Results of the
German Atmospheric Research
Programme – AFO 2000

AFO 2000

Contents

* This list contains only publications originating from results of AFO2000. References in the following contributions with regard to previous or other works or in not reviewed publications are listed at the end of the corresponding contribution.

Introduction

General aspects and the development of the German atmospheric research programme AFO 2000

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General Aspects of Atmospheric Research

The Earth's atmosphere is an essential, omnipresent element of our environment, occupying a seemingly inexhaustible space. Even though the atmosphere extends more than 100 km above, its total mass is only equivalent to a minute water layer of just 10 m height. Nearly invisible and volatile, it often remains unnoticed. But it is indispensable for life on Earth - fulfilling most important functions - e.g. delivering oxygen to animals and carbon dioxide to plants, transporting water, compensating global temperature differences, removing noxious exhaust fumes, protecting the Earth's surface from deadly impacts by meteorites and irradiation and providing a temperate climate.

There has always been a natural change of the atmosphere. But the anthropogenic influence on the atmospheric composition has increased so much, that since a few years it is clear that the anthropogenic activities are significantly shifting the physical properties which are controlling our climate and our environment.

Although much has been achieved to decouple growth of economy and consumption of natural resources, the trend towards an increased usage of environment is unbroken - worldwide.

There is a widespread understanding, that a sound environment is an essential basis for sustainable development. The high standards of environmental protection in Germany are accepted worldwide which have already resulted in significant improvements, especially of water and air quality. For example, the burden of the atmosphere with acidic components from sulphur and nitrogen oxides was reduced successfully; the number of days with "summer-smog" (high ozone concentrations in the ambient air) has dropped significantly. In spite of these improvements the state and trend of some environmental parameters cause concern and will demand action. So, the concentration of very fine particulate matter in the air (aerosols) will exceed the new EC-guidelines. The nitrate and ammonia burden in the ground water is increasing. The background concentration of ozone in the ambient air is going up. The concentrations of greenhouse gases are rising. Unexpectedly strong temperature trends have been detected recently in the higher atmosphere, which can't be explained yet. The transport of atmospheric pollutants which is happening locally, regionally and even from continent to continent is still not sufficiently understood.

We already know a lot of fundamental details from substantial previous efforts in atmospheric research. Most of the basic physical and chemical processes are already well investigated. They are taking place on an extreme wide scale of space and time ranging from molecular processes occurring in 10^{-15} sec in distances of 10^{-10} m to hemispherical mixing processes lasting several years or the atmospheric removal time of more stable trace gases, which can last more than 100 years as for some fluoro-compounds and nitrous oxide (N_2O). Additionally there are further complex interactions with the biosphere,

the oceans and the Earth's crust, as well as effects of changing solar activity and periodical variations of the Earth's rotation and orbit around the sun.

This complexity of dimensions and disciplines makes it very difficult to understand and to simulate these processes as a whole, which are acting in a highly non-linear system. This means that quite sudden and hardly reversible changes can occur like the ozone hole. Additional interactions with the hydrosphere and biosphere and the fact that there is also a natural variability, are hampering the understanding and the prognosis of the consequences of the anthropogenic impact.

The continuous improvement of the system understanding helps to identify alert indicators at an early stage and is needed to find possible action options. Moreover, the ability to predict the impact reliably and with sufficient accuracy helps to avoid costly wrong decisions in environmental politics. New technical developments open better observing and interpretation facilities, namely new satellite instruments (ENVISAT) and continuously increasing computing capacities open new possibilities to bear the ambitious challenge.

Development of the atmospheric research funding focus "AFO2000"

The programme of the German Federal Government "Research for the Environment" (effective 1997-2004) was the basis for "AFO 2000" (Atmospheric Research Programme 2000) supported by the German Federal Ministry of Education, Science, Research and Technology (BMBF). It basically resumes the previous four sectoral funding foci (tropospheric research, ozone research, pollutants from aviation, and aerosol research) in a more integral approach, considering the atmosphere as a whole interacting system. The main objectives of AFO 2000 are

+ **to improve the understanding of the atmospheric system,**

+ **to develop and supply instruments for the environmental policy, and**
+ **to support young scientists in the field of atmospheric research.**

The details of the scientific aims of the first objective are outlined in the "Concept for high priority German atmospheric research for the coming decade" (http://www.afo-2000.de/004/pdf_download/afo_concept.pdf). The intention of the second objective is to support projects which are adapted more close to the needs of the potential users, in order to ease and accelerate application and implementation of the results for an efficient environmental policy. The third objective addresses the expected extensive generation change in the German atmosphere science during the next years. There will be an increased demand for talented young scientists with appropriate experience.

The call of the funding focus AFO2000 was announced in September 1999. From about 500 contribution proposals one third was selected in several appraisal steps and several meetings of experts. Where appropriate, the contributions were coordinated in joint projects before the funding could begin about one year later. The projects supporting young scientists were realised first (they already began in 2000). The duration of most projects is prevalently three years and in some cases up to five years; the last projects will end in March 2006 (see Fig. 1).

In March 2001 all projects were granted and a kick-off-meeting was held in Jena. The different projects were introduced, networking opportunities were made available and the theme groups were established to improve cooperation and coordination among the projects.

At about midterm in October 2002, first results were presented on the status symposium which was organised in Schliersee. A first balance was drawn, and the projects were reviewed by the expert group.

In March 2004 the final symposium took place in Bad Tölz, providing all PIs the opportunity to present
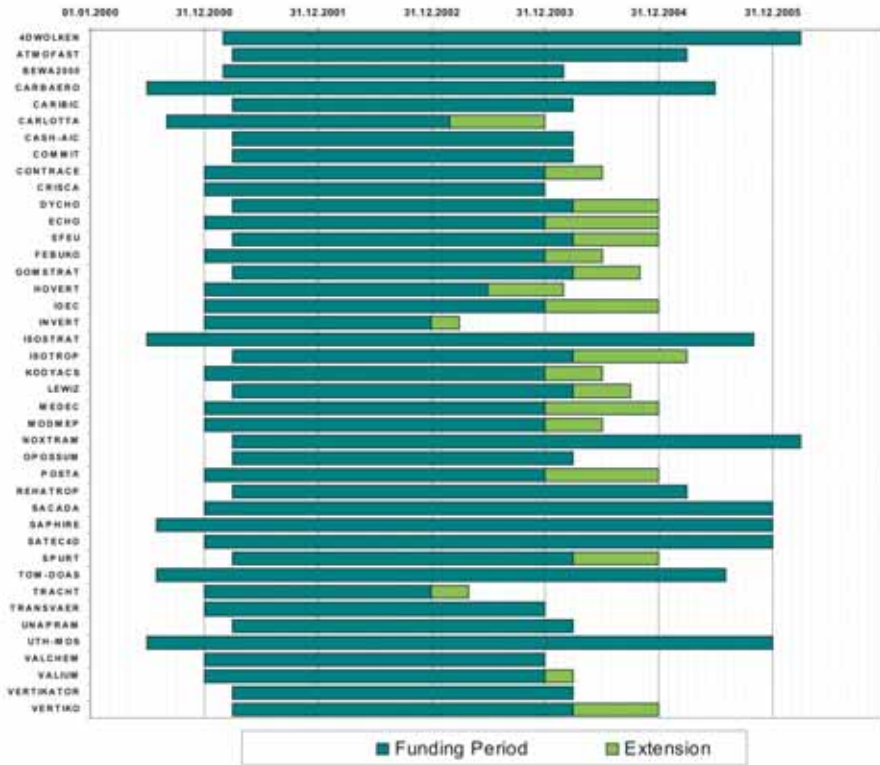
Figure 1: Funding periods of the projects of AFO 2000 in alphabetical order of the project acronyms

their results to the whole AFO-community for discussion and further use and development, application and implementation in science and policy.

The funding volume is about 10 Mio. € p.a. and amounts to ca. 42 Mio €. This sum is spread between different kinds of institutes as follows: Universities 43%, HGF[1] 30%, MPG[2] 16%, WLG[3] 10%, FhG[4] 1%. Most of the money is needed directly (45%) for personnel. The reasonable fraction 41% for subcontract work is explained by the fact, that the formation of administrative joint projects was favoured to ease cooperation. Because the institutes provided material and instruments for the most part, the additional support for these expenses, at approximately 5%, is quite low.

This publication compiles the results of the programme AFO2000 and gives an overview over the wide range of themes and the progress which has been achieved and partly is still to expect, as far as some projects haven't concluded their work yet.

More detailed information about AFO 2000 can be found in the web (http://www.afo2000.de).

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[1] HGF = Hermann von Helmholtz-Gemeinschaft Deutscher Großforschungseinrichtungen
[2] MPG = Max-Planck-Gesellschaft
[3] WGL = Wissenschaftsgemeinschaft Gottfried Wilhelm Leibniz
[4] FhG = Fraunhofer Gesellschaft

Matters of AFO 2000

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The study of atmospheric processes is an essential part of Earth system science. The most important aims of atmospheric research are an improved understanding of occurring processes as well as the forecast of the state of the atmosphere in the next decades in view of the global change presently observed. Clear symptoms for such global change can already today be identified in the environmental compartment 'atmosphere':

+ **The global background concentration of ozone in the troposphere steadily rises. The ozone concentration in regions influenced by human beings can grow by photochemical processes during the summer months to high levels establishing a possible health risk for humans and a possible danger for agriculture.**

+ **Stratospheric ozone still significantly decreases seasonally, particularly in high geographical latitudes in spring. Not only the area of the Antarctic ("ozone hole") is affected, but, to a smaller extent, the area of the higher northern latitudes. Reason for the ozone decrease has been the release of long-lasting halogen containing compounds from man-made (anthropogenic) activity. Although it is expected that ozone depletion is likely to be near its maximum (due to the control of ozone depleting substances under the Montreal Protocol and its subsequent amendments and adjustments), the timing and extent of ozone recovery in the stratosphere is uncertain (due to considerable natural variability and feedback with climate change).**

+ **The concentrations of important greenhouse gases significantly rise: Carbon dioxide (CO_2) is emitted from the burning of fossil fuels. Moreover, increasing emissions of nitrous oxide (N_2O) due to the worldwide application of nitrates as fertilisers and methane (CH_4) emissions from rice paddies and ruminants are observed.**

+ **Regions like Asia, which were not recognized as sources of strong air pollution in the past have been shown to be polluted with respect to both the gaseous phase as well as with regards to the particle phase according to experimental results in recent measurement campaigns.**

+ **Trace gases and particles formed due to anthropogenic activities and atmospheric chemical conversions affect the tropospheric and stratospheric composition, influence air pollution patterns, influence the radiative and energy budget and hence the climate and the hydrological cycle. Last, but not least, direct health risks for humans as well as for flora and fauna may arise.**

Atmospheric research on the one hand is a part of Earth system research and of research related to global change. On the other hand atmospheric research provides important contributions to the conservation of the basic living foundations of

people, flora and fauna, by trying to understand and to describe the operation of the natural atmosphere and by quantifying the consequences of anthropogenic activities as well. To this end, the following three fields, which are in close interaction with each other, are treated in atmospheric research:

+ **Field measurements: In experiments in the real atmosphere the available gases and particles are measured and the physical and chemical properties as well as meteorological parameters are characterised. A variety of platforms such as ground based and airborne techniques are employed.**
+ **Simulation and laboratory experiments: Single process sequences or elementary steps are comprehended and quantified in controlled experiments. The results are process parameters, which form the basis for the development of complex numerical models.**
+ **Modelling: Partial systems of the highly complex system atmosphere are described by models. This can happen on all space and time scales: In detailed form flow conditions in a single street canyon are modelled whereas on a much larger scale the worldwide physical and chemical processes occurring in the atmosphere are described. Model results are compared with the results of laboratory and field measurements and are used for their interpretation. Forecasts based on the analysis of the present actual state of the atmosphere lead to statements about possible future scenarios. The consistent combination of model results and observations by advanced inversion algorithms allow for better state and process analysis of the atmosphere.**

The natural variability of the atmosphere and climate system and the growing human influences particularly in the tropics lead to the expectation that the Earth's atmosphere will clearly change within the next decades. The most important factor influenced by human activities presumably is the change of the radiative forcing due to the increased emission of climatically active trace substances. The atmospheric content of halogen containing compounds, however, is declining. It will nevertheless remain on a high level and influence the stratospheric ozone concentration through the next decades. The emissions of nitric oxides and other pollutants also will change. While a certain tendency to smaller emissions is expected in the western industrial nations, similar reductions cannot be expected for Eastern Europe and especially not for the developing countries. Apart from such ground-based emissions air traffic is expected to drastically increase leading to considerably increased trace gas injections in the upper troposphere and in the lower stratosphere region. Increased NO_x emissions in high altitudes may influence ozone formation and destruction chemistry and may lead to a changed tropospheric ozone distribution pattern, which in turn will influence the self-cleaning capability of the troposphere.

Only recently the significant climatic effect of aerosols of anthropogenic origin has been recognised. These effects are expected to be highly variable due to considerable regional variations and a huge variety of contributing sources and processes. The geographic change of the main emission areas and the strong qualitative changes of aerosol characteristics due to the transition of traditional industrial areas to highly commutative post-industrial societies aggravate the forecast of trends and variability with regards to the atmospheric aerosol and its effects. The strong coupling between aerosols and clouds joins in as another complicating factor.

The development and use of suitable instrumentation for the characterisation of the actual state of the atmosphere has been taken into account in AFO 2000. Especially the height area between the planetary boundary layer and the lower stratosphere has

been included by more exact and full-coverage measurements of increased quality. Field measurements have contributed to in situ as well as remote sensing methods and satellite measurements have contributed on the global scale. The quality and availability of satellite measurements have become increasingly better and satellites have played a role for bridging the spatial scales mentioned before. The European environmental research satellite ENVISAT which was successfully inserted into orbit on March [1st], 2002 delivered first data sets, suitable for answering a variety of scientific questions. Additionally, other carriers of measurement instrumentation for the recording of atmospheric parameters, such as commercial aircrafts and balloons were used. Longer-term exact measurements are helpful for the detection of trends of atmospheric parameters.

By comparison of results of numerical models with measured (atmospheric) quantities the analysis of processes is driven forward and process understanding is improved. This applies to boundary layer processes, for convection and cloud processes in the troposphere as well as for chemical and microphysical processes in the stratosphere. The numerical models also serve the interpretation of the measuring data. Reference measurement stations have been installed, for which a standard has been set concerning quantity, temporal resolution and quality of the data (e.g. energy and substance fluxes). These will be used for the validation of satellite supported measurement methods as well as results at lattice points of high-resolution numerical models.

The research programme AFO 2000 integrated a large number of activities on atmospheric research in all compartments of the atmosphere. The physical and chemical phenomena which are of importance for a better system understanding were treated from the atmospheric boundary layer close to surface, in the free and upper troposphere, the lower and middle stratosphere up to the middle atmosphere (mesosphere). The dynamic exchange between the single compartments of the atmosphere was the

topic of various AFO 2000 research clusters avoiding a demarcation of single regions of the atmosphere.

Not only qualitative expansions of the existing scientific state of knowledge but also scientific first-rate contributions to the better understanding of the system atmosphere and to a quantitative description were obtained in the AFO 2000 programme. AFO 2000 operated at the top level of scientific research, pushed forward the boundaries of the scientific understanding and hence laid the foundations of political, perhaps far-reaching, measures. Research in atmospheric science and especially within AFO 2000 has moved along the border of what was technically possible involving the best available technologies. Hence, the AFO 2000 atmospheric research programme represented an innovative network core of first rank within the German science landscape.

Within the international research landscape, AFO 2000 moved along the front line of the atmospheric scientific research and considerably fostered scientific knowledge. The programme was one of the strongest activities in Europe in its area and was, by its participants, closely connected with the research supporting activities of the European Union within its fifth and sixth research framework. Atmospheric scientific research represented an important area of environmental and Earth system research in all western, highly industrialised countries. The programme intended to cooperate with such similar efforts not only throughout Europe but also with countries such as the USA or Japan. The AFO 2000 programme profited from active collaborations of its participants with international partners in the frameworks of international programmes such as IGBP, IGAC, or WCRP for the joint progress of better scientific understanding of global environmental problems.

The structure of AFO 2000 as an integrated programme has led to strong interdisciplinary activities in the programme itself, in its thematic sections and in the networking of the single research activities. A very narrow technical delimitation of the BMBF programme

promoting atmospheric scientific research was avoided. To be able to plan adequate precautionary measures, carry out and judge the success of measures based on extended scientific knowledge, the BMBF has followed three priority research political aims, i.e.:

+ **to improve the understanding of the complex system atmosphere,**
+ **to develop and provide scientifically sound foundations for environmental policy, and**
+ **to support the development of young scientists in the field of the atmospheric research.**

To reach these aims, the BMBF supported 26 research clusters, 5 groups of young scientists and 11 individual projects with about 43 Million EURO within AFO 2000. AFO 2000 was closely coordinated to the German climate research programme DEKLIM which focuses more on the reasons and effects of long term atmospheric changes.

The contributing scientific working groups within AFO 2000 were structured in four topic groups. Every group elected a speaker and a deputy from the participating scientists. The four theme groups of the AFO 2000 programme are:

(1)   Surface-Atmosphere Interactions
The studies in this area contained the analysis of energy and substance circulations in the atmosphere with the Earth's surface as a source or sink. A main topic of the supported research covered investigations of vertical exchange to better understand the impact of transport processes on the chemistry and dynamics of the atmosphere. An improved description and a better understanding of the role of forests as sources of organic compounds, their biochemical formation mechanisms, their main atmospheric transformations and an assessment of the role of the most important oxidants near the surface in forest areas were obtained.

(2)   Chemistry, Dynamics, Radiation and their Interaction
In this area studies of interaction processes of the different atmospheric layers mesosphere, stratosphere and troposphere as well as of the mutual influence of dynamical, physical and chemical processes were carried out. The studies also covered open questions with regards to the scientific understanding of the ozone loss over the Antarctic and the Arctic and continue the successful German ozone research programme. Transport processes and their interaction with chemical transformations were treated in detail by models of the latest generation.

(3)   Multiphase Processes
Studies on aerosols (i.e. particles of liquid and solid phase), cloud systems, and polar stratosphere clouds were carried out here. Ground based cloud field experiments were taken together with laboratory examinations and extensive modelling to better describe aerosol modifications due to cloud chemistry. Moreover, the role of the aerosol released from biomass burning and the role of stratospheric particles in stratospheric ozone loss were further elucidated. The degree of the understanding of the tropospheric and stratospheric multiphase system by the direct comparison of field measurements and model results was significantly improved.

(4)   Atmospheric System Analysis: Models and Data
The summarising analysis of observation data (e.g. of satellites) and of results of complex numeric model calculations was carried out in this thematic group. The technique of data assimilation, that is the consistent connection of measurements with model results, played a central role in many projects. Due to new data sources, improved numeric techniques and increased computer capacities a variety of new insights into the complex events in both the stratosphere and the troposphere was obtained.

Structure of the contributions

Results of Theme Group 1.1

Surface-Atmosphere Interactions: Reactive trace compounds in the lower troposphere. From process understanding to prognosis.

Regional biogenic emissions of reactive volatile organic compounds (BVOC) from forests: Process studies, modelling and validation experiments (BEWA2000)

Rainer Steinbrecher and BEWA2000 Partners[1]

Motivation

Current directives in environmental policy for the mitigation of photo-smog episodes appear to be not as effective as they should be. For example, in Europe, 68 % of air quality control station exceeded the 180 µg ozone per m^3 threshold for over 3 hours during summer 2003 (press release European Environmental Agency, October 2003). Surface ozone is formed from the main precursors nitrogen oxides and volatile organic compounds VOC. Through diverse degradation pathways they lead also to the formation of hazardous and climate relevant particles (HOFFMANN et al. 1997). Despite this great importance of VOC for the overall system of the atmosphere, our knowledge on the spatial and temporal behaviour of VOC emissions as well as of the mix of single emitted compounds is associated with great uncertainties (SIMPSON et al., 1999). Volatile organic compounds are emitted into the atmosphere by biogenic (e.g. by plants) and anthropogenic processes (e.g. car traffic and use of organic solvents). Two important aspects have to be considered when assessing the contribution of different VOC sources, in particular biogenic VOC, to regional photo-smog episodes:

+ **The biogenic contribution to the overall emission is strongly influenced by the seasonal fluctuation which is on one hand related to the physiological activity of plants and on the other hand to the strong dependence of the emission rates on temperature and light intensity. In the summer months, especially during sunny and warm high-pressure weather episodes, the estimated biogenic emission of Germany can exceed the anthropogenic VOC emission (RICHTER et al., 1998).**

+ **The emitted biogenic compounds are mostly unsaturated and partly oxygenated hydrocarbons which are highly reactive and have a higher ozone formation potential compared to most of the anthropogenic VOC (ATKINSON, 2000).**

Considering these facts, the contribution of biogenic VOC to the ozone formation in Germany is much more important as estimated from the relation of the single processes expressed in carbon per time step. As a consequence, biogenic VOC emissions are significantly contributing to the formation of photo-smog in the industrial part of the world with high NO_x loads. In spite of their important role in air pollution episodes, the knowledge about BVOC emission processes on leaf, canopy and landscape scale as well as knowledge of their fate in the atmosphere is rather

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[1] see Annex 1 of this contribution

vague. Integrative studies on the underlying biological, chemical and atmospheric processes that control sources and sinks of BVOC in the land surface layer are needed to improve prognostic surface exchange models for primary and secondary VOC.

Objectives

The overall objective of BEWA2000 is to significantly reduce the knowledge gaps related to BVOC and particle exchange in forested areas by focussing on:

+ **Canopy flux model development by implementing improved emission and chemical degrada-tion modules as well as improving landscape BVOC emission models.**
+ **Laboratory studies to determine (1) the relevant processes leading to the formation of VOC in plants, and (2) key oxidation processes of BVOC resulting in formation of particles.**
+ **Validation of the coupled model with field data.**
+ **Construction of validated regional emission inventories with high temporal and spatial resolution.**

This integrated approach is sketched in Figure 1. Nine institutions and more than 50 researches with

Figure 1: Overview of experiments and products executed and delivered in BEWA2000.
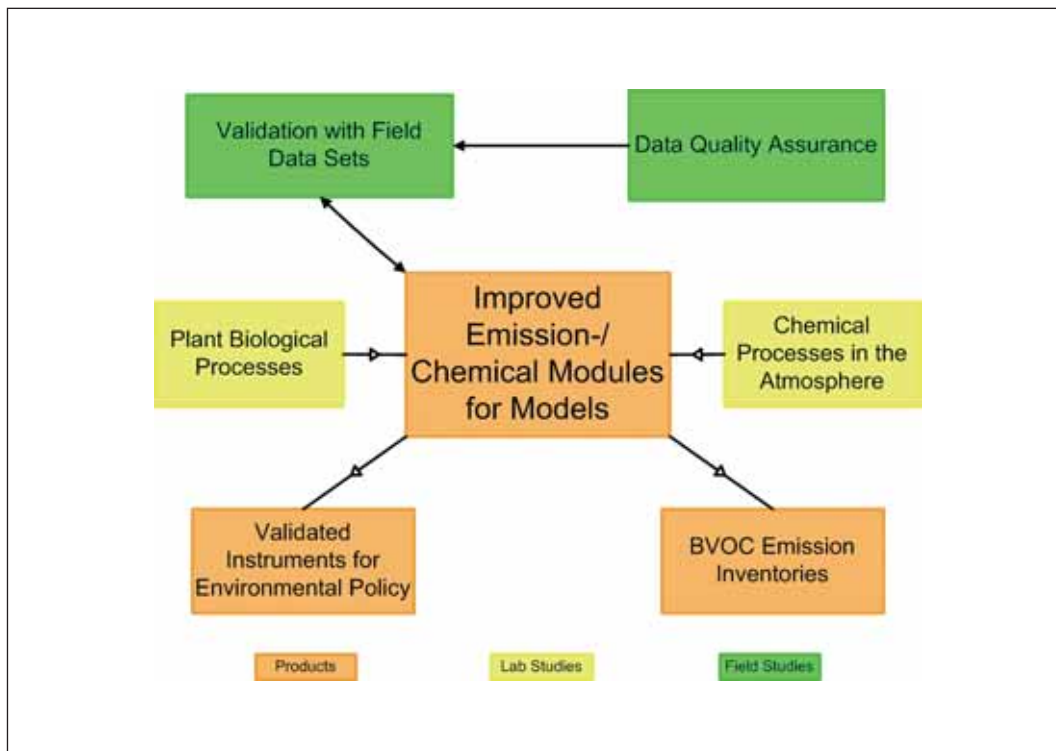
expertise in biology, chemistry, physics as well as meteorology joined together under the umbrella of BEWA2000 working on the common objectives (Figure 2). The tasks were focused in 17 projects (see Annex 1).

Experiments and Models

At the Norway spruce field-site Waldstein, Fichtelgebirge (50°08'32''N, 11°52'04''E, 775 m a.s.l.) of the Bayreuth Institute of Terrestrial Ecosystem Research (BITÖK), meteorological and air quality parameters (ozone, NO_x, SO_2), soil and leaf primary emission as well as the canopy exchange (net emission and net deposition) of gases (isoprenoids, carbonyls, peroxides, ozone) and particles were quantified using latest eddy covariance and analytical techniques (Klemm et al., 2004, Steinbrecher et al., 2004). Local and regional effects including advection on atmospheric trace gas compositions were studied. The

Figure 2: BEWA2000 partner institutions.

Waldstein data were used to validate and improve models on gas and particle exchange between the vegetation and the planetary boundary layer. The 1 dimensional canopy-chemistry-emission model CACHE (Forkel et al., 1999) considers new emission factors for isoprenoids and carbonyls as well as complex degradation pathways provided by an updated chemical mechanism from the ValCHEM group (Validation of CHEmical Mechanisms of the degradation of isoprene and α-pinene to be used in 3 dimensional chemistry-transport-models; Geiger et al., 2003). In smog chamber studies the processes were identified that lead to secondary organic aerosols (SOA) from limonene and α-pinene oxidation involving the NO_3-radical (Spittler, 2001). Furthermore, the impact of the acidity of the seed aerosol on the SOA yield during the oxidation of α-pinene was quantified (Iinuma et al., 2004). Biochemical studies on the formation of isoprenoids and carbonyls were used for improving and extending a process based isoprene emission model (Zimmer et al., 2000, Lehning et al., 2001). The stable isotope technique was applied to study the contribution of different carbon sources (in situ photosynthesis vs. stored carbon) to the biosynthesis of isoprenoids (Schnitzler et al., 2004). Regional scale BVOC emission estimates based on a semi-empirical approach (Guenther 1997, Steinbrecher et al. 1999) were significantly improved (Smiatek, 2004) including light extinction through the canopy, assessment of the leaf temperature, and seasonality of the emission factors as well as new emission factors, e.g. for beech calculated from data provided from the ECHO team (Emission and CHemical transformation of biogenic volatile Organic compounds). More detailed information on BEWA2000 is presented on the project web-page (http://imk-ifu.fzk.de/bewa2000). In the next month more than 15 papers will be published in peer reviewed journals highlighting the BEWA2000 research.
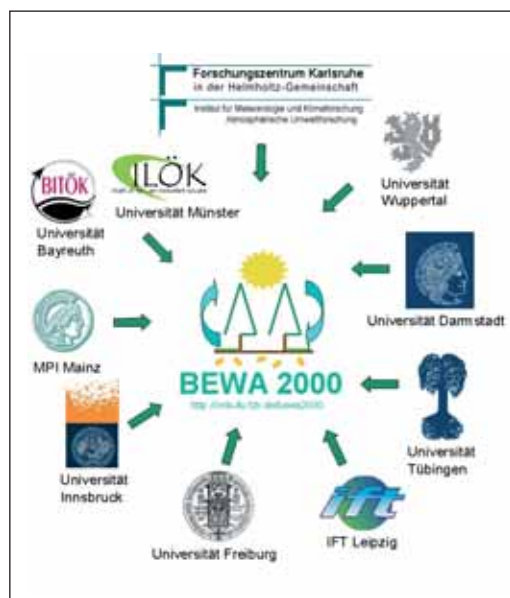
Biological Processes and VOC Production

In situ photosynthesis is one major carbon source for isoprenoid synthesis in plants. It was demonstrated that the flux of photosynthetic intermediates through the plastidic isoprenoid pathway is controlling short-term variability of the isoprene emission. The light dependence of isoprene emission is mainly due to a light-induced increase of flux of photosynthetic intermediates through the plastidic isoprenoid pathway, leading to enhanced chloroplastidic dimethylallyl diphosphate (DMADP) levels which are sufficient to saturate the isoprene synthase (ISPS) activity. Stopping photosynthetic processes by switching of the light (Figure 3) led to a rapid decline of isoprene emission rate which was accompanied by a decline of leaf DMADP levels. Further evidence that the flux of intermediates in the plastidic isoprenoid biosynthesis

pathway is the main short term control variable is provided by experiments on the temperature dependences. These experiments demonstrated that leaf DMADP levels increased as long as net assimilation rates increased. Beyond the photosynthetic optimum at approx. 30 °C the leaf DMADP pool became depleted due to a further increase of in vivo ISPS activity which peaks at approx. 45 °C. As consequence, isoprene emission rates dropped to almost zero at approx. 40 °C when DMADP leaf content reached night levels. Under CO_2 limiting conditions isoprenoid biosynthesis may serve as an alternative sink for primary products of net-photosynthesis, reductive (NADPH) as well as energy equivalents (ATP).

Just recently fixed carbon is not the only carbon (C) source used in isoprenoid synthesis as proven by exposing poplar leaves to a $^{13}CO_2$ atmosphere. Only about 75% of isoprene became 13C-labeled within minutes (Figure 4). In studies using ^{13}C-labelled compounds, it was tested if carbohydrates transported in the xylem and leaf internal C pools such as starch may act as C sources for isoprene synthesis. As shown by feeding poplar and oak leaves with [^{13}C]glucose, up to 10% of the C incorporated into isoprene emitted may be derived from xylem transported carbohydrates (KREUZWIESER et al., 2002). By depleting the internal C-pools of poplar leaves and refilling them with

Figure 3: Rapid decline of leaf DMADP levels after turn off the light from poplar leaves (Populus x canescens). (A) leaf DMADP content after onset of darkening (B) DMADP ratio dark to light and isoprene emission rate (n=8 ± SE).

Figure 4: Effect of $^{13}CO_2$ exposure on ^{13}C-incorporation into isoprene emitted by poplar leaves.
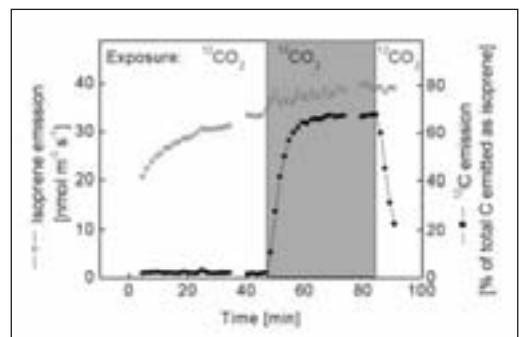
[13]C-labeled C (exposure to [13]CO_2) it was demonstrated that about 30% of isoprene carbon became ^{13}C labelled. This result clearly indicates that other C-sources, e.g. starch next to xylem transported carbon and in situ photosynthesis were used for isoprene formation, predominantly when net CO_2 assimilation was reduced due to closure of the stomata. The results suggest a dynamic exchange of carbon between different cellular precursors for isoprene biosynthesis as well as an increasing importance of these carbon pools under conditions of limited photosynthesis.

Reaction Chamber Studies

Two measurement campaigns were performed utilising the European photo-reactor EUPHORE in Valencia. Here, the reactions of α-pinene and limonene with NO_3 radicals were analyzed under conditions close to the atmosphere with respect to their oxidation mechanisms and aerosol formation (Figure 5). Additive experiments were performed in laboratory photo-reactors. Experiments were conducted in the absence and presence of both inorganic and organic seed aerosols. The observed formation of secondary organic aerosol was analyzed using a model aerosol which consists of three compounds identified as gas-phase products (two carbonyl compounds and one organic nitrate), and which allowed a physical characterization (surface tension, refractive index) of the SOA. Pinonaldehyde and endolim were identified as the major reaction products of NO_3 + α-pinene and limonene oxidation, together with large amounts of yet unidentified organic nitrates. But the reported compound α-pinene oxide could not be identified. Reaction mechanisms for NO_3 + limonene and α-pinene have been proposed. The work suggests that

Figure 5: Relationship between the product yields (estimated for nitrates) and SOA mass yields.
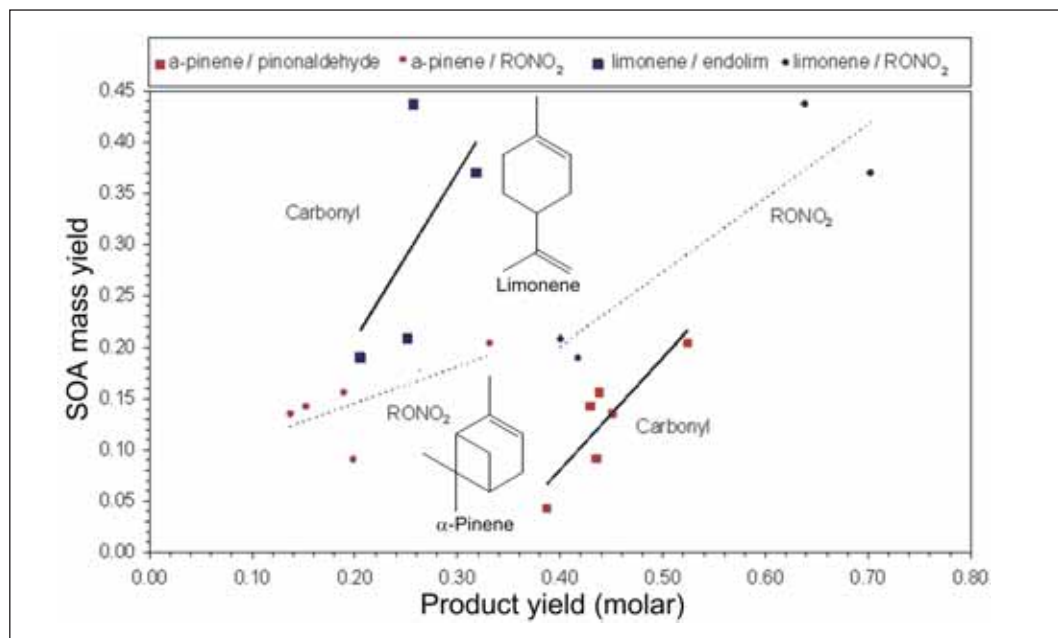
the total SOA mass observed from NO_3 + limonene originates mainly through the secondary chemistry of endolim, whereas for the NO_3 + α-pinene reaction pinonaldehyde has a small SOA mass yield, and so other compounds will have more impact on the total particle mass in α-pinene experiments.

An effect of aerosol acidity on SOA formation in the reaction cascade α-pinene/ozone/seed-aerosol is clearly observable resulting in higher SOA yields (IINUMA et al., 2003). Organic compounds with a molecular weight over 300 (possible dimers and oligomers) were identified on the sulphuric acid seed particles. The work suggests that ozonolysis of

monoterpenes will probably be the most important SOA source in forest canopies.

The "Waldstein" Field Experiments

During two summer field campaigns in 2001 and 2002, biosphere/atmosphere exchange of energy, gases, and particles were quantified in a Norway spruce forest in NE Bavaria in order to study the impact of the emissions of BVOC on chemical and physical processes in the atmosphere. A rigorous quality assurance/quality control plan was implemented that included also the 3D sonic anemometer

Figure 6: Five-day trajectories for the "Waldstein" site, Fichtelgebirge, 29.07.2002, 12:00 GMT.
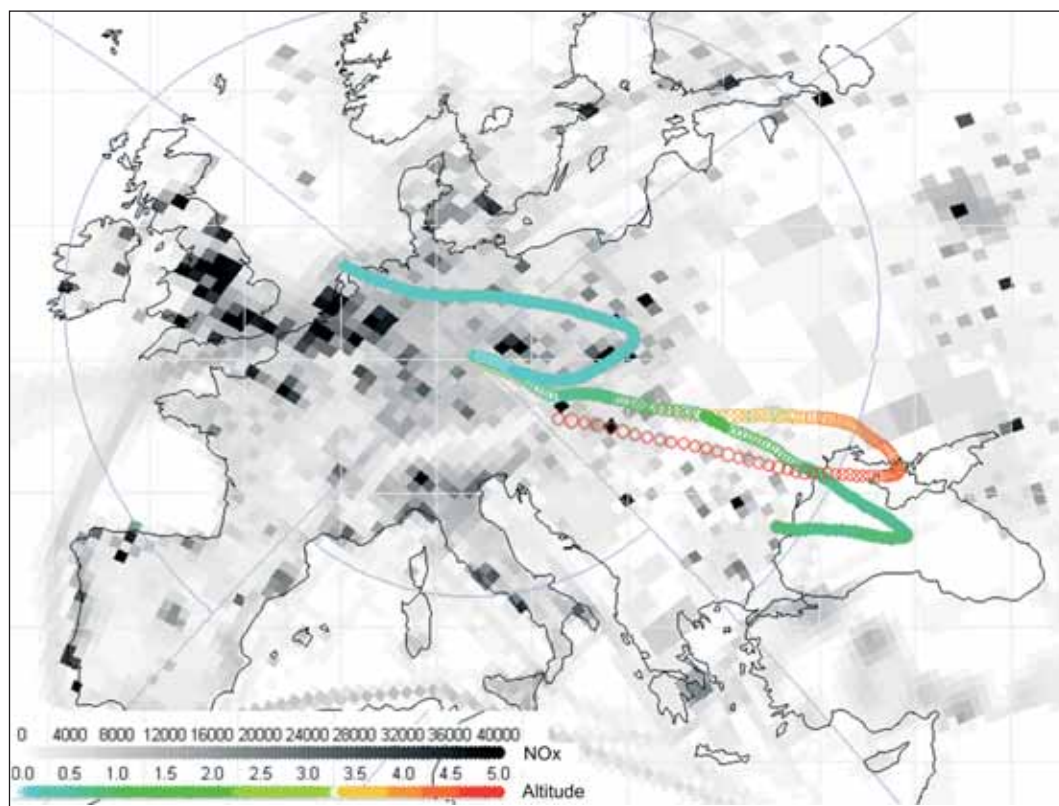
measurements. It could be demonstrated that all
instruments tested met the stated data quality objec-
tives. Periods under typical summertime conditions,
and representing various wind directions, were
selected for joint data analysis. Advection processes
were studied by a trajectory analysis (Figure 6). It was
shown that during the experiment clean as well as
polluted air masses arrived at the site.

Exchange rates were strongly correlated with
temperature and ambient carbonyl mixing ratios
(COJOCARIU et al., 2004). Compensation points of 6
ppb (acetaldehyde), 1.2 ppb (formaldehyde) and 5 ppb
(acetone) were determined (Figure 7). Acetaldehyde
exchange additionally correlated with mass flow of
ethanol in the xylem and ethanol emission support-
ing the idea that acetaldehyde emission results from
oxidation of xylem transported ethanol. Other path-
ways of acetaldehyde production such as during
light-dark transitions do not seem to be important in
spruce and other plant species (GRAUS et al., 2004).

Two algorithms developed by GUENTHER (1997)
considering (1) light and temperature as emission
controlling factors and (2) an algorithm describing
the exponential temperature dependence of emis-
sions were used to predict potential VOC emission

rates from Norway spruce twigs. For this experiment,
isoprene and acetaldehyde emission patterns were
best predicted by the Guenther algorithm (1), and the
exponential algorithm (2) described very well the
observed variation for the sum of monoterpenes
emitted as well as acetone and ethanol emissions.

