysis (Griffin et al., 1999). Subject of the studies reported here was the ozonolysis of α-pinene which was investigated under flow conditions at 295 ± 0.5 K and 1000 mbar synthetic air in the IfT-LFT (IfT - laminar flow tube). For the initial stage of the reaction, it is widely accepted that excited biradicals are formed, the so-called "Criegee Intermediates" labeled by the asterisk (Atkinson, 1997). They can decompose producing OH radicals or they can be collisionally stabilized by the carrier gas molecules M. The stabilized "Criegee Intermediates" represent the precursors for pinonaldehyde and other products.



First, experiments were performed without SO_2 additions. The formation of newly formed particles was investigated in dependence on the amount of reacted α-pinene, the humidity, the initial concentrations, the residence time, and in the presence and absence of *c*-hexane as an OH radical scavenger. Figure 1 shows the detected particle number in dependence on the amount of reacted α-pinene for three different residence times; initial [α-pinene] = 4.4 x 10^{12} molecule cm^{-3}, no further additions, r.h. < 0.3%.

Generally, particle formation was only detectable for an averaged α-pinene conversion rate above 7 x 10^8 molecule cm^{-3} s^{-1}. This observation is nearly in line with findings from a batch reactor where particle formation was visible for averaged

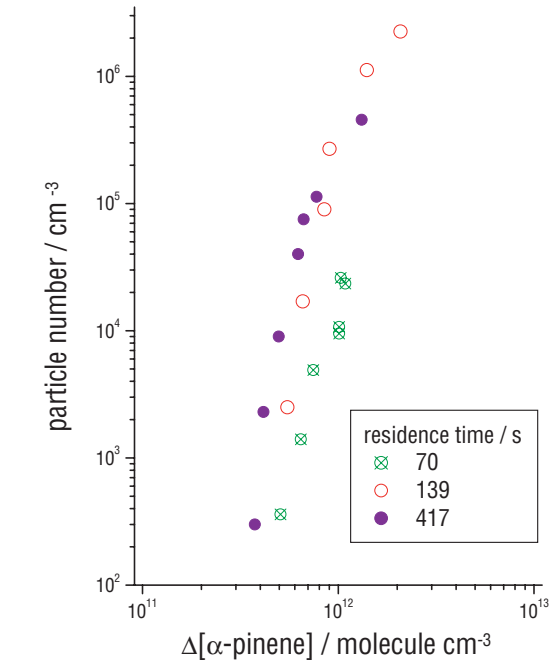
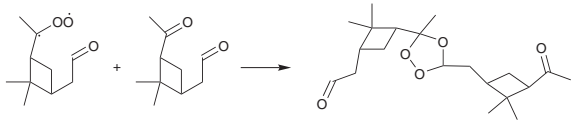


Fig. 1: *Measured particle number in dependence on the amount of reacted α-pinene for three different residence times; initial [α-pinene] = 4.4 x 10^{12} molecule cm^{-3}, no additions, r.h. < 0.3%.*

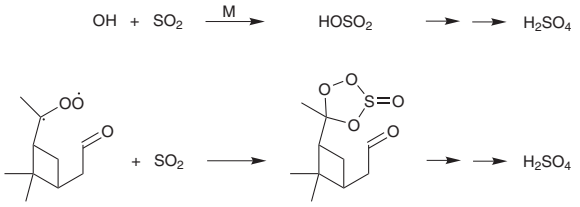
α-pinene conversion rates above (2 - 3) x 10^8 molecule cm^{-3} s^{-1} (Koch et al., 2000). From the mechanistic point of view, a possible precursor of the particles detected can be formed via the reaction of stabilized "Criegee Intermediates" with pinonaldehyde producing a C_{20} secondary ozonide. Pinonaldehyde represents a stable product of the stabilized "Criegee Intermediates" itself.



With decreasing amount of reacted α-pinene the concentrations of both reactants decreased reducing the formation of the C_{20} secondary ozonide and, therefore, suppressing the particle formation. Finally, the observations suggest that an occurrence of newly formed particles from the "pure" O_3/α-pinene reaction is unlikely under realistic atmospheric conditions.

Secondly, the particle formation was studied in the presence of SO_2 for nearly atmospheric concentrations of the reactants. Under these

conditions, OH radicals formed from the α-pinene ozonolysis reacted with SO_2 producing sulfuric acid. The reaction of the stabilized "Criegee Intermediates" with SO_2 represents a further pathway for sulfuric acid.



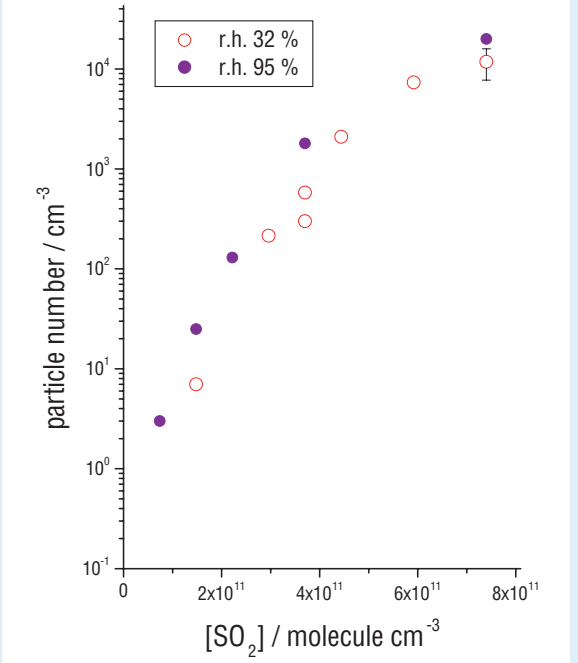


Fig. 2: Measured particle number for two different relative humidities in dependence on SO_2; [α-pinene] = 5 x 10^{10} molecule cm^{-3}, [O_3] = 7.6 x 10^{11} molecule cm^{-3}, residence time 417s.

Figure 2 shows the observed particle number for two different relative humidities in dependence on SO_2; [α-pinene] = 5 x 10^{10} molecule cm^{-3}, [O_3] = 7.6 x 10^{11} molecule cm^{-3}, residence time 417 s.

The calculated conversion rate for α-pinene was 3.5 x 10^6 molecule cm^{-3} s^{-1}, less than 1/100 of the value from the "pure" O_3/α-pinene reaction where particle formation was observable. In further experiments the amount of reacted α-pinene was varied for a constant SO_2 concentration at r.h. = 32%. For the description of the measured particle number from all experiments the following power equation was chosen:

$$N \sim (\Delta[SO_2])^a \, (\Delta[\alpha\text{-pinene}])^b$$

From a regression analysis the parameters were obtained to be a = 4.8 ± 0.3 (r.h. 32%), 3.9 ± 0.2 (r.h. 95%) and b = 1.35 ± 0.05 (r.h. 32%). That demonstrates the strong dependence of the number of newly formed particles on the amount of reacted SO_2 and thus on the produced sulfuric acid in the system. It can be speculated that the reaction products from the α-pinene ozonolysis condensed mainly on sulfuric acid / water particles and enhanced so the growth process. In conclusion, the presence of small amounts of SO_2 down to nearly atmospheric concentrations triggers the formation of newly formed particles from the α-pinene ozonolysis. Therefore, traces of SO_2 which are probably present in experiments concerning the "pure" O_3/α-pinene reaction can lead to an overestimation of the number of newly formed particles. As a result of a chamber study, the influence of SO_2 on the particle number was observed even for a SO_2 mixing ratio of 0.5 ppbV (Hoppel et al., 2001).

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The airborne spectral albedometer – A new tool for improved solar radiation energy budget studies

Manfred Wendisch, Evelyn Jäkel, Sebastian Schmidt, Dörthe Müller, Stephan Günnel

Introduction

Solar radiation, incident at the top of atmosphere is the driving force of weather and climate of the earth. Hence, the modification of solar radiation on its way through the atmosphere is of major importance for the global energy budget. Solar radiation is described by a physical quantity called irradiance, which is defined as the radiative energy flux through a horizontal surface. In order to characterize the atmospheric radiation field, accurate spectral irradiance measurements as well as validated radiative transfer calculations are required.

In the past the accuracy of the airborne irradiance measurements was significantly limited by calibration problems, distortions due to horizontal sensor misalignments, uncertainties in separating diffuse and direct irradiances, non-ideal cosine response of the sensors, or re-radiative thermal effects in the instruments. Also, the spectral resolution of the measurements was poor. As a consequence a new generation of airborne irradiance instruments with improved accuracy and higher spectral resolution was developed at IfT (Wendisch et al., 2001) and NASA (National Aeronautics and Space Agency; Pilewskie et al., 2002). The IfT-instrument, called albedometer, is unique due to

(a) its active horizontal stabilization of the up- and downward facing sensor heads (thus avoiding sensor misalignment problems), and
(b) its high spectral resolution (2-3 nm).