Diurnal patterns in canopy fluxes resulted from
the dynamics of the boundary layer, from regional
atmospheric processes (for example production of O_3
in the atmosphere), primary emission and deposition.
Horizontal advection of air masses into the trunk
space occurred sometimes due to the patchiness of
the forest. Isoprene as well as monoterpene canopy
fluxes reached about 3 nmol m^{-2} s^{-1} on warm and
sunny days (STEINBRECHER et al., 2004). The results of
the peroxid measurements revealed mostly a deposi-
tion of about 1 nmol m^{-2} s^{-1} for H_2O_2 as well as for
hydroxyl-methyl-hydro-peroxide (HMHP) (Figure 8).
Acetone, acetaldehyde, methyl-ethyl-ketone and
formaldehyde were the most important compounds
in the ambient air in and above the Norway spruce
canopy. Other carbonyls such as pinonaldehyde,
methacrolein - both are oxidation products of
α-pinene and isoprene respectively - and trans-2-
hexenal (from grass cutting). For pinonaldehyde the

*Figure 7: Influence of ambient formaldehyde (A) and acetone
mixing ratios (B) on their exchange between spruce and the
atmosphere. Results from experiments with 5 trees each are
shown. CP: compensation point.*
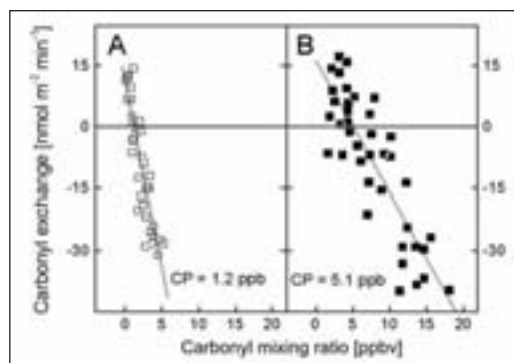
*Figure 8: H_2O_2 fluxes (A) and H_2O_2 mixing ratios (B). Hourly
(open symbols) and daily averages (closed symbols) in July and
August 2001 at the "Waldstein" site, Fichtelgebirge.*
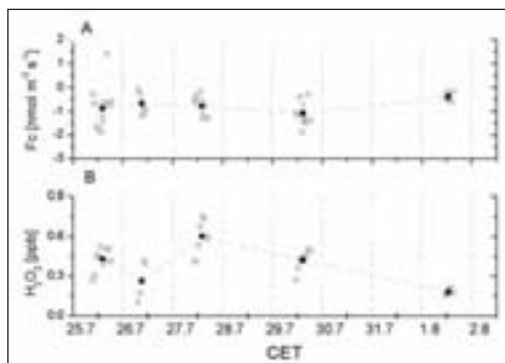
concentration maximum was observed in the second half of the night and very early morning. Other secondary biogenic compounds such as methyl-vinyl-ketone, glyoxal and methyl-glyoxal, were detected only in traces. A comparison of carbonyl data with back trajectories gave no evidence for advection from anthropogenic sources in the near environment.

Acetone, acetaldehyde and methyl-ethyl-ketone reached highest levels around noon at sunny days (DNPH-method). But using the PTR-MS for analysing carbonyls in ambient air, mixing ratios surprisingly were lowest at noon. During night and overcast days carbonyl mixing ratios resulting from PTR-MS and DNPH measurements agreed reasonable. The observed differences in the responses of the two methods measuring ambient carbonyl mixing ratios at a similar location under varying environmental conditions urgently needs further investigation and lead to the design of a laboratory intercomparison experiment. However, no differences were detected between the tow methods used for quantifying carbonyls in spiked ambient air. Thus the sometimes

observed large differences in carbonyl mixing ratio in the ambient air of a Norway spruce canopy using the DNPH and PTR-MS method, respectively, urgently needs clarification.

On airborne particles in the coniferous forest primary hydrocarbons as well as the inorganic ions chloride, sulphate, nitrate, ammonium and the organic (OC) and elemental carbon (EC) have been determined (PLEWKA et al., 2003). For alkanes no differences between day and night concentrations on particles from below and above the canopy was observed. Many terpene oxidation products were quantified but they account only for a small part of particle mass as a result of their high vapour pressure and possible photochemical degradation on the particle. Pinic-acid concentrations were higher on particles from above the canopy, probably due to photooxidation of precursors, e.g. resulting from the reaction of α-pinene with ozone. The isoprene oxidation products 2 methylthreitol and 2-methyl-erythritol, firstly detected by CLAEYS et al. (2004) were also quantified (Figure 9).

Figure 9: Formation of condensable products from the isoprene oxidation in ambient air. Filter sample PM 2.5, 28.-30.07.2002, " Waldstein", Fichtelgebirge.
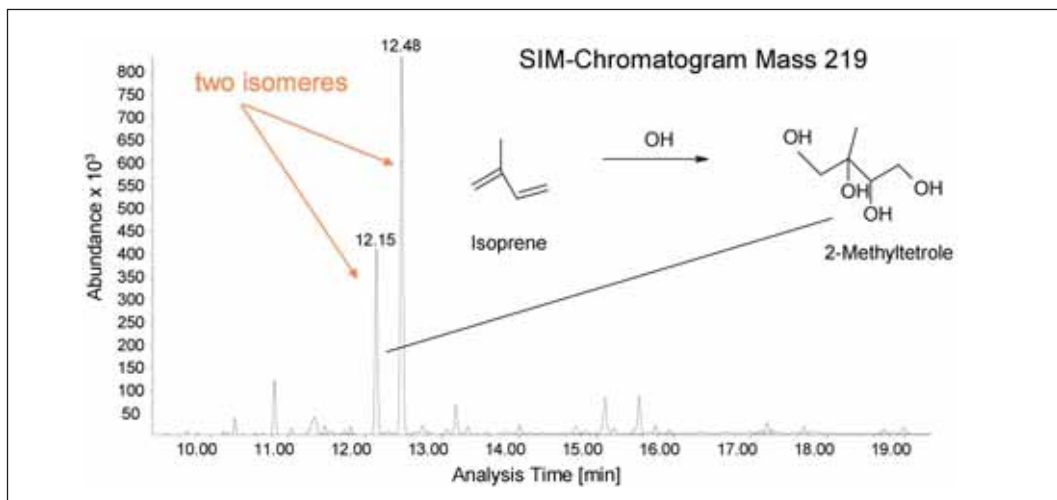
The average daytime concentrations of the isoprene oxidation products were higher than the night-time concentrations being in the same range as the terpene-acids. The concentrations of dicarboxylic-acids on the particles were decreasing with the number of C-atoms and were independent on sampling altitude and daytime. Some sugar compounds could be also quantified as particle components. Mass closure including OM (converted from OC by a factor of 2.1) and water (estimated by a growth factor determined in other experiments) was achieved in the range of ±20%.

On many days, the turbulent particle flux showed a diurnal pattern with almost no particle exchange

at night and strong fluxes during daytime (Figure 10). Particle deposition during daytime clearly dominated over emission with the strongest deposition fluxes in the order of 10^8 particles $m^{-2} s^{-1}$ occurring during particle formation events. For these events, an evolution of the particle size distribution is characteristic (Figure 10). Growth rates of the newly formed particles were quantified ranging from 2 to 6 nm h^{-1}. Calculations indicate a considerable contribution of BVOC oxidation products to the observed condensational growth.

Figure 10: Particle flux (lower panel) and particle size distribution (upper panel) on August 02, 2001.
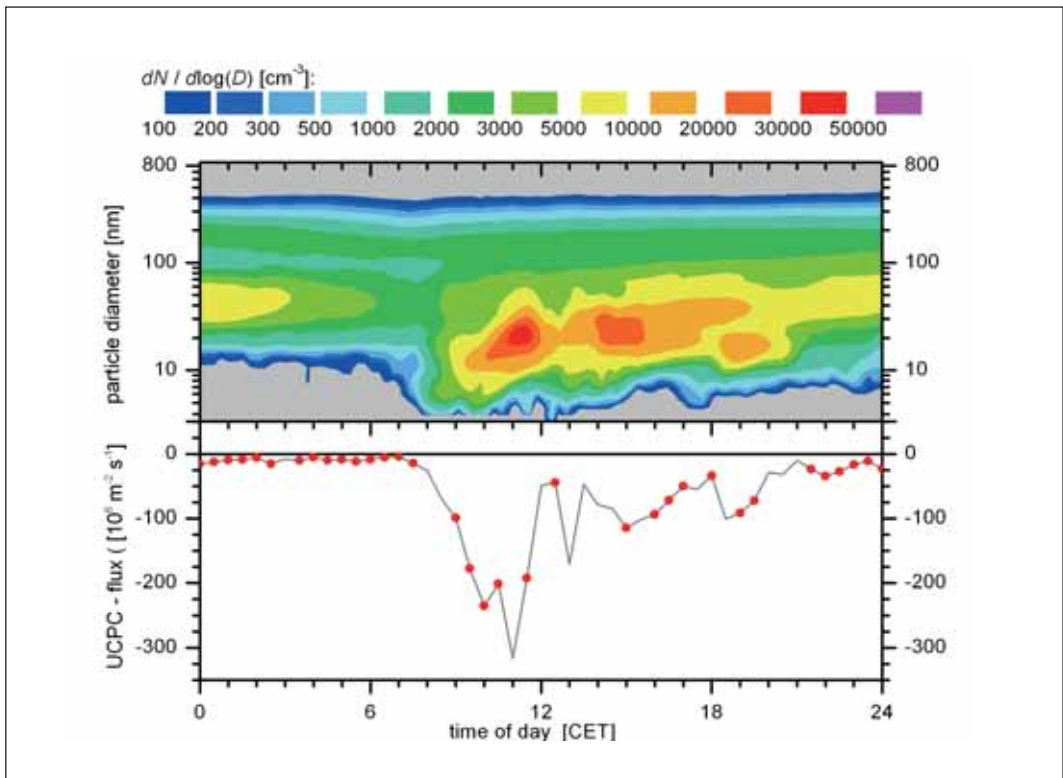
Model Development and Integration

The numerical leaf-level-isoprenoid-emission model has been continuously updated based on results of the biochemical studies on isoprenoid synthesis within BEWA2000. The structure of the model is shown in (Figure 11).

For model integration a new photosynthetic sub-module was developed. Employing data on the time courses of light, leaf temperature and air humidity as input, the model predicts leaf conductance, intracellular CO_2 concentration (p_i) and the rate of CO_2 fixation using physico-chemical principles. A biochemical sub-model is used to calculate (among others) the concentrations of the "isoprene precursors" triose-phosphates, ATP and NADPH. Validation of the model using data on *Quercus robur* and poplar (*Populus spec.*) indicate that photosynthesis and the levels of the isoprene precursors are reliably predicted. The photosynthesis model comprises a CO_2 diffusion module and a CO_2 fixation module which

are linked by the p_i-pool (NOE, 2004). The precursors for isoprenoid formation in the plant are further processed in an isoprenoid module. Phenological, light and temperature data are used to determine enzyme activities used for instantaneous emission rate calculations. The model has been parameterised for Hybrid poplar (*Populus alba x tremula*) and the evergreen tree species Holm oak (*Quercus ilex*) so far. The model performance has been evaluated with independent data indicating that photosynthesis and the levels of monoterpene emission are reliably predicted (Figure 12). This model has been successfully implemented into the regional potential emission model for specific studies on oak stands.

The application of CACHE to the Waldstein site (Norway spruce) showed good agreement between

Figure 12: Daily variation of simulated (solid line) and measured net assimilation (A) and total monoterpene emission rates (B) of Holm oak (Quercus ilex L.) leaves, measured in 2003 at the Wank experimental station, Garmisch-Partenkirchen.

Figure 11: Overview of model components of the process-based isoprenoid emission model. GAP: glycrin aldehyd phosphate; PGA: phospho glycerate; DXP: deoxy-D-xylulose phospate; IDP: isopentenyl diphosphate; DMADP: dimethylallyl diphosphate; GDP: geranyl diphosphate; GGDP: geranyl geranyl diphosphate; NADPH: nicotinamid dinucleotid phosphate; MEP: methyl erythritol cyclodiphosphate.

measurements and simulations for temperature, ozone, BVOC fluxes and BVOC ambient air mixing ratios as well as for secondary oxygenated compounds (Figure 13). For example the observed and simulated local maxima of the monoterpene and isoprene mixing ratios matched quite well in the morning and evening. Also the observed drop in the methacrolein mixing ratios during daytime when the chemical activity was at maximum was reproduced. The loss of BVOC within the canopy due to chemical degradation ranged between 10 to 15% of the total BVOC emitted around noon. Model results showed that BVOC degradation by the NO_3-radical was likely to proceed also during daytime in the shaded lower part of the canopy. Furthermore, the model simulations indicated that there are still gaps in our knowledge regarding the deposition processes of carbonyls in the forest canopy as well as in the temporal emission pattern of oxygenated VOC.

Figure 13: Measured and modelled (CACHE) mixing ratios of primary emitted BVOC and oxidation products for the Norway spruce site "Waldstein", Fichtelgebirge.
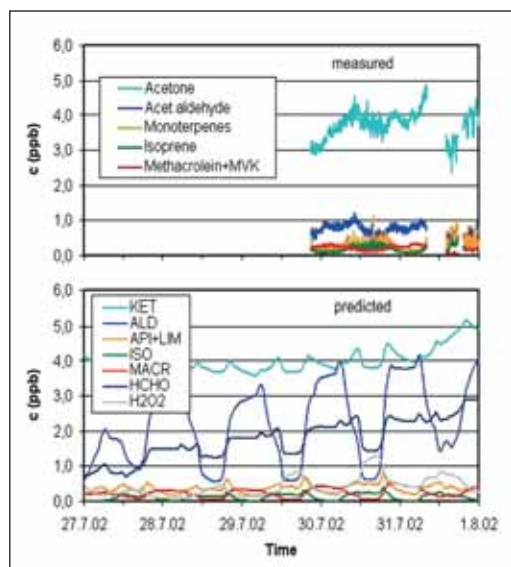
Regional Potential BVOC Emission Estimates

The new regional potential BVOC emission model has been employed to calculate hourly emission fluxes for each vegetation period (April - October) in Germany (10 km by 10 km grid) for the years 1994 to 2003. The yearly averaged VOC emission from forests amounted to 366 kt with 40 kt isoprene, 188 kt monoterpenes and 138 kt other VOC. Norway spruce is the main emitting tree species with a contribution of 40%. Annual BVCO emission showed a variation of about ± 20% from the 10-years-mean (Figure 14). The annual mean of BVOC emission in the year 2003 with a very long, hot period in August was in the same range as BVOC emissions in the years 1994/95, showing that on an annual basis the year 2003 was not an extraordinary high BVOC emission year. Highest year-to-year changes were observed for isoprene (up to +25%) whereas changes for monoterpenes, other BVOC as well as total BVOC were not exceeding 15%. This study shows that a detailed analysis of the BVOC pattern emitted by the forests is necessary when investigating the impact of BVOC on regional air chemistry on a year-to-year basis.

Figure 14: Variations of BVOC emissions from forests in Germany for the years 1994 – 2003. The baseline is the 10-years-mean.
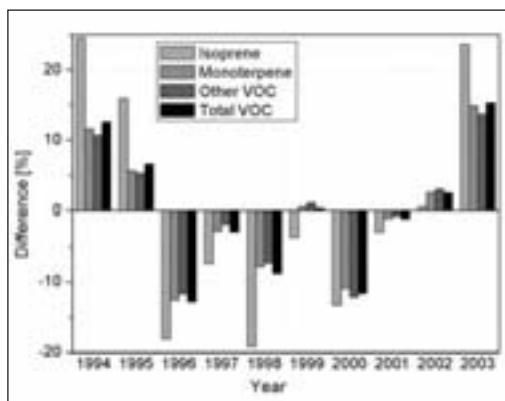
In a small scale application the BVOC emission rate for the ECHO field site (grid size: 125 m by 125 m) was estimated (Figure 15). For certain locations in the region more than 10 kg isoprene per km^2 per day were emitted in June 2002.

Figure 15: Regional potential isoprene emission for the ECHO field site in June 2002.
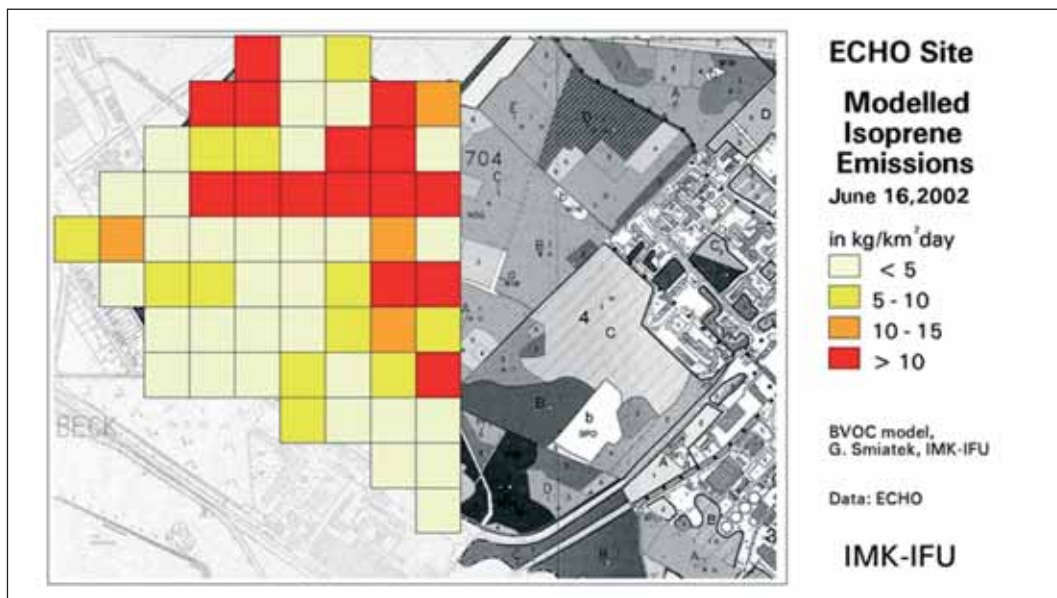
Conclusions

BVOC emissions play an important role not only in regional and global air chemistry but also in the global carbon and radiation budget that affect climate. Therefore, substantial efforts were undertaken to understand and estimate BVOC emissions from the earth surface. Despite of significant advances in the past 10 years, the comprehensive understanding of biosphere/atmosphere interactions in general, particularly the contribution of BVOC to particle exchange and the carbon cycle is still at its very beginning.

+ **BEWA2000 demonstrates that interdisciplinary field, laboratory and modelling experiments are necessary for understanding the biosphere/atmosphere exchange of reactive trace gases.**
+ **The results of BEWA2000 contribute significantly to the enhancement of our knowledge in this field.**
+ **The developed tools support environmental agencies to design more effective strategies for mitigating photo-smog episodes in the future.**
+ **However, secondary compound formation/deposition under low light conditions, eco-physio-**

logical control of leaf carbonyl emission as well as sometimes not comparable quantifications of carbonyls in ambient air by different methods require further studies.

+ **It will be a challenging task in the future to close the reactive carbon mass-balance for different forest ecosystems. This task is the prerequisite for the true assessment of carbon cycling in ecosystems.**

Global change (higher temperature, longer drought periods, altered nutrient conditions, etc.) is increasingly stressing vegetation. As a result enhanced emission rates of reactive VOC and a corresponding reduction of the carbon sink in forests is expected. The BEWA2000 results highlight the need for considering "biogenic reactive carbon" in future carbon budgets of ecosystems. Correct forest carbon budgets are a prerequisite for greenhouse gas trading in order to meet the Kyoto protocol's CO_2 reduction schedule.

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Annex 1

BEWA2000 Joint Projects:

Regional Emissions:

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Emission and chemical transformation of biogenic volatile organic compounds (ECHO) – Investigations in and above a mixed forest stand

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Scientific objectives

The objective of this project was to provide a better understanding of forest stands as a complex source of reactive trace gases into the troposphere. This was achieved by investigations in field and simulation experiments. Two field campaigns were carried out in the Stetternicher Forest on the area of the Research Center Jülich, Germany, in summer 2002 and 2003. To assure the quality of the data obtained by the different techniques three laboratory and one in-field intercomparison were carried out. As a speciality of the ECHO project, important aspects of the different processes determining the net emission from forest stands into the atmosphere were investigated in laboratory and simulation experiments. The chemical processing of the trace gas mixtures observed in the forest stand was investigated in the atmosphere simulation chamber SAPHIR. Emission and uptake of VOC by plants were investigated in plant chambers. The flow dynamics of the forest site was investigated in wind tunnel experiments. In the following, some representative results of the experiments are described. Additional information is avail-

able on the ECHO-web-page (http://www.fz-juelich.de/icg/icg-ii/echo/home). This project was closely collaborating with the AFO 2000 projects BEWA, IDEC, and VALCHEM.

Measurements of trace gases, radical concentrations, and photolysis frequencies

A large set of trace gases (VOC, CO, HCHO, HONO, O_3, NO_x, alkyl nitrates) was measured at two different towers at different heights in and above the forest stand. One important focus was the investigation of isoprene and its oxidation products methacrolein (MACR) and methyl vinyl ketone (MVK) (KOMENDA et al., 2003). Special attention was paid to the ratio of MVK/MACR which showed an unexpected behaviour that cannot be explained by photochemical processes alone. During the day isoprene reacts very fast with OH radicals resulting in a lifetime of ~ 30 minutes at OH concentrations of about 6×10^6 cm^{-3}. The expected MVK/MACR ratio would be on the order of ~ 1.4. In July 2003, this ratio varied between 0.6 to 2.1, in August 2003 between 0.6 and 1.4. During

September the MVK/MACR ratio did not exceed 1.0 (SCHAUB, PhD Thesis, in preparation). Figure 1 shows a plot of MVK/isoprene versus MACR/isoprene which can be used as a measure of the photochemical processing of the air mass. First analysis of the data shows a correlation between isoprene and its oxidation products during the day which points towards recent emissions and local photochemistry. Further investigation of the photochemistry of isoprene has been done in SAPHIR under the same conditions as

nating processes controlling the isoprene concentration in the forest quite well (Figure 2). The square of the correlation coefficient between model results and measurements (at all heights) for July, 2003, is 0.73. This means that three quarters of the variance of all isoprene measurements gained in July 2003 are in common with the model. Most of the remaining variance of isoprene is due to short time fluctuations probably caused by small scale inhomogeneities of the isoprene distribution inside and above the forest.

Figure 1: The ratio of MVK/isoprene versus MACR/isoprene based on data of July 2003 above canopy (red symbols; day, blue symbols: night). The green circle indicates the ratio for an isoprene lifetime of 30 minutes.

Figure 2: Comparison of measured and modelled isoprene mixing ratios for July 2003.
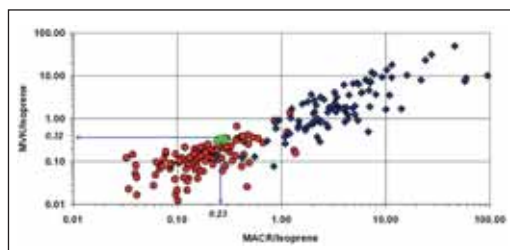
observed in the field experiment (KARL et al., 2004; Karl, PhD Thesis, 2004). From the measurements of isoprene and its oxidation products the dominant processes controlling the isoprene concentration could be extracted. With two factors describing the emission (temperature and light intensity) and two factors describing the lifetime of isoprene (OH reactivity and removal by transport) a simple model can be set up which is composed of three reactions:

precursor $\rightarrow$ isoprene   [precursor]*k=a * exp(b*(T-293)) * J(NO$_2$)^c
isoprene + OH $\rightarrow$ products   k=2.54 x10^{-11}*exp(410/T), [OH]=d*J(O^{1}D)
isoprene $\rightarrow$ k=1/e
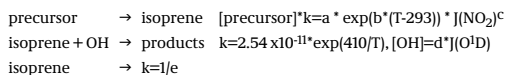
The five parameters a = 4.1 x 10^8 cm^{-3}s$^{-0.4}$, b = 0.15 K^{-1}, c = 0.6, d = 4 x 10^{11}cm^{3}s, and e = 1800 s are estimated to describe the measurements at the ECHO site. Despite its simplicity the model captures the domi-

A novel gas chromatographic system with coupled mass spectrometric and chemo-luminescence detection was set up to quantify isoprene nitrates in a field experiment for the first time. Although their concentration varied from day to day, the sum of the mixing ratios of the two identified isoprene nitrates was with 5-50 ppt on the same order of magnitude as the sum of the mixing ratio of the 4 main alkyl nitrates (methyl-, ethyl-, 2-propyl-, and 2-butyl nitrate).

Measurements with high time resolution (2 minutes) applying PTR-MS instruments operated by Agroscope-FAL and PSI were used to generate profiles of VOC in the forest canopy which are taken to characterize the temporal and spatial variation of both biogenic and anthropogenic compounds. Compounds of purely biogenic (e.g. monoterpenes) and anthropogenic origin (e.g. benzene) show comple-
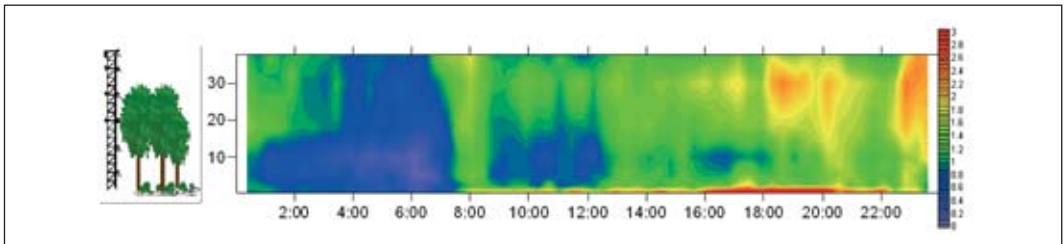
Figure 3: Contour plot of methanol concentrations (ppb) at the west tower on July 10, 2003. The contour is based on measurements at 8 heights (0.25 m , 0.5 m, 2 m, 5 m, 10 m, 18 m, 30 m, and 37 m above ground), each height was measured for 1.5 minutes, resulting in 5 complete profiles per hour.

mentary patterns, with high daytime concentrations of biogenic and high night time values of anthropogenic compounds. Figure 3 shows an interesting example for methanol.

During daytime, methanol concentrations were highest near the ground. A second accumulation of methanol can be seen in the crown region, whereas the "trunk area" in between appears like a methanol-free corridor. This picture would support emissions of methanol not only by the trees (accumulation in the crown region), but also by the vegetation close to the soil or the soil itself (AMMANN et al., 2004). PTR-MS instruments were also used in parallel to sonic anemometers to determine fluxes of biogenic VOC with eddy covariance (EC) technique (Figure 4)

(SPIRIG et al., 2004). Isoprene and monoterpene fluxes are consistent with light and temperature dependent emissions. Methanol emissions show a much lower increase during daytime, maybe an indication for "only temperature-dependent" emissions. The relative size of these fluxes is in agreement with the emissions of these VOC from the relevant tree species in the area around the tower. Biogenic VOC contribute 60 % to the total concentration of VOC, but on the order of 90 % of the photochemical turnover due to the reaction with OH radicals.

The ground-based measurements were supplemented by airborne measurements with a small research aircraft operated by MetAir under contract

Figure 4: Mean fluxes of biogenic VOC at the west tower from July 15 through July 23, 2003.
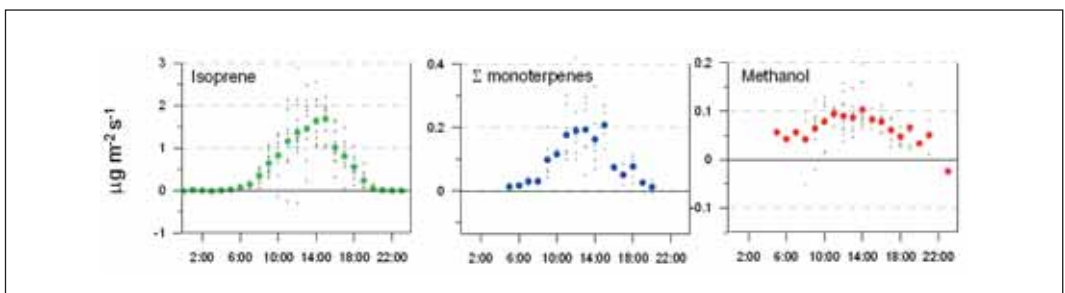
of the Research Center. As shown in Figure 5, the vertical gradient of isoprene over the canopy is significantly different from that of anthropogenic VOC, e.g. cis-pentene.

Figure 5: Vertical profiles of isoprene and cis-pentene on June 26, 2002.
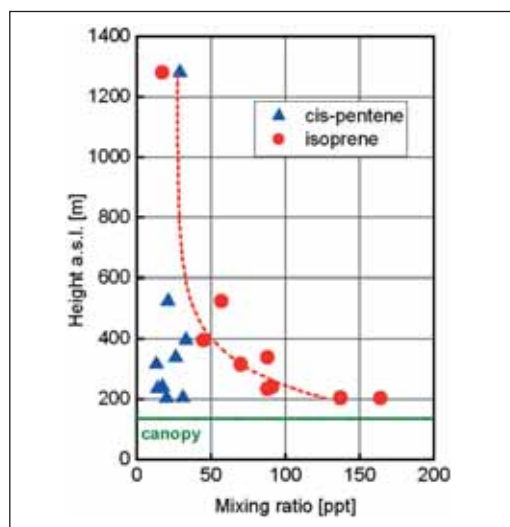
The vertical profiles of photolysis frequencies were intensively investigated. In order to quantify the photochemical activity in the forest stand, the solar actinic flux in the visible and ultraviolet was measured spectrally resolved to derive photolysis frequencies of relevant radical precursors (HONO, O_3, NO_2, HCHO, peroxides, oxygenated VOC).

Photolysis frequencies dropped to about 10% of above canopy values within the first 4-5 m below the canopy top and then gradually decreased to about 2% at ground level (Figure 6). Spectroradiometer measurements showed that the canopy acts as a grey filter in the spectral range below 500 nm. Figure 7 shows the ratios of photolysis frequencies at different heights rel-

Figure 6: Profile measurements of photolysis frequencies with a $J(NO_2)$-filter radiometer at two positions in the forest on June 23, 2003. The dotted line shows the average profile at the main tower which was located in a small clearing.
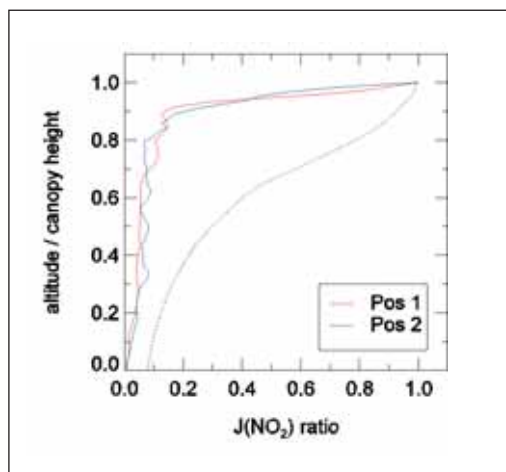
Figure 7: Ratio of daily averaged photolysis frequencies at different heights relative to above-canopy values. Circles: data of 2002, squares: data of 2003; closed symbols: cloudy days, open-filled symbols: variable cloudiness; open symbols: clear sky conditions; blue symbols: $J(O^1D)$ 19 m; black symbols: $J(O^1D)$ 4 m; red symbols: $J(NO_2)$ 4 m.
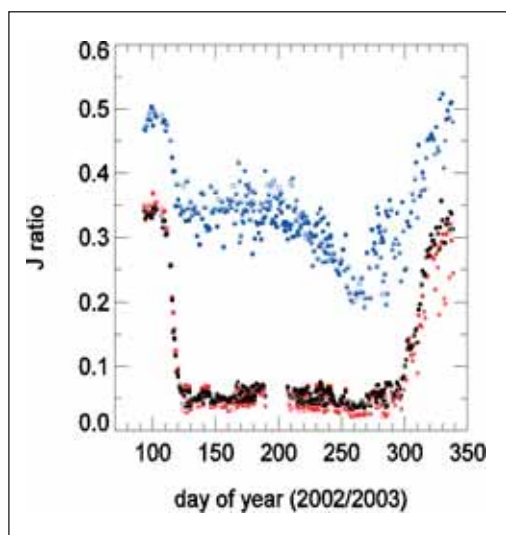
ative to above-canopy values. The drop in spring due to the leaf expansion occurs much faster than the increase in late fall due to the loss of the leaves.

For the first time, vertical distributions of HO_x radicals in and above a mixed forest stand have been measured during the campaign in summer 2003. A very compact LIF-instrument was specially developed for profile measurements on a portable platform. In Figure 8 typical daytime profiles of OH, HO_2 and $J(O^1D)$ are shown (July 14, 2003, 9:00-11:20 CET, 2-38 m, upwards), the dashed horizontal lines mark the lower and upper limit of the canopy. Frequently, significant amounts of HOx radicals were detected in the canopy and even in the trunk area. The observations show a clear correlation with $J(O^1D)$. During all night time intensives, surprisingly high concentrations of HO_2 were found even near the forest ground. Additionally, nocturnal OH concentrations around 1 x 10^6 cm^{-3} were observed above the canopy in situations characterized by high ozone and low NO_x concentrations. OH, HO_2 and RO_2 concentrations were modelled using the "master chemical mechanism" (MCM3). The calculated concentrations were in good agreement with the measurements during the

morning hours (around 11:00 CET) at high-NO_x (>2ppb). However, at low NO_x conditions the model underestimates OH by a factor 2-3 and overestimates HO_2 and RO_2 by a factor of 1.7.

Nitrous acid (HONO), a potential precursor of OH radicals, was measured with a very sensitive in-situ instrument (Long Path Absorption Photometer, LOPAP). HONO concentrations around noon were found to be more than one order of magnitude higher than expected from photostationary state considerations (Figure 9).

The measurements show clear evidence for a large, yet unexplained daytime source of HONO (~500 ppt/h), which accounts for about 35 % of OH radical production due to ongoing HONO photolysis. The evidence for a large HONO daytime source, which was postulated in previous studies for other environments, was for the first time completely constrained by measuring all parameters needed to determine the daytime budget of HONO. Thus, HONO could have an important impact on the photochemical transformation of biogenic VOC (KLEFFMANN et al., 2004).

Figure 8: Vertical distributions of OH, HO_2 and $J(O^1D)$ measured. from bottom to top. The dashed horizontal lines mark the lower and upper limit of the canopy, respectively measured on July 14, 2003, 9:00-11:20 CET between 2 and 38 m. Significant amounts of HOx radicals were detected in and even below the canopy.

Figure 9: Measured HONO (LOPAP), calculated photo stationary state of HONO (PSS), and ratio between measured and calculated values (LOPAP/PSS) for a one hour period above the canopy on July 29, 2003.
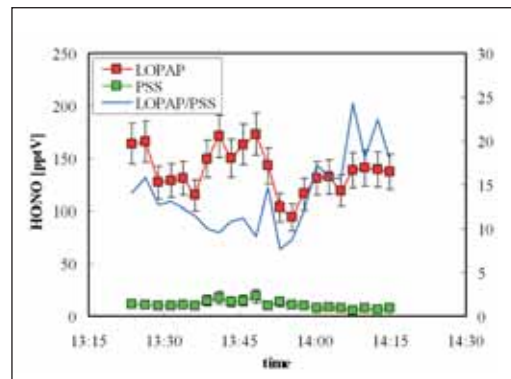
Primary emission and atmospheric fate of VOC species

During the field experiments VOC primary emissions of beech (sun and shade leaves) and oak (sun leaves) were investigated in close relation to physiological processes. Emission of isoprene, monoterpenes, as well as methanol from tree species was found to be a function of light and temperature and could be simulated by corresponding algorithms. In contrast to literature reports, substantial monoterpene emissions from beech were found (see Fig. 10). Total organic carbon emissions were shown to be a factor of 2 lower than those of the measured isoprenoids, indicating significant gaps in our understanding of the composition of the emitted VOC (DINDORF, PhD Thesis, in preparation).

Short-chain carbonyls were found to be mainly deposited, which could be confirmed by above canopy flux measurements (REA). Simultaneous profile measurements at two tower sites allowed the determination of the vertical and horizontal distribution of different VOC species. In close relation to the natural situation the impact of mildew on the release of VOC from young oaks were investigated in a plant chamber. We found a significant increase of the number of VOC species, among them several sesquiterpenes. Jasmonic acid turned out to act as an elicitor. Furthermore, the emission of several VOC (e.g. acetonitrile, isoprenoids, and methanol) as a function of stomatal aperture is under intensive investigation. First results of laboratory studies confirmed that isoprene emission from oak is not restricted by the stomata (BRACHO, Diploma Thesis, 2004).

The pronounced seasonality of the VOC emissions from beech were of special interest concerning the potential of short and long term adaptations to extreme heat periods. During 2002 and 2003 the seasonal variation of monoterpene emissions were investigated using mobile plant enclosure systems. The emission pattern did not change significantly over the year, the emission rates, however, showed a pronounced seasonal cycle with a maximum in July and August. The temperature dependence of the emissions was significantly lower in spring and fall than in summer (June, July, and August).

Figure 10: (a) Monoterpene emission (red circles) of beech (Fagus sylvatica) on a dry weight basis in correlation to the photosynthetically active radiation (PAR) on a hot summer day. Accompanying data represent time (MEST), mean leaf temperature (°C), stomatal conductance (cm/s) and net-assimilation ($\mu mol / (m^2 \, x \, min)$), each as 30 min averages. Green symbols and line indicate emissions as estimated according to the G93-Algorithm depending only on light and temperature.
(b) Methanol emission (measured by FAL, Switzerland) as observed with an enclosed oak branch (Quercus robur; red circles) in correlation with assimilation (green diamonds) and stomatal conductance (blue crosses).
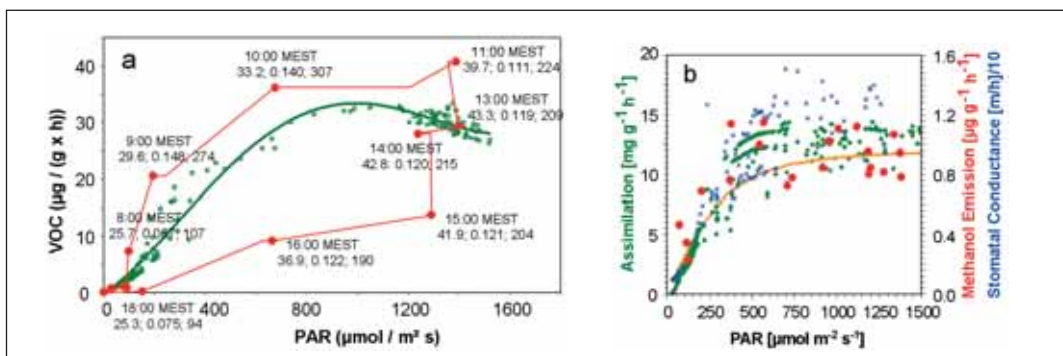
Investigations of the exchange of NO, NO$_2$, and Ozone

Applying the Membrane Tube Technique, concentration profiles (^{222}Rn, NO, CO$_2$, N$_2$O, CH$_4$) of soil air (0-12 cm) have been continuously measured during field experiments. These have been complemented by measurements of (a) NO-, NO$_2$-, O$_3$- und CO$_2$-fluxes (dynamic chambers), (b) meteorological parameters (forest floor), and (c) physical and chemical soil parameters. During the measurement period, the forest soil was characterised as a source of ^{222}Rn, CO$_2$, N$_2$O, and NO, but as a sink of CH$_4$ (LEHMANN, Diploma Thesis, 2003). Coming evaluation will focus on quantification of fluxes on the basis of measured soil profiles. In addition to these exchange studies, NO-, NO$_2$-, O$_3$-, CO$_2$- and H$_2$O-concentrations have been measured at eight levels within and above the forest canopy (see Figure 11); these measurements have been complemented by highly resolved vertical profiles of radiation, air temperature and humidity, wind speed and direction (up to 20 levels). Additional direct flux-measurements (*eddy covariance*) of NO, O$_3$, NO$_2$, and some VOCs have been performed 6 m above the canopy (external contribution to the project).

With the help of (dynamic) MPIC-cuvette systems we investigated the NO$_2$-specific exchange on sunlit and shadow beech-leaves. Following present state of data processing, a NO$_2$-compensation point could not be detected (no biogenic NO$_2$-emissions from beech).

In 2003, the investigation of structures in the turbulent exchange of energy and matter between the forest and the atmospheric boundary layer was started for the first time. Therefore, different techniques were combined: SODAR-RASS (operated by the University of Bayreuth) and tethered balloon soundings (0-600 m), as well as ultrasonic anemometry at 120 m above ground. Goal was the investigation of the spatial and temporal behaviour of coherent structures of the exchange of energy and matter, which (a) contribute substantially to the corresponding fluxes, but (b) can not be adequately quantified by classical micrometeorological techniques alone (*e.g. eddy covariance, gradient*). These data are not yet completely evaluated.

Chemical characterisation of aerosol particles

For chemical characterisation atmospheric aerosol particles were collected onto quartz fibre filters. These samples were used for the determination of the organic (OC) and elemental (EC) carbon content of the aerosol as well as the concentration of low

Figure 11: Typical diurnal evolution of vertical profiles of NO, NO$_2$ and O$_3$. These compounds drive the chemical transformation of VOCs within the canopy and thus determine the fluxes of VOCs and their oxidation products to the atmosphere. The measurements also allow to quantify the amount of soil emitted NO$_x$ finally emitted to the atmosphere.

volatile acidic oxidation products of terpenes (HOFF-MANN et al., 2002). These products add to the biogenic fraction of the secondary organic aerosol (SOA). Since the amount of SOA cannot be measured directly, the idea behind these measurements was to estimate the amount of the biogenic SOA using marker compounds. Using a capillary-HPLC-ESI-MS[n] method (WARSCHEID et al., 2002; WARNKE et al., 2003, WARNKE et al., 2004), which was continuously improved during the project, it was possible to quantify low volatile oxidation products of the most common terpenes, such as α-pinene, β-pinene, sabinene, 3-carene and limonene (see Fig. 12). The lowest detection limits obtained were around 20 pg. Since most of these analytes are not commercially available, it was necessary to run terpene oxidation experiments in the laboratory for the unambiguous identification of substances found in the field samples. At the same time new products from the α-pinene oxidation could be identified in the laboratory and in the field which have not been reported in the literature before. One of these substances is tentatively identified as an acidic ester with a molecular

mass of 232 u, the second one has a molecular mass of 204 u and probably the overall composition $C_9H_{16}O_5$, whereas the structure is not yet exactly known. The sum of all quantified products reaches concentrations from around 2 up to 15 ng/m[3] in 2002 and from 3 to 105 ng/m[3] in 2003 if only the same substances as in 2002 were taken into account. On the other hand, if all detectable terpene oxidation products were counted, the concentrations in 2003 ranged from 10 to around 130 ng/m[3]. The difference was mainly caused by the new product with a molecular mass of 204 u. Calculated mean values of total biogenic SOA derived from α-pinene, β-pinene, sabinene and 3-carene were 6 ng/m[3] in summer 2002 and 78 ng/m[3] in summer 2003. Surprisingly, the calculated biogenic SOA accounted only for 1% to the total SOA in 2002 and for 3% of the total SOA in 2003. Pinic and pinonic acid were used as markers for the SOA derived from α- and β-pinene, caric and caronic acid for 3-carene and sabinic acid for sabinene. The procedure for calculations can be found in (WARNKE, PhD Thesis, in preparation).