The NASA-instrument has a moderate spectral resolution (8-12 nm), and the sensor heads are fixed with the aircraft fuselage. The advantage of the NASA instrument is the larger wavelength range (300-1700 nm) compared to the albedometer (290-1000 nm).

In this contribution the albedometer is introduced and first measurements with this new type of irradiance sensor are presented. Furthermore, comparisons of the measured spectra with the output of two independent one-dimensional radiative transfer models are discussed shortly.

The new albedometer

The airborne spectral albedometer consists of two optical inlets (one upward-, the other one downward-looking) to measure down- and upwelling spectral irradiances. The optical inlets are connected via fiber optics to two identical multi-channel spectrometers (MCSs). Each MCS consists of a fixed grating for wavelength splitting and a 1024 pixel diode array for detection. It should be noted that for wavelengths smaller than about 310 nm and larger than 965 nm the data are uncertain due to the decreasing sensitivity of the diode array detectors towards their edges. The use of a fixed grating instead of a scanning spectrometer enables a high time resolution of the measurements (usually 0.3 s) and assures temporal wavelength stability during airborne operation. The two albedometer components were calibrated in absolute irradiance units using a 200 W tungsten halogen lamp, which is traceable to an absolute standard maintained at PTB (Physikalisch-Technische Bundesanstalt), the German National Calibration Authority. The deviations from the ideal cosine angular response were characterized in the laboratory as a function of incidence angle and wavelength, and were used to correct the irradiance measurements. The wavelength calibration of the two spectrometers has been checked and the FWHM (Full Width at Half Maximum) has been determined as a function of wavelength using different gas lamps (Hg, Ne, Kr, Xe, Ar) with distinct spectral peaks. FWHM values between 1.8 and 3.5 nm were obtained and the average value of 2.9 nm was used in the data analysis. The albedometer is equipped with a unique active horizontal stabilization unit, which significantly minimizes uncertainties related to horizontal misalignment of the up- and downward-looking optical inlets during the airplane movements (Wendisch et al., 2001). Altogether a measurement uncertainty for the spectral irradiance measurements with the albedometer of ±4% for wavelengths λ = 400-770 nm and of ±6% for $\lambda \leq 400$ nm and $\lambda \geq 770$ nm was estimated (Wendisch, 2002).

Measurements and comparison with model results

The presented data were gathered with an instrumented airplane during an experiment at the German North Sea coast (54.3°N, 9.4°E) in September and October 2000. Several profile measurements were collected over open sea and over land (hereafter referred to as “SEA” and “LAND”, respectively) under cloudless conditions. In particular, measurements of two descending

flight patterns performed on 30 September 2000 are shown in Figure 1.
During the slant aircraft descent the solar zenith angle θ_s varied between the values indicated in the figure. The average θ_s for the measurement over "SEA" was about 5° smaller than for the "LAND" measurements. Therefore the downwelling irradiance over "SEA" is higher (for the same altitude and wavelength) compared to the respective measurement over "LAND". The most obvious feature of Figure 1 is the systematic increase of both down- and upwelling irradiances with altitude. The spectral slope of the spectra increases with altitude due to Rayleigh scattering and the high amount of large particles at lower altitudes. The spectra clearly reveal the gas absorption bands of O_3, H_2O, and O_2 as well as some Fraunhofer line features, especially at the shortwave end of the spectrum. The upwelling irradiances show the typical reflection properties of the underlying surface. Over "SEA" the spectral slope is more or less continuous, whereas over "LAND" the typical vegetation step around $\lambda \approx 700$ nm is obvious.
The measurements are compared with the output of two radiative transfer models (Wendisch and Mayer, 2003). The differences between measurements and calculations of spectral downwelling irradiances are mostly less than ±10%, i.e., within the uncertainties of the measurements and calculations. However, the measured upwelling irradiance significantly deviates from the respective simulations, which is a clear effect of the unrealistic representation of the spectral surface properties in the models.

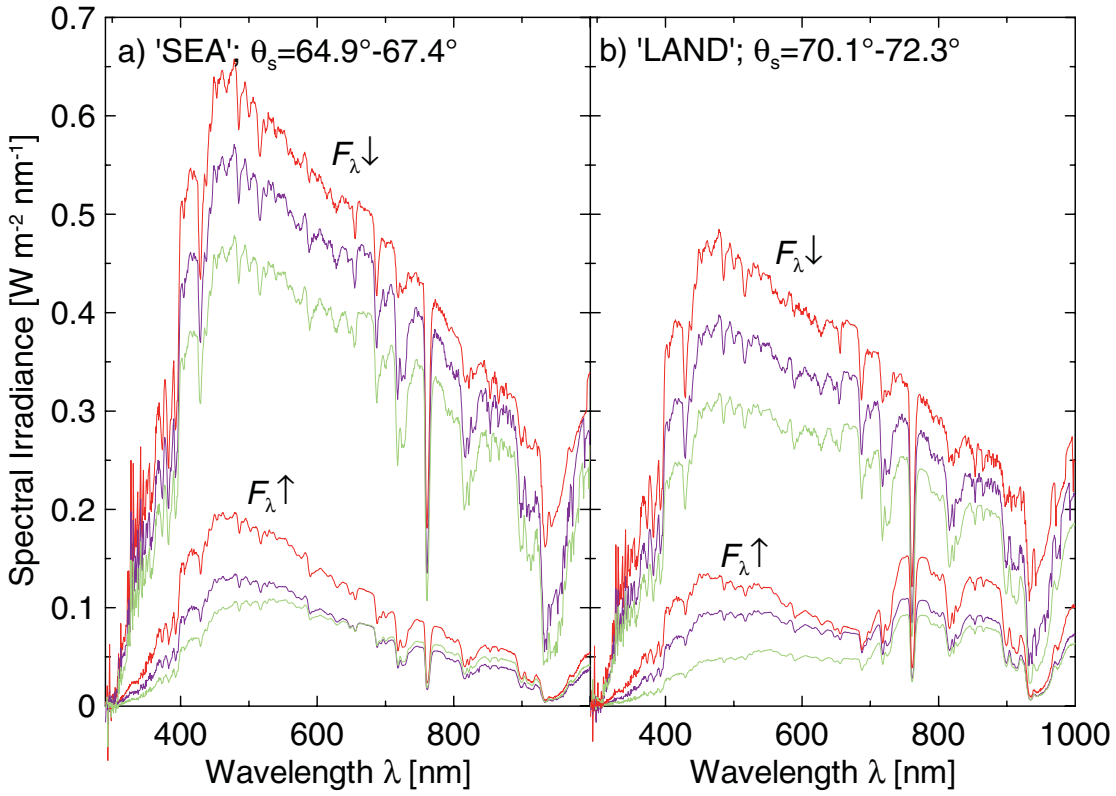

Fig. 1: *Down- and upwelling spectral irradiances (Fλ↓ and Fλ↑, respectively) measured at 3 km (red lines), 1.2 km (blue lines), and 0.4 km (green lines) altitude over "SEA" (Figure 1a) and "LAND" (Figure 1b).*

Conclusions

A new spectral albedometer is introduced, first airborne measurements of spectral down- and upwelling irradiances with a spectral resolution of 2-3 nm in the wavelength range between 290-1000 nm are presented. The level of the downwelling spectra as well as the their slope increases with altitude, showing the influence of Rayleigh scattering and aerosol particles in the planetary boundary layer. The upwelling spectra clearly represent the reflection features of the underlying surface. By comparing the measurements with respective radiative transfer calculations it is shown that it is necessary to determine surface albedo specifically for each location or campaign, rather than adopting literature values. With the albedometer a suitable experimental tool for this task is available.