Figure 12: Concentrations of acidic products from terpene oxidation measured by HPLC/MS during the field campaigns in Jülich during summer 2002 and 2003. In 2003 concentrations were significantly higher than in 2002. Furthermore more analytes could be detected by improvement of the analytical method.

Wind tunnel studies

The finite forest area surrounding the Research Center Jülich was built to a scale of 1:300 and studied in the large boundary layer wind tunnel at the University of Hamburg. Using rings of metallic mesh to represent the trees, preliminary tests were carried out to find the arrangement of these rings that provides the appropriate aerodynamic characteristics of a forest (SCHATZMANN AND LEITL, 2002; AUBRUN et al., 2004a). The agreement between wind and turbulence profiles measured in the wind tunnel and in the field ensured that the physical modelling of the complex forest area was realistic. The comparison of the turbulence properties of the flow, measured inside and above the canopy, with literature data concerning dense canopies showed strong similarities for the spectral densities and integral length scale profiles. During field campaigns, profiles of VOC concentrations were measured at three different measurement towers located in the forest stand. The origin of the sampled biogenic emissions is a very important parameter for the analysis of the field data. Obtaining footprint functions in such a complex configuration is nearly impossible with footprint numerical models. Therefore, footprint experiments were performed in the wind tunnel by moving a release point source all over the forest area, at 80 % of the tree height (height of maximum emission) upwind of the measurement towers, and measuring what fraction of tracer-gas reached the measurement towers. The concentration footprint functions can be deduced from these experiments, providing information about which part of the forest area contributes to the biogenic emissions sampled at the measurements towers and what proportion the contribution is (AUBRUN et al., 2004b).

Figure 13 presents two examples of these concentration footprint functions measured at the main tower location for a sampling height of 9 m and 27 m above ground, respectively. When sampling below the forest canopy, the origin of emissions is wide and its distribution is not aligned with the mean wind direction. Under the effect of the non-uniformity of the forest (tree heights, clearings, buildings, roads), the flow structure inside the forest is very complex, leading to the transport of biogenic emissions from the south towards the main tower. On the other hand, the origin of the emissions sampled at 27 m above ground (top of the canopy) is very limited in space and fairly aligned with the wind direction, signifying that the transport of the emissions is locally driven by the above-canopy flow.

Figure 13: Concentration footprint functions for a sampling point located at the main tower at 27 m above ground (top) and at 9 m above ground (bottom). Contours represent the isopleths (10% to 100% isopleths).
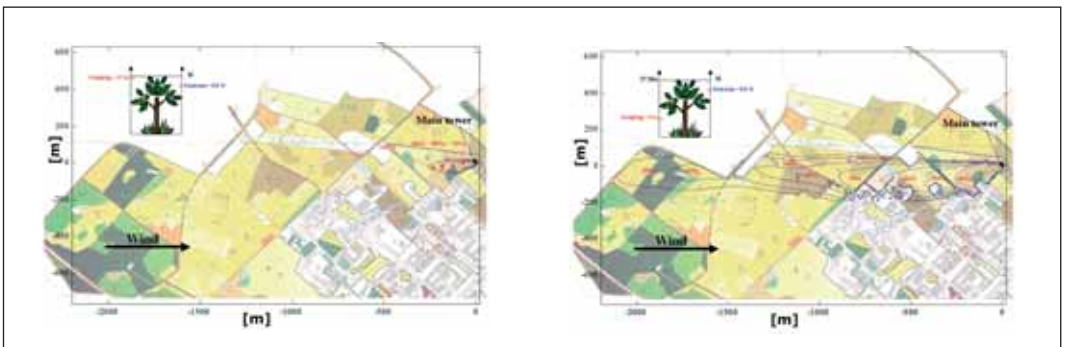
Micrometeorology

The micrometeorological measurements and data
analysis concept applied for ECHO is based on
balance equations for momentum, heat, latent heat
(water vapour) and other trace substances (e.g. CO_2,
VOC, O_3, NO_x) (BERGER et al., 2004; KRAMM et al.,
2002; KRAMM et al., 2004). To obtain all terms from
field studies vertically and horizontally resolved
measurements of mean and turbulent quantities e.g.
of wind vector components, temperature, humidity
and trace substances are performed in the forest. The
field experiments were carried out in a way that all
balance equations for the chemical parameters as
well as the meteorological parameters can be calcu-
lated and the impact of transport and turbulent

*Figure 14: Temporal variation of the specific humidity (top) and
the horizontal wind speed (bottom) measured at the main
tower between 12:00 and 18:00 CET on July 8, 2002 (DOY 189).*
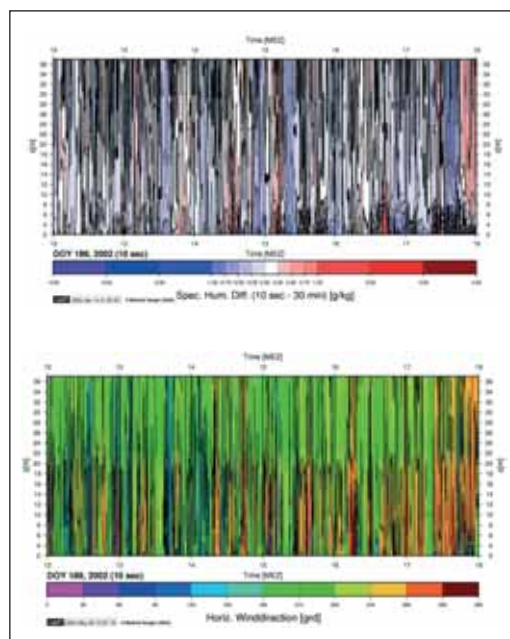
mixing can be separated from chemical transforma-
tion. The different parameters describing the ener-
getic and hydrological state of the system surface-
vegetation-atmosphere and the exchange processes
can be connected to those parameters describing the
emission of biogenic compounds like isoprene from
plants or NO from soil. An analysis of the data shows
that episodically the biogenic sources responsible for
the mixing ratios of biogenic trace gases above
canopy differ from those responsible for those in and
below canopy. This can be seen from differences in
the humidity and temperature profiles and their
dependence of the horizontal wind speed (see Figure
14). While above canopy the air is transported from
westerly direction, in canopy alternately warm dry
air masses are transported from SE to SW and cool
wet air is transported from north west from different
source regions to the field site. This indicates the
influence of a thermally induced secondary wind
system which overlays the larger scale wind system.

Emission and uptake of trace gases by plants

The emission behaviour of the most important tree
species at the site (beech, oak, birch) was investigated
in plant chamber experiments. The emissions could
be well parameterized with emission algorithms
(FOLKERS, PhD Thesis, 2002; KOCH, Diploma Thesis,
2002). An upscaling of the emission rates based on
these results is in good agreement with the emission
rates derived from the flux measurements described
above. For HONO a diffusion limited uptake through
the stomata was observed. A formation of HONO
from NO_2 deposited on the leaf surface or a compen-
sation point could not be observed. From our results
the trees can be seen as a sink for HONO (SCHIMANG,
Diploma Thesis, 2003). No uptake was observed for
isoprene, monoterpenes, MVK, and MACR.
Surprisingly, the experiments showed that there are
direct emissions of the isoprene oxidation products

MVK and MACR by birch. Experiments with a fumigation of the plants with $^{13}CO_2$ lead to a labelling of the emitted isoprene as well as MVK (see Figure 15). For oaks no such emissions were observed. For the species composition at the field site (4 % birches) the emission of MACR and MVK contribute to about 3 % of the ambient abundance of these compounds and is by far not sufficient to explain the observed ratios of the isoprene oxidation products in the atmosphere discussed above.

Figure 15: Emission of ^{13}C-labeled methyl vinyl ketone from birch after fumigation with $^{13}CO_2$ for 8 hours (black bar). Given is the part of labeled compounds relative to the totally emitted compounds. The green line indicates the photosynthetic active radiation.
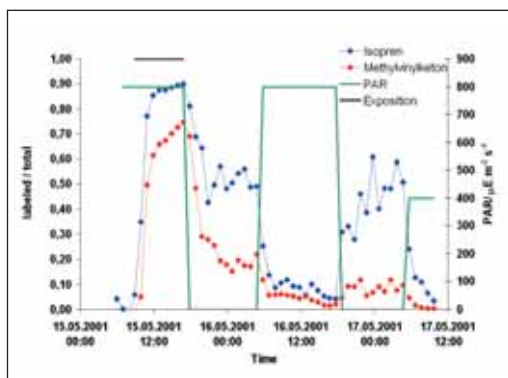
Summary

Although the data are not yet completely evaluated and interpreted, the most important results of this project can be summarized as follows:

+ **Significant concentrations of HO_x radicals were detected in the canopy and in the trunk area; nocturnal OH concentrations around 1 x 10^6 cm^{-3} were observed above the canopy in situations of high ozone and low NO_x concentrations.**

+ **Photolysis of HONO seems to contribute significantly to the OH radical concentration within the canopy.**
+ **Beech was found to be a substantial emitter of monoterpenes.**
+ **MVK and MACR are directly emitted from the trees und thus their presence in the atmosphere is not entirely due to isoprene oxidation.**
+ **Biogenic VOC emitted from the forest stand contribute up to 90 % to the total turnover of organic compounds.**
+ **A variety of terpene oxidation products have been identified in the organic coating of aerosols.**
+ **Fluxes of terpene oxidation products have been measured for the first time.**
+ **Wind tunnel studies were shown to be an ideal tool to understand transport processes in forests and to support the interpretation of field data.**
+ **The concepts of transport processes within the canopy and from the canopy into the internal boundary layer have to be changed considerably.**

The concept of combining field studies, simulation experiments, laboratory studies, and numerical simulations is a powerful method to investigate biosphere-atmosphere exchange processes in complex ecosystems. The results of the ECHO project together with the experience achieved will provide a basis for future studies on biosphere-atmosphere interactions.

Validation of chemical mechanisms for the description of isoprene and α-pinene degradation within 3-dimensional chemistry transport models (ValCheM)

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Introduction

Isoprene and monoterpenes play a major role in the tropospheric gas phase chemistry. Furthermore, monoterpenes are a relevant source of organic particulate matter. Within chemistry transport models the processes involved in isoprene and monoterpene oxidation must be described by highly condensed mechanisms due to limitations in computer time. Since the knowledge about the oxidation of biogenic VOC has increased significantly over the last years, a re-evaluation of the chemical mechanisms used within chemistry transport models appeared to be necessary.

Within the joint project ValCheM an updated condensed isoprene degradation mechanism was validated against smog chamber measurements and applied with a regional scale climate-chemistry model. Two monoterpene oxidation schemes, one for α-pinene and one for β-pinene were developed and applied in a global model combined with a new description of the formation process of secondary organic aerosols.

Isoprene oxidation

Mechanism validation against smog chamber data

A highly condensed reaction scheme for the tropos-

pheric oxidation of isoprene was implemented into the Regional Atmospheric Chemistry Mechanism RACM (STOCKWELL et al., 1997). The RACM is an established chemical module for regional air quality modelling but includes an isoprene chemistry which is no longer state-of-the-art. The new reaction scheme (GEIGER et al., 2003), which is based on the MIM ('Mainz Isoprene Mechanism', PÖSCHL et al., 2000) contains only seven additional reactants and seven additional chemical reactions as compared to the original RACM. It was successfully tested against the results of smog chamber experiments carried out in the European photoreactor EUPHORE. A model intercomparison between both the original and the updated RACM mechanism for a number of well-defined scenarios revealed large deviations in the concentration-time profiles of important reactants for isoprene degradation, particularly under 'low-NO_x' conditions.

Application of the new mechanism in a regional model

The RACM with updated isoprene chemistry was implemented into the regional meteorology-chemistry model MCCM (GRELL et al., 2000). Among others MCCM includes also the original RACM gas phase

chemistry. To facilitate the implementation of new additional atmospheric chemistry mechanisms into MCCM, the option of automatic code generation from a list of chemical reactions by the KPP pre-processor (DAMIAN et al., 2002) was added to the MCCM system. The up-dated mechanism was also implemented in the canopy-chemistry model CACHE (FORKEL et al., 1999) as part of the joint project BEWA2000 (STEINBRECHER et al., 2004).

In comparison to the original RACM, the regional simulations with the updated isoprene mechanism result in slightly lower ozone and PAN concentrations over Europe for most locations and meteorological situations. Especially in regions with high anthropogenic NO_x emissions, differences of the simulated ozone concentrations are only small (Figure 1). However, 5 to 10% higher ozone maxima can also

occur above forest regions in Europe under favorable meteorological conditions. In the tropics up to 60% difference was found by VON KUHLMANN et al. (2003) when different chemical mechanisms were applied.

The new mechanism shows only minor differences for the concentrations of iso-prene and OH. Higher concentrations of methacrolein and H_2O_2 are correlated with areas with high isoprene emissions. The largest differences between the results of the original RACM and the updated RACM were found for organic nitrates, where application of the new mechanism resulted in about 50% lower values. Generally, the new mechanism is recommended for the investigation of other products than ozone and for applications with significant isoprene emissions and moderate NO_x emissions.

Figure 1: Diurnal courses of near surface values of ozone and MACR (methacrolein + MVK) for one grid point of the model domain, corresponding to an area west of the city of Augsburg.
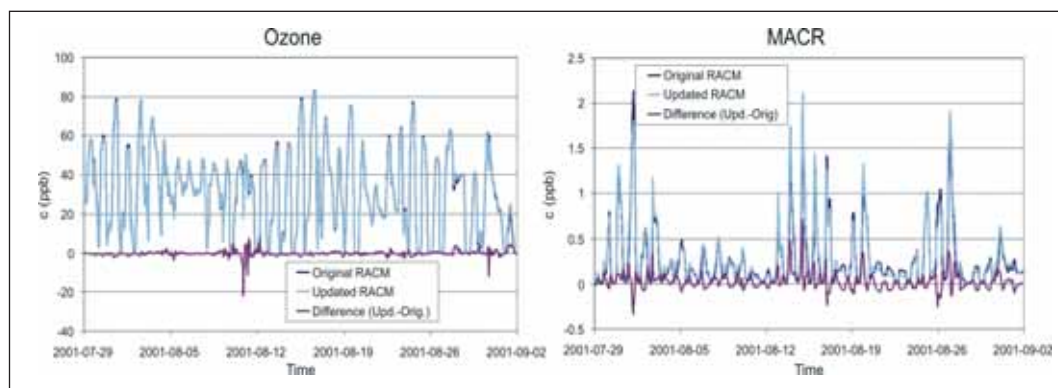
Monoterpene oxidation

Mechanism development

In order to investigate the impact of monoterpenes, which represent the second most important biogenic non-methane hydrocarbon class two reduced mechanisms for the most predominant monoterpenes

α-pinene and β-pinene were de-veloped. For the purpose of global modelling the highly detailed Master Chemical Mechanism MCM v 3.0 (SAUDERS et al., 2003) with approximately 1550 equations per monoterpene (JENKIN et al., 2004) have been condensed to 45 lumped reactions for each of the terpenes including the most important products with

respect to atmospheric chemistry and secondary organic aerosol formation. Therefore, twenty additional organic compounds, 10 stable compounds and 10 radicals, have been added to the pre-existing Mainz Isoprene Mechanism (MIM [PöSCHL et al., 2000]) and tropospheric background chemistry. The obtained two reduced schemes for α-pinene (Mainz A-pinene Mechanism, MAM) and β-pinene (Mainz B-pinene Mechanism) oxidation were tested against the MCM under different atmospheric conditions with a generally good agreement (BONN et al., 2004b).

Global modelling

Both newly developed mechanisms have been implemented individually in the global model MATCH-MPIC (VON KUHLMANN et al., 2003) by simulating the gas phase chemistry with an aerosol sink for the semi- and low volatile species considered in the reaction schemes.

With this set-up an annually averaged impact of monoterpenes on e.g. the tropospheric ozone burden of +3.5% was computed if aerosol formation was neg-

lected (BONN et al., 2004b). If secondary organic aerosol (SOA) formation was taken into account by either a partitioning approach (PANKOW, 1994) or a saturation vapor pressure approach (BONN et al., 2004a) the above mentioned effect on ozone was partially balanced. Thereby, the contribution of different species to SOA formation was simulated throughout the year indicating a previously not considered high contribution of hydroperoxide species. This compound class might be able to explain the high contribution by volatile carbonyl compounds to SOA mass observed in field studies (KAVOURAS et al., 2002) by decomposition as well as liquid phase reactions of the hydroperoxides in the atmosphere and during the sampling process. Because of the different temperature dependencies of the single compounds' saturation vapor pressures and the different chemical conditions in the tropics and in the mid-latitudes a clear annual course of the contributions to SOA production has been found (Figure 2). The contribution of hydroperoxides is highest during Northern hemispheric winter and nitrogen containing species were found to contribute significantly to SOA formation.

Figure 2: Annual contribution of individual compound classes to SOA formation on a regional scale in kg m^2 s^1 x 10^{10} (left). Calculated individual monthly contributions to total SOA production for 1998 (right plot): The individual fractions of the total SOA production are displayed in the following manner: dicarboxylic acids with vertical lines, monocarboxylic acids in gray, low volatile hydroperoxides in white, higher volatile hydroperoxides with horizontal lines, nitrates with slanted lines and PAN type species in black.
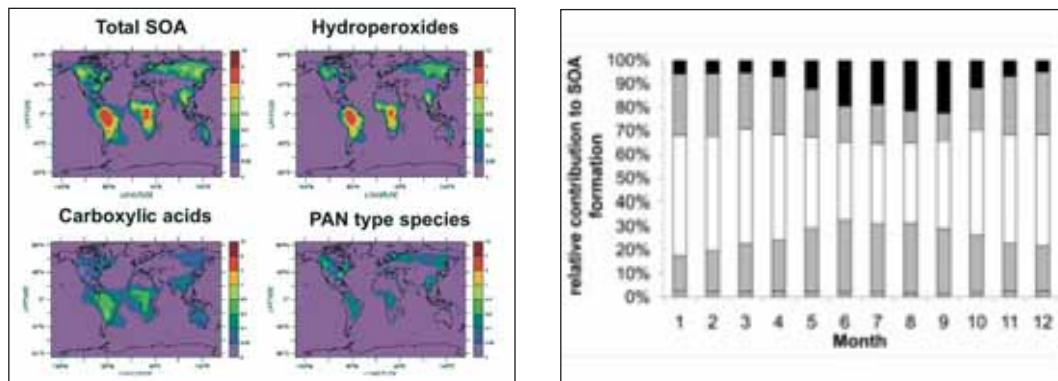
Concluding remarks

New condensed mechanisms for the oxidation of isoprene, α-pinene, and β-pinene including SOA formation were validated against the results of smog chamber experiments and simulations with the highly detailed Master Chemical Mechanism MCM, respectively. The new mechanisms were applied in regional and global chemistry transport models. The updated isoprene oxidation mechanism showed large differences as compared to the original mechanism for oxidation products like methacrolein and for organic nitrates. Significant differences in ozone concentrations were found for conditions with high isoprene emissions and moderate NO_x emissions. Global simulations showed a high contribution of hydroperoxides to the production of secondary organic aerosol from monoterpenes. This compound class might be able to explain the observed high contribution by volatile carbonyl compounds to SOA mass by decomposition as well as liquid phase reactions of the hydroperoxides.

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An integrated data archive of atmospheric chemical standard scenarios for the evaluation of chemistry-transport-models - IDEC

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Introduction and scientific objectives

In order to gain a better understanding of the complex coupled processes controlling the distribution and concentration of trace gases in the atmosphere 3-dimensional Chemistry-Transport-Models (CTM) are developed. Besides the treatment of the emission and transport processes in these models the photo-chemical gas phase reaction mechanism is the key component of CTMs. Due to the restricted computing power no explicit treatment of the full chemistry is possible. Thus condensed and necessarily simplified mechanisms are used which have to be carefully tested (DORN et al., 2002).

The main goal of IDEC is the evaluation and the improvement of the photochemical gas-phase reaction mechanism used in the air quality modelling system EURAD developed at the University of Cologne. The EURAD model is intensively used as an instrument for air pollution prediction ("chemical weather") and as a tool to investigate of the impact of changing emissions on air quality on regional and European scales.

The evaluation of the mechanism was achieved by comparison of the model with photo-chemical experiments under atmospheric conditions in the atmosphere simulation chamber SAPHIR.

Model development and simulation chamber results:

Within IDEC we have improved the RACM mechanism used so far in the EURAD model aiming to a better description of the ozone production. Special emphasis was laid on the detailed treatment of the photochemistry of isoprene and its oxidation products. Isoprene is the most important biogenic hydrocarbon and thus plays a major role with respect to ozone. In close cooperation with the AFO2000 project *ValCheM* we have extended and evaluated a new condensed chemical reaction module for the isoprene oxidation (RACM-MIM) developed by GEIGER et al. 2003. Using the reactions defined in the Master-Chemical-Mechanism (MCM V3, PILLING et al. 2004) we extended this module by the explicit treatment of the chemistry of the primary isoprene oxidation products methacroline, MACR, and methyl-vinyl-ketone, MVK, (RACM-MIM-2).

In chamber simulation experiments we investigated the OH radical induced oxidation of isoprene and studied the yields of its primary oxidation products MACR and MVK from low-to-high NO_x conditions by comparison of chamber data with corresponding model calculations.

The reaction rate constant of ozone and OH with isoprene was measured. Also the product yields of

MVK and MACR resulting from the OH oxidation of isoprene and their reaction rate with OH were studied.

The improved mechanism is already incorporated into the EURAD CTM and is being used for the interpretation of field data measured during the ECHO and BERLIOZ campaigns (Corsmeier et al., 2002). The results of the SAPHIR experiments can be summarised as follows:

Measured rate constants k(OH+ISO) and k(O$_3$+ISO)

Figure 1: (top): Isoprene oxidation by OH in SAPHIR simulating the atmospheric condition during the ECHO field campaign. Inset: OH concentration simultaneously measured by laser-induced fluorescence (LIF) and laser absorption (DOAS). The red line shows a model run using the rate coefficient k1=(1.00±0.12)·10^{-10}cm^3/(molec.·s) which was inferred from the measured decay of isoprene in these experiments. The currently recommended value (blue line) (Atkinson, 1997) was confirmed (Karl et al., 2004).

(bottom): Measured oxidation products MACR and MVK. Blue lines show model calculations using product yields and OH reaction constants from the literature. Red lines indicate model runs using kinetic parameters determined in SAPHIR experiments.
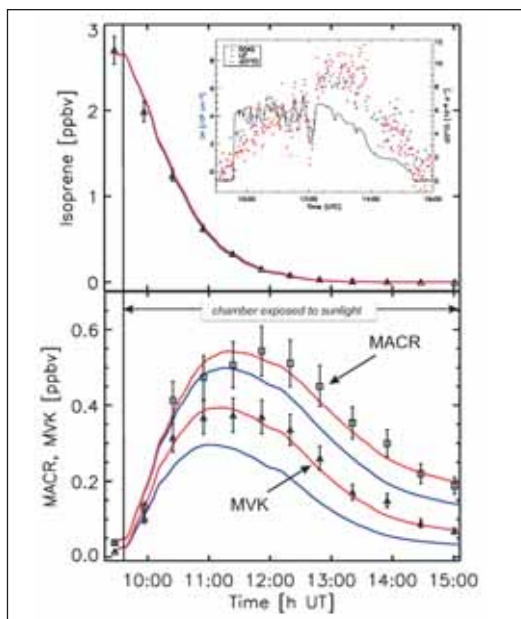
agree well with recommended values from the literature (Karl et al., 2004).

Product yields of the formation of MACR and MVK determined in SAPHIR under atmospheric conditions are 26% and 35%, respectively (Karl, 2004).

Compared to the literature the observed reaction rates for the OH oxidation of MACR and MVK are lower by 10% and 15%, respectively.

Using the simulation chamber it was possible to measure the MVK/MACR-ratio under nearly NO$_x$-free conditions. Below about 50 ppt of NO the MVK/MACR ratio reaches values around 0.7, whereas for NO above 100 ppt a constant value of about 1.4 is found. The measurements are in accord with the model (Figure 1).

EURAD CTM:

To meet the needs of the EU directives on air quality it is important to perform simulations on the time scale of one or even several years (Friese et al., 2002; Memmesheimer et al., 2004). The EURAD modelling system has been adapted to these demands. It has been developed to a quasi-operational system. Daily short term predictions are archived since 2001 and can be accessed by the public (www.eurad.uni-koeln.de) and can be used by the public and scientific applications (Jakobs et al., 2002, 2003; Lawrence et al., 2003). As one major goal of IDEC-SENEVA the gas phase chemical mechanism has been evaluated and extended. RACM-MIM has been used for log-term simulations over one year. The improvements in the treatment of gas-phase chemistry have broadened the scientific basis of EURAD as a reliable tool for the development of air pollution abatement strategies and the planning and analysis of measurement campaigns (e.g. ECHO (Figures 2 and 3); Berlioz and others) and "chemical weather" prediction.

Improved gas-phase chemistry and biogenic emissions lead to an increase of ozone, in particular the ozone maxima during summer episodes. The agreement between observations and measurements has been improved.
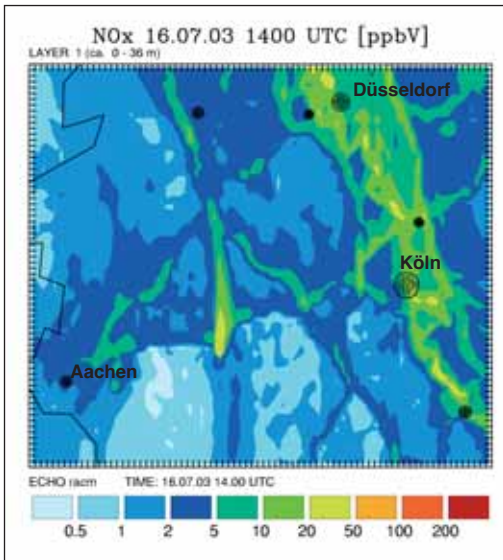
Figure 2: Modelled NO$_x$ mixing ratio (ppb) on July 16 2003, 14:00 UTC (ECHO field campaign) using the EURAD-CTM. Simulations were performed using sequential nesting from the European scale (125 km grid) to the local scale (1 km grid).

Results of the cluster analysis of long-term model data

A set of characteristic distributions of air constituents over Germany was determined from model results of the EURAD-CTM. A method to carry out a cluster analysis on the basis of spatial patterns was success-fully developed and applied to summer seasons of 1997, 2000 and 2003.

Typical compositions within a cluster are identi-fied through the accumulated sum of correlation coefficients of each day with the other days of the cluster. We identified 28 clusters which occur in at least 3 levels simultaneously, often consisting of less than 6 objects. The chemical composition of the Euro-pean troposphere has a high variability resulting from the superimposition of many local phenomena within Europe. Some features are: High or enhanced morning concentrations of CO, NO$_2$ and light alkanes (HC$_3$) over southern Great Britain, The Netherlands and northern France in 15 of the 28 clusters. Those

Figure 3: Time series of measured (black) and modelled ozone values (green: RADM2 with biogenic emissions from standard version; red: RACM-MIM2 with biogenic emissions implemented during AFO2000) at Niederzier (near Jülich, 1 km grid size). Ozone values increase with the more sophisticated version of the model. This, in general, leads to a better comparison between observed and mod-elled peak values of ozone. Measurements were provided by the Umweltbundesamt (UBA).
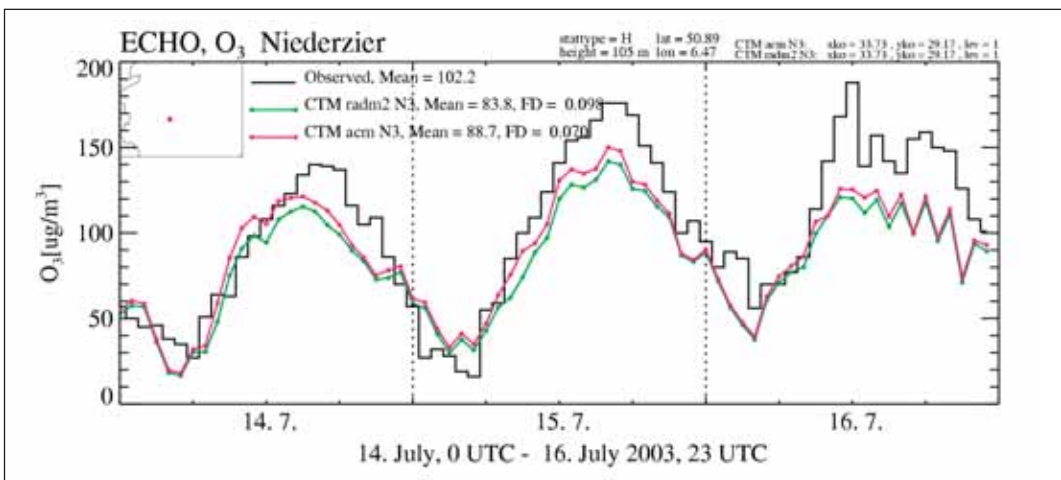
high pollution episodes coincide with low winds from the east or south. 12 clusters show a cloud of enhanced HCHO concentrations in northern Scandinavia.

The extremely hot year 2003 almost appears to be a cluster of itself in the surface level. Most of the clusters are linked to weather patterns. There are 8 clusters which are governed by transport phenomena with 2 remarkable cases: a vertical transport of ozone into the upper troposphere by a cyclonic system and a convergence of ozone in the south eastern Mediterranean area as a result of a northerly flow that pushes the air mass against a strong westerly wind field. This convergence does not occur in the surface level.

The results are used to design photo-chemical experiments in the atmosphere simulation chamber SAPHIR and promise to be useful for model evaluation and risk management.

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Results of Theme Group 1.2

Surface-Atmosphere Interactions: Processes for transport phenomena above inhomogeneous terrain

DOAS tomography: Mapping of trace gas distributions from ground and aircraft by UV-Vis spectroscopy

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Summary

DOAS Tomography is a novel technique developed for spatially resolved measurements of trace gas distributions. Column densities of one or several trace gases, measured by DOAS (Differential Optical Absorption Spectroscopy) along multiple light paths, are inverted to 2D or 3D concentration distributions by tomographic inversion. In the framework of the AFO Tom-DOAS project, first tomographic measurements from ground (section 1) and aircraft (4) have been carried out. New instruments have been developed (2) and the method was proven valid during an indoor validation campaign (3). In addition computer tools have been developed to optimize instrumental configurations and to invert the measurements (5).

Ground-based measurements

The first tomographic arrangement (Figure 1) was set up in the framework of a motorway emission campaign in April/May 2001 (CORSMEIER et al., 2004). It consisted of two Longpath telescope sites and eight retro-reflector sites. Each telescope emitted one light beam, which was successively directed to the eight retro-reflectors (PUNDT *et al*., 2004). The two light beams were reflected back to the telescopes and coupled into spectrometers (PLATT AND PERNER, 1984). Here 16 light paths were achieved in total. Since the reflectors were pointed at successively, a tomographic reconstruction is possible only for stable meteorological conditions and a constant vehicle flux during one measurement cycle, which varied between 15 and 45 minutes. In Figure 2, the two

Figure 1: Tomographic setup during the motorway emission campaign. From each telescope one light beam is directed successively towards the eight retro reflectors (PUNDT et al., 2004).
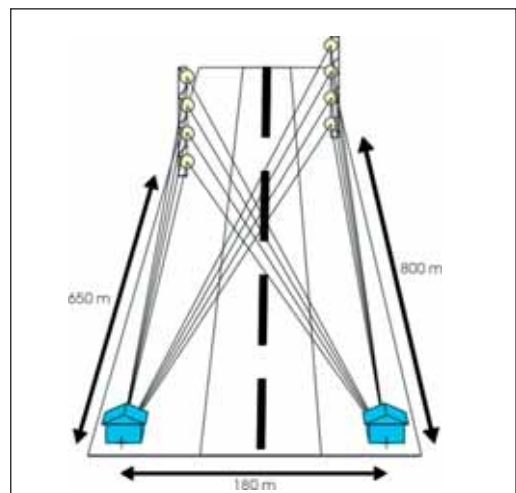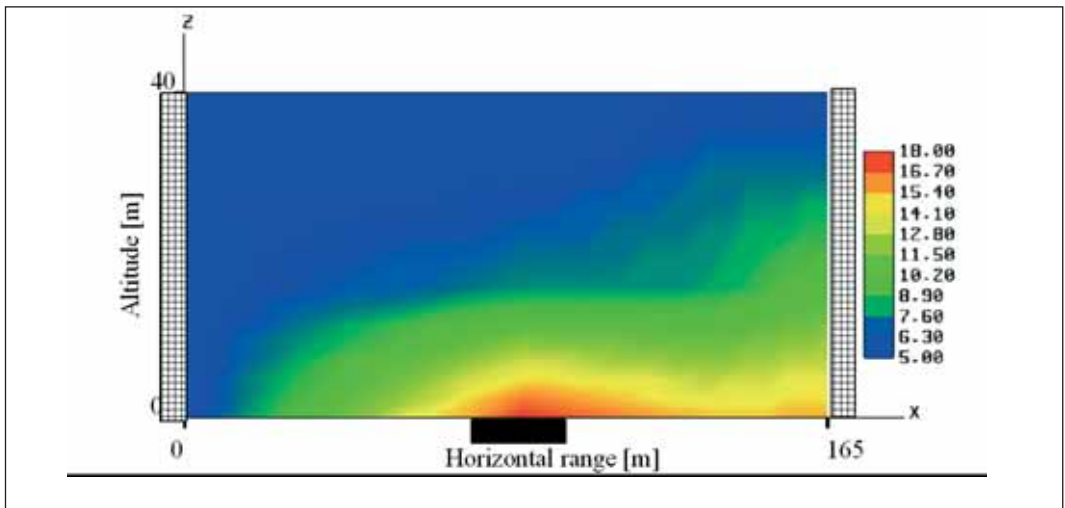
Figure 2: 2D reconstruction of the NO_2 emission plume (in ppbv) at right angles to the motorway, between the two towers and the two DOAS instruments. The reconstruction is carried out over a four hour average in the morning on May 10th 2001 (9:00-13:00 CET) using the SART inversion method. Black bar: Location of the motorway, shaded areas: the two towers.

dimensional NO_2 concentration distribution perpendicular to the motorway is shown for May 10th (average from 9:00 to 13:00). For the inversion, the Simultaneous Algebraic Reconstruction Technique (SART) was used. An emission plume from the location of the motorway towards the southern (right-hand) tower can be clearly seen in consistency with the wind direction measured during the presented period. The northern side represents the NO_2 background and the southern side the emission plus the background.

Instrumental developments

A new Longpath DOAS telescope type, the "Multibeam telescope", was developed for the simultaneous measurement along multiple paths (PUNDT AND METTENDORF, 2004). This telescope emits up to six light beams simultaneously, using only a single lamp as light source. Figure 3 shows the instrumental setup. Here, two light beams result from two 'virtual

lamps' created by additional mirrors inside the lamp housing (Figure 4). Three instruments were constructed and employed in Heidelberg and Italy, and during the validation experiment.

Figure 3: Instrumental setup of the novel Multibeam system developed during the Tom-DOAS project including telescope A, lamp housing B, mode mixer C, spectrograph D, CCD detector E, stepper motor controller F, mirror tower with four mirror units G, and the rotating disk retro reflector H, from PUNDT AND METTENDORF, submitted to Applied Optics.
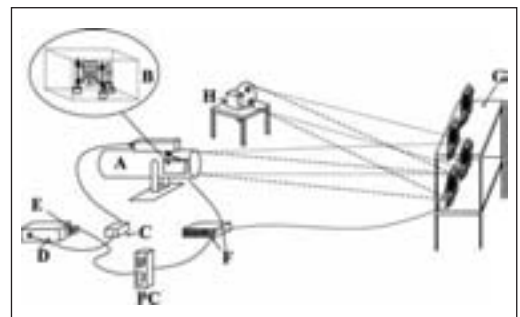
Figure 4: Picture of the Multibeam telescope from the front. Two virtual lamps appear on the telescope main mirror.

Figure 5: Picture of the Tom-DOAS validation experiment. One Multibeam-system (left), one polycarbonate cell (middle), and one mirror tower (right side) can be seen. Four light beams from another telescope are directed onto the experimentalists.

Figure 6: Measurement geometry of the Tom-DOAS validation experiment:

Tom-DOAS validation campaign

Following the suggestions of the AFO referee team a validation field experiment with specified concentration distributions was conducted in August 2003 (Figure 5). Above an area of 15 x 10 m^2 we simulated the atmospheric boundary layer in small scale. The setup corresponds to a small town with a ground surface of 1.5 x 1 km^2 containing two emission plumes with a diameter of 200 m. The emission plumes are simulated by one or two NO_2–filled cells, diameter of 2 meters each. The measurement geometry and one cell are displayed in Figure 6. Then the NO_2 2D concentration distribution was measured using 3 telescopes, 12 simultaneous light beams, and 39 light paths in total. An example of a reconstructed concentration distribution is shown in Figure 7. The comparisons between the given and the reconstructed distributions show very good agreement, proving the method valid for specific atmospheric conditions (METTENDORF et al., paper in preparation).

Figure 7: Tom-DOAS validation experiment: Reconstructed result from the measurement.

Aircraft measurements

The aircraft instruments were developed in co-opera-
tion with the Institut für Umweltphysik of the Univer-
sity of Bremen and installed in a small aircraft (Parte-
navia 68). The instruments consist of two spectrome-
ters (covering the UV and visible spectral ranges,
respectively) that are connected to ten telescopes
(PUNDT et al., paper in preparation). Onboard the air-
craft, three of the telescopes are pointing upwards
under different angles, the remaining seven are
pointing downwards into the boundary layer (Figure
8). 20 flights of about two hours each were per-
formed in the Milan area (Italy) in the framework of
the EU "FORMAT" project in summer 2002 and in
autumn 2003.

*Figure 9.: NO_2 slant column as function of geographic position
for 26th September 2003. The power plant of Sermide, which is
marked by the red cycle, was surrounded and its plume was over
flown three times*

*Figure 8: Partenavia Aircraft and the viewing directions of the
telescopes during the second Format campaign in
September/October 2003.*

*Figure 10: Aircraft measurement geometry near the Sermide
power plant on 26th September 2003. Only the solar light paths
from the ground towards the telescopes are shown for clearness.
The color of the beams presents the measured NO_2 column
density.*

Figure 9 shows the flight track of September 26th
2003 together with the measured NO_2 slant columns.
In Figure 10 an optical sketch of the overlapping solar
light paths is shown for downward looking tele-
scopes for a plume measurement near the Sermide
power plant. Figure 11 displays the NO_2 map derived
from the third pass over the Sermide emission plume.
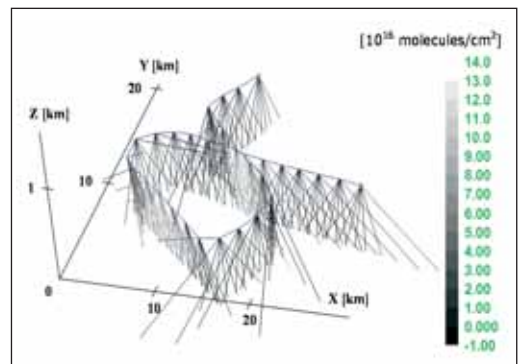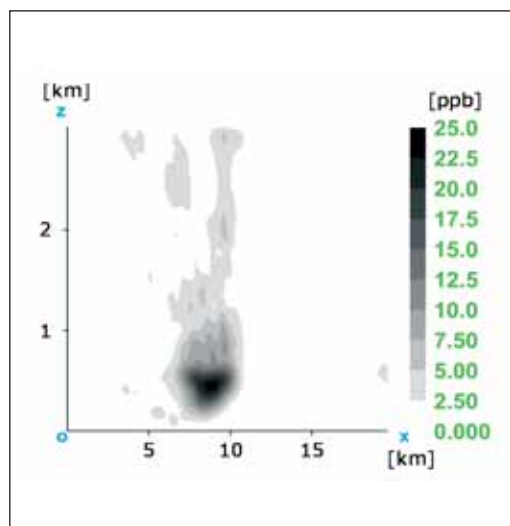
Figure 11: NO$_2$ 2D mixing ratio distribution after tomographic inversion for the third overpass, performed at 5 km distance from the chimney.