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Aerosol extinction measurements in the planetary boundary layer

Thomas Müller, René Dubois

Principle of operation of the DOAS extinction telescope

A system for measuring the trace gas absorption coefficient and the particle extinction coefficient in a broad spectral range (λ = 280 to 1000 nm) in the planetary boundary layer was developed. An enormous advantage of the new system is the capability to measure under truly ambient conditions. The optical arrangement is based on a DOAS (Differential Optical Absorption Spectroscopy) system for measuring trace gas concentrations of atmospheric absorbers in the ultraviolet and visible. It consists of a light source emitting the spectral intensity $I_0(\lambda)$, an emitting and receiving telescope, a reflector array and a detection unit. After a passage through the atmosphere with the optical measurement path length L_m, the transmitted spectral intensity $I_m(\lambda)$ is given by the Lambert Beer Law. Classical absorption spectroscopy cannot be used because atmospheric particle scattering overlaps with trace gas absorption. As the spectral characteristics of the particle extinction varies weakly with wavelength, DOAS analyses only narrow absorption bands with widths less than 5 nm. With this approach, a separation into a differential absorption cross section strongly varying with wavelength is used. The absolute intensity value is irrelevant in this case. In contrast to that, the absolute values of the extinction coefficient $b_{ext}(\lambda)$ and hence the absolute intensities are essential for particle extinction measurements. The slope of the smooth particle extinction spectra depends mostly on the particle size distribution, which can be retrieved from the extinction spectra by the use of inversion techniques. An absolute intensity measurement, however, is technically difficult to realize. The measured intensity $I_m(\lambda)$ is influenced by a dimensionless factor κ which considers the geometry of the optics and a factor $\gamma(\lambda)$ which includes all spectral system characteristics. The factor $\gamma(\lambda)$ can be eliminated by a reference measurement $I_r(\lambda)$. Hence for the absolute intensity measurement $I_m(\lambda)$ a calibration constant κ has to be derived experimentally. One example of how to perform such an experiment for special atmospheric conditions is presented below.

Construction of the DOAS extinction telescope

The DOAS extinction telescope is placed in a laboratory above the roof of the institute. The divergent light $I_0(\lambda)$ of a 450 W Xe arc high pressure lamp is reflected through a vertically positioned telescope to a parabolic mirror with diameter of 400 mm and 2000 mm focal length. The light beam can be directed in any horizontal direction by means of a scan mirror in an astronomical dome on the roof (Figure 1). The light passes through the atmosphere and is returned by a reflector array, e.g., in 3.2 km distance and 50 m height. The reflected light enters the telescope and is focused onto a beam divider of the detection unit where $I_m(\lambda)$ is measured by three spectrometers and a photodiode. The coupling of the two spectrometers (λ = 280 - 575 nm, λ = 530 - 1000 nm, respectively) for extinction and the third for trace gas measurements are realized by optical fibers. Fast spatial variations due to optical turbulence in the atmosphere generate on the optical fiber entrance an unstable light spot. Hence, there is an undefined loss of the intensity $I_m(\lambda)$ during the spectrometer detection. Consequently, not the absolute value of the spectral intensity but the spectral dependence of $I_m(\lambda)$ can be determined. Additionally, $I_m(\lambda)$ is measured at λ = 550 nm directly by a large-area photodiode, so that the light spot is always within its detection area and no such intensity loss happens. The highly variable atmospheric optical background, electronic background signals, and the dark current of the photodiode are eliminated by phase sensitive detection. For that purpose, the light beam is modulated with an optical chopper.

Fig. 1: *The DOAS laboratory with the dome.*

The spectrometer-measured spectra $I_m(\lambda)$ are fitted at λ = 550 nm to the corresponding absolute values I_m(550 nm) of the photodiode. Consequently, absolute spectral intensities are derived and used in combination with a reference measurement $I_r(\lambda)$ for calibration to calculate the absolute spectral extinction coefficient $b_{ext}(\lambda)$. After removing the Rayleigh scattering and trace gas absorption the spectral particle extinction coefficient $b_{p,ext}(\lambda)$ is given.

A measurement example

The 10th and 11th of October 2002 are selected to show a short measurement period which includes a calibration. Figure 2 shows a time series of the relative humidity, and the ambient extinction coefficient for the wavelength 550 nm, and the dry particle scattering coefficient, measured with an integrating nephelometer at a relative humidity (RH) of 20%. All data are hourly averages. The extinction data were recorded only in the daytime, because the necessary control for the automatic measurements during night time has not been realized yet. In contrast to the path-averaged extinction coefficient (3.2 km distance), the scattering coefficient and relative humidity are point measurements performed on the roof of the institute building. For data comparison especially for the calibration, relatively homogeneous atmospheric conditions are required.

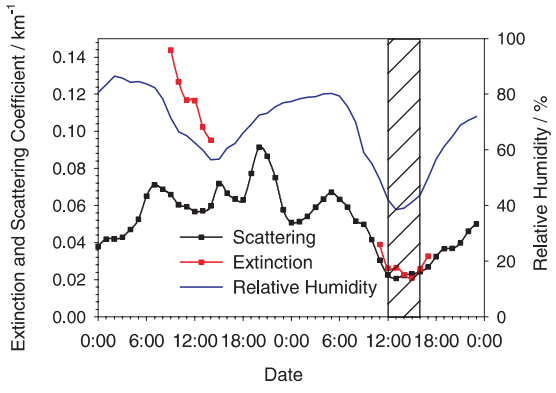

Fig. 2: *Hourly mean values measured at λ = 550 nm of ambient extinction coefficient (DOAS-extinction system) and scattering coefficient (Nepehlometer) at dry state (20% relative humidity) of the 10th and 11th of October 2002 in Leipzig. The time period for the calibration is labeled by shading.*

For the calibration, a period with low relative humidity around 40% and low scattering coefficients of 0.02 km^{-1} was chosen to minimize the uncertainty of the calibration. These low values were chosen, because the calculated correction for the hygroscopic particle growth has its smallest uncertainty for low relative humidities and small particle concentrations. The calibration was performed on the 11th October between 12:00 and 16:00 UTC by fitting the measured extinction coefficient b_{ext}(550 nm) to a calculated extinction coefficient in the following way. The measured particle scattering (at 20% RH) is corrected to 0% relative humidity with a hygroscopic growth factor from literature. Then a dry particle absorption coefficient of 10% of the dry scattering coefficient is assumed to calculate a dry extinction coefficient (0% RH). Likewise, the extinction coefficient at ambient relative humidity (40% RH) is derived with a hygroscopic growth factor from literature. Then the measured extinction coefficient (40% RH) is adjusted to the calculated extinction coefficient by changing the calibration constant κ. The four hourly means of the spectral extinction coefficient at λ = 550 nm during the calibration time have a standard deviation of 0.0076 km^{-1}. This value is used as the systems inherent and constant absolute accuracy for $b_{p,ext}$(550nm).

To show the dependence of the extinction coefficient on the relative humidity, the ratio of the extinction coefficient measured at ambient conditions and the dry state (RH=20%) scattering coefficient is shown in Figure 3. An increase of the extinction coefficient with relative humidity is clearly visible. The larger error bars at humidities below 50% are due to the assumed constant absolute accuracy in relation to the small extinction coefficients.

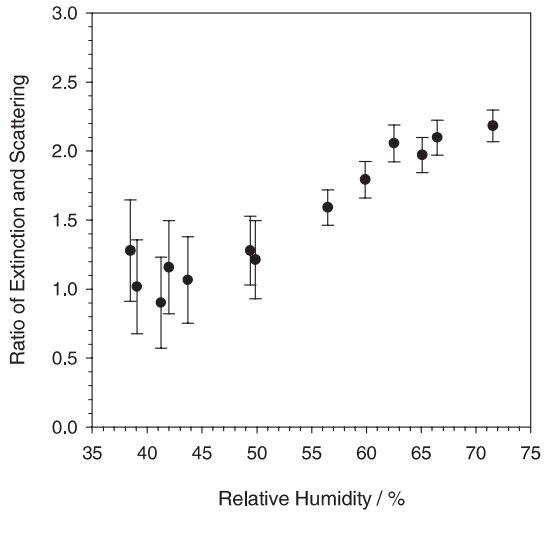

Fig. 3: *Ratio of ambient particle extinction and scattering coefficient (20% relative humidity) with error bars, both at wavelength 550 nm versus relative humidity from the data in Figure 2.*

Figure 4 shows changes of the spectral extinction coefficient $b_{ext}(\lambda)$ with respect to a reference spectrum measured during the calibration period at 14:25 UTC 11th of October 2002. The variability is caused by the particle and trace gas influence

on the spectral extinction coefficient $b_{ext}(\lambda)$ in the ultraviolet to near infrared region. In spectrum #1 to #4 the deviation in the near infrared region (800 nm - 1000 nm) is due to a higher water vapour concentration compared to the reference case. The contribution of ozone to the total extinction is noticeable in all cases. In the cases #1 to #4 an increase of the extinction in the ultraviolet region (280 nm - 320 nm) due to an increased ozone concentration occurred. In cases #5 and #6, the ozone concentration decreased.

This example points out how important it is for the absolute spectral particle extinction coefficient $b_{p,ext}(\lambda)$ to remove correctly the trace gas absorption and Rayleigh scattering. A correction by measuring trace gas concentrations with DOAS leads to an improved particle extinction spectrum from 280 to 1000 nm.

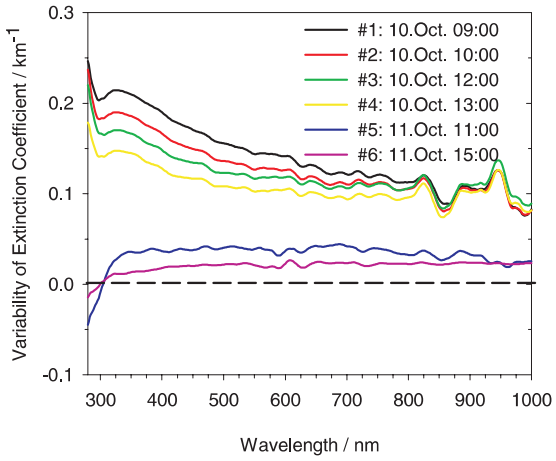

Fig. 4: *Change of ambient spectral extinction coefficient with respect to a reference spectrum at 14:25 UTC 11th of October 2002. Values above 0.0 km^{-1} are due to a relative increase of gas absorption and particle extinction. Values below 0.0 km^{-1} are indicating a decrease of the absorbing species relative to the reference spectra.*

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Leipzig Aerosol Cloud Interaction Simulator (LACIS): First results

Frank Stratmann, Alexej Kiselev, Heike Wex, Sabine Wurzler, Manfred Wendisch, Jost Heintzenberg, Karoline Diehl and Sebastian Schmidt

To validate experimentally the basic theory and the recent extensions of the theory of cloud droplet activation and growth (e.g., Kulmala et al., 1993), a new device to study the formation and growth of cloud droplets under realistic super-saturations has been designed and constructed at the IfT. Specific scientific tasks to be adressed utilizing the new device are:

- Comparison of the measured equilibrium size of pure salt particles and mixtures of salts with insoluble (or slightly soluble) material of various original sizes and compositions as a function of the water vapor saturation with Köhler theory.
- Comparison of the measured growth of aerosol particles and droplets by water vapor diffusion with known theoretical growth equations.
- Experimental investigation of the influence of atmospheric trace gases (e.g., HCl, HNO_3) on the growth of aerosol particles (soluble, insoluble and mixed).
- Experimental determination of the competition effect when aerosol particles of different sizes grow simultaneously as compared to the growth of the individual sizes.
- Experimental verification of parameterizations of the relationship between particle number/mass concentration and droplet number concentration as a function of the chemical and microphysical environment of the cloud.

On the basis of a careful review of the available literature it was concluded, that a flow diffusion tube seems to be the most suitable tool to perform such investigations (Stratmann et al., 2002). Consequently, the Leipzig Aerosol Cloud Interaction Simulator (LACIS) was realized as a laminar flow tube. It has been designed to reproduce the thermodynamic conditions of atmospheric clouds in a realistic manner and to control and adjust these conditions as accurately as needed. LACIS was constructed utilizing a numerical particle/droplet growth model which was realized in the framework of the Fine Particle Model (FPM, Wilck et al., 2002), a newly developed Eulerian particle dynamical add-on module to FLUENT 6 (Fluent, 2001). The numerical simulations performed with this model showed that the thermodynamic fields (temperature, Figure 1a and saturation, Figure 1b) inside the laminar flow tube of LACIS are rather inhomogeneous within the tube. This finding necessitated injecting the particles/droplets near the centerline of the tube with a particle free sheath air surrounding the resulting particle beam (Figure 1c). In this way, injected particles experience similar thermodynamic conditions during their growth.

A series of experiments was conducted to evaluate the behavior of LACIS and to prove that it can be applied to investigate the growth of particles/

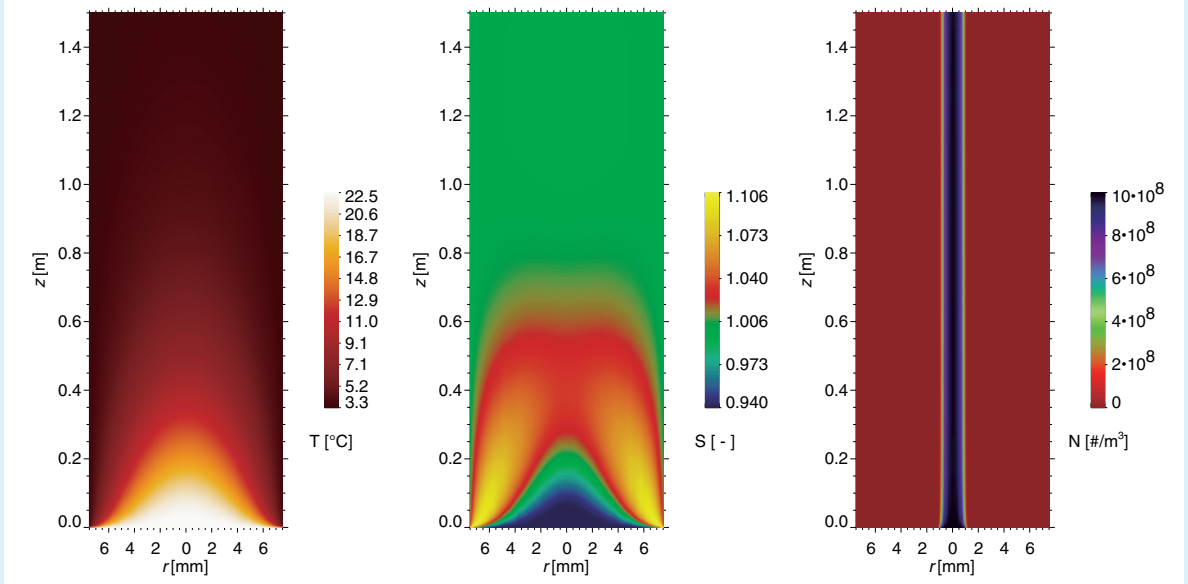

Fig. 1: Temperature (a), saturation (b) and particle/droplet number concentration (c) profiles inside the flow tube (flow direction bottom to top).

droplets under conditions close to those observed in natural clouds (Stratmann et al., 2003). The flow field inside the LACIS flow tube was characterized. The measurements showed a well-defined and fully developed laminar flow inside LACIS. Visualization experiments proved that the generated particle beam could be maintained stable, well-defined and undisturbed for wet and dry conditions on the time scale of days.

Furthermore, monodisperse seed particles of known size (here 100 nm in diameter) and composition (NaCl) were injected into LACIS and mean droplet diameters were measured at the outlet of the LACIS tube with a Fast-FSSP (Forward Scattering Spectrometer Probe, Brenguier et al., 1998) for a variety of thermodynamic conditions. Figure 2 shows calculated (lines) and measured (symbols) droplet diameters as function of saturator temperature, i.e. different relative humidities, and for two different LACIS wall temperatures.

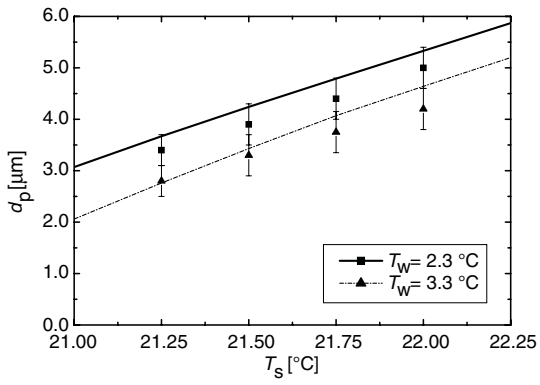

Fig. 2: *Calculated (lines) and measured (symbols) mean droplet diameters d_p as function of saturator temperature T_S and for two different wall temperatures T_W.*

Calculated and measured droplet diameters in Figure 2 agree within the range given by the measurement uncertainties (vertical bars). The measurement uncertainties were found to be approximately +/- 0.4 µm. Considering the more than four orders of magnitude particle/droplet volume change taking place inside LACIS, the agreement between theory and experiment is excellent.

Summarizing the measurement results it is concluded:

- For constant thermodynamic conditions measured droplet sizes were constant and stable on the time scale of hours.
- Measured and numerically predicted droplet sizes agreed within the level of experimental uncertainty.
- In accordance with the theoretical model, the droplet diameter could be systematically and reproducibly varied by changing the critical thermodynamic conditions of the set-up.