Software Development

The experimental software TOMOLAB has been developed and used for the optimization and inversion of tomographic measurements. It comprises different inversion techniques, and allows the choice of a large variety of parameters (e.g. basis functions, grid, and constraints). For theoretical studies, the software is able to calculate slant column data from CT-Model concentration fields and assumed errors. The reconstruction results of these theoretical data are then evaluated by comparison with the original data (LAEPPLE et al., 2004, KNAB, 2003).

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Vertical transport of particles between planetary boundary layer and free troposphere

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Goals

Vertical and horizontal transport processes, which control the large-scale distribution of particles and trace gases in the atmosphere, are of major interest in climate and air-pollution research. Lidar instruments give us the opportunity to investigate these processes because of their ability to provide the parameters of interest, i.e., wind speed, particle properties, temperature, and humidity, with high spatial and temporal resolution. In this project, emphasis was put on the development of instruments and methods to study the vertical transport of particles in the planetary boundary layer (PBL) and the lower free troposphere. One of the key issues was the development of a Doppler lidar with coherent detection for the measurement of the vertical wind speed. This instrument shall be applied together with the two IfT aerosol lidar systems in order to determine turbulent aerosol fluxes, temperature and humidity. Parallel to the instrumental developments we conducted two measurement campaigns with the IfT aerosol Raman lidar and the MPI-M Doppler wind lidar in order to elaborate methods for the evaluation of combined lidar data sets, especially with respect to the derivation of vertical aerosol mass fluxes.

Wind lidar development

The Doppler wind lidar operates in an atmospheric transmission window at an eye-safe wavelength of 2.02 μm. It consists of a continuous-wave, single-mode, single-frequency master oscillator, a pulsed power oscillator, a transceiver, a heterodyne-detection and a data-processing unit (Žeromskis, 2004; Žeromskis et al. 2004). In both lasers Tm:LuAG crystals are pumped with fiber-coupled laser diodes. An acousto-optical modulator serves as Q-switch and as injection seeder. It also delivers a frequency shift of 80 MHz between master oscillator and power oscillator for the heterodyne detection. An off-axis Mersenne-Cassegrain telescope of 15 cm diameter is used for both transmitting and receiving radiation (Engelmann, 2003). An InGaAs photodiode with extended spectral range detects the superimposed radiation from the atmosphere and from the master oscillator. The heterodyne signal is amplified, band-pass filtered, and digitized with an analogue-to-digital converter at a rate of 250 Megasamples per second. For each range bin, the main peak of the FFT spectrum is found with a power-spectral algorithm (Rhone, 2004). The Doppler lidar has been mounted in a transportable frame and can be used for smaller campaigns at the institute's site. This instrument gives us the opportunity to measure the vertical wind speed with an accuracy of a few centimetres per second and a resolution of 50 m in space and 10 s in time.

Turbulent aerosol mass fluxes

The MPI-M Doppler wind lidar and the IfT aerosol Raman lidar were operated next to each other at the

IfT site during two measurement campaigns in October 2002 and in September 2003. With both instruments, we performed measurements in the convective boundary layer under a variety of meteorological conditions and we made first attempts to use the eddy correlation technique on the basis of highly resolved aerosol and vertical-wind data to calculate turbulent aerosol fluxes.

The turbulent flux f_b of an atmospheric parameter b is given by the covariance between b and the vertical wind speed w, $f_b = \overline{b'w'}$ where positive values of w and w' imply upward winds and positive values of f_b upward fluxes. Fig. 1 (left panel) shows the time series of aerosol backscatter ratio b and vertical wind speed w obtained with the two instruments in a dry, convective boundary layer at a height of 1300 m on September 22, 2003. In the first time interval from 12.40-14.20 UTC the PBL height increased from 1.6 to 2.2 km and upward aerosol fluxes were found, see Fig. 1 (right panel). After 14.20 UTC, when the convective motion slowed down and the PBL height decreased to 2.0 km, downward fluxes were obtained.

A parameterization was used to convert aerosol fluxes observed as function of backscatter ratio into aerosol mass fluxes. The multiwavelength lidar technique developed at IfT has been applied, i.e., the mean particle volume concentration was calculated from the mean optical data (backscatter coefficients at 355, 532, 1064 nm, extinction coefficients at 355, 532 nm). From the mean volume concentration, we estimated the mean mass concentration $\overline{m}$ by using a typical particle density of 1.9 g/cm^3. We further assumed that the fluctuations of the scattering properties used for the flux determination are mainly caused by turbulent motion and not by a change of the aerosol properties, i.e., $b'/\overline{b} = m'/\overline{m}$. Then we can estimate the turbulent aerosol mass flux to $f_m = \overline{m'w'} = \overline{m}/\overline{b} f_b$. The ratio $\overline{m}/\overline{b}$ was found to be 5 μg/m^3 in the present case and was used for the conversion of the lower into the upper axis of Fig. 1 (right panel). an extended descritption of the methodology and the measurements can be found in Wandinger et al. (2004).

Figure 1: Time series of vertical wind speed and aerosol backscatter ratio (BR) at 1300 m height on September 22, 2003 (left), and corresponding aerosol flux profiles for the indicated time periods (right). For both time series mean value and linear trend have been subtracted. The BR values are scaled so that the distance between minimum and maximum values is the same as for the wind data.
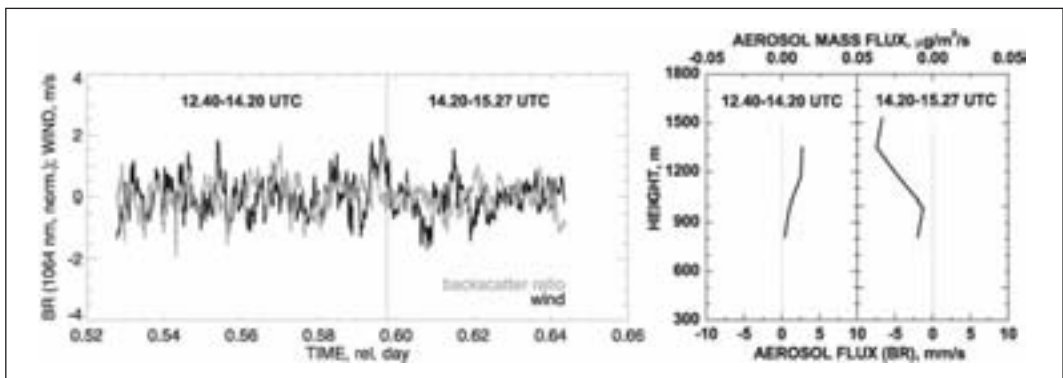
Development and validation of tools for the implementation of European air quality policy in Germany (Project VALIUM)

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Scientific Objectives

Project VALIUM aimed on the development and validation of tools as they are needed for the execution of the European Air Quality Guideline 96/62/EU and its daughter directives. This set of regulations requires from the EU member states to submit maps that show the spatial distribution of air pollutants

+ **for the member state in total,**
+ **for conurbations with more than 250.000 inhabitants and**
+ **for micro environments as, e.g., city districts subject to high pollutant concentrations.**

Project VALIUM comprised the following tasks:

+ **the development of the numerical model system M-SYS which is capable to provide pollutant concentrations in spatial resolutions matched to the regional and local scales.**
+ **the development of a methodology to generate emission data with resolutions as required by the models,**
+ **the generation of a high quality data set based on a combination of field measurements and wind tunnel simulations in a city district, and finally**

+ **the presentation of the measured data in the form of a generally accessible data base and utilization of the data for the validation of M-SYS and its parameterisations.**

Project VALIUM was designed as the German core project for EUROTRAC-SATURN 'Studying Atmospheric Pollution in Urban Areas'.

Experiments

The field measurements were made at the test site 'Goettinger Strasse' in Hannover/Germany (Fig. 1). This side includes a busy street canyon with approximately 30000 vehicles per day, a large percentage of them being trucks. Since nearly two decades the Lower Saxony State Agency for Ecology (NLÖ) operates a monitoring station at this site. Long time series of data are available which offer the opportunity to approach the urban air pollution problem also by statistical methods.

Continuous measurements of air pollutants inside the street canyon and in the surrounding area of 1 km x 1 km (Goettinger Strasse in Hannover) were performed in addition to the routine NLÖ monitoring from early 2001 until the end of 2003. Both air pollutants and meteorological parameters were

measured by in-situ instruments at four locations inside the street canyon and at three locations in the surroundings of the canyon (one of those being a roof-top station at the highest building in that area). Path-averaging optical measurement techniques (two, some times three DOAS systems) were used continuously at the ground and at building roof level. The meteorological background (vertical profiles of wind and turbulence plus mixing layer height) came from a SODAR system that was positioned about 500 m apart from the street canyon.

Three intensive observation periods (IOPs) in different seasons were successfully executed. Path-averaged concentrations of air pollutants were measured at both sides of the street using FTIR spectrometry. In addition, vertical gradients of air pollutants in the street canyon were determined applying a DOAS-system. During selected periods, the standard meteorological parameters measured continuously at the roof top station and by the SODAR system were complemented by (1) a ceilometer (operated by Vaisala) that was located at the roof of a building, and (2) a Wind-Temperature-RADAR (WTR, operated by IMK-ASF). The ceilometer (aerosol backscatter profile) and WTR data (temperature profile) were used for com-

Figure 1: Street Canyon 'Goettinger Strasse with instrumentation. The picture shows the continuous monitoring stations HRVI to HRV3 (white) and the SF$_6$-line-source at the median strip plus the sampling bag locations (green). The path-averaged measurements are indicated by the blue (FTIR) and red (DOAS) lines.

parisons with the SODAR data. All measurement systems were carefully calibrated. Simultaneous operation of the different instruments carried out at one site before or after each IOP showed only differences in the order of the specified accuracy of the instruments.

Most of the previous field experiments carried out in urban areas suffer from the fact that the source term is not properly known. There is the choice between several emission models that link the traffic flow to the source strength. Since the concentration fields predicted in dispersion calculations are directly proportional to the source strength, the results of numerical models may to a large degree depend on the particular choice of the emission model. In order to circumvent this problem, the meteorological and pollutant measurements in the Goettinger Strasse were complemented by some tracer experiments. An artificial line source was installed on the median strip dividing the four traffic lanes, and controlled amounts of SF$_6$ were released. The line source had a length of 96 meters. Air probes were collected at 15 sampling points within the street canyon and at roof level. The SF$_6$ content of the probes was subsequently analysed in the laboratory. For altogether 8 days within the period 2001 to 2003, 100 half-hourly averaged concentration values were determined at each monitoring position. Additionally path averaged concentrations of tracer SF$_6$ were measured at both sides of the street using FTIR spectrometry. Prevailing wind directions during the experiments were westerly for five days and northerly, easterly and southerly, respectively, for the remaining 3 days.

The tracer experiments provided the opportunity to validate not only dispersion models but emission models as well. During the intensive observation periods, manual traffic counts were carried out together with traffic speed measurements. In addition, the vehicles were subject to an automated plate number registration. The subsequent consultation of the German vehicle register allowed the exact deter-

mination of the vehicle engines and, by means of emission factors, the best possible quantification of the traffic emission rate.

Field measurements cannot properly be controlled. During the measurement campaigns one has to cope with the weather as it is, the boundary conditions for the experiments are only partly known and, even worse, they change continuously due to the diurnal cycle. Therefore, VALIUM comprised also a wind tunnel sub-project that was tasked to support the field measurement campaigns with corresponding flow visualization experiments and with some systematic sensitivity studies which help to analyse and to understand the data. In addition complete fields of velocity and concentration were measured within the street canyon with a spatial resolution which corresponds to that of micro-scale numerical models. A detailed aerodynamic wind tunnel model of the urban site was built, the complete model covering an area of about 1 km x 1 km. A wind tunnel boundary layer corresponding to the model scale (1: 250) was generated utilizing a combination of vortex generators and floor roughness elements. The complete mean and turbulent boundary layer properties were determined. Subsequently numerous and until now unique experiments have been carried out from which only a few examples can be presented in the results chapter.

Numerical model development

The meso- and micro-scale meteorological and chemistry transport models METRAS, MITRAS, MECTM and MICTM were combined to the model system M-SYS. For M-SYS the necessary input data (building inventory, emission inventory) have been processed and an analysis procedure on the basis of routine meteorological observations was developed. The gas phase chemical mechanism was complemented with simple aerosol chemistry, and a nudging method for chemical data assimilation was implemented and applied. The sensitivity of model results

on the limited horizontal resolution was reduced by calculating aggregated surfaces fluxes. All these measures lead to a significant improvement of model performance in comparison with observations. After completion of the development phase a large number of simulations were made. First it was shown that the model results are in good agreement with those of the measurements before subsequently selected air pollution episodes were simulated (Fig. 2).

Figure 2: Horizontal cross section of NO_2 concentration at 1,5 m above ground for April 11th, 2003, 07:00-07:30 as simulated with the MITRAS/MICTM model system. NO_2 is formed by chemical reaction between the primarily emitted NO and O_3 advected into the street from outside. The meandering of the maximum concentrations in the street canyon is caused by heterogeneities in the wind field. A convergence zone in the northernmost part of the street causes the peak concentration.
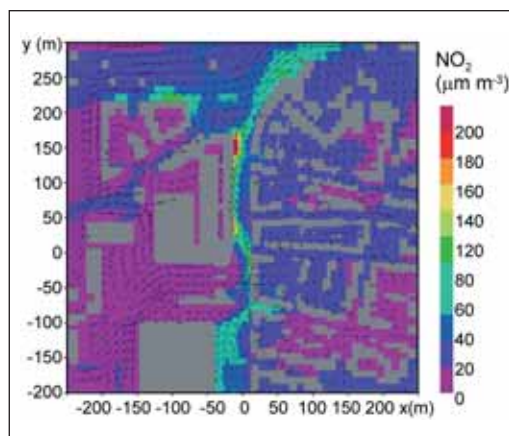
Results

The VALIUM experiments are up to now the probably worldwide largest and most detailed of its kind. In order to make the data generally accessible, the 'intelligent' database VALIDATA was created that comprises all the measurements in a well-documented and easily understandable form. Before the data entered the database they have carefully been checked for plausibility and consistency. Part of the examination was the comparison of the spot concentrations at the monitoring positions with the line-averaged measurements along the Goettinger Strasse. Furthermore the dilution of the tracer gas SF_6 was compared with that of NO_x. It was checked whether the pattern of path-averaged air pollutant concentrations at both sides of the Goettinger Strasse shows the rotor-like circulation inside the street canyon. Whenever possible the data were compared with the results of the wind tunnel that were obtained under carefully controlled and steady boundary conditions. In all cases the data agreed reasonably well with each other and showed the expected qualitative and quantitative behaviour.

From the numerous results obtained only a few can be mentioned here. The analysis of NO_x concentrations measured at the roof top station revealed that about one third of the variance of NO_x concentrations was caused by regional transport and depends on the mixing layer height. At ground-level stations this dependence has not been found. The correlation between mixing layer height and PM_{10} is much weaker and not different for ground and roof top level data, but the PM_{10} concentrations at ground level inside the street canyon are about twice as high as those at roof level. That confirms the expectation that the concentrations measured at roof top are representative for the urban quarter surrounding the street canyon. In contrast to that, the concentration measurements at ground level are dominated by traffic emissions inside the canyon.

The comparison of wind vectors determined by the SODAR system with those from the roof top station (10 m above roof level) showed that for this quantity the roof station measurements are not fully representative for the site. The velocity data are influenced by the building itself and by the building structure surrounding the site. This finding is confirmed by the wind tunnel measurements that allowed quantifying the wind vector modification.

The traffic measurements in the street canyon Goettinger Strasse in Hannover showed, that the driving behaviour and the driving patterns are subject to strong temporal variations. The common procedure to assign a rigid "traffic situation" to a given road segment results in considerable errors and should not be applied in numerical simulations with high temporal resolution. The quality of currently published emission factors (including systematic errors) has been assessed by comparisons between the emission rates, modelled on the basis of the extended traffic measurements, measured pollutant concentrations and the data from the tracer gas experiment.

Air pollution in the Goettinger Strasse is predominantly caused by traffic emissions. As is typical for so-called hot spots in cities, the sources and the receptor points are close together. The values measured at the pedestrian walkway must depend on the source location. The VALIUM tracer experiment was carried out with an artificial SF_6 line source, which was located at the median strip at the centre of the street canyon, whereas the vehicles emit along the four traffic lanes. To study how the values measured depend on the source conditions, in the wind tunnel the following scenarios were carried out, (a) all traffic lanes emitted equally, (b) the lanes emitted according to their traffic density and (c) each lane emitted separately. Since in the field experiments the line source was only 96 m long, the effect of the finite length of the source was also studied. Additional laboratory experiments investigated the influence of

different averaging times on mean concentration values (Fig. 3).

During measurement campaigns only individual cases are covered. The characterisation of the general pollutant situation at a site requires numerical modelling. It is known that substantial parts of the pollutant concentrations measured at an urban site can be caused by long-range and regional transport. Therefore it is necessary to apply a hierarchy of meso- and micro-scale models nested into each other. 40 days of the year 2000 which showed high NO_2-concentrations in the Hanover region were simulated, the results were in good agreement with routine observations of DWD and NLÖ. Maps showing concentration distributions were compiled at the various scales.

For the VALIUM project the micro-scale flow simulations were of particular interest. Good agreement can be reported between the numerical model results and the data taken within and around the street canyon or in the wind tunnel. The micro-scale concentration fields are very heterogeneous; for westerly flow they show two local peaks some 60 m apart, which is in agreement with SF_6 tracer experiments. For easterly winds it was found that tracers are exported from the street canyon over the roofs into the backyards.

Figure 3: Physical model of the site in the Boundary Layer Wind Tunnel of Hamburg University
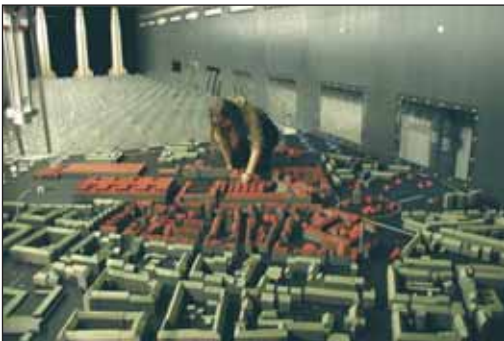
It has been demonstrated that the model system M-SYS is fit for its purpose. M-SYS is presently able to simulate the following pollutants considered in the EC daughter directives: SO_2, NO_2, NO_x, some of the PM_{10}-substances (secondary ammonium aerosols), CO, O_3. The prototype version of M-SYS is generally available. Due to the fact that the originally planned second period of AFO2000 was cancelled, the fully documented PC version of M-SYS could not be developed.

Conclusions

The combination of field studies, wind tunnel experiments and numerical model simulations within one project proved to be a very successful strategy to investigate the complex flow and transport processes that occur in urban environments. The main objectives of VALIUM, the development of a prototype model system that provides pollutant concentrations in spatial resolutions matched to the requirements of the new European regulations and the generation of a high quality data set based on a combination of field measurements and wind tunnel simulations in a city district, were reached.

Additionally, project VALIUM can gave answers to a number of questions that are of particular importance for urban air pollution studies:

+ **How accurate are present emission models for micro-scale applications?**
+ **What are the chances of ground-based remote sensing techniques for the determination of vertical profiles of atmospheric variables in urban areas?**
+ **What is the influence of surrounding buildings on roof-top measurements at urban sites?**
+ **How much dependent are air pollution levels in urban environments on the height of the mixing layer?**
+ **To which degree are local concentrations influenced by long-range and regional pollutant transport?**

+ **How representative are concentrations measured at positions within the canopy layer with respect to time and space?**
+ **Is the commonly used non-dimensional concentration c* a suitable parameter for the generalisation of street canyon concentrations from one situation to the other?**
+ **To which extent depend numerical or physical model results on the choice of the domain size?**
+ **How sensitive are street canyon measurements to the particular source pattern?**

Detailed answers to these questions will be given in a special edition of the "Meteorologische Zeitschrift" which is about to appear in the year 2005.

However, there are also open questions for which final answers have not yet been found. These comprise the contribution of vehicle-induced turbulence to pollutant dispersion in street canyons, the representativeness of episodic measurements within the urban canopy layer and the concepts for a unified presentation of concentration data.

Investigation of vertical and horizontal transport processes and their influence on the concentrations of ozone and aerosols near the surface (HOVERT)

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Overview

A one year measurement campaign in and around Berlin has been carried out from September 2001 to September 2002 to observe concentrations of PM10, major aerosol-ions, ozone and some precursors. The most important challenge was to improve the knowledge about the contribution of anthropogenic urban sources and of long term transports of anthropogenic and natural constituents of air to local concentrations in order to give advice to authorities to elaborate reduction strategies for ozone and PM10

concentrations. The validation of the chemical transport model (CTM) REM_Calgrid (Stern et al, 2003) by means of observed data was propaedeutic to any further use of it in determining possible sources of air pollution.

Aerosol measurements

By means of a DIGITAL Hi-Vol-Sampler, daily samples of atmospheric PM10-fractions have been collected on quartz- filters at 15 sites in and around the city of Berlin. These filters were conditioned according to

usual regulations, in order to be able to determine gravimetrically the total dust content. These probes were chemically analysed by means of ion chromatography in order to measure main ions (Sulphate, Nitrate, Chloride, Potassium, Magnesium, Calcium, Ammonium) as well as by means of thermogravimetry to measure EC (Elemental Carbon) and OC (Organic Carbon).

Measurement positions have been decided in cooperation to local authorities under considerations of magnitude of local emissions and meteorological aspects (Fig. 1). PM10 measurements by means of a High Volume Sampler were taken at MP27 (Schichauweg), MP42 (Nansenstr.), MP77 (Buch) and MP174 (Frankfurter Allee) and after a few months extended to further measurement stations. Ion analysis of water soluble aerosol components as well as OC/EC analysis has been performed continuously at station MP42, MP174 and BS (Beusselstr.), whereas samples from MP27 and MP77 have been analysed depending on the wind direction in order to have insight into the city plume. At the rural stations HH (Hasenholz) and PA (Paulinenaue) aerosols were measured in 2002 by High-Vol-Samplers to obtain a rural counterpart to the urban samples.

Fig. 2 shows the annual average concentration of PM10 (left) and of Nitrate, Sulphate, Ammonium, Elemental and Organic Carbon (right) over the whole measurement period. While total PM10 and Elemental Carbon concentrations show a net decrease from traffic sites (30-35 μg/m^3 for PM10 and 4.6 μg/m^3 for EC) toward rural measurement locations (20 μg/m^3 for PM10 and 1.3 μg/m^3 for EC) and toward upper air concentrations (15 μg/m^3 for PM10 and 1 μg/m^3 for EC), secondary inorganic aerosol components show more homogeneous concentrations between traffic to rural sites.

Figure 2: Annual mean concentrations of total PM10 (top) and main aerosol components (bottom) in μg/m^3.

Figure 1: Aerosol Monitoring Stations in and around Berlin. High Volume Sampler at MP27 (Schichauweg), MP42 (Nansenstr.), MP77 (Buch) and MP174 (Frankfurter Allee) and at the rural stations HH (Hasenholz) and PA (Paulinenaue); Ion analysis of water soluble aerosol components as well as OC/EC analysis at station MP42, MP174 and BS (Beusselstr.); samples from MP27 and MP77 have been analysed depending on the wind direction.

Fig. 3 shows the mean PM composition at different measurement sites. Secondary inorganic aerosol components Sulphate, Nitrate and Ammonium are more or less similar in both regimes, while traffic sites show much higher concentrations of Elemental Carbon contribution to PM10 than rural sites.

Figure 3: PM-composition at traffic sites (left) and at rural stations (right).
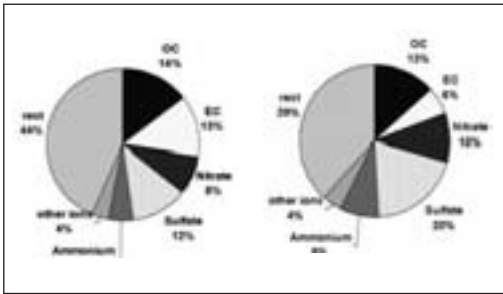
Elight and Physics Department FU Berlin: Ozone measurements by a ozone-LIDAR and PM measurements by means of backscatter coefficient analysis

For long term monitoring Elight GmbH has developed a new DAS-Lidar system or DIAL (differential absorption) to observe profiles of ozone concentrations and extinction. In comparison to the Frohnau Lidar an additional multiphase Lidar from FU Berlin was used episodically on the Charité in the centre of Berlin observing back scatter of aerosols. In addition mixing height was determined from the back scatter profiles and compared to the meteorological fields from analyses and measurements.

The LIDAR signal represents a qualitative measurement of aerosol concentrations. The vertical measurements of ozone and PM extinction coefficients were performed by the DIAL-LIDAR near the Frohnau tower (321m) in the northern part of Berlin (Fig. 4). The measurements have been compared to the observations at the observed PM10 and ozone concentrations for the one year campaign to calibrate the extinction observed by the Lidar as an integral value.

The Lidar measurement at the Frohnau tower were automatically carried through and depending on weather the measurements were useful for 75% of the observation period.

Air mass transport analysis and application of the chemical transport model REM_Calgrid.

These one year data sets were used to analyse the levels of air pollution in and around Berlin area. In order to provide a chemical air mass classification depending on meteorological situations, trajectories have been calculated. A dense set of 3D backward trajectories were used to analyse the seasonal and episodic relationship between measurements and possible source areas. The surface contacts below 50 m height of the 3D trajectories are integrated over a 1 km^2 grid and weighted by the locally measured concentrations of all species and the given time sections. The resulting concentration matrices for all observational sites are combined and, in result, special source areas or transport paths are determined (Fig. 5).

Figure 4: Aerosol concentrations near the Frohnau tower measured by LIDAR operating at 532 nm. Backscattering coefficients range from 0 to 3x10^{-5} 1/m sr.
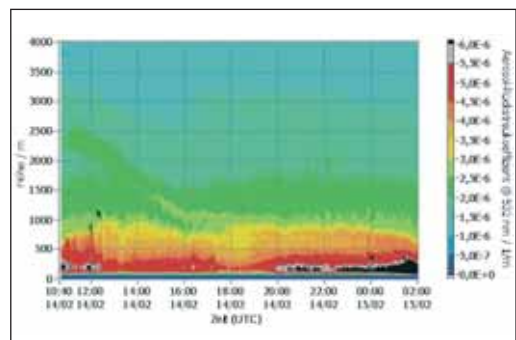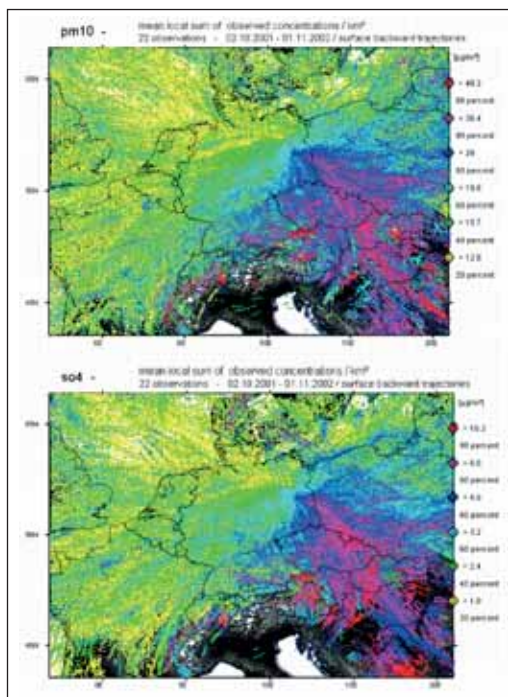
Figure.5:: PM10- (top) and Sulphate- (bottom) weighted back-trajectory statistics over all measurement sites in Berlin.

PM10 transport into the Berlin area shows distinctive seasonal and spatial patterns: besides the typical source areas South-East of Berlin down to Katowice, mainly during autumn and winter. Different species (ions, Organic-, Elemental Carbon) show distinct transport paths. Sulphate paths are predominantly determined by long range transport from South-East. The relative contribution of transport and of local production of aerosol components in the greater Berlin area is estimated by a direct compari-son of influence matrices of suburban and urban observations.

REM_CALGRID was run for the measurement period over Europe with a nest over the Berlin-Bran-denburg area with a 4 km horizontal resolution. Sec-ondary inorganic aerosol components have been cal-culated using ISORROPIA (Nenes et al., 1999) while

organic PM-constituents have been simulated using SORGAM (Schell et al., 2001). The model was applied at a modelled resolution of approximately 25 km for an area that covers Central Europe and a nested domain of about 300 km x 300 km around Berlin with a horizontal resolution of about 4 km. The model was run with 4 dynamically changing layers with the ref-erence layer the mixing height.

Comparisons between REM_Calgrid simulations and annual means of PM10 at different measurement locations show an underestimation of PM10 at traffic sites, but a good agreement at urban background sta-tions. Rural sites as well as upper air concentrations are underestimated by the CTM, as well. Sulphate fraction concentrations are for all comparison sites underestimated by about 30 percent, while Nitrate concentrations show a good agreement with the annual mean concentration. Ammonium is also well depicted at urban background stations as well as at the measurement site at 324 m height. Elemental and Organic Carbon concentrations are underestimated at traffic sites and overestimated at the urban back-ground location.

The seasonal variations of the PM-compounds are well depicted by the model.

Correlations between simulated and observed daily averages were especially large for the autumn and winter season (between 0.7 to 0.8 for Sulphate, Ammonium and PM10). Minimum correlations between observed and simulated concentrations were found for Elemental Carbon (0.4) and for Organic Carbon concentrations (0.5).

A systematic budget study for a control volume over the greater Berlin area has been performed for aerosol components using the CTM REM_Calgrid quantifying the contribution of horizontal and verti-cal advection, mixing, emissions, secondary aerosol formation, and dry and wet deposition to pollution accumulation and loss in the city of Berlin (Panitz et al., 1999).

Primary aerosols are accumulated during the whole year due to emissions, and lost due to horizon-

tal advection through the lateral boundaries of the control volume and due to dry and wet deposition. Coarse PM is accumulated mostly due to emissions and reduced mostly due to horizontal transport. Fine particles show the same behaviour.

Secondary inorganic aerosol-components, (Sulphate-, Nitrate- and Ammonium ions) are mainly accumulated by chemical production and to a much lesser extent by emissions; loss processes are mainly wet deposition processes and transport through the upper boundary of the control volume. There is also an overall net accumulation of secondary inorganic aerosols due to horizontal advection processes within the control volume over the city.

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Uncertainty analysis on the parameterization of processes at the biosphere and hydrosphere in atmospheric models (UNAPRAM)

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Objectives and Method

The goal was to examine the uncertainty of predicted trace-gas-, water- and energy-fluxes that is caused by parameterizations of processes at the biosphere-hydrosphere-atmosphere interface and by empirical plant and soil parameters. Several land surface models (LSMs) used in Germany were analyzed and categorized according to their complexity. Out of these classes representatives were chosen for detailed evaluation, sensitivity studies, and uncertainty analysis (e.g. MÖLDERS et al. 2003). The equations used in these LSMs to simulate the fluxes and state variables at the surface and within the soil were differentiated with respect to the empirical parameters the predicted quantities depend on. These derivatives together with the variance of the empirical parameters served within the framework of Gaussian Error Propagation (GEP) principles to calculate the statistical error inherent in the predicted quantities, i.e. the parameter-induced uncertainty in state variables and fluxes.

Results and Conclusions

Parameter-induced uncertainty decreases with depth. At the surface and in the upper soil, the uncertainty of predicted state variables and fluxes shows a diurnal cycle with greater values by day than at night. Soil temperatures and volumetric water content uncertainty plays a minor role except during phase transition. Then the freeze/thaw term dominates the uncertainty.

Analysis showed that not necessarily the empirical parameters having the highest percentage variance contribute the greatest to the statistical uncertainty in the predicted quantity. Saturated water potential, for instance, has a high percentage variance, but contributes less to ground-heat-flux uncertainty than porosity or pore-size distribution index. Uncertainty in hydraulic parameters, generally, contributes more to soil-temperature uncertainty than that of the thermal parameters. Thus, increasing the accuracy of hydraulic parameters offers a greater potential for improvement of soil-temperature predictions than doing so for thermal parameters. Since in many LSMs (e.g. OSULSM, HTSVS) the parameterization of thermal conductivity depends on hydraulic parameters, increased accuracy of hydraulic parameters will even reduce uncertainty in soil-heat fluxes. Some parameterizations were identified as being extremely parameter-sensitive and should be replaced in the future.

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Vertical transports of energy and trace gases at anchor stations and their spatial and temporal extrapolation under complex natural conditions

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G. Spindler[7], B. Stiller[3], J. D. Tenhunen[13], R. Vogt[11], M. Wagner[1], H.-J. Weigel[4], A. Ziemann[15], F. Zimmermann[10],
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Introduction

Improved knowledge on the atmosphere as part of the climate system, on regional weather as well as on effects of land use change and management requires better understanding of land-surface atmosphere interactions at different scales. The research project VERTIKO aimed at measuring and modelling vertical transports (momentum, energy, trace gases) in the atmospheric boundary layer as a spatial mean above inhomogeneous terrain. It questioned the appropriate spatial and temporal resolution to describe land-surface atmosphere interactions adequately. This

was investigated for a specific target area in eastern Germany comprising 30,000 km^2 from the Erzgebirge (Ore Mountains) to the Oder-Spree lake district (Figure 1). Continuous measurements and measurement campaigns were conducted at anchor stations of different land use types in the target area supplemented by data sets from external sites (test sites). Spatial integration is achieved by remote sensing (AVHRR, Landsat TM, Meteosat 6/7/8) in connection with a model hierarchy of SVAT/BGC models, a boundary-layer model and the mesoscale model (Lokalmodell) of the German Meteorological Service, DWD. Data on meteorological variables, measured

and modelled fluxes as well as site descriptions are provided by the VERTIKO data bank (to be published in 2005). Research activities included 17 individual projects (*see table in appendix*) at 13 institutions covering various disciplines such as meteorology, atmospheric chemistry, plant ecology, agricultural sciences and geosciences.

Measurements at Anchor Stations and Test Sites

Measurements at Anchor Stations
A main focus was laid on measurements at anchor stations, which are sites within the target area with continuous long-term data records including radiant and turbulent fluxes. Anchor stations (see Table 1) are Lindenberg (grassland, pine forest), Melpitz (grassland), Tharandt (mixed conifer forest, grassland) and Oberbärenburg/ Rotherdbach (spruce forest). Fluxes of momentum, sensible heat, water vapour, and carbon dioxide are determined by the eddy-covariance technique (BALDOCCHI et al., 2001).

A prerequisite of site comparisons and model validations are standardised high quality data. It was therefore important to develop procedures of quality controls applied to all anchor stations (*task UBT1, see appendix*). An approach has been developed that provides an additional tool for flux data quality evalua-

Figure 1: Schematic overview of the VERTIKO concept illustrating measuring and modelling tasks related to the target area, anchor stations, and test sites. Background: NDVI, NOAA-AVHRR).
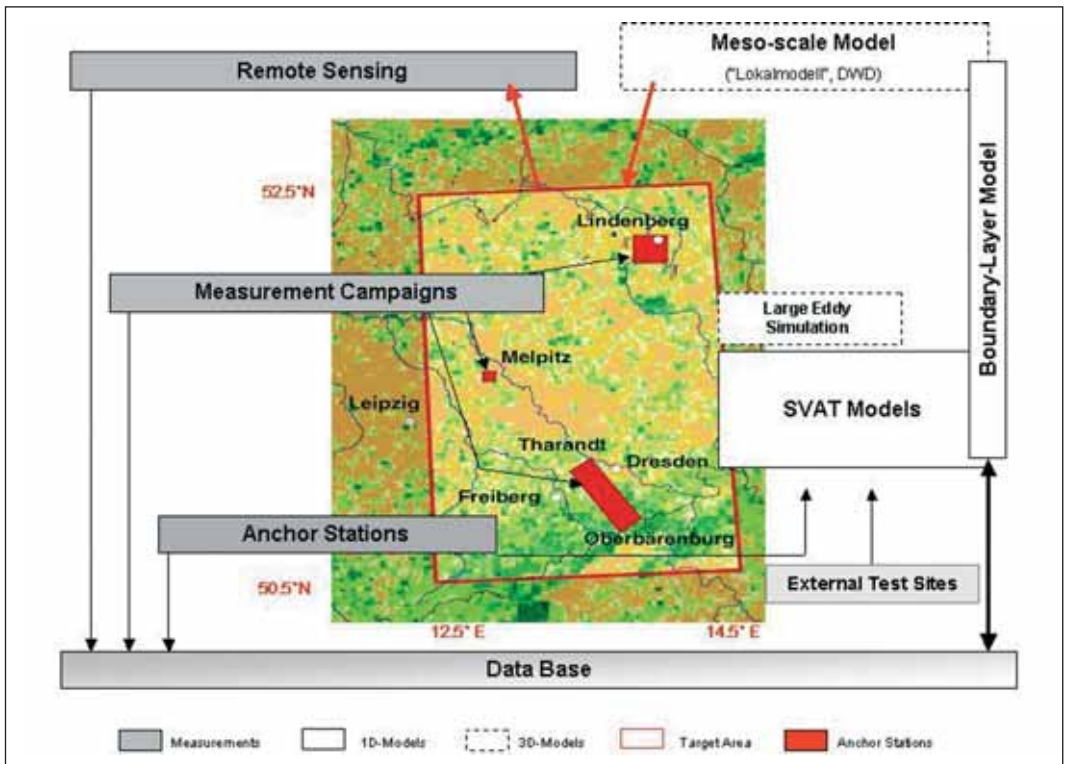
Table 1: Anchor stations in the area of investigation, ranked by altitude (Fluxes: net radiation Rn, soil heat G, momentum τ, sensible heat H, latent heat LE, carbon dioxide CO_2, nitrogen oxides NO_x).

Station	Measurement height	Operated since	Height a.s.l.	Type of surface	Fluxes measured
Lindenberg Pine	30.5 m	2000	49 m	Forest (pine)	Rn, G, τ, H (, LE)
Lindenberg Grass	12 m	2000	73 m	Grass	Rn, G, τ, H (, LE)
Melpitz	12 m	1993	86 m	Grass	Rn, G, τ, H, LE
Tharandt	42 m	1992	370 m	Forest (mixed)	Rn, G, τ H, LE, CO_2, NO_x
Grillenburg	3 m	2002	385 m	Grass	Rn, G, τ, H, LE, CO_2
Klingenberg	3 m	2004	480 m	Crop rotation	Rn, G, τ, H, LE, CO_2
Rotherdbach	24 m	1996	720 m	Forest (spruce)	Rn, G, τ, H
Oberbärenburg	18 m	1994	735 m	Forest (spruce)	Rn, τ, H
Oberbärenburg	30 m	2003	735 m	Forest (spruce)	Rn, τ, H

tion at meteorological measurement sites in complex terrain (GÖDECKE et al., 2004a, 2004b). It combines the quality assessment tools for eddy covariance measurements of FOKEN AND WICHURA (1996) with the forward Lagrangian stochastic footprint model of RANNIK et al. (2003). In a pre-processing step, the microscale aggregation model of HASAGER AND JENSEN (1999) is implemented to provide effective roughness lengths as input for the footprint analyses. This combination yields the dominating quality flag for the different observed fluxes and the relative flux contribution of each cell to the total measured flux.

The analysis can provide results for different stratification regimes, and may also be applied to produce maps of footprint averaged meteorological parameters such as the vertical wind component w. Another important output option is the determination of the contribution of each land use type to the measured flux. The procedure presented is especially useful for the interpretation of results from monitoring stations situated in heterogeneous terrain. The contribution of the target land use type to the total flux can be assessed for any user-defined period, indicating how representative the measurements are for that specific kind of surface cover. The approach can be employed

to evaluate the performance of a coordinate rotation method such as the Planar Fit approach, and in addition proves to be a powerful tool for the identification and visualisation of factors distorting the measurements. Within the VERTIKO project, this site evaluation tool has been applied to the test site Waldstein Weidenbrunnen, and to the Anchor Stations Tharandt, Lindenberg (pine forest) and Oberbärenburg. The grassland Anchor Station Melpitz was evaluated with a limited version of the approach. In close cooperation with the DEKLIM project EVA-GRIPS and partners within the European project CARBOEUROPE-IP, a state-of-the-art software tool was developed to calculate turbulent fluxes from eddy covariance raw data. Besides the usual set of correction methods, this system includes the Planar-Fit coordinate rotation method, footprint applications, and a number of additional tools to evaluate the flux data quality. The developed quality control and site evaluation tools have been chosen as reference procedures to evaluate and compare more than 100 flux monitoring sites within the CARBOEUROPE-IP.

Footprint approaches were also realised to compare different measurement systems, and to assess the data quality within measurement campaigns. For example, the scatter between CO_2-fluxes derived by a closed soil chamber system on the one hand, and eddy covariance measurements on the other, could be significantly reduced by use of a footprint filter (RETH et al., 2004). Further studies concerned the development of an approach to estimate the source areas for line source measurements, such as scintillometers. For all measurement complexes involved in the STINHO experiments (see below), footprint studies were conducted in order to assess their spatial representativeness, and thus to improve the input data set for the subsequent large eddy simulation study.