Thus, LACIS is capable of addressing a variety of crucial open questions concerning the dynamical physical and chemical processes affecting cloud formation under controlled laboratory conditions. Consequently, it is a suitable tool to improve the understanding of cloud formation and the effects of clouds on processes of global significance.

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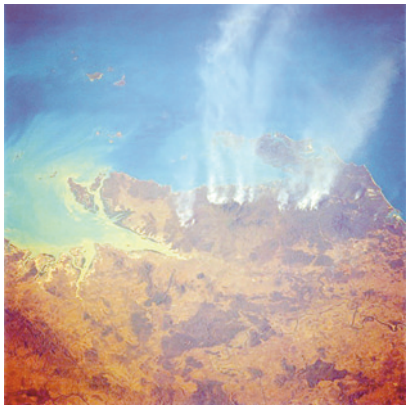

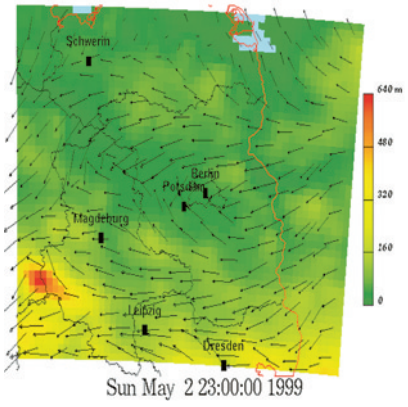
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Appendix

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Book section

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Lectures

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Heintzenberg, J. 2001. *Das atmosphärische Aerosol oder wie gut verstehen wir den Dunst, in dem wir leben?* Universität Bayreuth, Deutschland. 1. Februar. *)

Heintzenberg, J. 2001. *Aerosolprozesse in der Grenzschicht.* MPI für Meteorologie Hamburg, Deutschland. 12. Februar.

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Heintzenberg, J. 2001. *Wendet das atmosphärische Aerosol die Klimakatastrophe ab?* Berlin-Brandenburgische Akademie der Wissenschaften, Berlin, Deutschland. 28. September. *)

Hesper, J. und Herrmann, H. 2001. *OH- und RO_2-Kinetik in flüssiger wässriger Phase.* Strahlenchemikertreffen. Naumburg, Deutschland. 17.-20. September.

Knoth, O. 2001. *A nonhydrostatic model using z-coordinate numerics, parameterisations and tests.* DWD, Seminar of the LM-User Group, Langen, Germany. Langen. 21-23 May.

Knoth, O. 2001. *A non-hydrostatic model using z-coordinate numerics.* DWD, Fourth International SRNWP-Workshop on Non-Hydrostatic Modelling, Bad Orb, Germany. Bad Orb. 24-26 September.

Lammel, G., Brüggemann, E., Gnauk, T., Müller, K., Neusüß, C. und Röhrl, A. 2001. *Das bodennahe Aerosol: Zusammensetzung und Variabilität.* Abschlusskolloquium Aerosolforschungsschwerpunkt (AFS), Köln, Deutschland. Köln. 23. März.

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Wendisch, M. 2002. *Globale Klimaänderung - Was geht uns das an?* Naunhofer Kulturwerkstatt. Naunhof, Deutschland. 9. März. *)

Wendisch, M. 2002. *Flugzeuggetragene Strahlungsmessungen mit einem spektralen Albedometer.* Alfred-Wegener-Insitut. Bremerhaven, Deutschland. 10. Juni. *)

Wendisch, M. 2002. *Globale Klimaänderung - Was geht uns das an?* Gymnasium Engelsdorf, Deutschland. 11. Oktober. *)

Wendisch, M. und Coe, H. 2002. *Results of the EURAR-Workshop on „Airborne Particle Inlets“.* 4th EUFAR Progress Meeting, Instituto Nacional De Tecnica Aerospacial (INTA). Madrid, Espania. 12 June. *)

Wendisch, M., Jäkel, E. und Schmidt, S. 2002. *Airborne solar irradiance measurements with a spectral albedometer.* EUFAR-Workshop on European Airborne Atmospheric Radiation Measurements, Meteorological Research Flight (MRF). Farnborough, England, UK. 1 October. *)

Wendisch, M., Jäkel, E., Schmidt, S. und Trautmann, T. 2002. *Inhomogeneities of aerosols, clouds, and surface albedo - Influence on irradiances and actinic flux density.* INSPECTRO Kick-off meeting. Norwich, UK. 7-10 January.

Wendisch, M., Schmidt, S., Jäkel, E. und Trautmann, T. 2002. *Cloud inhomogeneities and their influence on spectral irradiances and optical cloud properties.* AFO 2000. The German Atmospheric Research Programme 2000 - 2006. Statusseminar. Schliersee, Germany. 7-9 October.

Wiedensohler, A., Wehner, B., Maßling, A., Birmili, W. und Herrmann, H. 2002. *Feine und ultrafeine Partikel - Außenluftbelastung und Exposition. Wirkung und Erfassung von Fein- und Ultrafeinstäuben.* Institut für Toxikologie und Umwelthygiene der TU München, Deutschland. 12. Februar. *)

Wiedensohler, A., Wehner, B., Maßling, A. und Tuch, T. 2002. *Physical properties of urban aerosole.* 19th Symposium on Aerosol Science and Technology. Kyoto, Japan. 7 August. *)

Wolke, R., Herrmann, H., Diehl, K., Hinneburg, D., Knoth, O., Majdik, Z.-T., Sehili, A. M., Wurzler, S., Zech, G., Mauersberger, G. und Müller, F. 2002. *Modeling of tropospheric multiphase processes: Tools and chemical mechanisms (MODMEP).* AFO 2000. The German Atmospheric Research Programme 2000 - 2006. Statusseminar. Schliersee, Germany. 7-9 October.

Wurzler, S. 2002. *Welche Rolle spielen unlösliche Partikel in Wolken bei der Bildung der Eisphase?* DLR Oberpfaffenhofen, Deutschland. 13.-14. Juni. *)

Wurzler, S. 2002. *Gute Luft, schlechte Luft.* „Das Jahr der Geowissenschaften“, Veranstaltung „Lufttage“ im Hauptbahnhof Leipzig (BMBF). Leipzig, Deutschland. 19. April. *)

Wurzler, S. 2002. *Wechselwirkungen Aerosolpartikel und Wolken: Die Rolle unlöslicher Partikel in troposphärischen Wolken.* Institut für Physik der Atmosphäre, DLR. Oberpfaffenhofen, Deutschland. 4. November. *)

Wurzler, S. 2002. *Wechselwirkungen Aerosolpartikel und Wolken: Die Rolle unlöslicher Partikel in troposphärischen Wolken.* Zentrum für Umweltforschung (ZUF), Universität Frankfurt, Deutschland. 27. November. *)

Wurzler, S., Herrmann, H., Iinuma, Y., Simmel, M., Stratmann, F., Wiedensohler, A., Zech, G., Zeromskiene, K., Hungershöfer, K., Trautmann, T., Andreae, M. O., Helas, G., Trentmann, J., Graf, H., Langmann, B., Nober, F. und Textor, C. 2002. *Impact of vegetation fires on the composition and circulation of the atmosphere: The AFO2000 joint research project EFEU.* AFO 2000. The German Atmospheric Research Programme 2000 - 2006. Statusseminar. Schliersee, Germany. 7-9 October.

*) invited lectures

Patent

2002

Berndt, T., Böge, O. und Heintzenberg, J., **2002**, Verfahren zur Herstellung von Epoxiden durch Oxidation von Olefinen (Patentantrag), Deutschland: DE 100 44 538 A 1. Int.: PCT/EP01/10231 (WO 02/20502 A1)

University courses

Winter semester	2000/2001	University Leipzig Faculty for Physics and Geosciences
Heintzenberg, J.	Frontlines of Atmospheric Research	1 HPW*)
Heintzenberg, J.	Radiation on the Atmosphere	1 HPW
Heintzenberg, J. Stratmann, F. Wiedensohler, A.	Atmospheric Aerosols I	2 HPW
Herrmann, H.	Atmospheric Chemistry I Atmospheric Chemistry Seminar Atmospheric Chemistry Lab	2 HPW 2 HPW block course
Renner, E.	Modeling of transport and chemical transformation of air pollutants	2 HPW
Wendisch, M.	Airborne Physical Measurement Methods	1 HPW
Althausen, D.	Optical Measurement Technique	1 HPW
Wandinger, U.	Atmospheric Optics	1 HPW

Winter semester	2000/2001	Friedrich-Schiller-University Jena Faculty for Chemistry and Geosciences
Renner, E.	Environmental Meteorology Modeling of transport and chemical transformation of air pollutants	

Summer semester	2001	University Leipzig Faculty for Physics and Geosciences
Heintzenberg, J. Ansmann, A. Dubois, R. Klugmann, D. Siebert, H.	Modern Meteorological Instruments I	1 HPW
Heintzenberg, J. Stratmann, F. Wiedensohler A.	Atmospheric Aerosols II	2 HPW
Herrmann, H.	Atmospheric Chemistry II Atmospheric Chemistry Seminar Atmospheric Chemistry Exercises	2 HPW 2 HPW 1 HPW
Renner, E. Hellmuth, O. Knoth, O. Wolke, R.	Mesoscale Meteorological Modeling	2 HPW
Wendisch, M.	Airborne Physical Measurement Methods	1 HPW

Winter semester	2001/2002	University Leipzig Faculty for Physics and Geosciences
Heintzenberg, J.	Frontlines of Atmospheric Research	1 HPW*)
Heintzenberg, J.	Radiation on the Atmosphere	1 HPW
Heintzenberg, J. Stratmann, F. Wiedensohler, A.	Atmospheric Aerosols I	2 HPW
Heintzenberg, J. Wendisch, M. Wurzler, S.	Cloud Physics I	2 HPW
Herrmann, H.	Atmospheric Chemistry I Atmospheric Chemistry Seminar Atmospheric Chemistry Lab	2 HPW 2 HPW block course
Renner, E.	Modeling of transport and chemical transformation of air pollutants	2 HPW
Wendisch, M.	Airborne Physical Measurement Methods	1 HPW
Althausen, D.	Optical Measurement Technique	1 HPW
Wandinger, U.	Atmospheric Optics	1 HPW

Winter semester	2001/2002	Friedrich-Schiller-University Jena Faculty for Chemistry and Geosciences
Renner, E.	Environmental Meteorology Modeling of transport and chemical transformation of air pollutants	

Summer semester	2002	University Leipzig Faculty for Physics and Geosciences
Heintzenberg, J. Ansmann, A. Dubois, R. Klugmann, D. Siebert, H.	Modern Meteorological Instruments I	1 HPW
Heintzenberg, J. Stratmann, F. Wiedensohler A. Hellmuth, O.	Atmospheric Aerosols II	2 HPW
Herrmann, H.	Atmospheric Chemistry II Atmospheric Chemistry Seminar Atmospheric Chemistry Exercises	2 HPW 2 HPW 1 HPW
Renner, E. Hellmuth, O. Knoth, O. Wolke, R.	Mesoscale Meteorological Modeling	2 HPW

Summer semester	2002	University Leipzig Faculty for Physics and Geosciences
Wendisch, M.	Airborne Physical Measurement Methods	1 HPW
Trautmann, Th. Wendisch, M. Heintzenberg, J.	Radiative Transfer in Clouds	2 HPW

Summer semester	2002	University Hohenheim Graduiertenkolleg 768 Strategien zur Vermeidung der Emission klimarelevanter Gase und umwelttoxischer Stoffe aus Landwirtschaft und Landschaftsnutzung
Heintzenberg, J.	The Role of Aerosol Particles in the Climate System of the Earth	

Summer semester	2002	Universidad Técnica Federico Santa Maria Valparaiso, Chile
Heintzenberg, J.	Atmospheric Aerosols	

Master thesis 2001

University Leipzig Faculty for Physics and Geosciences	
Galgon, Diane	Die Größenverteilung feiner atmosphärischer Partikel in der Abluftfahne einer Großstadt
Bräsel, Sylvia	Untersuchung von Partikelneubildung und -wachstum im atmosphärisch relevanten H_2So_4-H_2O System im laminaren Strömungsrohr

Master thesis 2002

University Leipzig Faculty for Physics and Geosciences	
Fischer, Corinne	Partikelgrößenverteilung und Bildung ultrafeiner Partikel in der Arktis
Chemnitzer, Renè	Analytik von Metallen in troposphärischen Partikeln und Wolkentröpfchen mit Atomabsorptionsspektroskopie

Doctoral theses 2001

University Leipzig Faculty for Physics and Geosciences	
Müller, Thomas	Bestimmung streckenintegrierter Aerosolparameter und Wasserdampf-konzentrationen aus spektralen Extinktionsmessungen
Lohrmann, Birgit	Spurenstoffmessung in der kontinentalen Grenzschicht mittels Differentieller Optischer Absorptionsspektroskopie-Untersuchung zur salpetrigen Säure und zum No_3-Radikal
Siebert, Holger.	Tethered-Balloon Borne Turbulence Measurments in the Cloudy Boundary Layer
Maßling, Andrea	Hygroscopic Properties of Atmospheric Particles over the Atlantic and Indian Oceans

University Leipzig Faculty for Chemistry and Mineralogy	
Ervens, Barbara	Durchführung und Interpretation von Multiphasen-Modellrechnungen. Ermittlung physikochemischer Parameter zu Flüssigphasen-reaktionen des OH-Radikals
Schütze, Matthias	Entwicklung und Anwendung einer spektroskopischen Anordnung zur Ermittlung von “Up-take” Koeffizienten verschiedener Spezies an Tröpfchen
Plewka, Antje	Untersuchung des organischen Anteils im atmosphärischen Aerosol

Doctoral theses 2002

University Leipzig Faculty for Physics and Geosciences	
Mattis, Ina	Charakterisierung des troposphärischen Aerosols über einem europäischen Ballungsgebiet mit einem Ramanlidar
Wex, Heike	Closure and Sensitivity Studies on Physical Parameters of Rual Continental Aerosols
Sierau, Berko	Entwicklung eines Interfaces zwischen einem differentiellen Mobilitäts-analysator und einem Massenspektrometer
Jaenisch, Volker	Der einfluss turbulenter Mischungsprozesse auf die Bildungsraten atmo-sphärischer Aerosolpartikel

University Leipzig Faculty for Chemistry and Mineralogy	
Donati, Andreas	Zeitaufgelöste kinetische Untersuchungen halogenhaltiger Oxidantien in wässriger Phase

Guest scientists

2001		
Assoc.Prof. Min Hu	1./31. Jan. 01	Peking University, China Center for Environmental Sciences
Dr. Francisco Cereceda Balic	25. Febr./2. March 01	Univ. Técnica Federico, Santa Maria, Chile
Prof. Dr. Robert J. Charlson	9./14. April 01	University of Washington, Seattle, USA Dept. of Atmospheric Sciences
Prof. Dr. Robert J. Charlson	30. Aug./4. Sept.	University of Washington, Seattle, USA Dept. of Atmospheric Sciences
Dr. Yinon Rudich	2./7. Sept. 01	Weizmann Institute Rehovot, Dept. of Environmental Sciences, Israel
Dr. Christian Carrico	9./20. Sept. 01	Georgia Institute of Technology, Atlanta, USA
Prof. Dr. Jeffrey L. Collett	3./9.Nov. 01	Colorado State University, Colorado, USA Dept. of Atmospheric Sciences
Prof. Dr. I. Morozov	18. Nov./13. Dec. 01	Russian Academy of Sciences, Inst. of Chemical Physics, Moscow, Russia

2002		
Dr. Y. Arshinov	1. March/30. April 02	Russ. Academy of Sciences, Inst.f. Atmosph. Optics, Tomsk, Russia
Dr. S. Bobrovnikow I. Serikov	1. March/30. April 02	Russ. Academy of Sciences, Inst.f. Atmosph. Optics, Tomsk, Russia
Prof. Dr. I. Morozov	3. March/30. April 02	Russ. Academy of Sciences, Inst.f. Atmosph. Optics, Tomsk, Russia
Dr. Fred Brechtel	21. July/7. Sept. 02	Brookhaven National Laboratory, Upton, USA
Prof. Dr. J. Collett	11. Sept./21. Dec. 02	Colorado State University, Colorado, USA Dept. of Atmospheric Sciences
Dr. J. Barett	5. Aug./6. Oct. 02	Nuclear Department, HMS Sultan Gosport, UK