The Anchor Station Lindenberg, operated by the Meteorological Observatory of the German Meteorological Service (DWD) contributed continuous long-term measurements of energy fluxes (short wave and long wave radiation, turbulent heat fluxes, soil heat flux) over different land use types (grassland and forest) in flat terrain (*task DWD1*). Further, basic micrometeorological parameters (wind, temperature, humidity, precipitation, soil parameters) over grass and at a forest site (pine) have been performed continuously over the project period. In addition, seasonal measurements (between April and November) were carried out over water regularly every year.

After intensive quality controls the data were used to force and to validate different types of numerical models within VERTIKO (SVAT models, LES, LM, cf. chap. "4. Model Integration").

Figure 2 shows an example of the analysis of the flux measurements over the different surface types. Significant differences occur for the net radiation and the sensible heat flux while the latent heat flux is comparable in magnitude. Sensible heat fluxes about double as high as over grassland have been found over forest during different seasons in several years. Obviously, the higher available energy of pine (lower albedo) is not used for evaporation. This is also indicated by the lower evaporative fraction of forests compared to grassland.

At Anchor Station Tharandt, continuous flux measurements of water vapour and carbon dioxide are performed since 1996 within the EU project CARBOEUROPE (GRÜNWALD, 2003, Figure 3). Continuous measurements of carbon fluxes are important to assess interannual variability of terrestrial carbon sinks and to indicate important biological controls on exchange processes between land surface and atmosphere.

During the VERTIKO period (2001-2003) intermediate annual fluxes were measured in 2001/ 2002 (-576/-572 gC m^{-2}a^{-1}) and minimum uptake in the year 2003 (-438 gC m^{-2}a^{-1}) with lowest annual precipitation (501 mm a^{-1}) and a drought period in summer.

Figure 2: Mean diurnal course of net radiation, sensible and latent (turbulent) heat flux during July 2003 at GM Falkenberg (grass), at the Kehrigk (pine) forest site and over the shallow lake Großer Kossenblatter See
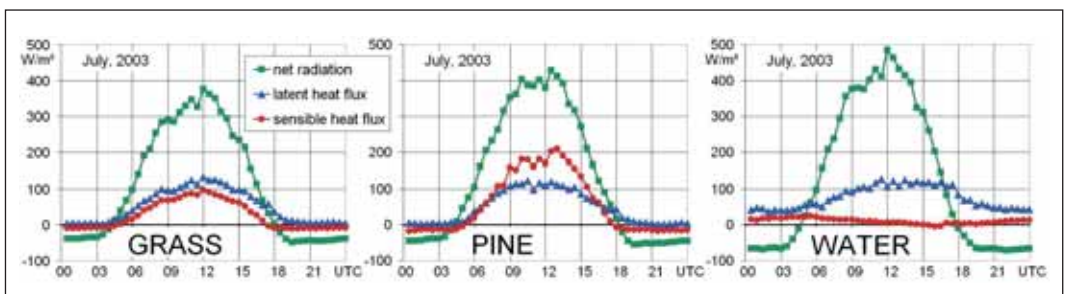
During the remaining years with sufficient water availability fluxes were best related with length of the growing season with a maximum in 1999 (192 days; mean annual T_a 9.0°C; NEE -729 gC m^{-2}a^{-1}) and a minimum in 1996 (171 days; mean annual T_a 6.1°C; NEE -449 gC m^{-2}a^{-1}). The evaporative fraction (Rn-H/Rn) was lowest at Anchor Station Tharandt when compared with the spruce site at higher elevation (Oberbärenburg) and even lower than the pine forest at Lindenberg (see Figure 12).

At Anchor Station Melpitz (grassland site) a special focus was laid on measuring and modelling nitrogen fluxes (*task IfT*). A micrometeorological aerodynamic profile technique was used to determine the net NH_3 flux (Figure 4). Concentrations of NO_X, NH_3, HNO_2 and HNO_3 have been measured by chemiluminescence and wet annular denuder techniques, respectively. Dry deposition of HNO_3, HNO_2 and also NH_3, was estimated by resistance analogy (SUTTON et al., 1998, SPINDLER et al., 2001). The N-input by precipitation (NO_3^-, NH_4^+) was quantified using a wet-only sampler. NO_3^- and NH_4^+ concentrations of aerosol particles were determined with a low flow filter sampler (upstream impactors for PM_1, $PM_{2.5}$ and PM_{10}). The size dependent deposition

Figure 3: Carbon net ecosystem exchange (NEE) from 1996 to 2003 measured at Anchor Station Tharandt. Variation of daily values (above), cumulated values (below) and numbers of annual sums (g m^{-2} a^{-1}) are shown.

velocities were modelled on the basis of the friction velocities (SLINN, 1982). The N-impact shows a high interannual variability between 8 and 24 kg per hectare and year. The N-impact by wet deposition shows relatively constant yearly amounts in the range about 8 kg. The dry N-impact caused from NH_3 shows the strongest variability. The deposition by particulate matter realizes less than 10% of the dry N-impact. A comparison with model results shows a good agreement for the Anchor Station Melpitz (GAUGER et al. 2003).

At the Anchor Stations Tharandt and Oberbärenburg in the eastern Erzgebirge, ambient concentrations of acidic and basic N-compounds were measured continuously from Sep 2001 to Dec 2003 based on simultaneous denuder and filter pack sampling (*task TUBAF*, PLESSOW et al., 2003). Precipitation and fog droplet chemistry were investigated in parallel. Average NH_3-N, HNO_3-N, HNO_2-N, and SO_2-S concentrations in air were about 0.45, 0.16, 0.06, and 1.4 µg/m^3 at Oberbärenburg and about 1.2, 0.4, 0.2, and 0.9 µg/m^3 at Tharandt. The NH_3-concentrations correlated with air temperature, global radiation, and relative air and leaf surface humidity. Concentration

Figure 4: One day of ammonia concentration measurements (2002-04-21) at Anchor Station Melpitz in three levels above ground. The flux (emission) was calculated using measurements of horizontal wind velocity and temperature in different levels over ground.
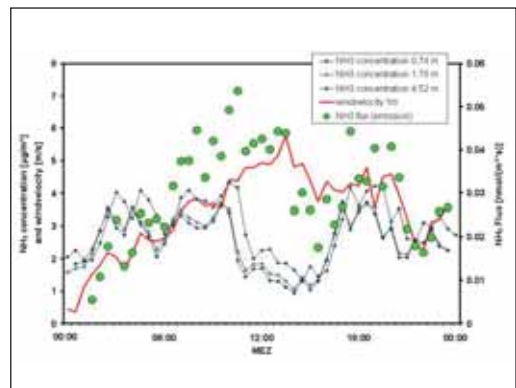
measurements above and below the canopy indicate a significant gradient for NH_3 and HNO_3. Values decreased to about 60% after passing the canopy.

Dry deposition rates for gaseous and particulate N-species have been estimated using the new developed resistance model SPRUCE-DEP (Zimmermann et al. 2004). SPRUCE-DEP is mainly based on the one dimensional model PLATIN (Grünhagel et al., 1997), but was adapted to forest sites and the special type of vegetation. Furthermore, parameterisations derived from direct micrometeorological measurements at the anchor stations were used (Queck, 2003). The modelled monthly N-fluxes at Oberbärenburg are presented in Figure 5. Fluxes of NH_3, NH_4^+ and HNO_3 exhibit distinct seasonal variations. Calculated mean deposition velocities (in cm s^{-1}) were about 1.8 for both NH_3 and NH_4^+, and 3.5 for HNO_3. Maximum deposition rates were observed for NH_3 with up to 60 mg m^{-2} $month^{-1}$.

The annual N-Flux, as sum of dry and wet deposition, can be estimated to about 30-40 kg N ha^{-1} a^{-1} for both forest sites. In 2003, the total N-Flux was considerable lower with 27 kg ha^{-1} a^{-1} at Tharandt and 24 kg ha^{-1} a^{-1} at Oberbärenburg (Plessow et al., 2004). The year 2003 was very dry with a precipitation deficit of about 40%. This resulted in reduced wet and fog deposition rates. The contribution of dry deposition (gases and particles) to the total nitrogen budget was

about 50-65% for both anchor stations.

Additional to the continuous measurements, a joint nitrogen experiment of six VERTIKO subprojects was performed in summer 2003 at Anchor Station Tharandt in order to compare measurement techniques and to complete parameter derivations for N deposition modelling. The gaseous species dominated the studied compounds with mean atmospheric concentrations of 1.2 $\mu g/m^3$, 0.4 $\mu g/m^3$ and 0.2 $\mu g/m^3$ for NH_3-N, HNO_3-N and HNO_2-N, respectively. Ambient concentrations of the particulate NH_4^+-N and NO_3-N were about 0.9 $\mu g/m^3$ and 0.3 $\mu g/m^3$. Concentration measurements above and below the canopy indicate a significant gradient for NH_3 and HNO_3. Values decreased to about 60% after passing the canopy. Dry deposition fluxes of various species were estimated from measured concentrations by using resistance models of different VERTIKO subprojects: MixForSVAT (Gaug, Oltchev et al., 2002), PLATIN (FAL, Grünhage et al., 1997), Gas_DEP (TUD1, Queck, 2004), and Spruce_DEP (TUBAF, Zimmermann, 2003). The N-fluxes, computed by Spruce_DEP for the gases NH_3, HNO_3, HNO_2 and by PLATIN for particulate NH_4^+, NO_3^-, were used for an estimation of the annual N-budget (Figure 6). Together with wet deposition, the total N-flux

Figure 5: Modelled monthly fluxes for measured N-species at Oberbärenburg, December 2001 to January 2003.
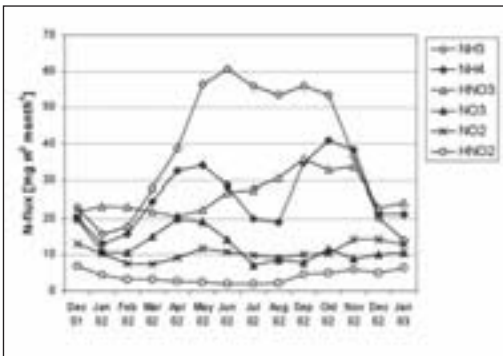
Figure 6: Different deposition pathways with an estimated annual N-budget of about 28 ± 5 kg N ha^{-1} a^{-1} in the year 2003 at Tharandt
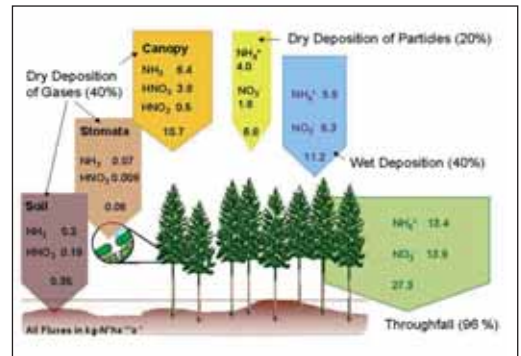
accounted for 28.2 kg N ha^{-1} a^{-1}. The campaign was a dry observation period with a cumulated precipitation rate of 25 mm in six weeks. Model simulations indicate that in such drought periods only a small amount of N-species is internally taken up through the stomata. Thus, most of the N-deposition was represented by canopy throughfall. Additional to N deposition, efflux of N species (and CO_2) from the forest soil was measured by chamber techniques (IFU, UBT2), which supported BGC and SVAT-CN model development. The role of N-deposition as suspect to support additional forest growth and C sink strength of forests is underlined by the VERTIKO results.

Supplementary Measurements at Test Sites

Test sites outside the target area (Braunschweig, Göttingen/Solling, Freiburg/Hartheim) delivered additional flux measurements and surface characteristics of land use types (esp. crops and urban areas) for model parameterisations.

The Federal Agricultural Research Centre in Braunschweig contributed measurements on a 20 ha experimental field to assess gas and energy exchange between cropped land and the atmosphere under typical agricultural management (task FAL). For the determination of high resolution fluxes, the field site was equipped with micrometeorological and analytical instrumentation (closed and open path eddy covariance systems, denuder filter samplers) as well as dynamic chambers to assess vertical fluxes of sensible and latent heat, trace gases and aerosols (for details see WEIGEL AND DÄMMGEN, 2000). Gradient and chamber measurements were in the correct order of magnitude, whereas the closed path eddy covariance system showed unacceptably small fluxes. Spectral analysis of high frequency measurements of water vapour and carbon dioxide pointed out that these flux losses were a result of inadequate performance of the turbulence criteria inside the closed path system. Correction methods based on co-

spectral analysis yielded increased fluxes. However, the energy balance could not be closed satisfactorily. The application of the open path system proved to be successful (for details see SCHAAF et al., 2003, 2004). With regard to quality assurance of the micrometeorological measurements, accuracy and representativeness of flux measurements was studied in detail. Their deviation from "the truth" is in the order of magnitude of 20% (DÄMMGEN AND SCHAAF, 2002; DÄMMGEN et al., 2004). This is at least partly due to small scale inhomogeneity of the plant soil system to which the fluxes can be attributed by foot print analysis.

At the Solling test site, an eddy covariance measuring system was developed and tested to determine vertical NH_3-fluxes above a spruce forest (task GAUG). This system allows to measure NO_x and NO_x+NH_3 alternately with time separation of a few seconds. The differences of these two concentrations are assigned to the NH_3-concentration. The measurements of vertical fluxes showed that power spectra have a maximum around 100 seconds. The missing high-frequency part of the NH_3 power spectrum is determined by comparison with the power spectrum of sensible heat flux. Measured NH_3-concentrations varied around 1.0 ppb and NH_3-fluxes ranged between –0.01 and 0.10 $\mu g/m^2 s$. Long-term NH_3-balances can not be disposed yet.

At the test site Hartheim in the southern upper Rhine plain near Freiburg, long-term forest- and hydrometeorological data (1978-2003) are recorded above and within a Scots pine forest (*Pinus sylvestris* L.). The objectives have been to analyse the impacts of the growth dynamics of the forest (height growth and thinnings) on both its energy fluxes and variables for the aerodynamic surface roughness. In addition, the weather conditions during the VERTIKO period 2001-2003 were evaluated using the long-term Hartheim data. A specific hierarchical test model was developed for the quality control and management of the Hartheim data, which permits to mask missing and critical values and to mark them by different quality flags. Turbulent heat fluxes were

calculated by both the Bowen ratio energy balance method and the aerodynamic profile method. The aerodynamic surface variables roughness length z0, zero-point displacement d and friction velocity u_* were calculated by an iterative linear-logarithmic approach.

Average annual air temperature above the Hartheim Scots pine forest increased in the period 1978-2002 (0.069 K·a^{-1}). Though the average annual net radiation Q^* decreased (KESSLER AND JAEGER, 2003), the annual average of the turbulent latent heat flux λE did not reveal a significant trend, which could be explained by the slight increase of the annual gross precipitation and the reduction of the leaf area index of the Scots pine forest by thinnings. As a result the annual average of the turbulent sensible heat flux H decreased, which led to a decrease of annual averages of the Bowen ratio β from 0.76 (1978) to 0.55 (2002). Temporarily occurring summer drought of variable intensity was characteristic of the Hartheim Scots pine forest during the investigation period.

Long-term growth dynamics caused a significant increase of d with small fluctuations of z_0 and u_*, whereas for periods after thinnings a short-term

Figure 7: The test site Hartheim is used to compare the measurements years of VERTIKO with the long-term trend of available energy, sensible and latent heat.

reduction of d with a simultaneous increase of u_* and z_0 could be detected. Altogether linear relationships with the mean stand height h were found in the investigation period: $d = 0.72h$ ($R^2 = 0.77$) and $z_0 = 0.09h$ ($R^2 = 0.13$).

Despite the comparatively large distance between the Hartheim site and the VERTIKO target area, the time series of the Hartheim energy fluxes could be used as a reference to evaluate the representativeness of weather conditions during the VERTIKO period 2001-2003 (Figure 7). While the year 2002 was characterised by average weather conditions, the extreme heat in summer 2003 led to comparatively high annual averages of Q^*, H and β as well as a lower annual average of λE than for the reference period. In 2001, the annual averages of Q^*, H and λE were lower (Q^*, H and β) resp. higher (λE) than for the reference period.

Investigation of Heterogeneity Effects during Measurement Campaigns

Measurements at anchor stations were intensified during special observation periods (SOPs). They included the experiment STINHO ("Structure of Turbulent fluxes under INHOmogenous surface conditions") in 2001 and 2002 investigating effects of microscale heterogeneity on surface energy fluxes, and the advection experiment in the Ore Mountains (MORE) in 2001 and 2003 examining effects of advection on fluxes through a forest volume. Further, campaigns based on Sodar measurements accompanied the measurement campaigns and included also urban areas in the southern upper Rhine plain near the test site Hartheim (task ALUF1).

The STINHO Experiment

The STINHO experiment consisted of three parts: the acoustical and optical probing (task ULLIM), airborne turbulence measurements with the Helipod (task TUB) and highly resolved large eddy simulations (task UH). This experiment should investigate how
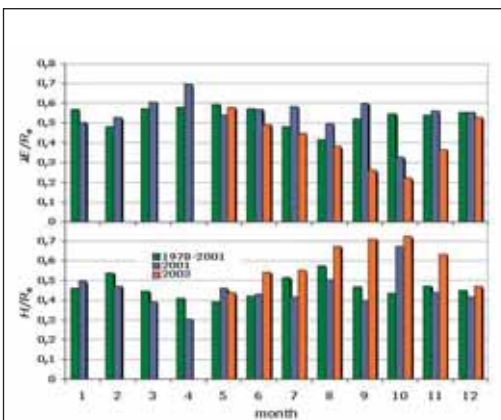
the heterogeneity of the surface modifies the vertical turbulent exchange and specifies the amount of the horizontal heat fluxes. Combined observations and simulations are used to quantify the horizontal divergence of sensible heat fluxes under heterogeneous surface conditions.

Two measurement campaigns were realised inside the VERTIKO target area, STINHO-1 at Anchor Station Melpitz and STINHO-2 at the observatory of the German Meteorological Service in Lindenberg.

During STINHO-2 intensive observations concentrated on the morning boundary layer development above a heterogeneous surface. Helipod flights on four different days were carried out in the early morning shallow boundary layer. The structure of the ground-based inversion and the development of flat convection during the morning transition were observed on grid-flights at low altitude. Highly non-stationary low-level jets were observed in connection with intermittent turbulence and steep ramps with

Figure 8: First illustrations of the high-resolution study for the STINHO2 experiment:
A) Photo of the measurement area Lindenberg;
B) Tomogram of acoustic virtual temperature and wind speed (arrows);
C) near surface (z=2 m) cross section through the field of the simulated vertical heat flux.
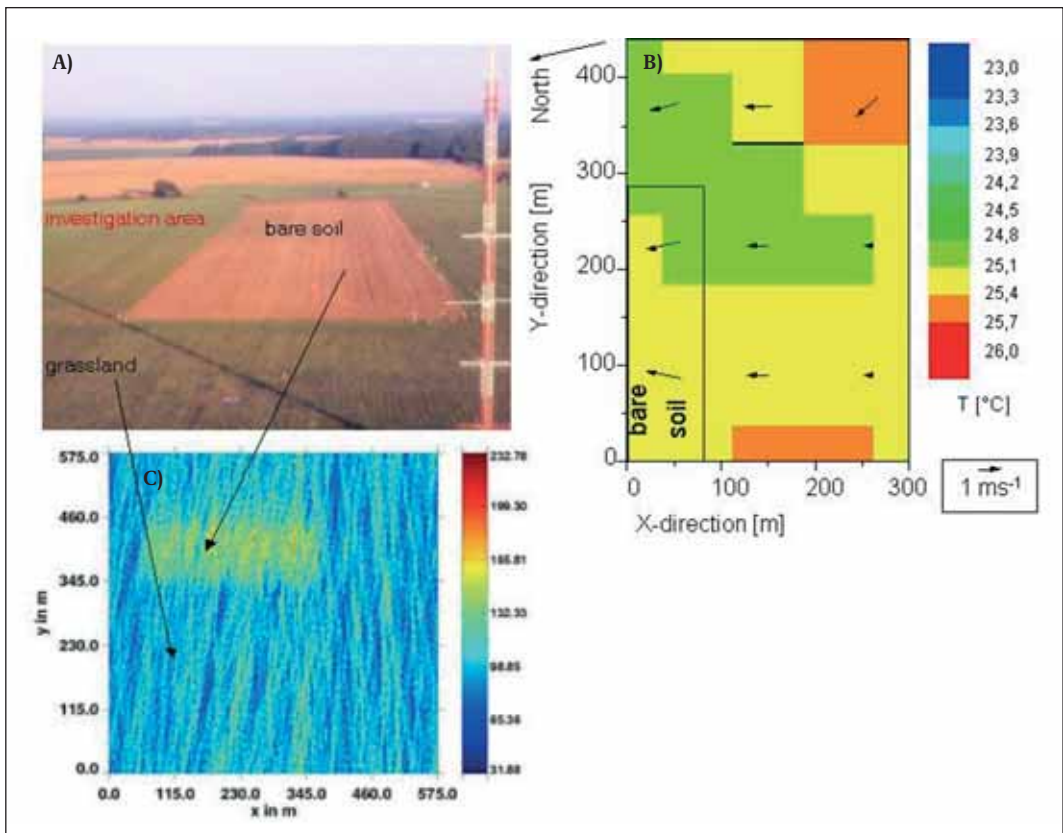
extremely large vertical gradients of potential temperature and humidity.

The database contains all ground based and airborne measurements as well the radiation and energy balance, and the height of the convective boundary layer. These parameters were summed-up in particular for the LES initialisation and validation. The realised set-up of the measurement techniques provided an image of surface/atmosphere interaction parameters and processes which corresponded with the (horizontal) grid structure of the LES model. Combining the experimental with the highly resolved numerical data, the energy transfer under inhomogeneous surface conditions was analysed (Figure 8). These investigations at the micro-α scale demonstrate how heterogeneous surface conditions modify the vertical turbulent heat fluxes.

The combination of the STINHO database and the simulation results enables the verification of the representativeness of long-term flux measurements at the anchor stations inside a heterogeneous landscape (ARNOLD et al., 2004), and to generalise the experimental data (see also "4 Model Integration").

The Advection Experiment

The standard methodology used by the global community (FLUXNET) measuring net ecosystem exchange (*NEE*) of CO_2 is relying on the eddy covariance (EC) technique and usually ignores the advection terms in the conservation equation. Recent studies however show that the neglected terms play an important role in the carbon budget especially in forest canopies and that they may be responsible for the underestimation of nighttime fluxes of CO_2 in forests rather than measurement errors. Within the frame of VERTIKO two extensive measurement campaigns have been performed in September/October 2001 and from May to October 2003 to investigate the influence of non-turbulent horizontal and vertical advection processes on the NEE of CO_2. During the campaigns the permanent measurement facilities at the Anchor Station Tharandt have been substantially enhanced by additional towers to accurately acquire the horizontal and vertical CO_2 concentration gradients in a control volume.

The results of the first (short term) campaign, published in FEIGENWINTER et al. (2004), show that, by considering the advection terms into the conservation equation, *NEE* was reduced by 15% during a 20 day period (Table 2).

The second (long term) campaign established the fact that the advective fluxes significantly increase the nightly CO_2 source and thus reduce the carbon sink of the forest.

Mean advective fluxes are in the same order of magnitude as the turbulent EC-flux of CO_2, however there is a large scatter originating from a large day to day variability. The horizontal advection turned out to be height dependent with its maximum in the lower trunk space and practically disappeared at and above the canopy top.

Table 2: Mean sums (DOY 263–283) of Carbon flux in g C m^{-2} per period of relevant variables for NEE for MORE I.

	00:00-08:00	08:00-16:00	16:00-24:00	**Total day**
horizontal advection	-1.78	0.80	-1.25	**-2.23**
vertical advection	1.52	-0.20	1.19	**2.51**
storage change	-0.08	-0.19	0.27	**0.00**
EC-flux	0.73	-3.00	0.43	**-1.84**
Total	**0.39**	**-2.59**	**0.64**	**-1.56**

There are still many methodological problems in this subject and only a few studies for comparison. The results may therefore only be characteristic for the specific site. However, the current trend in experimental micrometeorology to design measurement set-ups to probe an entire soil-vegetation-atmosphere volume with more than one single tower confirms that including advection processes into budget considerations is one of the most important issues of recent micrometeorological research activities.

Model Integration

Model integration in VERTIKO aims at both integrating physical and chemical processes as well as integrating fluxes from heterogeneous surfaces (microscale to mesoscale). The atmosphere integrates processes at all scales but feeds back to the full heterogeneity of real landscapes. In order to understand and predict processes at the mesoscale, models should therefore account for all potentially relevant controlling factors resolved for heterogeneous surfaces at smaller scales. The model hierarchy being developed in VERTIKO will serve as a tool to test appropriate spatial resolutions and sufficiently detailed parameterisation of patchy landscapes.

SVAT and BGC Models

In a first step, SVAT ("soil vegetation atmosphere transfer") and BGC (biogeochemical) models specified for different land use types and predicting various physical and chemical factors were described and compared (Falge et al., 2004). Results of the investigated energy fluxes were within an acceptable range for all models. However, the comparisons also revealed that model validations by measurements (taken from field campaigns) require assumptions on energy balance closure. The energy balance is a priori closed in the models but this was not the case for measurements. Within integrating physical and chemical fluxes in models, the goal of one sub-project was providing simple, biologically-based models of soil CO_2 and N_2O emissions as lower input in atmospheric transport models for the VERTIKO target area (*task UBT2*). For model development and validation CO_2 and N_2O emissions of meadow soil, bare soil, and forest soil were measured with a photo-acoustic infrared monitor in closed chambers. Both, the level of CO_2 emissions and N_2O emissions varied between the different land use types. In general the meadow and bare soils showed higher CO_2 and N_2O fluxes than forest soils.

A non-linear regression model for the calculation of CO_2 emission was adopted for the use on different land use types, and for regional application. In addition to soil temperature and soil moisture, the parameters pH and root mass were included in the model. As an unexpected parameter the time after a rain event was identified to affect the soil CO_2 emission. The model overestimated the observed fluxes during and within four hours of the last rain event. Conversely, after more than 72 hours without rain, the model underestimated the fluxes. Between four and 72 hours after rainfall, the regression model of soil CO_2 emission explained up to 91% of the variance. For N_2O emissions a non-linear regression model (DenNit) was developed for meadow, forest, and bare soils. Only six parameters (soil temperature, soil moisture, pH, nitrate and ammonium availability, and the time to the last rain) were needed as model input. The model explained 81% of the variability in soil N_2O emission of all field measurements, except for data with short-term soil water changes, namely during and up to two hours after rain (Figure 9).

The models provide a sound description of soil CO_2 and N_2O emissions in a biologically based framework, and can be adapted for future use in boundary layer or in atmospheric transport models.

An one-dimensional multi-layer SVAT Model (MixFor-SVAT) was further developed to describe the energy-, water-, CO_2- and NH_3-exchange between vertically structured tree stands and the atmosphere (*task*
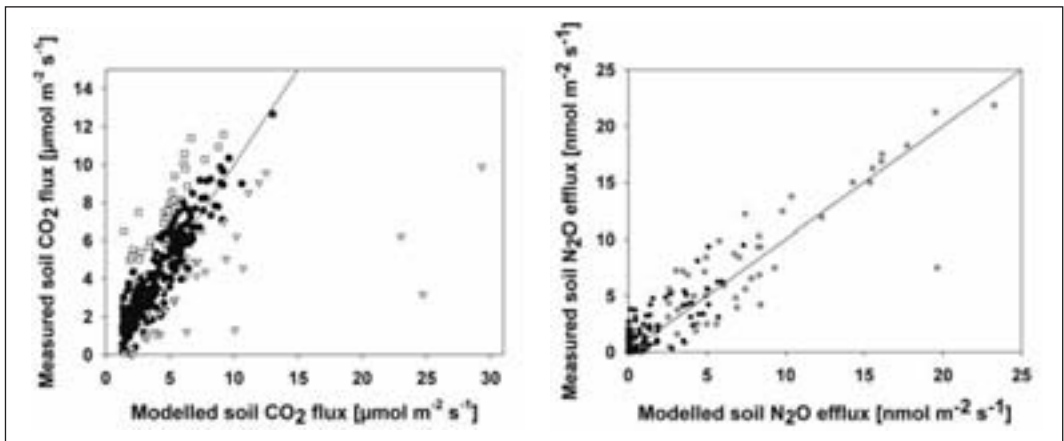
Figure 9: Comparison of modeled and measured soil CO_2 (A) and N_2O (B) efflux evaluating the temporal effect of last rain event. Lines are 1:1 lines. Slightly modified from RETH et al. 2004a,b. (A) Root mean square error (RMSE) of the model results was 0.89 μmol $m^{-2} s^{-1}$. Data not effected by rain or drought (dots, n=240). During and up to 4 hours after a rain event (triangles, n=44) the model overestimated the measured CO_2 fluxes. After a dry period of more than 72 hours (squares, n=39). B) RSME was 0.58 nmol $m^{-2} s^{-1}$. Data between 2 and 8 h after the last rain (squares, n=132), more than 8 h after the last rain (dots, n=271).

GAUG; OLTCHEV et al., 2002, 2003, GRAVENHORST et al., 2004). Simulation of NH_3 exchange between different forest canopy layers and the atmosphere is based on a modified Sutton-approach of NH_3 "compensation point" concentrations. It allows to take into account both emission and dry deposition of NH_3 (cuticular and stomatal transfer of NH_3 by different sublayers of overstorey and understorey vegetations, and soil surface transfer). Comparisons of the MixFor-SVAT model results with experimental results at anchor stations during field campaigns SOP1 and SOP2 (2001-02) showed that it allows to describe energy and water fluxes very well under various environmental conditions (FALGE et al. 2004). The modelling approach for simulation of NH_3-fluxes was tested for the experimental site in Solling and the Anchor Stations Tharandt and Oberbärenburg (see under 2.1: nitrogen experiment). It showed good correlation with measured data. The annual deposition rate of NH_3 into forests ranged between 3 and 10 kg NH_3/ha year.

The SVAT model PLATIN (Plant Atmosphere Interaction, see GRÜNHAGE AND HAENEL, 1997) was

upgraded and applied to simulate energy, gas and particle exchange between vegetation and atmosphere (*task FAL*). PLATIN was able to reproduce the energy balance for different ecosystems appropriately (FALGE et al., 2004). It was used to determine depositions of atmospheric N and S species to arable crops. A yearly mean deposition of 27 kg $ha^{-1} a^{-1}$ N and 9 kg $ha^{-1} a^{-1}$ S was calculated (SCHAAF et al., 2005). PLATIN was also used to estimate ammonia losses from the vegetation/soil system after a herbicide treatment of the plants (MANDERSCHEID et al., 2004).

Biogeochemical (BGC) models were used for spatial scaling of nitrogen fluxes from forests and agricultural soils (task FZK). The project focused on the further development and application of two process oriented models, here the DNDC (for agricultural soils) and the PnET-N-DNDC (for forest soils) models, which allow the simulation of the biosphere-atmosphere exchange between soils of terrestrial ecosystems and the atmosphere. Both models were improved with regard to the numerical description of soil hydrology and of processes involved in the

production and consumption of N trace gases in soils. After successful validation of the models (BUTTER-BACH-BAHL et al., 2004; KIESE et al., 2004), for which partly datasets obtained in the VERTIKO project were used, the models were coupled to Geographic Information System (GIS) for calculating a regional inventory of N trace gas emissions from soils in Saxony (KESIK, 2002; MIEHLE, 2002; BUTTERBACH-BAHL et al., 2002, 2004).

The GIS provided all relevant datasets necessary for model initialisation (e.g., soil properties and field management) and driving (e.g., daily climate data, atmospheric N deposition) in detailed spatial and temporal resolution (BUTTERBACH-BAHL et al., 2004). Total annual N_2O-emissions from agricultural soils in Saxony ranged from 0.5–26.0 kg N_2O-N ha^{-1} yr^{-1} and

were calculated to amount to about 5475 t N_2O-N yr^{-1} in the year 1995, which compares quite well with previous estimates based on the IPCC approach (4892t N_2O-N yr^{-1}). Compared to the agricultural soils, N_2O-emissions from forest soils in Saxony (range: 0.04–19.7 kg N_2O-N ha^{-1} yr^{-1}) were much lower and amounted to 1011 t N_2O-N yr^{-1}. In comparison with other sources of N_2O in Saxony our estimates show, that - even in such a highly industrialised region like Saxony – soils contribute more than 50% to the total regional N_2O source strength. Simulated emissions of NO from the agricultural and forest soils were approx. in the same magnitude than for N_2O (Figure 10).

The modelled NO-emission rates ranged from 0.4–26.3 kg NO-N ha^{-1} yr^{-1} for the agricultural soils and 0.04 28.3 kg NO-N ha^{-1} yr^{-1} for the forest soils with total

Figure 10: Estimated mean annual NO-emission rates from soils in Saxony in the year 1995.
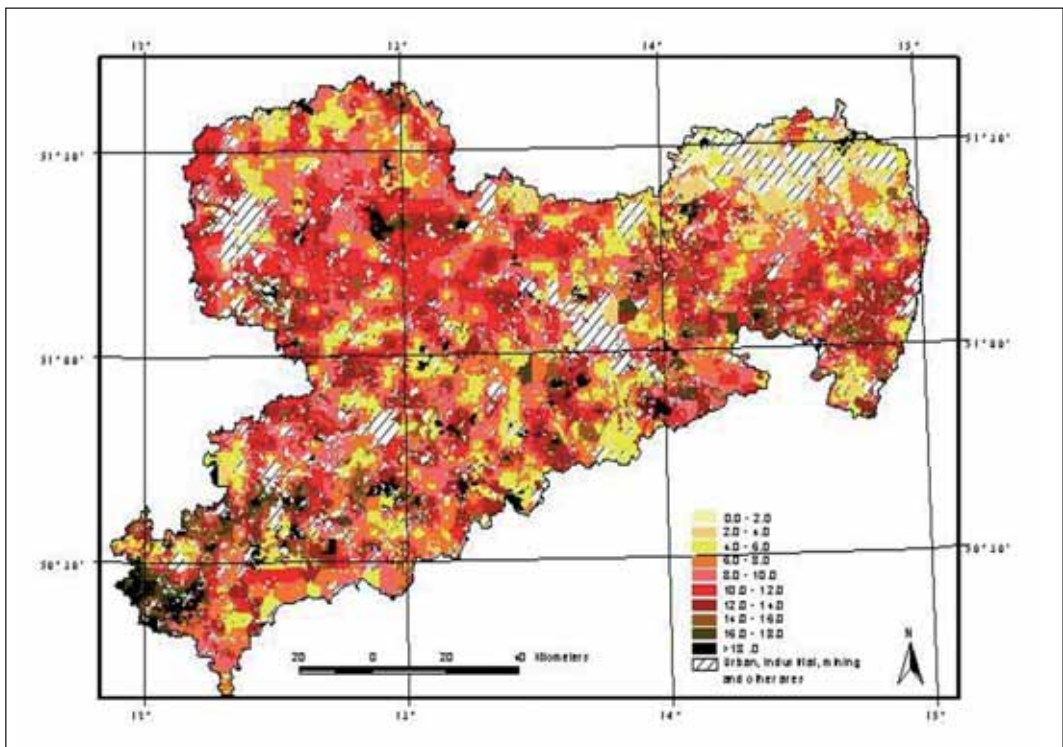
emissions of 8868 t NO-N yr^{-1} (agricultural soils) and 4155 t NO-N yr^{-1} (forest soils). Our results indicate that the agricultural and forest soils were a significant source, which contributed 17.9% of the total NO$_x$ emissions from various sources in Saxony. Furthermore, a series of sensitivity tests were carried out, which demonstrated that variations in soil organic carbon content (SOC) and soil texture significantly effect the modelled N-trace gas emissions from agricultural soils at the regional scale, whereas, in addition, for forest soils also the soil pH is a sensitive factor. Finally, multi-year simulations were conducted for the region

with observed meteorological data from 1994-1996. The results demonstrated that the modelled interannual variations, which were obviously induced by only the climate conditions, in the N-gas emissions were as high as 36%. The high interannual variations imply that multi-year (e.g. 5-10 years), instead of single baseline year, simulations would produce more reliable estimates of mean soil N$_2$O emissions at regional scale. With respect to the Kyoto protocol this means that the mean N$_2$O-emissions from soils in the period 1988-1992 should be evaluated instead of focusing on a single year, 1990.

Figure 11. Modelling effects of the structure of vegetation on turbulent fluxes H and LE as demonstrated with the ABL model HIRVAC: (left panels) – radiation transfer according to Beer's law, (right panels) – radiation transfer with randomly distrubuted sun flecks.
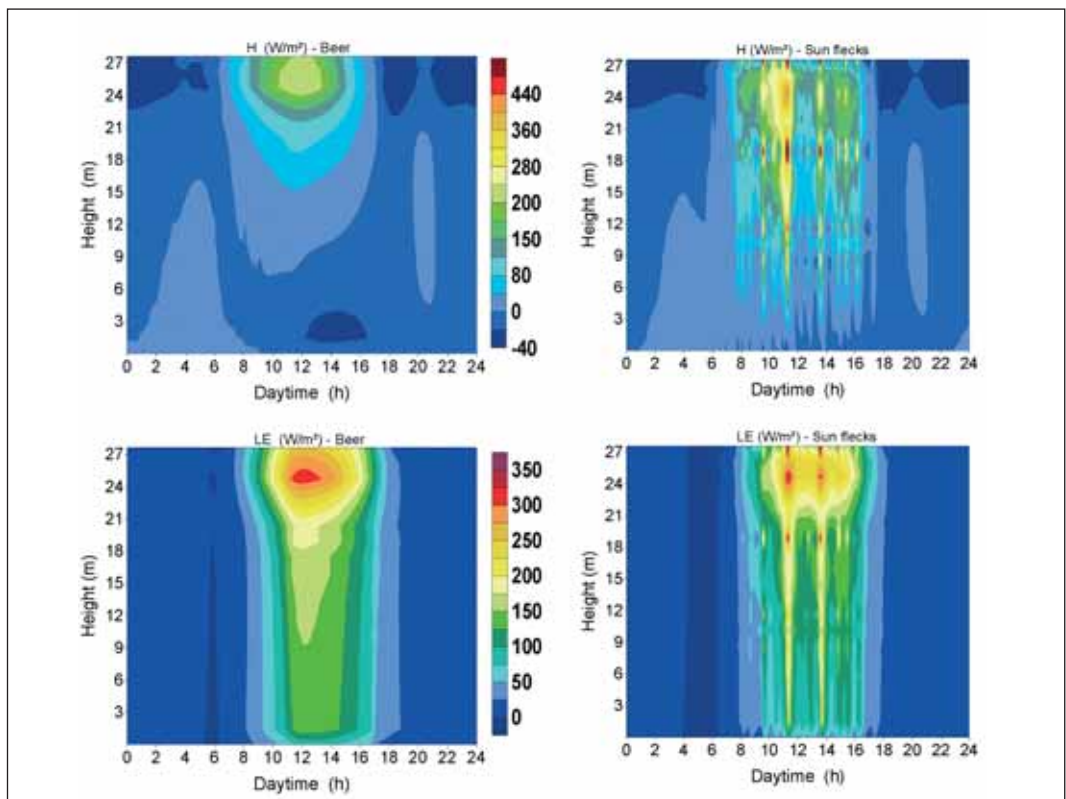
Boundary-Layer Model

The PBL-model HIRVAC (HIgh Resolution Vegetation Atmosphere Coupler; GOLDBERG AN BERNHOFER, 2001) is used as a test platform for SVATs as applied in mesoscale modelling. It consists of up to 120 horizontal layers from soil to the lower troposphere including layers of different leaf area densities coupled to the leaf gas exchange model PSN6 (from partner project *UBT2*, see HARLEY AND TENHUNEN, 1991). Within VERTIKO, an interception module and a multi-layer ground water module (FEDERER, 1995) has been included. HIRVAC was then applied to investigate the variability of energy and trace gas fluxes (mainly H_2O and CO_2) for dominant land uses. Measurement sites that provide data for parameterisation and validation were the Anchor Stations Tharandt in the lower mountain range (spruce), the grassland site Melpitz and two sites at Lindenberg (grass and pine), situated in the lowlands of northern Saxony and eastern Brandenburg, respectively. To consider effects of sunflecks in canopies a vertical and temporal variable random function was applied (Figure 11). Based on co-operation with UBT2 and the DEKLIM project EVA_GRIPS new parameter sets of the PSN6 model for beech and different crops were applied. The turbulent fluxes of latent heat L.E, sensible heat H and carbon dioxide F_{CO2} were simulated for the SOP2 period and compared with measurements of the anchor stations. The results are in a good agreement for H and L.E, derived from energy balance residuum (L.E - measured) and flux-gradient-relationship (model), as well as for F_{CO2}. Comparison between directly measured L.E and simulated latent heat from gas exchange module PSN6 showed an adequate agreement with more scattered data due to cumulative variance of single atmospheric input parameters. Besides, the comparison between measurements and simulation showed that the inclusion of a sunfleck parameterisation in the light transfer scheme seems to be a plausible way to simulate more realistic sensible and latent heat fluxes.

The comparison with measurements also showed that it is reasonable to distinguish two methods in HIRVAC for calculation of the latent heat flux – the "SVAT mode" and the "ABL mode". The first mode is based on the scaling up of single-leaf transpiration to canopy scale calculated with the gas exchange model which is comparable with outcomes from SVAT-BIGLEAF approaches and therefore suitable for integration in SVAT comparisons. The second one uses the flux gradient relationship to get the latent heat flux towards to the gradient of humidity, which is comparable with outcomes from other ABL models. In further studies results from SVAT models should be compared with HIRVAC output to assess the effect of high resolved canopies on area-averaged fluxes in contrast to the output of a common one-layer SVAT / BIGLEAF approach. In addition, results from canopy and SVAT models must be compared with HIRVAC output to quantify effects of interaction between the vegetated surface and the ABL environment on the control parameters of canopy transpiration.

Spatial Integration and Mesoscale Modelling

A prerequisite of spatial integration was the examination and comparison of parameters and fluxes related to different land use types. This was performed using the continuous measurements at anchor stations. For the surface parameters of different vegetation types clear dependencies of d and z_o on wind speed and surface characteristics in the vicinity of the towers could be described. Further, consistent differences in the evaporative fraction between forest and grassland could be demonstrated (Figure 12): Smaller albedo of forests leads to larger net radiation, larger sensible heat flux and larger Bowen ratios – despite very similar evapotranspiration. Thus, latent heat flux differs only slightly between land use types. Whether this holds also for periods of water shortage has to be clarified.
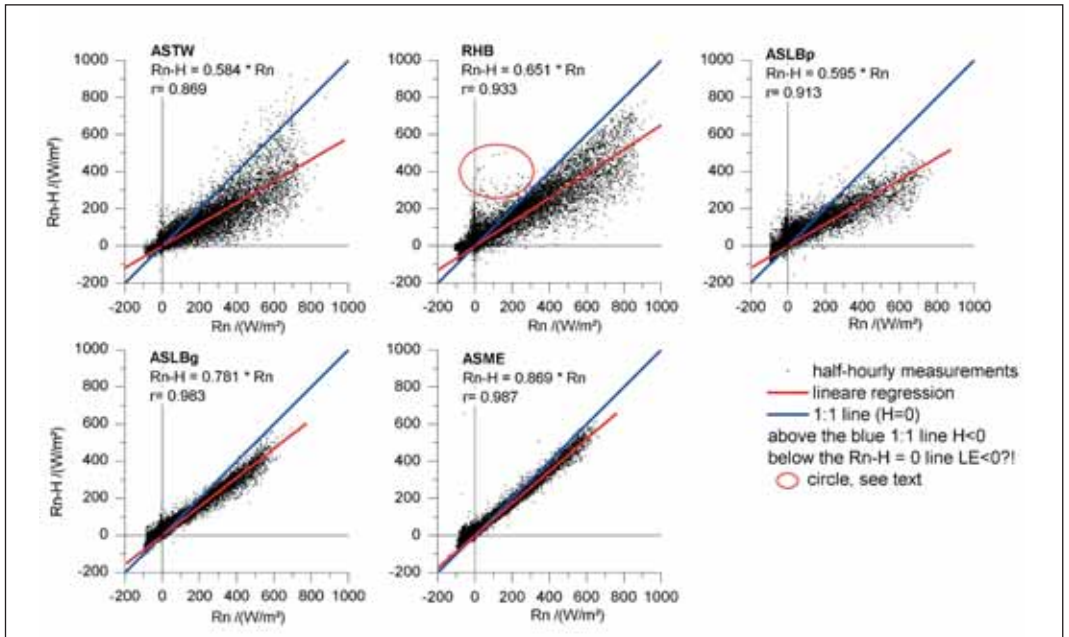
Figure 12: Evaporative fraction measured above spruce forests (Tharandt and Rotherdbach/Oberbärenburg), pine forest (Lindenberg) and grass (Lindenberg and Melpitz); red circle indicates fog at Rotherdbach/Oberbärenburg.

Figure 13: Tharandt 2001: Airborne measurements during STINHO-1 campaign with the aircraft Do 128.
The first step for calculating area-averaged turbulent surface fluxes over different surface types in a heterogeneous terrain was the distinction of e.g., wood and grassland by surface temperature.
In the second step we calculated the area averaged turbulent surface heat flux using the inverse method. The heat flux above forest was larger than over grassland. The heat flux above the forest decreased more rapidly with height than over grassland.
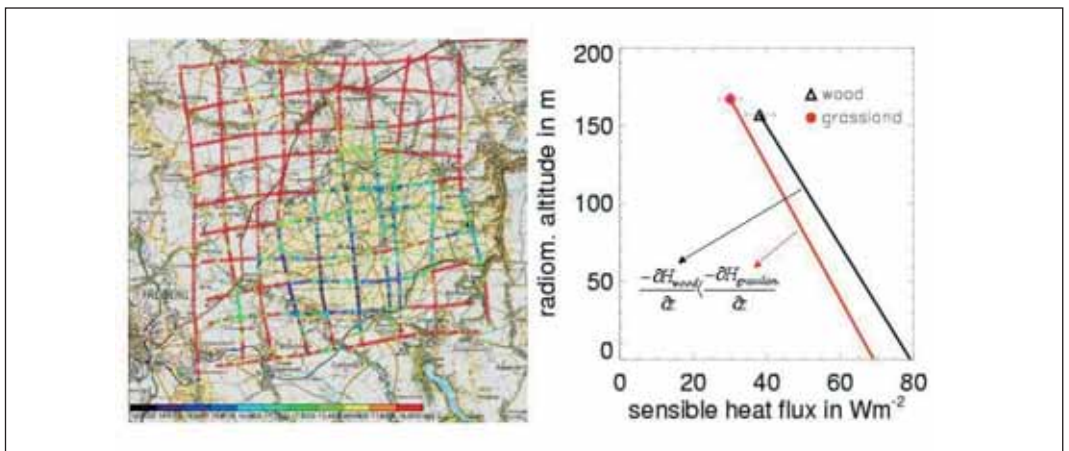
In the vicinity of the anchor stations effects of land-scape heterogeneity on surface parameters and fluxes were described by air-borne measurements linking the anchor stations with the surrounding landscape and spatial modelling (*task TUB*). The flight patterns support area-averaged turbulent fluxes and were performed during VERTIKO campaigns in 2001 and 2002 as well as in cooperation with the project DEKLIM/EVA-GRIPS at LINDENBERG in 2003 (LITFASS 2003). In total 40 hours of flight measurements with the helicopter-borne turbulence probe Helipod (Figure 13) and the research aircraft Do 128 were performed. The flights were carried out over heterogeneous terrain near the LITFASS site and the anchor stations of Melpitz and Tharandt. During the flights the turbulent fluxes, statistics, and characteristics of the atmospheric boundary layer (ABL) were measured with high accuracy. Several different flight patterns at various heights were performed in order to quantify the influence of the heterogeneous surface on the energy transport (heat, humidity, momentum) in the lower atmosphere. In addition to the horizontal flights, vertical sounding was performed using slant flights at several locations within the investigation area. The flights cover large areas in comparatively short time. Therefore, the representativeness of ground-based observations (stations, a tower, scintillometers, windprofilers, and other remote sensors) could be checked. Area-averaged surface fluxes derived from the Helipod measurements are necessary to define an averaging strategy for ground-based flux measurements. The results are also needed for the initialisation and verification of numerical atmospheric models.

The flights near Tharandt revealed that the influence of different surfaces (forest and grassland) was detectable well within the convective boundary layer (CBL). This result is contradictory to the 'textbook idea' of a low blending height and a well mixed CBL. Using the recently developed, very accurate LLF+IM method (low-level flights combined with inverse modelling), for the first time the surface fluxes of

individual surface types within a heterogeneous terrain were derived from flight measurements. The spatial representativeness of heat fluxes on the basis of single-tower measurements and the mechanism of the energy imbalance problem are investigated using the LES model PALM (*task UH*; KANDA et al., 2003). The results of this study suggest the limitation of single-tower measurements with temporal eddy-correlation even above homogeneous surfaces and the necessity of an up-scaling algorithm derived by additional investigations with advection experiments and a multi-tower approach, respectively.

The typical eddy correlation flux based on a point measurement systematically underestimates the 'true' flux (~20% for low wind speeds). Increasing wind speed reduces the imbalance as well as an increasing averaging time, whereas increasing the observation height increases the imbalance (Figure 14). The imbalance could be attributed to local advection effect caused by slowly moving turbulent organised structures (TOS). These TOS are linked with divergences of the horizontal heat flux (ZIEMANN et al., 2004, RAABE et al., 2004), which produce an additional term in the energy balance observed for single grid-volumes.

Figure 14: The imbalance study with LES has demonstrated that horizontally averaged turbulent fluxes based on point measurements systematically underestimates the 'true' flux (negative imbalance) in the convective boundary layer even under ideal site conditions.
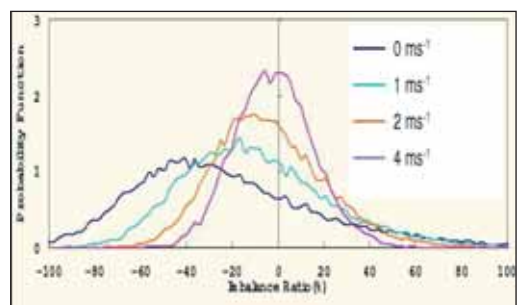
Under heterogeneous surface conditions the mesoscale transport, induced by the surface inhomogeneities, has been detected as one of the significant reasons for the energy imbalance problem (INAGAKI et al, 2004).

The additional mesoscale transport is part of the thermally induced mesoscale circulation (TMC) induced by surface flux heterogeneities. TMC's on scales larger than the boundary layer height significantly modifies the boundary layer structure:

+ **The surface (1D-)inhomogeneities induce periodic mesoscale circulations (LETZEL und RAASCH, 2003) whose amplitude depend on the thermal surface forcing and intensity.**
+ **The turbulent kinetic energy increases markedly and remains significantly higher than in the homogeneous case.**
+ **The horizontal turbulent exchange processes are significantly increased Increasing wavelength as well as increasing amplitude of the inhomogeneities increase the horizontal turbulent exchange (HEROLD, 2002; UHLENBROCK et al., 2004).**

Against previous assumptions a marked influence of surface heat flux inhomogeneities on vertical exchange processes within the ABL was not detected. Spatial integration up to the VERTIKO target area was achieved by measurements from satellites combined with modelling schemes of radiation and energy fluxes. One of the main targets was the quantification of energy exchange between surface and atmosphere with remotely sensed data and the designation of areal means for specific regions within the VERTIKO target area as well as their spatial and temporal variability (*task TUD2*). Special emphasis was put on validation with ground measurements of anchor stations. The knowledge about the accuracy of the applied analysis tools is an important requirement for model improvement and allows the specification of uncertainty ranges.

Based on NOAA-16 AVHRR data instantaneous energy balance components were calculated for 3 special observation periods within 2001-2003 for the target area with SESAT. SESAT is a modular structured analysis scheme for meteorological satellite data described in detail in BERGER (2001). For the determination of surface properties a 7-day sampling of the spectral reflectances removes the impact of clouds and enhances the surface parameterisation for the energy flux determination.

The validation of the SESAT results was carried out with half-hourly ground truth data of the Anchor Stations Lindenberg, Melpitz and Tharandt and the 11x11 kmÇ pixel average around the central 1x1 km^2 pixel for each site. The comparisons are summarized in Table 3, where the overall RMSEs [Wm^{-2}] for the single components are listed. They vary for the single sites and decrease for cloudless cases. The deviations seem high despite several improvements within the analysis tool.

Table 3: Validation results for satellite derived and ground based measurements, respectively, of radiant and turbulent fluxes at anchor stations of VERTIKO.

number RMSE	number RMSE	number RMSE	number RMSE	number RMSE	number RMSE
insolation	net radiation	latent heat residual*	latent heat	sensible heat	soil heat flux
433 153	373 115	246 116	302 112	395 119	338 21.1

residual of ground measurements

Once more it became apparent that the validation of pixel-based data with point measurements at ground is problematic for many reasons. Beside the need to use a pixel average instead of the exact location (and risking a mixed information in case of heterogeneous terrain) the dimension of the error of ground truth data is mostly unknown. Especially for turbulent flux data the uncertainties are high and the closure problem further complicates validation. On the other hand sensitivity studies showed that uncertainties within the computed surface parameters (LAI, z_0, vegetation height) may lead to considerable deviations, too.

Determination of the vertical turbulent fluxes for the entire target area is achieved by mesoscale modelling. For this the "Lokalmodell" (Local Model, LM) of the German Meteorological Service (DWD) has been applied to the VERTIKO domain (*task DWD2*). Further the error bars of these fluxes due to faulty external parameters and model resolution should be investigated using surface parameters provided by remote sensing. The sensitivity studies showed that, e.g., the surface albedo is mainly dependent on the value of the plant albedo during the vegetation period. The influence of the soil albedo plays only a marginal role. The plant albedo in the LM is relatively high with a value of 0.15, especially for forest areas. By reducing the value to 0.1 the derived surface albedo decrease by about 25% in the areal average. This change has the strongest influence on the sensible heat flux, which increases at about 8%. The effects are lower on the latent heat flux and the short-wave radiation balance. The averages of this values rise only at approximately 3.5%. The change of the plant albedo does not have relevant effects on the long-wave radiation balance and the temperature. A better agreement with the measurements from the anchor station Tharandt (land use: spruce) was obtained for the radiation balance with lower albedo. The comparison of the surface albedo from remote sensing with the values of the LM showed that the surface albedo is particularly too high in the model areas

with high forest coverage. Because of the missing differentiation of vegetation by the value of plant albedo, the heterogeneity of the area is not represented adequately.

From all investigated parameters the change of plant cover has the largest effects on the turbulent heat fluxes. An increase of 10% of the plant cover leads to lower sensible heat flux of around 2% on average. The latent heat flux rises at about 3.3%. The radiation balance is affected only insignificantly and the temperature is practically independent on the degree of plant coverage. The investigation of the soil parameters pore volume, field capacity, permanent wilting point and minimum infiltration rate shows that the influence of the pore volume on the turbulent heat fluxes is the largest. The influence of permanent wilting point and field capacity is smaller. The long-wave radiation balance reacts particularly to the pore volume. However the short-wave radiation balance and the temperature remain practically uninfluenced. The effect on the LM results depends strongly on the prevailing soil type. The largest range of the latent heat flux (LE) was found for sand relative to the pore volume. In contrast, the soil type loam leads to only small fluctuations of LE for all used parameters. So the latent heat flux can be adapted on average for sand, sandy loam and loam, if for sand the maximum pore volume and for the other soil types the minimum pore volume is used. Only by use of unrealistic soil parameters the evaporation over sand could become actually smaller than over sandy loam and loam – which is in contrast to expectation and suggests need for model improvement. The different resolutions of LM lead to only small differences of the area-averaged fluxes. The largest influence of grid resolution could be found for the momentum fluxes, which increase clearly with the higher resolution of 2.8 km in relation to 7 km resolution. This effect is caused by the increased roughness. The radiation balance is also affected by the resolution but only marginal. The two turbulent heat fluxes (Figure 15) and the temperature seem to be

independent on the model resolution. Large changes of the LM results could be observed in the VERTIKO area on grid points with water coverage. This land use disappears in the coarse resolution of 7 km.

Figure 15: LM-results for the VERTIKO target area on May 30, 2003 for 7 km (left) and 2,8 km (right) resolution; a) latent heat, b) sensible heat.
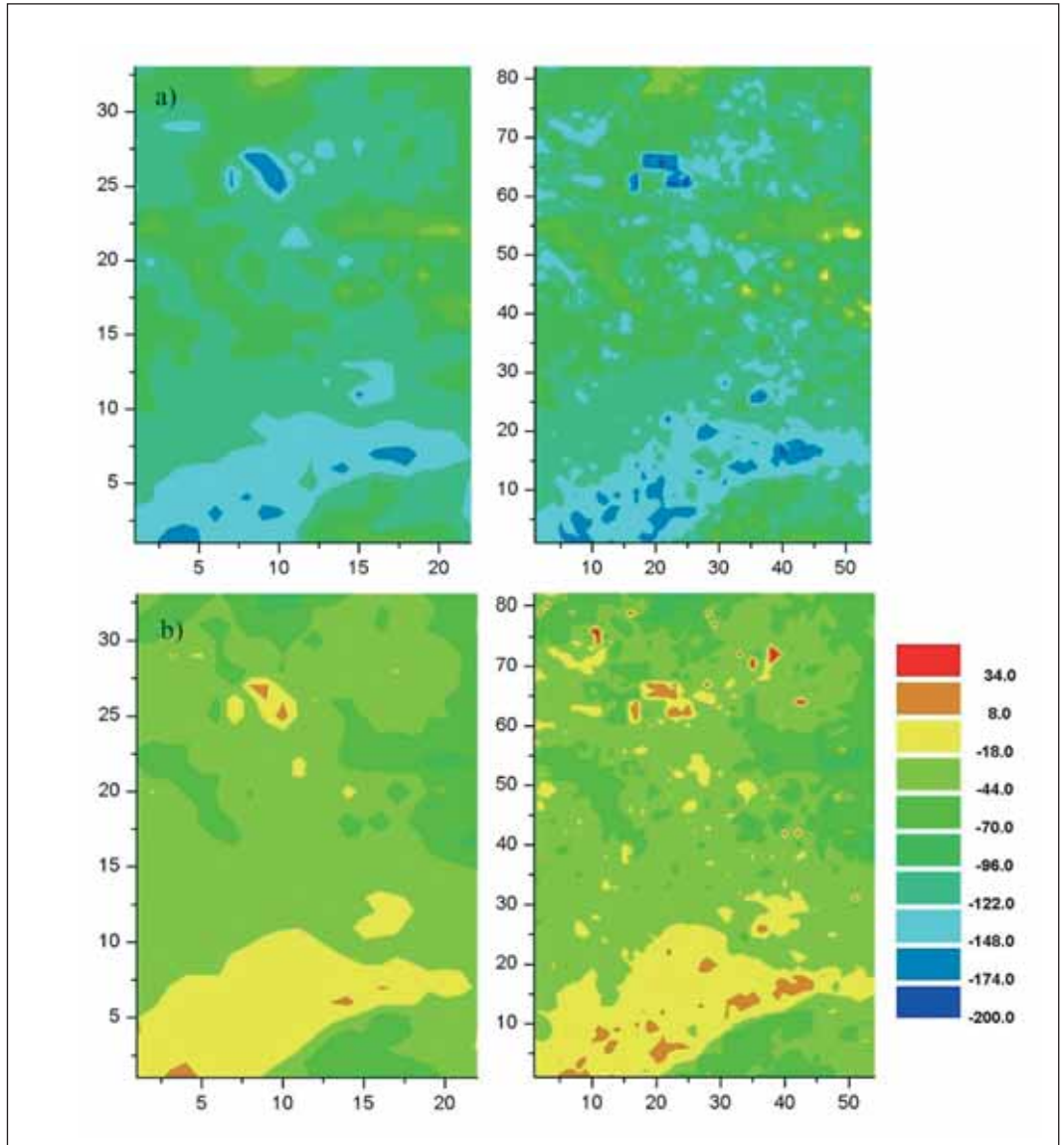
Conclusions and Perspectives

Results of the VERTIKO research have confirmed
that continuous flux measurements are crucial to
understand surface-atmosphere interactions and
improve model parameterisation. However, SVAT-
model comparisons also revealed that model valida-
tions by measurements require assumptions on
energy balance closure (closed in models but not in
measurements). Campaigns have improved method-
ology of C and N flux measurements and have
shown high importance of advective fluxes in inho-
mogeneous terrain. For spatial integration it is possi-
ble to derive surface energy fluxes from satellite
with remaining inaccuracies being related to both
ground-based and remote measurements. There
exists small-scale heterogeneity in canopies that
needs to be integrated in a hierarchy of models.
Atmospheric characteristics above the active surface
might be less affected. The use of effective parame-
ters in mesoscale modelling should be challenged by
a more complete consideration of heterogeneous
landscapes to allow for a realistic feed-back between
land surfaces and the atmosphere.

Improvement of measurement accuracy and spatial
representation, specification of heterogeneity effects
on horizontal and vertical fluxes and better under-
standing of the energy balance of heterogeneous sur-
faces are future important research issues. Long-term
observation programmes reflecting seasonal and
interannual variability and landscape heterogeneity
should be continued for validation of SVAT and BGC
Models as well as regional climate models.

Acknowledgement

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Appendix

Acronym	Institution	Anchor Station Test Site	Task
ALUF1	Albert-Ludwigs-Universität Freiburg		Surface parameters
ALUF2	Albert-Ludwigs-Universität Freiburg	Hartheim	Longterm interannual variability
DWD1	Deutscher Wetterdienst	**Lindenberg**	Land surface heterogeneity
DWD2	Deutscher Wetterdienst		Mesoscale modelling, LM
FAL	Bundesforschungsanstalt für Landwirtschaft	Braunschweig	Crop parameterization
GAUG	Universität Göttingen	Solling	N-fluxes, forest/soil
IfT	Institut für Troposphärenforschung e.V.	**Melpitz**	N-fluxes, grassland
IMK	Forschungszentrum Karlsruhe GmbH		N-BGC
TUB	Technische Universität Braunschweig		Helipod (STINHO)
TUBAF	Technische Universität Bergakademie Freiberg		N-fluxes, forest canopy
TUD1	Technische Universität Dresden	Tharandt	Coupling SVAT-CN-PBL-LM
TUD2	Technische Universität Dresden	**Oberbärenburg**	Remote sensing, target area
UBAS	Universität Basel		Advection experiments
UBT1	Universität Bayreuth	Weidenbrunnen	Qa/Qc, footprint modelling
UBT2	Universität Bayreuth		SVAT-CN
UH	Universität Hannover		LES (STINHO)
ULLIM	Universität Leipzig		Tomography (STINHO)

Vertical transport and orography (VERTIKATOR)

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Summary

Within the AFO2000 project VERTIKATOR the chain of
processes, especially convection and secondary flow
systems induced by mountains, which contribute to
the vertical distribution of momentum, heat, mois-
ture and trace substances from the earth surface to
higher levels of the atmosphere have been studied.
Large field campaigns and advanced mesoscale
models have been used to study these processes in the
region of the northern Alps and of the Black Forest. It
has been substantiated, that secondary flow systems
like the 'Alpine Pumping' and similar flow systems in
medium range orography contribute to a high degree
to the feeding of the atmosphere with heat and mois-
ture from the lower levels close to the earth surface.
The triggering effect of mountains for deep convec-
tion and the formation of precipitation is an impor-
tant part, which has not been included in weather
forecast and climate prediction models so far. Verifi-
cations with observed data have shown that these
effects can be included only in spatially sufficiently
high resolved models.

Measurement Campaigns and Model Studies

In June and July 2002 two extended field campaigns
have been conducted in the area of the Northern
Black Forest (mid-sized orography) and in the region
of the Northern Alps (high orography). By surface
observations, vertical soundings, modern remote
sensing instruments, radar systems and aircraft
observations data sets of completeness as much as
possible have been gathered in order to be able to
analyse far reaching processes in the surrounding
areas and above the mountain regions.

Fig. 1 and Fig. 2 demonstrate an overview of the
flight paths within the experimental areas in the
northern Black Forest and within the northern
Alpine regions respectively. The measurements were

Figure 1: Experimental area over the northern Black Forest.
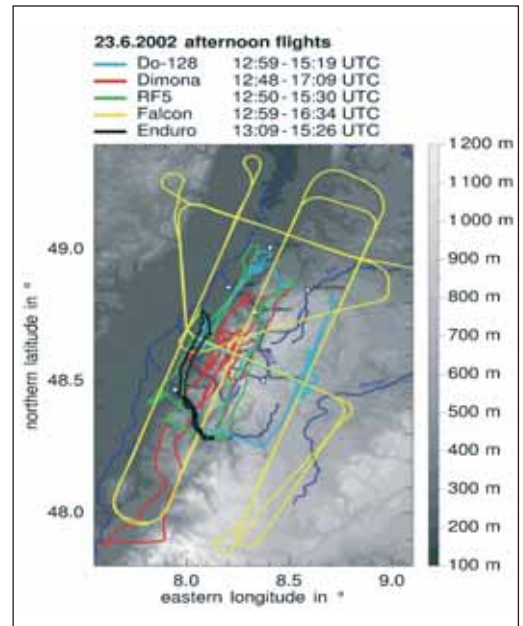
performed, so that the life cycle during days of the pumping processes could be documented. Also the horizontal influence zone of the mountains and the vertical extension of secondary flow systems were of primary interest.

In order to follow the life cycle of the energy and moisture input from the ground, of the boundary layer development and the convergence of energy supply over the mountain ridges intensive measurement periods have been used to cover the related variables.

Figure 2: Experimental area in the northern part of the Alps. Measurement sites and flight paths on Aug 7th 2002.
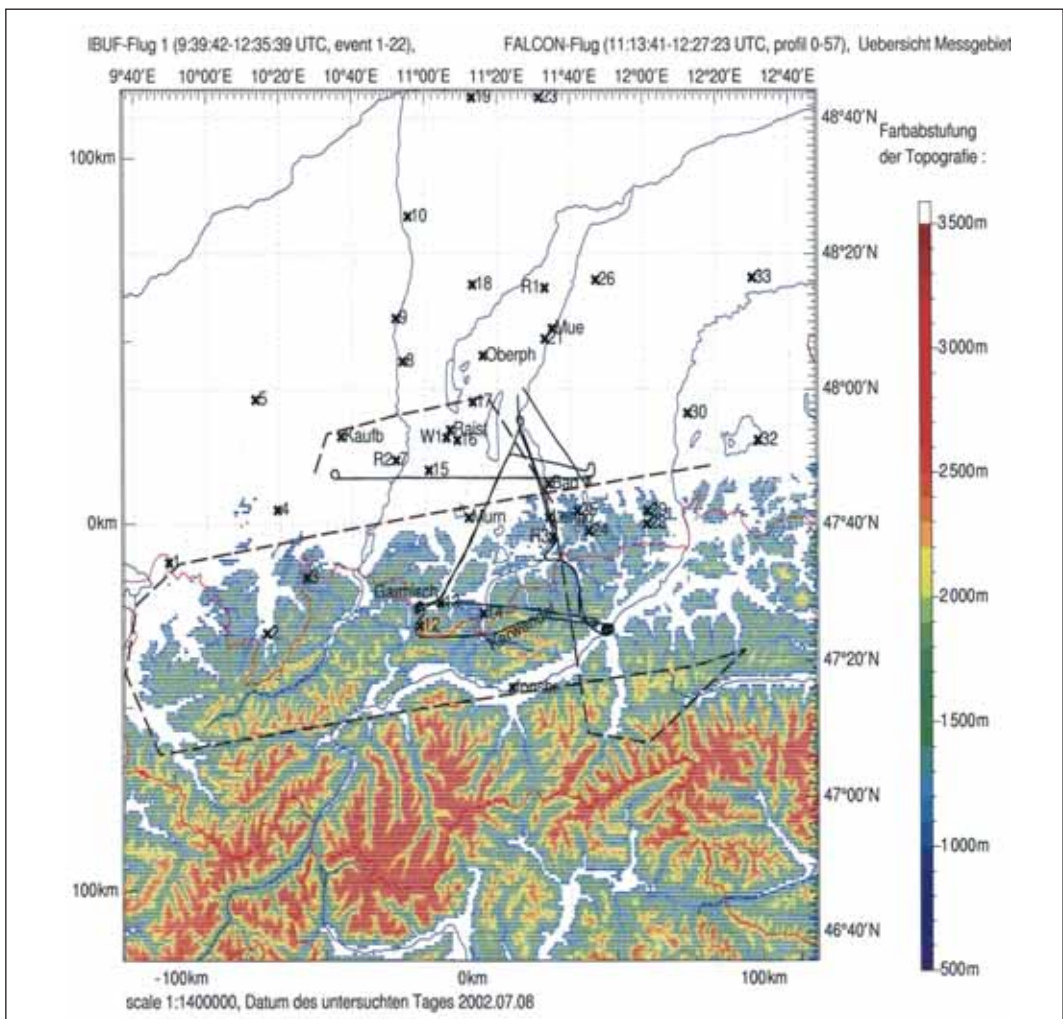
Highlights

Experimental Results

Through the joint action of the participating groups the development of the daily cycle of secondary flow systems could be well documented in medium high (Black Forest) and in high mountainous (Alps) regions. The chain of processes includes near surface boundary layer development, converging low level mountain and valley wind systems and the triggering effect of deep convection over the mountains.

Figure 3: Mean surface wind field during days with Alpine Pumping (from Lugauer, Winkler)
Always a strong converging zone of low level wind fields above the mountain crests act as a trigger mechanism for deep convection.
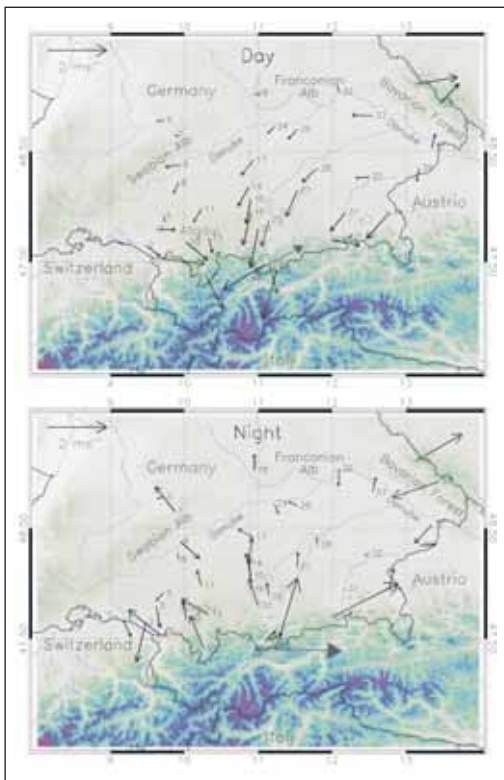
In the high mountain area of the Alps a clear developed and important flow system could be recognized. The so called "Alpine Pumping" is a far reaching flow system covering a region of more than 100 km in front of the mountains. So far the existence of such a system had been anticipated. However the VERTIKATOR – experiment has presented a clear picture of those developments with horizontal extensions of more than 100 km to the north and a vertical extension of about 1.5 km. The hand-over system of energy and moisture from the ground to the free atmosphere must be locked at in a completely new frame compared to the classical one-dimensional boundary layer transport in the vertical direction.

Fig. 3 shows the mean surface wind field during days with Alpine Pumping. It covers the total area up to the Danube River valley to the north of the Alps. Similar structures however of smaller horizontal and vertical extensions have been found in the Black Forest regions.

So a rapid hand over of energy and moisture from the earth surface to the higher troposphere is initiated.

Model Simulations

The success of the VERTIKATOR project was based on a broad experience of the participating groups in developing and applying most advanced mesoscale atmospheric models.

Through the comparison of three different models deficiencies in some directions could be first eliminated. Most important is however the resolving power of the secondary flow systems in the mountainous areas with different horizontal scales both in steep mountains (Zugspitz-area) and in mid-sized mountains (Black Forest).

A clear result delivered by the model studies is that the inclusion of a normal classical convection parameterization leads to strong deviating precipitation distributions compared to observations. Also the onset of convection over the mountains can only be

found in the right spatial structure when the horizontal resolution of the mesoscale models is high enough (in the order of 2 km and better).

Fig. 4 shows the temporal development of convection over the Black Forest triggered by the small scale low level wind fields as a carrier of heat and moisture.

Modern Instrumentation and Analysis Techniques within the Field Campaign of VERTIKATOR

Modern instrumentation from aircraft measurements, from remote sensing systems, vertical soundings (drop sounds), from Radar and from classical surface measurement systems has been used. From these observations many important relations could be quantified. An important quantity is the vertical mass transport of the liquid phases in convective systems. From those studies new relations between

Radar-reflectivity and liquid water or ice content has been derived, as shown in Fig. 5.

Figure 5: Radar-reflectivity and liquid water content.
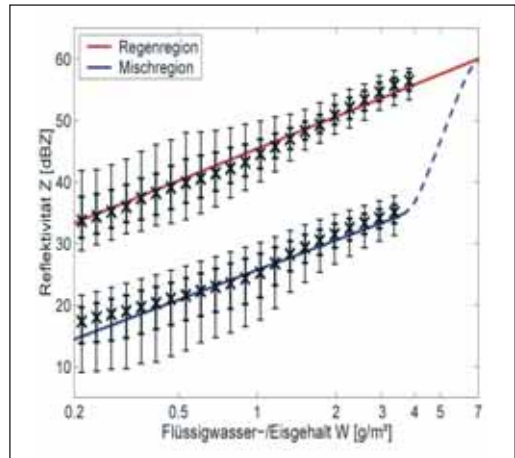
Figure 4: Surface wind field and convective clouds triggered over the Black Forest and Vosges mountains for 19/06/2003 from model simulations. (Noppel, IMK, Kolloquium Leipzig, 08.01.2004).
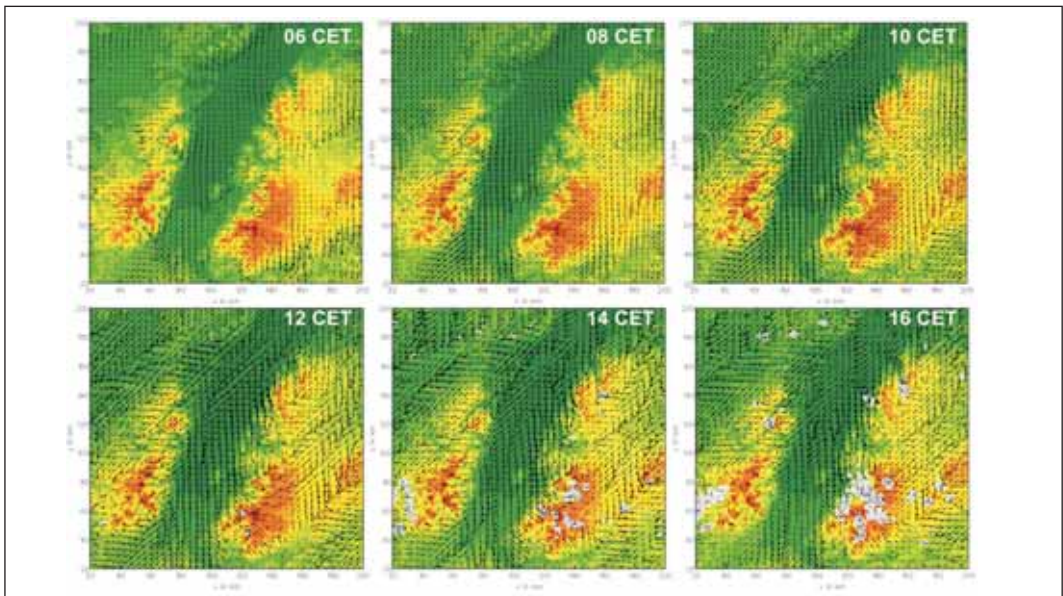
Through these results Radar-observations could be combined with numerical simulations to study also regions in convective systems which are normally not accessible by classical measurement platforms like aircraft or other observing system due to extremely strong and dangerous internal turbulence.

Conclusion

In this project the existence and importance of secondary flow system originated by the presence of mountains has impressively shown. Through those motion systems a considerable part of energy and moisture is carried a way from the earth surface and handed over to higher levels of the atmosphere. A close link between boundary layer processes and deep convection is active. Model simulations contrasted to the observations show that present day parameterisations of convection do not cover this effect in correct way. Only models with spatially high enough resolution show convective developments and hence distributions of precipitations with sufficiently good comparison to the observations. The project results show that a completely new handling of boundary layer and convection link is needed in order to provide accurate energy and moisture supply to the atmosphere in mountainous regions in weather prediction and climate models.

Results of Theme Group 2.1

Chemistry, Dynamics, Radiation and their Interaction: Tropopause and Middle Atmosphere

Investigation of chemical and dynamical processes in the stratosphere and upper troposphere using isotopes as process tracers (ISOSTRAT)

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The aim of the project ISOSTRAT was to combine concentration and high precision isotope ratio measurements on atmospheric trace gases to examine important physical and chemical processes in the strato-

sphere and the upper troposphere. The focus is on three individual projects: 1) The isotopic fractionation of long-lived trace gases (CO_2, CH_4, N_2O) in the stratosphere as tracer for stratospheric chemical and

Figure 1: Concentration and average ^{15}N fractionation of N_2O from more than 200 stratospheric air samples obtained from balloons and the high altitude aircraft Geofizika at various latitudes from polar (blue) to tropical (red). The Geofizika samples were obtained during the EUPLEX campaign. As N_2O is removed in the stratosphere, it acquires strong isotope enrichment in all signatures. These stratospheric enrichments are very important for the global N_2O isotope budget.
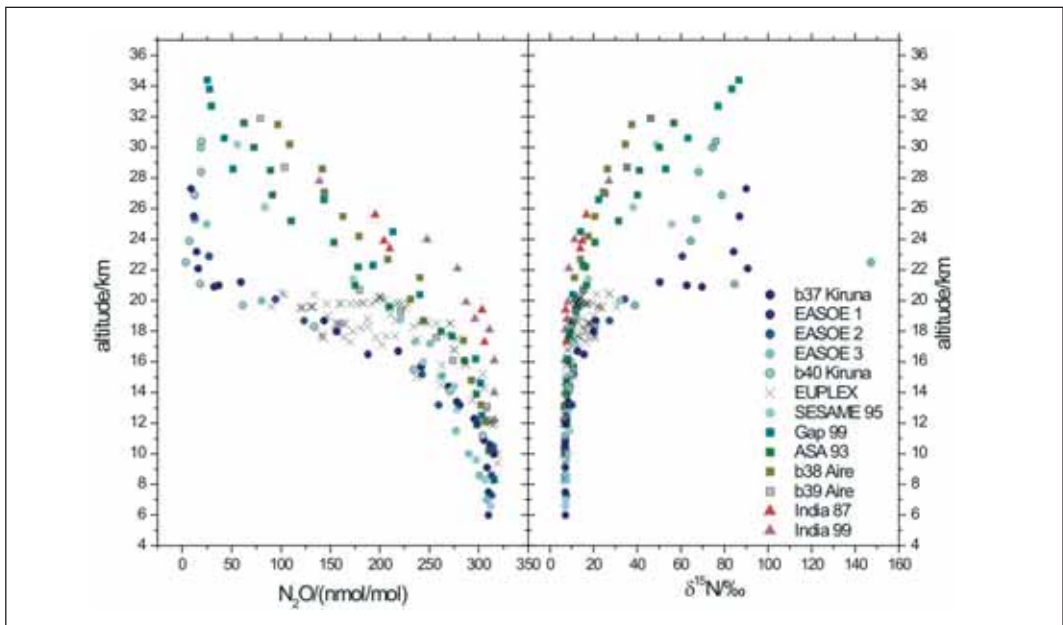
photochemical sink and exchange reactions, and the importance of the stratospheric sinks for the global isotope budgets of these trace gases. 2) The transfer of the heavy oxygen isotope anomaly from ozone to other oxygen-bearing compounds in chemical reactions. 3) The investigation of the stratospheric water budget via measurement of the isotopic composition of water in the stratosphere and upper troposphere. This report summarizes some of the results.

Within the project ISOSTRAT a state-of-the-art isotope laboratory has been set up. Analytical extraction and preparation techniques for high-precision measurements of all stable isotope signatures of CO_2, CH_4, N_2O, H_2 and H_2O samples have been developed. Sampling devices for whole air and water from the middle atmosphere have been constructed. Atmospheric samples collected on aircraft and balloon platforms have been analyzed for the isotopic composition of the long-lived trace gases. In addition, isotope exchange and fractionation processes were investigated in detail in the laboratory.

The largest existing dataset of isotope measurements from stratospheric air samples has been

obtained. The results allow assessing in detail the role of the removal and exchange processes in the stratosphere for the isotope budget of the long-lived trace gases. As an example, Figure 1 shows concentration and isotope measurements of N_2O in more than 200 air samples from 12 stratospheric balloon flights and from the high-altitude aircraft Geofizika.

Similar information is achievable from methane isotope ratio measurements. Here, isotope information provides also information about stratospheric free Cl levels, since the reaction of CH_4 with Cl is associated with an unusually large carbon isotope fractionation. Based on a comparison of our measurements with a 2D-model, we estimate that 28% of stratospheric CH_4 is removed by Cl, 31% by $O(^1D)$ and 41% by OH. Driven by this result, we carried out model calculations in collaboration with the Research Center Jülich (J.-U. Grooß and R. Müller) to investigate the effect of changing atmospheric Cl, OH and $O(^1D)$ levels on stratospheric water, based on observations of the precursor substances, and found that those changes significantly affected water vapor levels in the upper stratosphere in the past century (Figure 2).