Scientific events

2001

Event	Place and date	Number of participants
European Aerosol Conference (EAC)	Leipzig 3-7 Sept.	581 international
International Multiphase Workshop (within the scope of the EAC)	Leipzig 4 Sept.	100 international
Preparation Meeting of the ESD-Proposal “Transistor Radar”	Leipzig 23-24 Aug.	6 national
Opening Meeting of the AFO-Project FEBUKO (Field investigations of budgets and conversations of particle phase organics in tropospheric cloud processes)	Gehlberg (Schmücke) 26-27 March	13 national
Field Measurement Campaign Tracer Experiment AFO-Projekt FEBUKO	Gehlberg (Schmücke) 14-31 May	14 national
Project Meeting AFO-Project FEBUKO	Weißenstadt/Bayreuth 10 Sept.	15 national
FEBUKO Field Measurement Campaign (I)	Gehlberg (Schmücke) 24 Sept.- 8 Nov.	32 national
BEWA 2000 Field Measurement Campaign (Regional biogenic emissions of reactive volatile organic compounds)	Waldstein (Bayreuth) 2 July - 4 Aug.	9 national
UAVL-Meeting (Unmanned Airborne Vehicles Laboratory)	Leipzig 2-4 July	4 national 9 international
Project Meeting EFEU (Impact of Vegetation Fires on the Compositionand Circulation of the Atmosphere)	Leipzig 28-29 May Hamburg 29-30 Nov.	16 national 14 national
Project Meeting MODMEP (Modelling of Tropospheric Multiphase Processes: Tools and Chemical Mechanisms)	Leipzig 15-16 March	11 national

2002

Event	Place and date	Number of participants
Projekt Meeting MODMEP	Leipzig 28 Febr.- 1 March	12 national
EUFAR- International Workshop on Airborne Particle Inlets (European Fleet for Atmospheric Research)	Leipzig 12-13 April	20 national 15 international
Projekt Meeting FEBUKO	Berlin 25-26 April	20 national
DFG-Workshop Atmospheric Forecast 2020	Leipzig 2-3 May	22 national
Data Analyzis Workshop (BBC-Measurement Campaign)	Leipzig 13-16 May	20 national 40 international
Project Meeting EFEU	Mainz 13-14 June	14 national
BEWA Field Measurement Campaign	Waldberg (Bayreuth) 1 July- 2 Aug.	9 national
FEBUKO Field Measurement Campaign (II)	Thür. Wald 16 Sept.- 30 Oct.	34 national 1 international
Project Meeting MODMEP	Gehlberg (Schmücke) 23-24 Oct	11 national
Project Meeting FEBUKO/MODMEP	Gehlberg (Schmücke) 24-25 Oct.	32 national 1 international

Memberships

Ansmann, A.

- NASDA-ESA EarthCARE Joint Mission Advisory Group

Heintzenberg, Jost

- Nationales Komitee für Forschung auf dem Gebiet globaler Umweltveränderungen der Deutschen Forschungsgemeinschaft
- Ordentliches Mitglied der Sächsischen Akademie der Wissenschaften
- Außerordentliches Mitglied Berlin-Brandenburgische Akademie der Wissenschaften (ab 2002)
- Wissenschaftlicher Beirat des Deutschen Wetterdienstes
- VDI-Ausschuss “Messen von Ruß”
- Sprecher der Sektion E der Wissenschaftsgemeinschaft “Gottfried Wilhelm Leibniz”
- Editor of Geophysical Research Letters (bis 2001)
- Editorial Board Tellus B, Atmospheric Research
- Scientific Advisory Board of the Research Institute for Humanity and Nature, Kyoto, Japan (ab 2002)
- ESF Scientific User Committee for EUFAR (European Fleet for Atmospheric Research)
- Permanent Scientific Advisory Committee of the Centro de Geofisica de Evora, Portugal

Herrmann, Hartmut

- Sprecher der Gruppe “Mehrphasenprozesse” im BMBF-Programm AFO 2000
- IUPAC Committee “Aqueous Solution Kinetics Data for Atmospheric Chemistry”
- Koordinator des CMD-Teilprojekts CMD-APP (Aqueous Phase Processes)
- CMD Data Evaluation Panel
- GdCh, Arbeitskreis “Atmosphärenchemie”

Renner, Eberhard

- DECHEMA/GVC-Arbeitsausschuss “Schadstoffausbreitung”
- Scientific Committee of the NATO/CCMS ITM conference series, German member

Stratmann, Frank

- “Scientific Programm Committee” der Europäischen Aerosolkonferenz

Wandinger, Ulla

- International Coordination-group on Laser Atmospheric Studies (ICLAS)
- Committee on Laser Atmospheric Studies of the American Meteorological Society (CLAS)
- COST-720, European Co-operation in the field of Scientific and Technical Research - 720: Integrated Ground-Based Remote-Sensing Stations for Atmospheric Profiling, Working group 1

Wendisch, Manfred

- Koordinator (Aerosol Mikrophysik) im EUFAR (European Fleet for Atmospheric Research) Projekt

Wiedensohler, Alfred

- “Scientific Advisory Group” für Aerosole innerhalb des “Global Watch”-Programmes der World Meteorological Organization
- Editorial Board Member “Atmosphere, Water, Air and Soil Pollution”

National and international Cooperation

2001/2002

Research project	Cooperation partners
AEROSOL "The Aerosol Balance in Europe" EUROTRAC 2	• European partners
Characterization of aerosols in strong anthropogenically influenced industry and agrarregions	• Max-Planck-Institut für Meteorologie, Hamburg • Universität Hohenheim • Umweltforschungszentrum Leipzig/Halle GmbH
Characterization of aerosols over the Indian Ocean Indian Ocean Experiment (INDOEX)	• University Washington, USA • NOAA Antlantic Oceanographic and Meteorological Lab., Miami, USA • Indian Institute of Technology, Mumbai, India • Laboratoire d'Optique Atmospherique, Universite des Sciences et Technologies de Lille, Lille, France
New particle formation	• Deutscher Wetterdienst Met. Oberservatorium Hohenpeißenberg
Ground-based local closure experiment	• German partners
Multiwavelength lidar and aircraft observations during an aerosol closure experiment	• Max-Planck-Institut für Meteorologie, Hamburg • Max-Planck-Institut für Chemie, Mainz • GSF - Forschungszentrum für Umwelt und Gesundheit, München • Deutscher Wetterdienst, Met. Observatorium Lindenberg • Deutsches Zentrum für Luft- und Raumfahrt, Oberfaffenhofen / Neustrelitz Universitäten: • Berlin, Darmstadt, Potsdam, Frankfurt, Bremen München, Mainz, Würzburg, Hohenheim, Wien
Characterization of the vertical aerosol distribution over an anthropogenically influenced region	• Max-Planck-Institut für Meteorologie, Hamburg • Leibniz-Institut für Atmosphärenphysik, Kühlungsborn • Universität Potsdam • Fraunhofer-Institut für Atmosphären- und Umweltforschung, Garmisch-Patenkirchen
Optimisation of methods for the reduction emissions in the black triangle	• Hochschule Zittau/Görlitz • Sächs. Landesamt für Umwelt und Geologie, Dresden
Modelling of unimolecular reactions Model Development	• Universität Halle, Institut für Physikalische Chemie
Reservoir distribution of atmospheric trace gases	• Brandenburgische Technische Universität Cottbus • FI für Toxikol. und Aerosolforschung Hannover
CMD-APP "Interactions of free radicals with organics within the tropospheric aqueous phase"	• European partners

Research project	Cooperation partners
CMD-MPM "Modelling calculations of tropospheric gasphase and aqueous phase chemistry"	• European partners
The influence of heterogeneous reactions In seasalt-aerosol on regional and global photooxidant chemistry	• Max-Planck-Institut für Chemie, Mainz • University of Utrecht, Netherlands
Atmospheric aerosol chemistry	• Harvard University, Boston, USA
MITRAS, Development of a numerical scheme for high resolution models under consideration of transport and deposition of diesel soot	• Universität Hamburg, Meteorologisches Institut • Alfred-Wegener-Institut, Bremerhaven • Fraunhofer Institut f. Atmosphären- und Umweltforschung, Garmisch-Partenkirchen
Modelling of mixing height and of entrainment processes	• Universität Hamburg, Meteorologisches Institut • Deutscher Wetterdienst, Offenbach • Deutscher Wetterdienst, Met. Observatorium Lindenberg
Parallelization of meteorological and chemical transport models	• Universität Leipzig, Institut für Meteorologie • Universität Leipzig, Institut für Informatik • Technische Universität Chemnitz, Fakultät für Mathematik
Modelling of micro-physical and chemical processes in cloud droplets	• Brandenburgische Technische Universität Cottbus • Universität Hamburg, Meteorologisches Institut
Implicit-explicit methods for integration of chemistry-transport-models	• Universität Halle, Fachbereich Mathematik und Informatik • Centrum for Wiskunde en Informatica, Amsterdam, Netherlands
GLOREAM (Regional modelling of air pollution in Europe)	• European partners
SATURN (Studying atmospheric pollution in urban areas)	• European partners
Development of a dynamical high resolution emission inventory for Saxony	• Technische Universität Dresden • Hochschule Zittau/Görlitz
The role of cloud processing and of organic matter on the formation of soluble layers on mineral dust particles and the impact on cloud characteristics	• Universität Bonn, Meteorologisches Institut • Max-Planck-Institut für Chemie, Mainz • Tel Aviv University, Israel Dept. of Geophysics and Planetary Sciences • Weizmann Institut Rehovot, Israel
Effects of aerosol particles on drop size distributions and drop freezing: A numerical investigation with detailed cloud microphysics and derivation of parameterizations	• Universität Bonn, Institut für Meteorologie
Influence of anthropogenically polluted clouds on the radiation budget of the atmosphere and the formation of photochemical smog.	• Universität Bonn, Meteorologisches Institut • Universität Mainz, Institut für Physik der Atmosphäre • Universität Leipzig, Institut für Meteorologie