A highly surprising result was obtained for the deuterium content of molecular hydrogen in the stratosphere (Figure 3). Although the H_2 mixing ratio stays constant through most of the stratosphere, deuterium gets extremely enriched in stratospheric H_2. This can only be explained by a source of isotopically heavy hydrogen from the oxidation of CH_4, which is the main H_2 source in the stratosphere. This allows for the first time to understand the isotopic composition of H_2 in the troposphere, since a heavy source was formerly missing to close the iso-tope budget.

Results from an additional balloon flight that intercepted mesospheric air in the Arctic vortex in 2003 (not shown) reveal another very interesting feature: In the mesosphere, along with increasing concentrations, the deuterium content of H_2 decreases dramatically again, and even reaches tropospheric values. This is a clear signature of H_2 produc-

Figure 2: Decadal trends of stratospheric H_2O (dashed lines) and CH_4 (solid lines) for the period 1980-1990 due to the increase in CH_4 only (green), and the additional effects of increased anthropogenic CFC emissions, the increased influx of H_2O from the troposphere (which itself has been subtracted in the diagram for better reading) and changes in the upper stratospheric ozone column during that period. Details in [RÖCKMANN et al., 2004].

tion from photolysis of water in the mesosphere. These data are being evaluated.

In addition to the stratospheric samples, for N_2O we have analyzed air from Antarctic firn as well as archived atmospheric air samples from Antarctica and determined the present rate of change of the N_2O isotopic composition, as well as the total change since pre-industrial times. This allowed for the first time to derive the isotope signature of the global average N_2O source. Analysis of the data in a global isotope budget calculations shows that the observed increase in N_2O since pre-industrial times is primarily caused by increasing soil emissions, likely due to intensified agriculture.

Where no data were available before, the pertinent reactions were studied in laboratory experiments, which have led in most cases to a complete and quantitative understanding of the origin of the isotope fractionation in the stratosphere. As an example, Figure 4 shows the wavelength dependence of the isotope fractionation in the UV photolysis of N_2O. One important result is that the fractionation in UV photolysis and the one in the reaction with $O(^1D)$, the second important sink of N_2O, are clearly different, and both are furthermore different from

isotope patterns that are for example caused by mixing of different air masses, i.e., physical and chemical processes leave different isotope signatures in the remaining gas.

In the stratosphere, also CO_2, which is generally thought to be chemically inert, is at the center of a particularly interesting isotope effect: CO_2 can form a short lived CO_3 complex with $O(^1D)$ produced from O_3 photolysis, which promotes isotope exchange. O_3 is well known to have a strongly anomalous oxygen isotope composition (MAUERSBERGER et al., 1999). This anomaly is transferred to CO_2, which leads to an isotope anomaly in stratospheric CO_2, but the details of the exchange process are not yet understood. Our stratospheric observations show that this is not a simple transfer, but that the isotope anomaly is even enhanced in the CO_2 product, i.e., for stratospheric CO_2 we find $\delta^{17}O \approx 1.7 \times \delta^{18}O$ [LÄMMERZAHL et al., 2002]. Laboratory isotope exchange experiments between CO_2 and $O(^1D)$ are in progress to study this effect in detail (Figure 5).

Regarding isotope measurements on atmospheric water vapor with the aim of ultimately carry-

Figure 3: Hydrogen mixing ratio and $\delta D(H_2)$ in the stratosphere as a function of altitude. Although the mixing ratio stays virtually constant, $\delta D(H_2)$ increases almost 300‰. These are to date the highest observed isotope enrichments in the terrestrial atmosphere. From [RÖCKMANN et al., 2003].
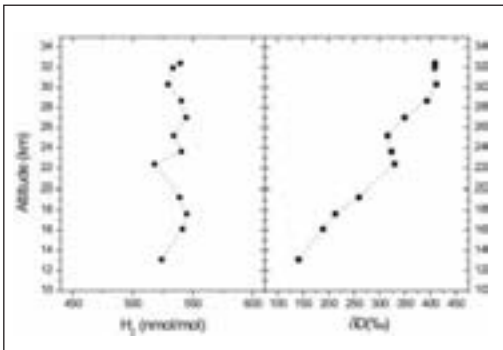
Figure 4: Isotopic fractionation constants for the central N atom in the UV photolysis of N_2O as a function of photolysis wavelength; measurements (symbols) and modeling results (lines). The dashed line is a linear fit through the results from the direct fractionation experiments. The model results deviate strongly for wavelengths >200nm. Further explanations and references in KAISER et al., 2003].
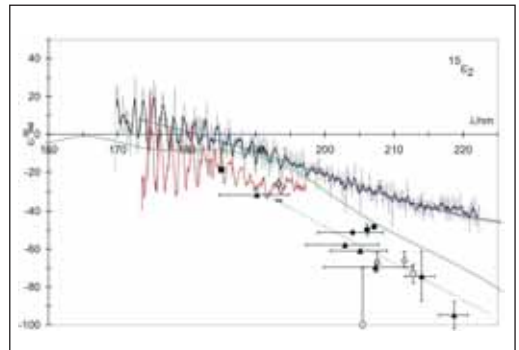
ing out isotope ratio measurements on stratospheric water, analytical methods for laboratory analysis of

Figure 5: Laboratory studies of the exchange process between CO_2 and $O(^1D)$ were carried out with natural (blue) and artificially prepared (magenta and green) CO_2 samples of varying isotopic composition. With increasing extent of reaction the CO_2 approaches a well-defined isotope equilibrium point, which is different from the O_2 and O_3 used in the experiments (yellow ellipses). Different slopes can be generated depending on the isotopic composition of the initial CO_2.
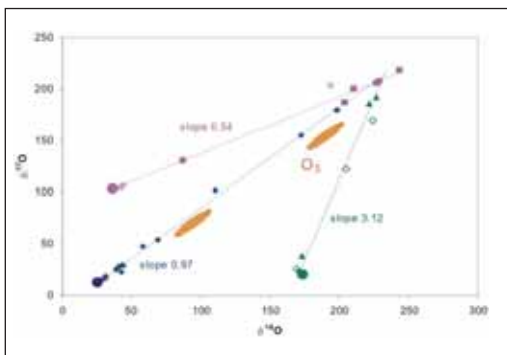
very small water samples have been developed. In particular, with this method we are able to determine the oxygen isotope anomaly of a water vapor sample with high precision, probably unique compared to several other methods that are being developed in other labs for isotope measurements on water vapor with optical absorption techniques. A first-generation water vapor sampler for 100 nL samples has been constructed, has operated on a first aircraft flight and will be tested soon on a stratospheric balloon flight. Results are expected within the remaining 15 months of the project.

Highly interesting scientific results have been obtained from atmospheric and laboratory measurements of the isotopic composition of atmospheric trace gases. Isotope data help to understand physical and chemical processes in the atmosphere, and can provide much insight into global trace gas budgets. Studies of the oxygen isotope anomaly of atmospheric trace gases provide interesting new tracers to follow reaction mechanisms and to quantify exchange processes related to ozone.

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The realization of long-term near global-scale, regular, extensive, detailed, *in situ* assay and sampling, of atmospheric trace gases and aerosols with commercial passenger aircraft

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Introduction

Civil aircraft can be excellent platforms for obtaining detailed global scale atmospheric data. CARIBIC combines important aspects of research aircraft, land based observatories, and satellite based sensors. Specific advantages are; low operational costs, long time horizon, near global scale coverage, and a detailed high resolution dataset. The CARIBIC system comprises 2 elements, namely an automated measurement container (1200-1400 kg) - basically a compact flying laboratory - and a special air intake system designed for contamination and loss-free, representative sampling of air and aerosol particles. The optimal flight frequency equals typically 1-2 flights/month. CARIBIC operated from 1997 to 2002 using an LTU Boeing-767, and resumes its task fall 2004 as a frequent flyer with Lufthansa using a new Airbus A340-600, carrying a powerful luggage package with 20 analytical instruments. We first show a few only of the many highlights of phase 1 of CARIBIC, and then aspects of the AFO 2000 supported development of the new platform implemented on a Lufthansa Airbus A340-600.

Sensitive NO and NOy measurements and aircraft exhaust

AFO 2000 enabled the incorporation of a DLR NO/NOy analyser, which signified a fundamental extension of the analytical capability. Reactive nitrogen species play a key role in controlling ozone concentrations in the tropopause region. Questions pertaining to the contribution of air traffic to the nitrogen budget of the UTLS, of deep convection, and of lightning are of great importance. Figure 1 shows the detection of aged aircraft exhaust plumes just above the tropopause, at O_3 levels of 250 nmole/mole and PVU values of 5-6 (flight March 2002, Cuba-Düsseldorf, 5°E). The coincident sharp increases (~10s) in NO, NOy, and number densities of particles of over 4 and 12 nm, well above values in the surrounding air masses that are free of fresh input of NOx, allow the calculation of emission ratios of condensation nuclei relative to NOx emission by jet engines. The average emission index derived from these CARIBIC observations equals $1.1*10^{17}$ particles of greater than 4 nm per kg fuel. This value agrees with the main value of $1*10^{17}$ from the SONEX campaign using the NASA DC-8 research aircraft.
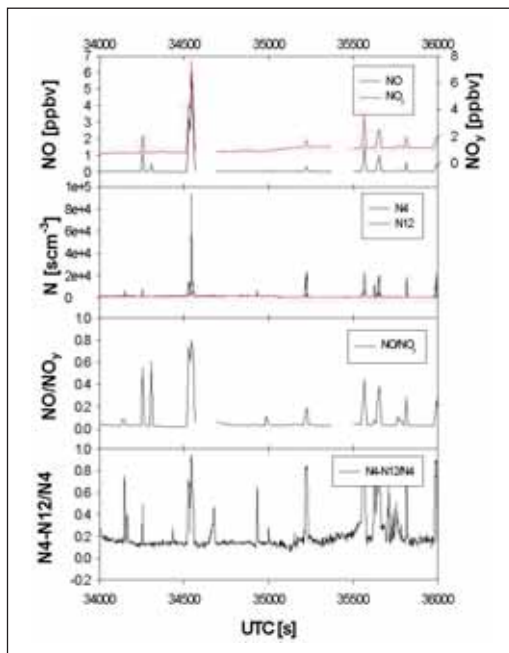
Figure 1: NO, NOy, and particle data.

Aerosol particle size and abundance measurements

Representative aerosol measurements (IfT) using a passenger aircraft cruising in excess of 250 m/s require a well designed and characterized air inlet and transmission system.

Figure 2 shows results for ultrafine (4nm $<\emptyset<$ 12nm, N_{4-12}) and Aitken mode plus accumulation mode particles (12nm $<\emptyset<$ 1300nm, N_{12}). The data are for the upper troposphere and lower stratosphere for a series of flights between Germany and the Indian Ocean, in the form of a latitudinal probability distribution for the summer season. At sub-tropical latitudes, in generally subsiding air, lower con-centrations of 1500 to 3000 are most frequent, whereas over the Arabian Sea and Europe high particle concentrations of up to 8000 prevail. In particular near the ITCZ, rapid vertical transport in regions of strong convection

entrains precursor gases, which leads to the rapid for-mation of N_{12} particles in the intercepted air masses.

Figure 2: Aerosol climatology.
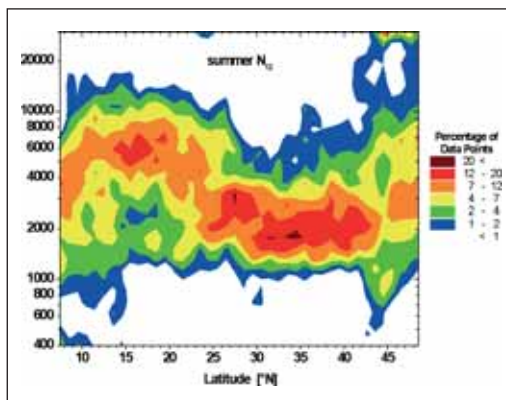
Tropospheric O$_3$ budget-transport vs. chemistry- using CO

The systematic precise observations by CARIBIC of characteristic changes in O_3 and CO across the tropopause (IMK and MPI) have led to the definition of a chemical tropopause. Furthermore, these obser-vations have lead to a far more representative insight of processes inside the mixing layer above the extra-tropical tropopause. Figure 3 illustrates the power of regular long distance measurements (Germany-Indian Ocean route, 1997-2001) by means of the his-togram of UT O_3 -CO correlation slopes indicative for stratospheric related effects as compared to changes caused by photochemical O_3 production. In the tropics the positive correlations of ~ 0.22 (fall and winter) and ~ 0.5 (spring and summer) imply photo-chemical production with little if any stratospheric import. These results support recent model results and allow calculation of the annual photochemical O_3 production NH tropics leading to 564 Tg. In the NH extra-tropics (> 30°N) a non-skewed distribution

with positive values only occurs in summer, confirming mainly photochemical production. In winter and

spring input of stratospheric O_3 origin prevails at the cruising altitude of ~10 km.

Figure 3: Seasonal variation of O_3-CO slopes .
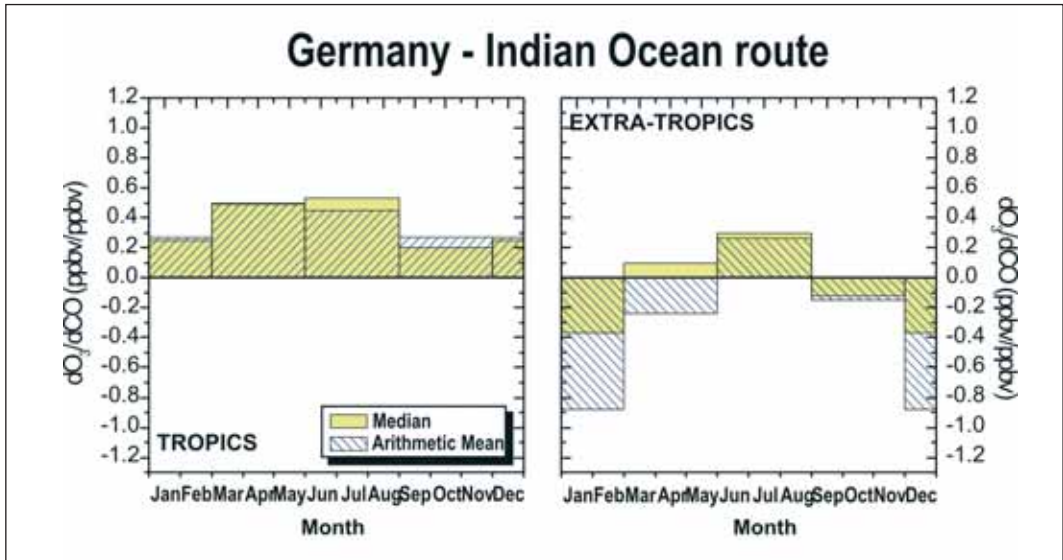
The Lufthansa A340-600 instrumentation and inlet system

Supported by AFO 2000, one of Lufthansa's ten A340-600er (a new series of long range, very long aircraft equipped by LHT with onboard broadband wireless LAN) is provided with a sophisticated inlet system (3 separate inlet probes one of which with double inlet orifice, containing a camera and DOAS system), tubing systems (partly heated, with specific surface coatings/treatments) between the inlet and container (aft of the forward cargo compartment in front of the belly fairing), and a 3 meter full sized instrumented container (1450 kg).

The inlet system (IFT aerodynamics and IMK heating systems, in conjunction with Kolt and Garner GCT, and manufacturing by Heggemann Aerospace)

features 3 air inlet probes (fig. 4). The lowermost 0.5 m long aerosol diffuser tube has a leading aerodynamic shroud which minimizes dependence of the aerosol sampling characteristics on speed and angle of attack. The air intake for H_2O measurement is placed outside the area of pressure disturbances inflicted by the entire inlet itself. A frontal intake orifice is for total H_2O measurement, whereas a second inlet protruding slightly outside the probe collects gaseous H_2O only. The inlet probe for trace gases is large and in conjunction with 2 parallel tubes inside the aircraft ensures a high throughput of uncontaminated air with minimum wall interactions. A system of heaters prevents icing and condensation/wall interactions for all trace gas inlets. A video camera observing the inlet tips and the surrounding air, and a 3 axis DOAS telescope (Platt, Envi-

ronmental Physics, Heidelberg) are accommodated in the main spar.

The container (MPI-Chemie) accommodates 8 racks (mostly 19 inch) mounted on vibration dampers (fig. 5). Mechanical compliance (up to 9 g) is achieved (Enviscope Frankfurt). The power supply units (nominal 6 kW) are compatible with the aircraft's sophisticated power management system. Each analyser/set has its dedicated computer whereas overall control and data transfer are governed by a central computer (including pressure measurement, ARINC flight data, and temperatures). Data transfer is based on a new flexible, universal bus system. The video camera data are recorded in the optical parti-

cle counter unit (IFT). Calibration and gas supply cylinders are housed in a separate rack (back of container), with the oxygen bottles protected against engine burst failure. Incorporation of smoke detectors, a dual forced ventilation system, and temperature sensors comply with the safety requirements. Coupling of the container to the electrical, electronic and air tubing system is optimized using mainly quickfit connectors. Table 1 summarizes the analytical - including the DOAS remote sensing - and sampling (air samples, OVOC samples, aerosol samples) capabilities. (For info on the individual consortium partners please see website www.caribic-atmos pheric.com).

Figure 4: Photograph of the CARIBIC inlet system.

Figure 5: Photograph of the CARIBIC measurement container system.

Trace Compound	Instrument
O_3 ultra-fast	Chemi-luminescence on an organic dye
O_3 accurate and precise	UV Photometer
CO	Vacuum UV-fluorescence
H_2O gas phase	Photo-acoustic detector with diode laser
H_2O total	Chilled mirror and photo-acoustic detector
NO	Chemi-luminescence reaction with O_3
NOy	Chemi-luminescence (conversion to NO)
Hg	Enrichment and atomic fluorescence
CO_2	NDIR
O_2 ultra high precision, 10 per meg	Electrochemical cells
Methanol, Acetone, Acetaldehyde	Proton Transf. React. Mass-Spec.(PTR-MS)
Aerosol with diameter > 4 nm	Condensation Particle Counter (CPC)
Aerosol with diameter >12 nm	Condensation Particle Counter (CPC)
Aerosol with diameter >18 nm	Condensation Particle Counter (CPC)
Aerosol size distribution 150-5000 nm	Optical Particle Counter (OPC)
Aerosol elemental composition	Impactor sampler, and analysis with PIXE
Particle morphology	Impactor sampler, and electron microscopy
VOC samples	Absorption tubes and GC-MS analysis
NMHC, halogens, greenhouse gases	Air sampling in glass bottles and GC analysis
BrO, HCHO, OClO, O_4	DOAS
Cirrus (under certain conditions)	Camera

Table 1: Instrumentation for trace gas (plus O_2) and aerosol particle analyses.

SPURT:
Trace gas transport in the tropopause region

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Introduction

SPURT (**SPUR**enstofftransport in der **T**ropopausenregion, trace gas transport in the tropopause region) was concerned with the tropopause region and the transport processes influencing the chemical composition in this region. While the focus was on airborne tracer measurements with a Learjet 35A provided by enviscope/GFD, flying at altitudes up to 13.7 km, there was also a theoretical part to the project. Eight measurement campaigns were performed, probing the Upper Troposphere/Lower Stratosphere (UT/LS) region from the subtropics to high northern latitudes. Two campaigns were performed during each season. The observations were used to characterise transport pathways and time scales for air masses mixed into the lowermost stratosphere. Experimental observations were performed by the groups at the

Fig. 1: (a) Vertical cross section along a segment of the flight to Iceland during SPURT 4 showing potential vorticity PV (in colors) and the flight path (black line) at about 200 hPa above a strongly disturbed tropopause (2 pvu surface shown in red). (b): 10-day backward trajectories from a flight segment during SPURT 4 that illustrates the sometimes complex history of the sampled airmasses.
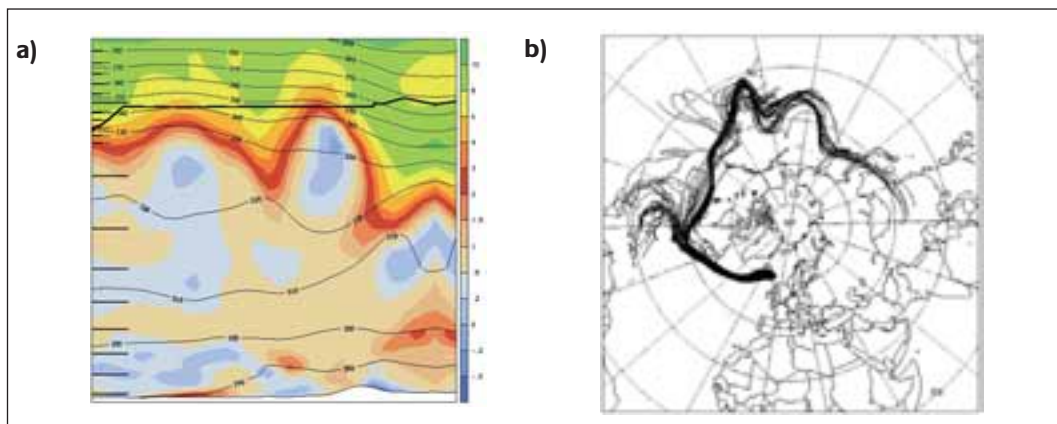
University of Frankfurt, the Max-Planck-Institute for Chemistry and at the Research Centre Jülich. Theoretical studies were contributed by the group at the University of Mainz.

Meteorological support for campaign preparation and data analysis was provided by the ETH Zürich. A fixed set of diagnostic products, based upon operational ECMWF analysis data, was produced after every campaign. They include standard meteorological charts to characterize the synoptic setting of the campaign, vertical cross sections of potential vorticity and temperature along the exact flight path to illustrate the location of the flight relative to the local tropopause (see Fig. 1a) and 10-day backward trajectories starting every 10 seconds from the aircraft location to identify the recent history of the sampled air parcels (cf. Fig. 1b). These products were helpful for the interpretation of the tracer measurements and a starting point for further data evaluations.

Lag time in the lowermost stratosphere

The University of Frankfurt provided an in-situ gas chromatograph (GhOST II) for the measurements of several long lived tracers. GhOST II is able to make high precision measurements of N_2O, CFC-12, SF_6, CO and H_2 with a time resolution of 70 seconds (WETTER, 2002). One of the main focuses was to investigate transport times in the UT/LS region. For this purpose SF_6 is an ideal tracer, because it has no atmospheric sinks up to about 50 km altitude and its atmospheric abundance increases monotonically with a near linear growth rate. SF_6 can, therefore, be used as a tracer for the mean age of air. The mean age of air describes how fast tropospheric air is transported to a certain stratospheric location on the average. As tropospheric air is supplied to the lowermost stratosphere from the extratropical upper troposphere and the TTL (tropical Tropopause Layer), it is only possible to determine a lag time rather than an exact mean age. However, the influence of extratropical tropo-

sphere to stratosphere transport (TST) is most pronounced in a layer above the local tropopause (see next chapter). Above this layer the lag time is expected to be a very good proxy of mean age.

Figure 2 displays the distribution of the lag time observed during the SPURT campaigns in Fall 2002 and Spring 2003 in a reference frame of equivalent latitude and potential temperature (see HOOR et al., 2004a). Much higher lag times observed in spring 2003 show the descent of aged stratospheric air into the lowermost stratosphere.

Figure 2: Lag times derived from the SF_6 observations during SPURT campaigns 5 and 7. The thin lines show the isolines of potential vorticity PV (2, 4, 6, and 8 PVU).
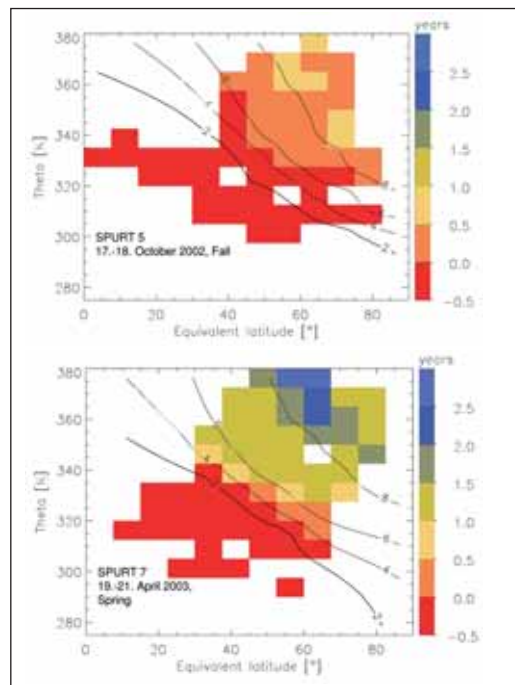
Tropospheric air in the lowermost stratosphere:
Extratropical versus tropical origin

The MPI-group provided measurements of CO, CH_4 and N_2O using tunable diode laser absorption spectroscopy and of CO_2 employing a modified commercial non-dispersive infrared instrument. The high precision measurements of CO_2 with its well known tropospheric seasonal cycle facilitated to infer different pathways of tropospheric air, which were mixed into the lowermost stratosphere (Diploma thesis CHRISTIAN GURK, 2003). Additional NO and NO_y measurements were made in cooperation with the ETH Zürich, Switzerland (HEGGLIN et al., 2004; HEGGLIN, 2004).

One important result was the observation of comparable vertical CO-gradients relative to the local tropopause independent from potential temperature at the local tropopause itself. Taking into account that the tropopause in the extratropics is tilted against isentropic surfaces, we could show that in the extratropics tropospheric air mainly affects a layer parallel to the local tropopause (HOOR et al., 2004a). This so-called mixing layer exhibits the photochemical characteristics of both, the troposphere and the stratosphere. However, even at large distances from the local tropopause CO-values were well above the stratospheric photo-chemical background indicating a significant tropospheric contribution. The unique CO_2 dataset obtained during SPURT allowed us to identify the source region of this tropospheric air using the propagation of the CO_2-seasonal cycle.

Figure 3 shows the temporal change of CO_2 for potential temperature levels relative to the local tropopause. The black curve reflects the seasonal variation of CO_2 in the extratropical upper troposphere. Above the local tropopause a similar seasonal cycle is evident (green curve) illustrating a strong coupling with the local upper troposphere. However, at larger distances from the local tropopause the

spring maximum occurs three months later (blue and red curves). This phase transition indicates a surprisingly clear separation between the tropopause mixing layer and the region above. As shown in HOOR et al. (2004b) the delayed phase is the result of transport of tropospheric air from the tropical tropopause layer, which most likely is a major source region for tropospheric air in the lowermost stratosphere even at mid and high latitudes. The estimated transport times are only of the order of weeks, highlighting the importance of the impact of tropical tropospheric composition even on the lowermost stratosphere over Central Europe.

Figure 3: CO_2 seasonal cycle at different distances from the local tropopause.
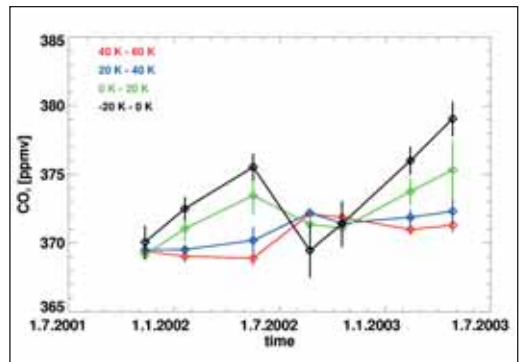
Water vapour in the lowermost stratosphere

Water vapour in the tropopause region is a crucial parameter for the radiation budget of the atmosphere. Transport of moist tropospheric air across the extratropical tropopause leads to significant moistening of the lowermost stratosphere. Measurements of H_2O in the lowermost stratosphere made by the group of the Research Centre Jülich often showed significantly enhanced mixing ratios of several 10 ppmv, well above the typical stratospheric back-

ground. Similar as derived from CO measurements, a mixing layer following the tropopause was identified. Lagrangian studies indicated that this layer is influenced by transport from the troposphere on time scales of days to weeks. But even above this layer, H_2O mixing ratios were frequently influenced by the troposphere. Long-term simulations using the Jülich CLaMS model could reproduce these enhanced H_2O values which originate from transport on longer time scales.

The H_2O data obtained in the UTLS region were analysed using probability density distributions for each season. These distributions are most compact when displayed as a function of PV. Figure 4 shows the H_2O-distribution for the two summer campaigns and compares its mean with those of the other seasons. In the lowermost stratosphere, i.e. at PV > 2 PVU, a clear seasonal cycle is apparent with a maximum H_2O-abundance during summer (orange line). We can

Figure 4: Frequency distribution of H_2O for both SPURT summer campaigns. The lines show the mean H_2O distribution for summer (orange), autumn (red), winter (blue) and spring (green).
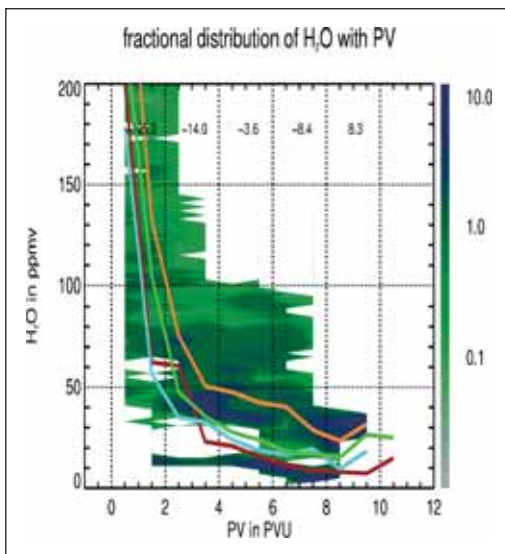
show that this seasonal cycle is mainly determined by the entry value of H_2O at the extratropical tropopause. The SPURT data show, that transport of air across the tropopause is often accompanied by the formation of clouds. In the extratropics, the tropopause thus acts as a cold trap and freeze-dries the air crossing the tropopause, dependent on the actual local temperature. The latitudinal gradients of tropopause temperature explain the finding, that air which has crossed the tropopause at higher potential temperature in the subtropics is generally dryer than that passing the tropopause at mid latitudes.

Spatial tracer structures in the tropopause region

The evolution of tropopause displacements and the advection of dynamically active and passive tracers can approximately be modelled by Juckes' model (Martin Juckes, developed at Meteorological Institute, University Munich) for quasi-geostrophic tropopause dynamics. The Juckes model with the contour advective semi-Lagrangian (CASL) algorithm made it possible to analyse the formation and evolution of small scale structures in the tropopause region with very high spatial resolution. Idealized experiments were carried out at the University of Mainz starting from two cyclonic anomalies with column-like tracer distributions in the tropopause region. The evolution of the tracer distribution was examined by different diagnostic methods. The "contour surgery" algorithm was used to remove filaments below a predefined cut-off scale, which allowed us to quantify the rate of filamentation by "coarse graining". Furthermore, contour length diagnostic was calculated as a measure of the amount of filamentary structures. Both the contour length and the rate of filamentation have a maximum at an intermediate distance from the tropopause (Figure 5), which can be explained in terms of a transition between dynamically active and passive tracer advection (WIRTH et al., 2004). This result indicates that stirring and mixing efficiency sig-

nificantly depend on the vertical distance from the tropopause.

A channel version of the "Lokal-Modell" (LM) of the German Weather Service (DWD) was applied to examine the behaviour of the tropopause in a setting which is more realistic than Juckes' idealized model. Numerical experiments were performed starting from an initial state, which consists of a perturbed jet flow. The sharpness of the tropopause was studied during the nonlinear stage of the baroclinic wave development. The motivation stems from previous theoretical studies suggesting that the observed tropopause sharpening can be caused by dynamical processes. The results show several features expected by the theoretical considerations. The effect of vertical convergence and divergence proved to be significant for the sharpening and smoothing of the tropopause locally. Bands with sharp and smooth buoyancy frequency profiles along the jet indicate the effect of transverse secondary circulations. Cut-off

cyclones and anticyclones contain smooth and sharp profiles, respectively, corresponding to earlier results with balanced dynamics.

Figure 5: Total length of tracer contours as a function of distance from the tropopause at different times (scaled units)
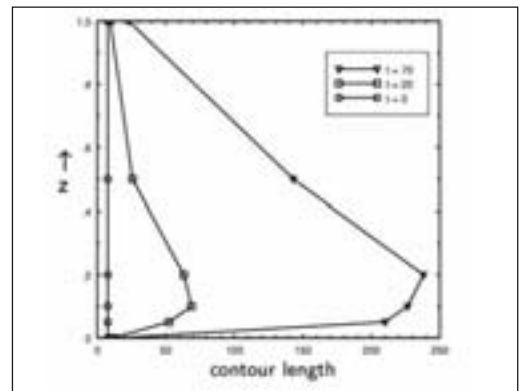
CASH-AIC: An automatic in-flight calibration method for on-line humidity measurements aboard commercial aircraft based on the MOZAIC humidity sensor

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Introduction

MOZAIC (Measurement of Ozone and Water Vapour by Airbus In-service Aircraft) has demonstrated that commercial aircraft are a reliable and cost efficient

platform for accurate humidity measurements (HELTEN et al., 1998). Pre-requisite for operational use in meteorological networks is the on-line data provision, which hitherto cannot be achieved in MOZAIC because of the need for recalibration of the sensor in

the laboratory after each month of flight operation. Aim of CASH-AIC (Compact Airborne Sampling Humidity with Automatic In-flight Calibration) was the development of an in-flight calibration method for continuous and real time measurements on in-service aircraft.

Characteristics of the MOZAIC-Humidity Sensor

The capacitive MOZAIC-humidity sensor is mounted in an aeronautic housing close to the nose of the aircraft. Due to the strong speed reduction in the housing the sampled air is subject to an adiabatic temperature increase of about 30°C at cruise altitude. Therefore, the detected relative humidity (RH_D) at the sensor is much lower than the actual relative humidity of the ambient air. At constant sensor temperature (T_D), RH_D is a linear function of the sensor signal (U_D):

$$RH_D = A(T_D) + B(T_D) \bullet U_D \qquad (1)$$

Offset (A) and slope (B) are determined in the calibration procedure as a function of T_D. (HELTEN et al., 1998). Calibrations during several years of MOZAIC demonstrated that the long term drift of (A) is the critical parameter determining the accuracy. Therefore, it must be monitored and adjusted in-situ, while the slope (B) can be determined at much longer intervals in the laboratory.

Methodology

The method developed for automatic in-flight calibration is to determine the offset $A(T_D)$ in Eq.(1) from the measurements themselves whenever the aircraft is flying in the lower stratosphere, where the water vapour mixing ratio is low and rather well defined at values of 5±1 ppmv (KLEY et al., 2000). Figure 1 shows the humidity sensor's raw signal, obtained during

one month of flight operation, as a function of sensor temperature. The stratospheric portion of the data (red dots) is determined from potential temperature, which are calculated in situ from pressure and temperature, against a threshold given by the average potential temperature at the tropopause as a function of latitude north of 40°N.

To determine the sensor offset $A(T_D)$, an *a-priori* calibration curve is fitted in the temperature interval 245-265 K to the lowest signal values identified as stratospheric. The *a-priori* calibration curve is a composition of $A(T_D)$ and the signal produced by a water vapour mixing ratio of 5 ppmv. The MOZAIC calibration record shows that $A(T_D)$ can be described by a third order polynomial. The functional dependence is the same for all sensors. Hence, only the offset of the polynomial needs to be adjusted by the fitting proce-

Figure 1: Raw signal of the MOZAIC humidity sensor as function of sensor temperature during one month of flight operation. Stratospheric measurements are indicated by the red dots. The black line is the empirical calibration curve for the sensor offset including the signal for 5 ppmv of water vapour.
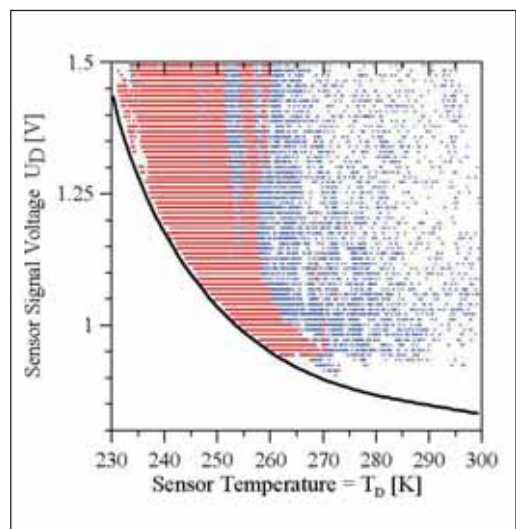
dure. The thus derived offset A is used for the subsequent real time retrieval of atmospheric humidity data and serves as the *a-priori* curve in the next evaluation, which is conducted whenever a sufficient ensemble of new stratospheric data are available.

Evaluation of the Method

For testing of its robustness, the AIC-algorithm was applied month by month to two years of raw data from MOZAIC flights and compared with the RH data based on the laboratory calibrations conducted after every 500 hours of flight operation. Figure 2 shows that the AIC-method has no significant bias ($< 0.5\%$ RH). The standard deviation is better than $\pm 4\%$ RH in the upper troposphere and $\pm 6\%$ in the boundary layer.

The good precision and accuracy of the MOZAIC humidity sensor was confirmed by in-flight comparison with a Lyman(α)-fluorescence hygrometer on board a research aircraft in collaboration with the AFO2000 project SPURT showing agreement within $\pm(3-6)\%$ RH. In conclusion, through regular (14 weeks) in-flight correction of the offset A with the AIC-

method, the MOZAIC sensor can provide real time RH-measurements with an uncertainty of $\pm(5-10)\%$ RH.

Figure 2: Deviation between the CASH-AIC method and the calibrated MOZAIC data as a function of altitude. Red line: average; error bars: standard deviation.
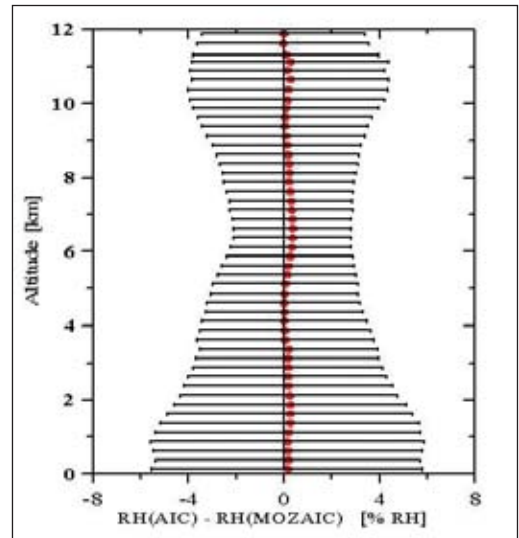
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Scaling behavior and gravity waves in CRISTA data (CRISCA)

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Motivation

We best can motivate the CRISCA project by quoting the recent Fritts and Alexander (2003) review paper on atmospheric gravity waves (GWs):
"Parameterizations of middle atmosphere gravity wave effects presently suffer acutely from the lack of constraints on their inputs and tuneable parameters. ... The process of tuning gravity wave parameterisations in global models to attempt to infer the properties of the gravity wave input parameters is unwise at present because the range of parameters is too broad, and the solutions are not unique. As a result, GCMs tuned for one result (and at one resolution) using a gravity wave parameterization cannot confidently be used in any prognostic way." The global measurement of GW momentum flux and the inference of GW sources is therefore an important open issue for reliable climate predictions. This is also true for other GCM-subgrid processes.

Scaling behaviour

The so-called CRISTA staring mode provides radiance data of unprecedented horizontal resolution at four different altitudes. Using wavelet technique power spectra were deduced for two different regions of the globe. Spectral slopes are close to -5/3, but vary with region. However, staring mode data are only a very small subset of the CRISTA data (EIDMANN et al., 2001). The standard data modes are sampled too irregularly to obtain good results by standard wave analyses. Instead, an autocorrelation method was applied. A consistent scaling behaviour of along-track data, random pairs, and radiosonde pairs was obtained (EIDMANN et al., 2002). The results are of practical relevance for satellite validations.

Gravity wave analysis

The analysis technique used in various previous CRISTA studies was improved to estimate the vertical flux of horizontal momentum due to GWs. Because of strong aliasing effects only the absolute values of GW momentum flux (GW-MF) can be deduced from CRISTA data. These correlate well with modelling results employing the Warner and McIntyre GW parameterisation scheme. Two main results are obtained (ERN et al., 2004a,b):

1. **GW variances are not necessarily a proxy for the global distribution of GW-MF. This is illustrated in Figure 1.**
2. **The parameterisation scheme describes the measurements best if the waves are launched at a low launch level.**

In a case study CRISTA data have been utilized to investigate a large scale stratospheric GW above supertyphoon Winnie. Cloud top brightness temperatures measured by the GMS-5 nadir observing satellite

indicate that a 24 h period oscillation in the convection of the typhoon system is the source of the wave (PREUSSE, 2001). Motivated by the experimental findings a model study on the basis of the MM5 mesoscale model was initiated. In this model study a dominating period of 15 h was found. The typhoon clearly acted as GW source and the stratospheric phase fronts closely resembled the structures of the tropospheric cloud bands (cf. Preusse et al., 2003 and Figure 2, simulated GWs in colour, measured cloudbands in grey).