Research project	Cooperation partners
MOST (Multiphase chemistry of Oxygenated Species in the Troposphere)	• Laboratoire d'Application de la Chimie à l'Environnement (LACE), France • University College of Dublin • CNRS-LCSR, Orléans France • Bergische University Wuppertal • University of Leeds • University of Provence, France • EPFL Lausanne • Fundaciòn CEAM EUPHORE, Spain • Fraunhofer Institut für Toxicologie und Aerosolforschung, Hannover • University of Crete, Greece • Laboratoire Inter-universitaire des Systèmes atmosphériques, France
FEBUKO (Field investigations of budgets and conversations of particle phase organics in tropospheric cloud processes)	• Brandenburgische Technische Universität Cottbus • Zentrum für Umweltforschung Frankfurt • Technische Universität Darmstadt
BEWA 2000 (Regional biogenic emissions of reactive volatile organic compounds)	• Universität Innsbruck, Österreich • Institut für Meteorologie und Klimaforschung Karlsruhe • Technische Universität Darmstadt • Universität Freiburg • Universität Tübingen • Bergische Universität - Gesamthochschule Wuppertal • Max-Planck-Institut für Biogeochemie, Jena • Bayreuther Institut für Terrestrische Ökosystemforschung • Max-Planck-Institut für Chemie, Mainz
INTERCOMP 2000 (Aerosol intercomparison experiment Melpitz 2000)	• ECN Petten, Niederlande • Universität Wien, Österreich • Universität Genf, Belgien • Paul-Scherrer-Institut, Villingen, Schweiz • Finnish Meteorological Institut, Finnland • National Institut Chemistry, Ljubljana, Slovenien
Characterization of mineral dust from lidar and sunphotometer observations	• AERONET, NASA - Goddard Space Flight Center, Greenbelt, USA
Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO)	• NASA - Langley Research Center, Hampton, USA
Measurements of vertical temperature	• Institute for Atmospheric profile with a Raman lidar Optics, Tomsk, Russia
4D-CLOUDS (Inhomogene Bewölkung - Ihr Einfluss auf die Austausch- und Transportprozesse in der Atmosphäre)	• German partners
BBC (Baltex Bridge Campaign)	• European partners

Research project	Cooperation partners
INSPECTRO (Influence of clouds on the spectral actinic flux in the lower troposphere)	• European partners
ICRCCM-III International Project (Inter Comparison of Radiation Codes in Climate Models, Phase III)	• Partners from USA, Russia, Australia, UK, Canada, France, Japan, Finland
EUFAR (European Fleet for Airborne Research)	• European partners
EARLINET (European Aerosol Research Lidar Network)	• European partners
TRANSVAER (Vertical transport of particles between planetary boundary layer and free troposphere)	• Max-Planck-Institut für Meteorologie Hamburg
ATLID (EarthCARE lidar performance study)	• Meteorologisches Institut der Universität München
ACE-ASIA Aerosol Charaterization Experiment)	• University of Washington, Seattle, USA • Pacific Marine Environmental Laboratory, Seattle, USA
EFEU (Impact of vegetation fires on the compositionans circulation of the atmosphere)	• German partners
Urban Aerosol	• Umweltforschungszentrum Leipzig-Halle, Leipzig
World Calibration Center	• World Meteorological Organization, Geneva, Switzerland • Umweltbundesamt, Berlin
CARIBIC (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container)	• German partners • European partners
Hygroscopicity of supermicron particle	• University of Washington, Seattle, USA • Pacific Marine Environmental Laboratory, Seattle, USA
STINHO (Structur of turbulent transport over inhomogeneous surfaces)	• German partners
The influence of turbulence on particle formation in the meso-scale	• Deutscher Wetterdienst, Abt. Forschung u. Entwicklung
Particle transport and deposition	• Gerhard-Mercator-Universität, Duisburg • Inst. für Energie- und Umwelttechnik, Duisburg

Research project	Cooperation partners
CITY-DELTA, An inter-comparison of long-term model responses to urban-scale emission reduction scenarios	• Joint Research Centre, Institute for Environment and Sustainability, Ispra, Italy • International Institute for Applied Systems Analysis, Laxenburg, Austria • EMEP • TNO, Environment, Energy and Process Innovation, Apeldoorn, Netherlands
On the impact of LES derived turbulence length-scales on predicted mesoscale fields using the Local Model	• Max-Planck-Institut für Meteorologie, Hamburg • Deutscher Wetterdienst , Abt. Forschung u. Entwicklung, Potsdam
Development of a detailed and a moment based cloud microphysical modul for the investigation of the effects of the biomass burning on clouds AFO 2000, EFEU	• Universität Leipzig, Institut für Meteorologie • Max-Planck-Institute für Chemie, Biochemie, Mainz • Max- Planck-Institut für Meteorologie, Hamburg
Numerical simulations of the cloud – aerosol - gas interactions in the overflow over a mountain AFO 2000, MODMEP	Projektpartner MODMEP: • Max- Planck-Institut für Meteorologie, Hamburg • Brandenburgische Technische Universität Cottbus
Coupled time integration of multiphase processes AFO 2000, MODMEP	• Projektpartner MODMEP: • Max- Planck-Institut für Meteorologie, Hamburg • Brandenburgische Technische Universität Cottbus
Spatial description of clouds and their boundaries in multi-dimensional Eulerian grid models AFO 2000, MODMEP	• Projektpartner MODMEP: • Max- Planck-Institut für Meteorologie, Hamburg • Brandenburgische Technische Universität Cottbus

Boards

Scientific advisory board **2001/2002**

Name	Institution
Prof. Dr. E. Schaller (Chair)	Brandenburgische Technische Universität Cottbus Lehrstuhl für Umweltmeteorologie
Prof. Dr. J. Lelieveld	Max-Planck-Institut für Chemie, Mainz
Prof Dr. P. Lemke	Stiftung Alfred-Wegner-Institut für Polar- und Meeresforschung, Bremerhaven
Prof. Dr. U. Schurath	Forschungszentrum Karlsruhe GmbH Institut für Meteorologie und Klimaforschung
Prof. Dr. F.-J. Lübken	Leibniz-Institut für Atmosphärenphysik an der Universität Rostock
Prof. Dr. Th. Peter	Institut für Atmosphäre und Klima ETH, Zürich, Schweiz
Privat-Dozent Dr. G. Adrian	Deutscher Wetterdienst Geschäftsbereich Forschung und Entwicklung, Offenbach
Dr. R. Delmas	Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), St. Martin d'Here Cedex, Frankreich
Dr. I. Tegen	Max-Planck-Institut für Biochemie, Jena

Boards of trustees **2001/2002**

Name	Institution
MinR`in Dr. P. Karl	Sächsisches Staatsministerium für Wissenschaft und Kunst, Dresden
Dr. G. Hahn	Bundesministerium für Bildung und Forschung, Bonn
Prof. Dr. H. Graßl	Max-Planck-Institut für Meteorologie, Hamburg

Member of the IfT-Association (e.V.) **2001/2002**

Name	Institution
Prof. Dr. P. Warneck (Chair)	Max-Planck-Institut für Chemie, Mainz
MinR`in Dr. P. Karl	Sächsisches Staatsministerium für Wissenschaft und Kunst, Dresden
Dr. G. Hahn	Bundesministerium für Bildung und Forschung, Bonn
Prof. Dr. B. Brümmer	Universität Hamburg Meteorologisches Institut
Prof. Dr. W. Engewald	Universität Leipzig Fakutät für Chemie und Mineralogie
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