Motivated by previous analyses of CRISTA data (Preusse et al., 2001), we used a 1.5 year data series of CLAES GW temperature variances to statistically investigate convective generation of GWs. Regions of high SST in the northern subtropics could be clearly identified as important GW source regions by correlational analysis (PREUSSE AND ERN, 2004). The combination of CRISTA and CLAES results shows that a new satellite mission providing even higher resolution than CRISTA for a time period of several years could provide fascinating new insight in the

global distribution of GW-MF as well as in different GW source mechanisms. Such a new instrument appears feasible based on current technology. Current best resolution achieved by CRISTA (coloured dots, left side) and resolution aimed at by the new instrument (right side) are compared in Figure 2 (see also: http://www.fz-juelich.de/icg/icg-i/gloria).

Figure 2: Simulated GWs (MM5) above Typhoon Winnie.

Figure 1: CRISTA-2 (August 1997) global GW distribution at 25 km. Figure adapted from ERN et al. 2004.
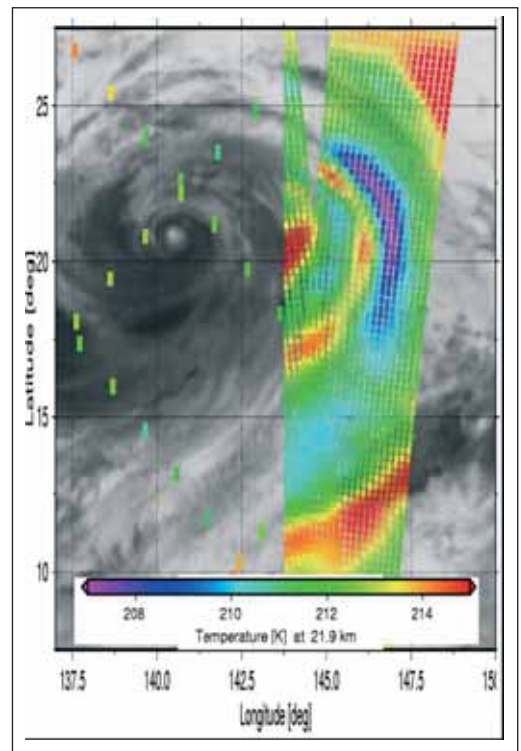
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Inertia - gravity waves and their connection to breaking Rossby waves (LEWIZ*)

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The investigation of the generation and propagation processes of inertia-gravity waves (IGW) in the troposphere and stratosphere in connection with poleward Rossby wave breaking (RWB) events is the main aim of this project (Peters & Waugh, 1996, O`Sullivan & Dunkerton, 1995).

Climatology based on ERA-40 dataset

The mean winter circulation based on ERA-40 data (1991-2001) over Northern Germany was studied. The mean zonal wind shows a value of 15 m/s around the tropopause region and increases up to 70 m/s with height. A high variability is found in the jet regions. The tropopause is placed at about 11 km height but the stratospheric cold point temperature (67° C) at about 25 km. Based on the dispersion relation good conditions for IGW propagation occurred over Northern Germany. By counting RWB-events during our winter campaigns 1999-2003 we found that for more than 40 % of winter days, Northern Germany was under the influence of RWB events. In 30 % of winter days a poleward RWB event was observed. Our campaigns covered eight poleward downstream RWB events, with or without polar vortex, and capture 3 upstream RWB events.

Radiosonde and VHF radar measurements over Kühlungsborn

During the campaigns we measured parallel the intrinsic frequency of IGW by the radiosondes and the fixed-observer-frequency by radar for the first time and estimated the horizontal wavelength over the Doppler shifting equation. For studying the structure of RWB events and their connections to IGW, 11 field campaigns were prepared and conducted in the winter seasons 1999-2003. The gathered campaign-data were analyzed with standard methods in order to estimate energy spectra and phase diagrams. Closer inspection of the campaigns showed IGW with 600-850 km zonal wavelength, 2-4 km vertical wavelength and intrinsic periods of 12-12.5 hours. The waves are generated at the exit of tropospheric jet streaks by geostrophic adjustment and are propagating upward and downward but upstream. In Figure 1 four typical horizontal positions of the jet streak in the tropopause region and the evolution of various RWB events of the LEWIZ-campaigns are shown. The specific location of the tropopause jet streak and the appearance of the stratospheric jet cause a different IGW generation and propagation.

MM5 – mesoscale model simulations

For modelling of the respective dynamics a non-hydrostatic version of the MM5 (NCAR / Penn State Fifth-generation Mesoscale Model) was successfully implemented. The MM5 has been used to get a consistent dataset.

The known evolution of the Rossby wave was included in an area of 5000 km extend, the IGW were resolved with a spatial resolution down to 8 km horizontally and 100 m vertically. We run the model also for the above mentioned campaigns and found IGW with parameters: 360-860 km, 2.9-6.2 km and 3.7-8.7

hours. These jet-generated waves contribute 40 %, orography 40 % and convection 20 % to the overall wave energy (Fig. 2). For a run without orography, moisture and surface friction we found long IGW too, which confirms the active role of the jet stream in the generation process. It is also shown, that shorter IGW

(about 200 km) excited by orography and deep convection travel faster in the troposphere and stratosphere. More detailed and additional results are available on the LEWIZ webpage[+] and in the papers of PETERS et al. (2003), SERAFIMOVICH et al. (2004) and ZÜLICKE & PETERS (2004).

Figure 1: Schematic diagram of RWB events and jet streak position (marked by an arrow). Ertel´s potential vorticity maps at 330 K isentropic layers as well as mean zonal wind (at 330 K and 500 K layer) calculated from ECMWF analysis data are shown. A star marks the position of Kühlungsborn. The evolution of the Rossby wave shows a strong northeasterly transport of air masses.
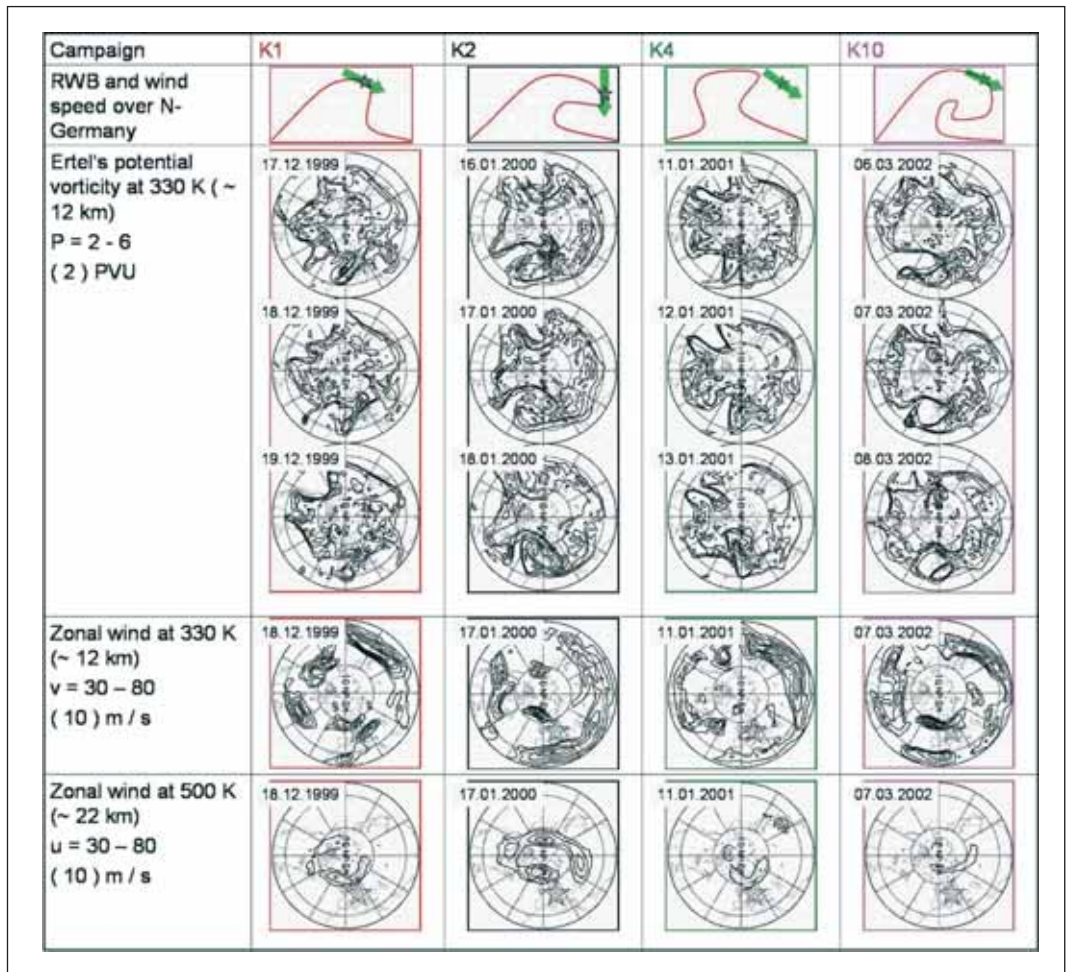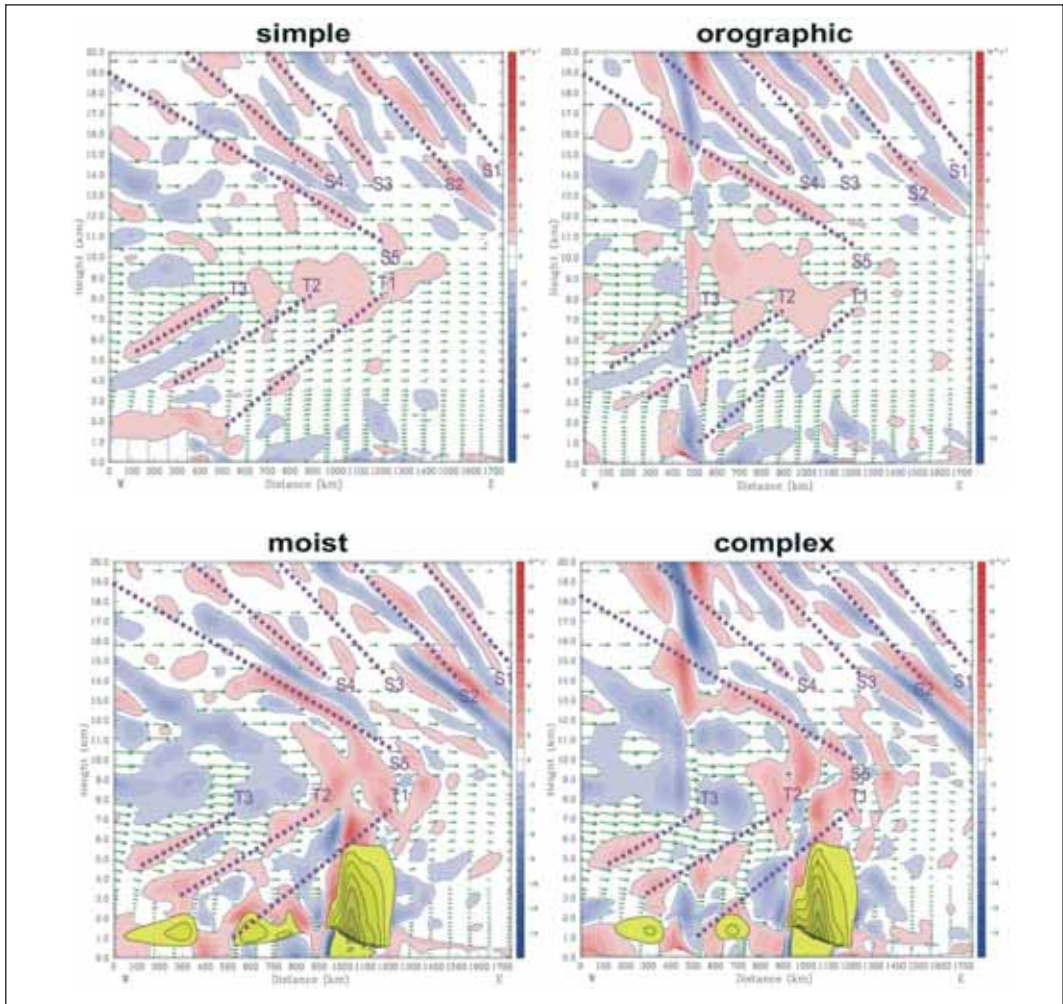
Figure 2: Vertical cross section through the medium-resolution run (x = 24 km, z = 250 m) with different physics for campaign KI (simple, orographic, moist, complex). It is shown the horizontal divergence (red/blue), wind vectors (green) and precipitation mixing ratio (yellow). Some IGW are marked with a dotted violet line.

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* **Le**tzte **Wi**nter im **Z**wanzigsten Jahrhundert

\+ http://www.iap-kborn.de/modell/PETERS/LEWIZ/lewizafo.html

Coupling of dynamics and atmospheric chemistry in the stratosphere (KODYACS)

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Summary

The primary objective of the AFO 2000 project KODYACS was to identify and quantify the coupling of dynamical, chemical, and (micro-)physical processes in the upper troposphere / lower stratosphere (UT/LS) and the middle atmosphere (i.e. stratosphere and mesosphere). Also, the interaction of the different atmospheric layers themselves was examined. Investigations were based on a hierarchy of atmospheric models and on long-term observations. Coupled Chemistry-Climate Models (CCMs) were employed to simulate dynamic (climate) and chemical changes during the recent four decades. Model results have been intensively validated with multi-year observations derived from ground-based and satellite measurements, including new data products of MIPAS/ENVISAT, and with results derived from Chemical-Transport Model (CTM) calculations. A detailed inter-comparison study has been carried out to quantify the individual natural and anthropogenic components of dynamic and chemical variability of the stratosphere. The agreement between analyses of model results and observations is surprisingly good, although there are obvious indications for model deficiencies which emphasises the need for further improvements of the model systems.

Strategy

During recent years, fully coupled CCMs have become available for multi-year simulations (e.g., HEIN et al., 2001; STEIL et al., 2003). They have been used for various scientific investigations, for example to study the interaction of climate change and atmospheric chemistry (BRÜHL et al., 2001; SCHNADT et al., 2002, MANZINI et al., 2003), aerosol chemistry interactions after large volcanic eruptions (TIMM-RECK et al., 2003) or dynamic coupling of different atmospheric layers (SCHNADT AND DAMERIS, 2003; MAGER, 2004). Uncertainties of CCMs have been analysed (AUSTIN et al., 2003; SHINE et al., 2003). First results have been used to estimate the future development of the stratospheric ozone layer (WMO,

Figure 1: Left panels: Geographical distribution of the long-term trend in total column ozone in winter (December, January, February), derived from TOMS/SBUV satellite data for 1979 to 2002, and from simulations performed with MAECHAM4/CHEM and E39/C for 1978 to 1999. Right panels: Same, but for the size of ozone variations related to the 11-year solar cycle. Here the size is defined by two standard deviations of ozone variations attributed to the 11-year solar cycle.
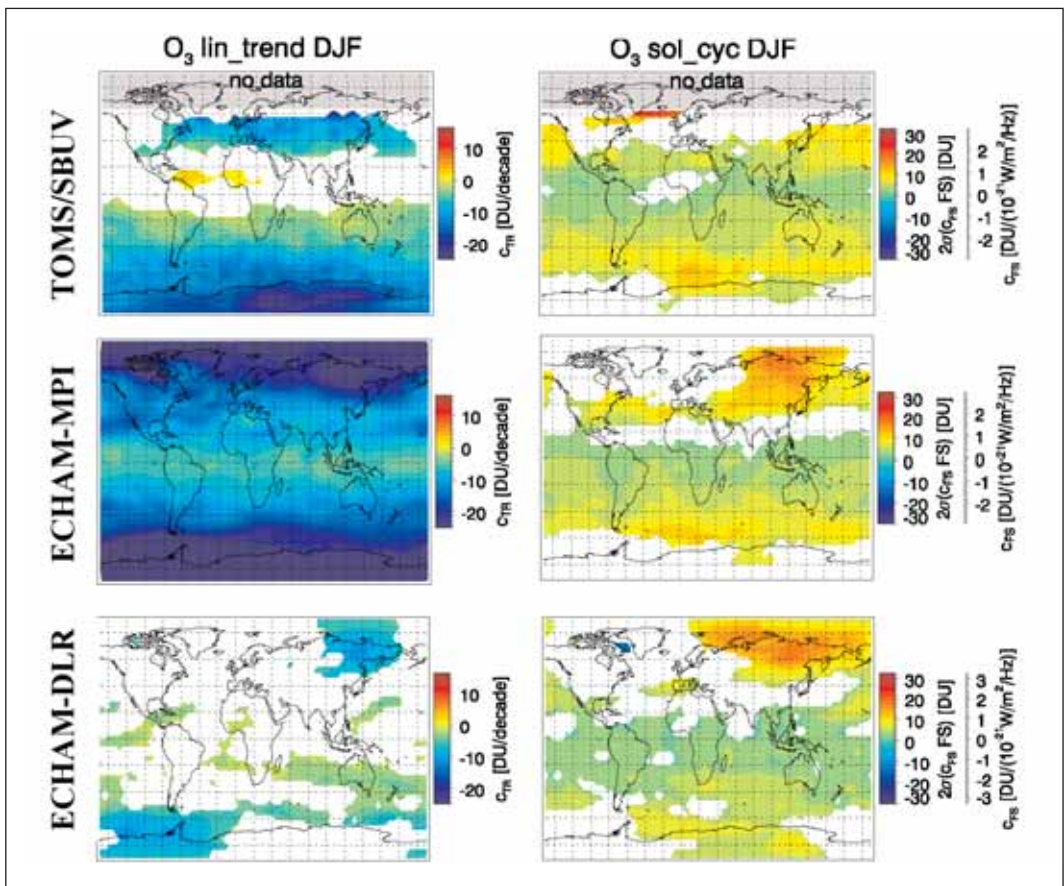
2003). Within KODYACS, two versions of the ECHAM4/CHEM model system were employed for 40-year transient simulations (1960 to 1999): E39/C (39 model layers between the surface and 10 hPa), and MAECHAM4/CHEM (39 model layers between the surface and 0.01 hPa). For details see HEIN et al. (2001) and STEIL et al. (2003). For both model simulations identical boundary conditions were adopted, to account for natural factors as the 11-year cycle of solar irradiance, the quasi-biennial oscillation (QBO), the effect of sea surface temperature variations like El Niño, and the chemical and direct radiative effects of the major volcanic eruptions. The anthropogenic influence was represented by specifying atmospheric concentrations of the most important greenhouse gases, chlorofluorocarbons (CFCs), and nitrogen oxide emissions.

Highlights

Inter-annual variations of ozone and temperature in observations and model simulations

Anthropogenic emissions of chlorine, bromine and greenhouse gases lead to a decline of stratospheric ozone concentrations and to stratospheric cooling during the last 25 years. In contrast, ozone and temperature have generally increased in the troposphere. These long-term changes are masked by a variety of natural variations, which can be quite large. Detection of any anthropogenic signal, e.g. a beginning recovery of the ozone layer (STEINBRECHT et al., 2004a,b), requires accurate knowledge of these natural variations.

A multiple linear regression scheme was used to quantify the natural variations and the anthropogenic components of the long-term trend. For the first time, not only the best available long-term observations from satellites, ozone-sondes and -lidars were analysed (STEINBRECHT et al., 2003), but also results from two 40-year transient CCM simulations. As one example, Figure 1 shows the global distribution of the overall long-term total column

ozone trend and the amplitude of variations due to the 11-year solar cycle. Despite differences between observations and models in the magnitude of the ozone trend, the results for most other modes of variability are very similar. In particular, very realistic QBO- and solar cycle-related variations are now reproduced by the model simulations. This is a major step forward.

Next steps could be a much more reliable simulation of the future evolution of the middle atmosphere, a detailed analysis of the underlying processes, or the study of observed and modelled stratosphere to troposphere couplings for better long-range weather forecasting.

Evaluation of CCM results with CTM calculations

The recovery of Arctic polar stratospheric ozone is uncertain. Its first signs are predicted between 2004 and 2019 (AUSTIN et al., 2003). The CTM CLaMS was employed, adopting meteorological and chemical boundary conditions derived from E39/C timeslice experiments (SCHNADT et al., 2002). Thus, offline-CTM calculations were performed on the basis of coupled CCM calculations. E39/C underestimates cold-winter ozone depletion compared to observations and to detailed CTM studies. Among other factors, bromine chemistry is not included in E39/C. Bromine chemistry accounts for up to 30% of the ozone depletion in CTM simulations. The coarse model resolution (T30) does not resolve filaments and the isolation of the vortex, which enhances the import of nitrogen species (NO_x and HNO_3) into the polar vortex. Together with insufficient denitrification this leads to enhanced deactivation of active chlorine species. One of the main results of the offline simulations carried out with CLaMS was to quantify the sensitivity of vortex ozone loss to mixing and resolution. The resolution was varied from 50-400 km, mixing intensity was varied over a range of critical mixing parameters (KONOPKA et al., 2003). However, even considering

exaggerated mixing, low model resolution and no denitrification, CLaMS ozone loss is higher than ozone loss found in E39/C. In addition, the sensitivity of simulated ozone loss to neglected bromine chemistry was assessed and is shown in Figure 2. The results of the CTM simulations will help to correct and improve E39/C and other CCMs in the near future.

Water vapour distribution in the TTL derived from MIPAS/ENVISAT

MIPAS on ENVISAT is a high-spectral resolution infrared limb emission sounder on a sun-synchronous polar orbit which provides about 1000 vertical pro-

Figure 2: Arctic chemical ozone loss inside the polar vortex on the 475 K isentropic surface, integrated over the time period from January 1 to April 1, 2015. The vortex edge was defined by the 36 PVU contour line. Chemical depletion in E39/C (green bar) was analysed using a methane-ozone correlation for the whole timeslice, with the coldest winter of the timeslice being on the far right of the bar. Sensitivity studies on this particular winter were performed with CLaMS. The contribution of ozone destruction cycles are shown in yellow. With regards to a reference simulation (grey line), sensitivities to horizontal resolution, mixing intensity and denitrification were performed (variabilities shown in cyan, combined variability including Br cycles in blue). None of these sensitivities can fully explain the underestimated ozone loss in E39/C. For the coldest winter the E39/C underestimation of ozone destruction can be quantified as 600 ppbv or 55%.
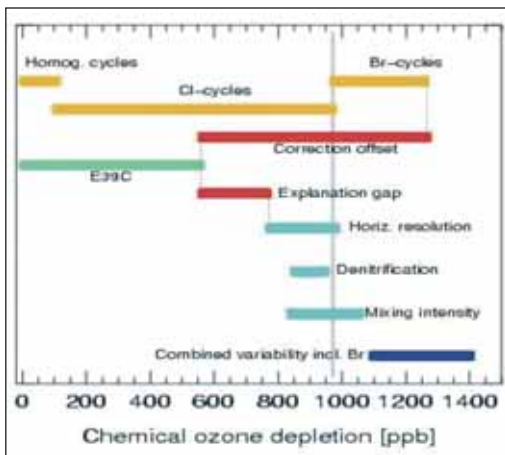
files per day of the atmospheric radiance in the spectral region from 4.15 to 14.6 μm since summer 2002. Vertical profiles of up to 35 various atmospheric parameters, temperature and cloud top heights are retrieved between 6 and 68 km altitude. Within this data set, the period September 2002 to November 2003 is covered by at least 3 days of observations per month. The retrieval of water vapour concentrations in the tropopause region is a challenging task, since the hygropause and the strong tropospheric increase of the water vapour profile is difficult to catch within the instrument's vertical resolution of 3 to 4 km. The high tropospheric water vapour mixing ratios lead to rapid saturation of the radiance data. In addition, signatures of thin clouds have to be distinguished carefully from water vapour signatures (MILZ, 2003; MILZ et al., 2004). From the retrieved data, mean altitude-latitude cross-sections for 10° latitude bands, as well as global distributions averaged over bins of 15° longitude x 5° latitude, were generated for several altitudes. The global distribution of water vapour averaged for June, July and August 2003 at 18 km altitude is shown in Figure 3 (top part). First inter-comparisons with model results have been started. Further studies should improve our understanding of the transport through the TTL, of dehydration processes, of the role of cirrus clouds, and of seasonal and interannual variations and the possible trend of stratospheric water vapour.

Conclusion

A wealth of significant, leading edge results have been achieved in KODYACS (see http://www.pa.op. dlr.de/kodyacs). A solid basis has been laid for improved and more reliable predictions of dynamics (climate) and chemical composition of the lower and middle atmosphere. Upgraded numerical tools are now available for better estimates of the future development (recovery) of the stratospheric ozone layer because the interactions of changes in climate and chemical composition can be considered. Moreover,

the investigations carried out in this project indicate a potential for the application to seasonal weather forecasts. KODYACS has been instrumental in creating a capacity for state-of-the-art chemistry-climate modelling in Germany. The links and interactions between modelling and observations groups have been strengthened and greatly improved. KODAYCS has contributed to create substantial know-how in Germany.

Figure 3: Upper panel: Mean global distribution of water vapour (in ppmv) at 18 km altitude for June, July and August 2003 derived from MIPAS/ENVISAT observations. Lower panel: Corresponding result derived from the CCM MAECHAM4/CHEM. Lowest water vapour distributions are found inside the Antarctic polar vortex, due to dehydration by formation of PSCs, as well as in the TTL over South-East Asia, which is correlated with lowest temperatures and highest-reaching convection there. In the subtropics, belts of high water vapour mixing ratios are found, with maximum values over the Arabic peninsula.
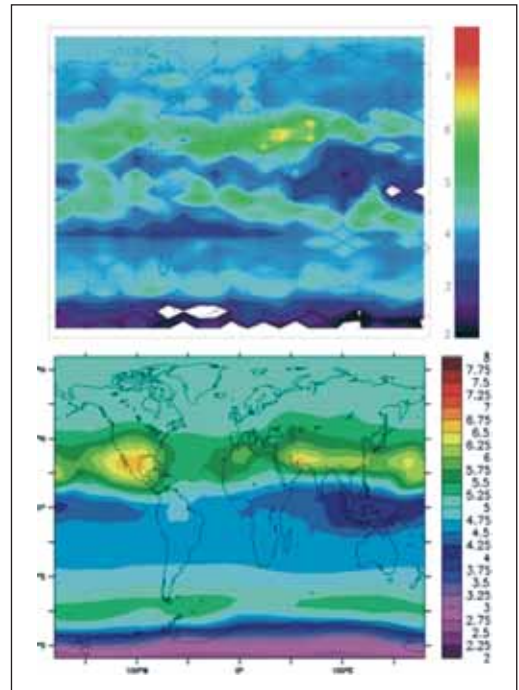
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Mesospheric dynamics, energetics and chemistry (MEDEC)

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Introduction

The atmospheric region above the stratopause is in general much less understood than the region below. Or, as Paul Crutzen (1997) stated, a number of mysterious issues remain to be solved in the mesosphere. The overall objectives of the MEDEC project were (1) to improve our under-standing of the coupling between the dynamics, energetics and chemistry of the mesosphere, (2) to assess how these processes affect the global atmosphere, and (3) to estimate the vulnerability of the mesosphere to natural and human-induced perturbations. These questions were tackled by collecting and analyzing ground-based, rocket-borne and satellite observations as well as by developing and ap-plying complex three-dimensional numerical models of the entire atmosphere. In the following, some of the scientific progress obtained within MEDEC will be highlighted.

Temperature and Dynamics

During the summer months the upper mesosphere is characterized by the lowest temperatures in the Earths atmosphere leading to such spectacular phenomena as noctilucent clouds (NLC) and polar mesosphere summer echoes (PMSE). Different ground-based and rocket-borne measurements in the upper mesosphere and lower thermosphere performed by IAP enhance our knowledge of temperature and dynamics in this region. For example, in 2001, first in-situ and remote measurements of the thermal structure of the summer mesosphere at very high latitudes were performed from the north polar island Spitsbergen (78°N, LÜBKEN AND MÜLLEMANN, 2003). This database was complemented by intensive lidar measurements in 2002 and 2003. We now have a quasi-permanent temperature coverage for the period from February to October. The observations were used in an iterative process to evaluate and improve results of the newly developed chemistry and climate model HAMMONIA of MPI-M (see section 5). The comparison of temperatures simulated for the upper mesosphere at 78°N and observed with the "falling sphere" technique is shown in the left part of Figure 1. The current model version reproduces the seasonal variation of temperatures in the summer mesosphere provided that the model altitudes are corrected by ~3 km. Observations of winds (right part of Figure 1) and turbulence parameters (e.g. MÜLLE-MANN et al., 2002) complete the data sets.

The global picture of mesospheric temperatures is improved by the analysis of infrared spectra observed during the CRISTA (CRyogenic Infrared Spectrometers and Telescopes for the Atmosphere) satellite missions in November 1994 and August 1997 by UNI-W
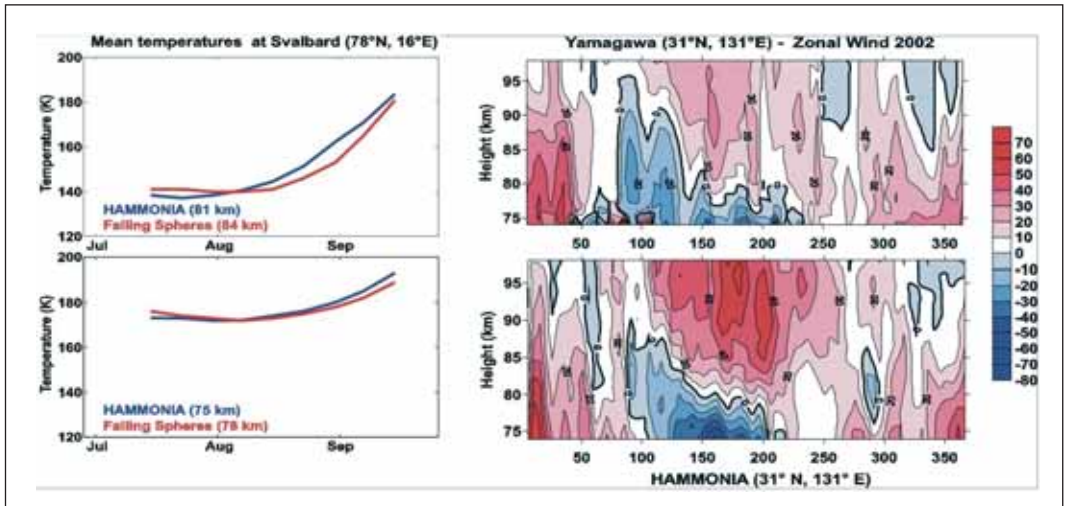
Figure 1: (Left panel) Mean temperature variation at 78°N. Seven day mean values are displayed. They show very good conformities between the "Falling Sphere" data (red lines) and the HAMMONIA model output (blue lines), shifted by values of 3 km. (Right panel) Annual cycle of zonal winds (m/s) observed by MF-radar at Yamagawa (31°N, 131°E) in the year 2002, and simulated for the respective location by the HAMMONIA model for an arbitrarily chosen year.

(GROSSMANN et al., 2004). Figure 2 shows a polar view on temperatures at 82 km in August 1997. The zonal structures observed by CRISTA in the mesosphere indicate that the now commonly accepted dynamical control of the stratospheric trace gas distribution by

Figure 2: Temperature at an altitude of 82 km around the North Pole (August 1997) observed by CRISTA.

planetary waves also governs the mesosphere. Respective structures were found in temperatures and ozone concentrations in the winter hemisphere as well as in the summer hemisphere in August 1997.

Chemical composition
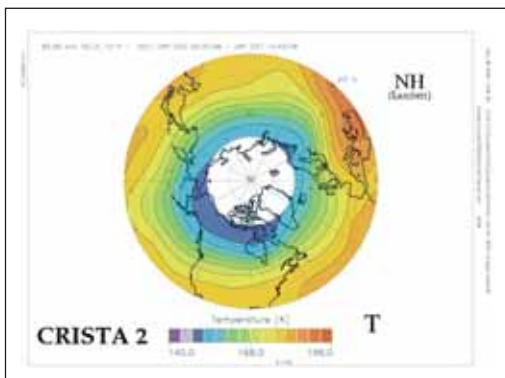
CRISTA is a triple telescope cryogenically cooled infrared spectrometer which senses the earth limb from a Shuttle orbit. The geographical coverage was -57°/+68° and -74°/+74° during the two missions of slightly more than one week duration each. Besides temperature, CRISTA data allowed the analysis of pres-sure, winds (derived from temperatures), and a set of trace gases including ozone (KAUFMANN et al., 2003), carbon dioxide (KAUFMANN et al., 2002), carbon monoxide, methane, water vapour and atomic oxygen.

An important finding regarding mesospheric trace gases was the discovery of the tertiary ozone maximum in the winter mesosphere at about 70 km

in a study performed jointly with colleagues from NCAR (National Center for Atmospheric Research, Boulder, USA, MARSH et al., 2001). This feature is the result of a decrease in atomic oxygen loss by catalytic cycles involving odd hydrogen in the vicinity of the terminator where the atmosphere becomes optically thick for the radiation (Lyman-α) leading to water vapour photolysis. Figure 3 shows the tertiary maximum in both CRISTA observations and HAMMONIA simulations.

Figure 3: Ozone volume mixing ratios as observed during August 1997 by CRISTA (from Marsh et al., 2001, data averages for local times of approximately 0000, 0400 and 0500 hours for 70S (black), 30S (red) and 2S (green), respectively) and simulated by HAMMONIA (monthly mean zonal average for August). Both data sets show the tertiary ozone maximum in the mesosphere at high southern latitudes.
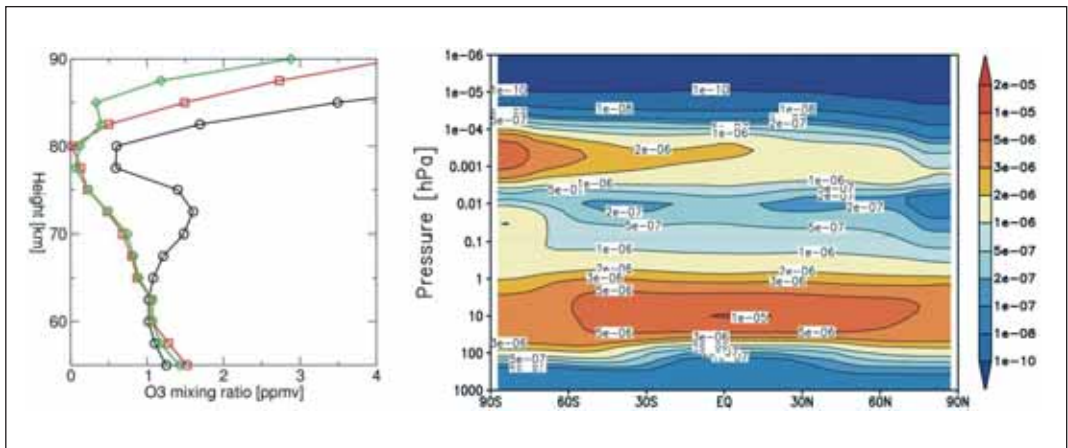
The quasi two-day wave: investigations on the origin and propagation in the middle atmosphere

The quasi two-day wave (QTDW) is one of the most prominent features of the upper middle atmosphere. This planetary wave with a zonal wave number of 3 and a period of about two days is regularly observed in the summer hemisphere during solstice conditions where it exhibits strong amplitudes. The phenomenon lasts for few weeks, while during the rest of the year the wave amplitudes are small but still observable. Two mechanisms of origin and excitation of the QTDW are in discussion. The wave can be iden-

tified as a solution of the Laplace tidal equations but this can not explain their amplification at each summer season. It is therefore assumed that the QTDW can be enhanced or even excited via instability processes. Likewise, interactions with gravity waves, solar tides and other planetary waves can play a role. The behavior of the QTDW was investigated at UNI-L using the non-linear mechanistic circulation model COMMA-LIM (Cologne Model of the Middle Atmosphere - Leipzig Institute for Meteorology, FRÖHLICH et al., 2003b).

The QTDW is excited in the model as an additional heating term. The latitudinal structure is calculated as Hough-Mode of the appropriate Rossby-

gravity wave (3.0). The propagation conditions of this free traveling wave were investigated as well as the interactions with other waves (FRÖHLICH et al., 2003a) and with the zonal mean flow. To investigate the possibility of wave generation via instability processes a spectral model was used (MERZLYAKOV AND JACOBI, 2004). It could be shown that indeed a QTDW develops depending on an artificially increased summer mesospheric jet (Figure 4).

Figure 4: Dependence of the amplitude (in m/s) of the 2-day wave (right panels) on the velocity of the easterly jet (left panels, values given in m/s). Axis labels are approximate (log-pressure) altitude (km) and co-latitude.
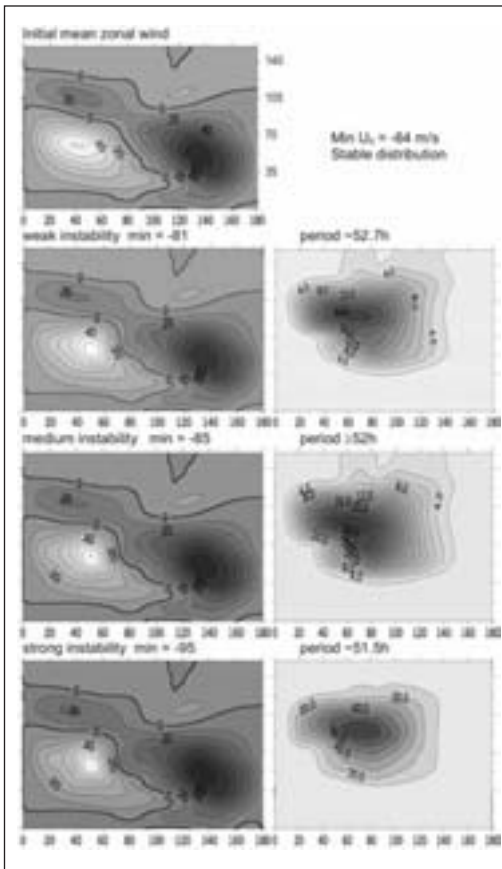
The response of the mesopause region to natural and anthropogenic climate forcing

Before the start of this project coupled general circulation and chemistry models of the whole atmosphere did not exist. However, such a model is an essential tool for studying the mesosphere, its response to climate variability, and interactions with other height regions. The main task of MPI-M within the MEDEC framework was the development and application of such a model, HAMMONIA (HAMburg MOdel of the Neutral and Ionized Atmosphere), which covers the altitude range from the surface to the thermosphere. The model combines dynamics and physics from the ECHAM5 general circulation model (ROECKNER et al., 2003) in a fully interactive way with the MOZART3 chemistry scheme (48 compounds, 152 reactions). The model physics were extended by parameterizations accounting for important processes in the upper atmosphere: e.g. heat conduction, solar heating in the UV and extreme UV, solar heating by CO_2 in the near IR, non-LTE IR cooling, the ion drag and the gravity wave forcing (CHARRON AND MANZINI, 2002).

Model applications concentrate on the response of dynamics and trace gases in the MLT region to solar and anthropogenic climate forcing. Different simulations with HAMMONIA for low and high solar activity on the one hand, and for pre-sent day and doubled CO_2 concentration on the other hand were performed (SCHMIDT AND BRASSEUR, 2004, SCHMIDT et al., 2004). Results of the solar cycle experiments are e.g. an ozone increase for high solar activity up to 25% around the mesopause (Figure 5). The temperature is increased by 3 to 10 K (Figure 6) which corresponds well to observations listed by BEIG et al. (2003). Together with the simulated decrease in water vapor this should have implications for the NLC formation. CO_2 doubling (not shown here) leads to a temperature decrease everywhere above the
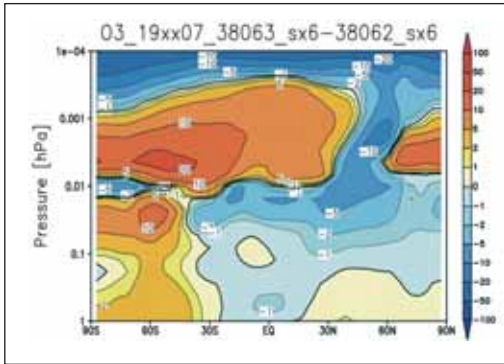
Figure 5: Relative difference of ozone volume mixing ratio between the simulations for solar maximum and solar minimum conditions. Differences are given in % as zonal mean values for the month of July.
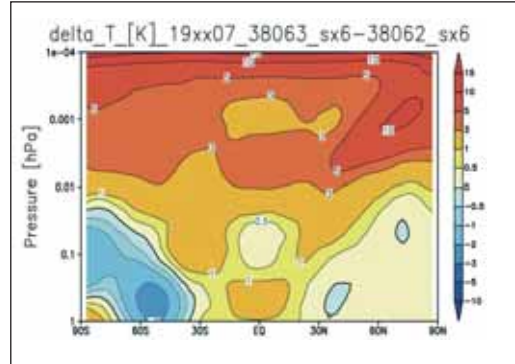
Figure 6: Temperature difference between the simulations for solar maximum and solar minimum conditions. Differences are given in K as zonal mean values for the month of July.

tropopause but by only very small and sometimes even insignificant values in the mesopause region.

A new method to compute non-LTE infrared cooling

A new efficient subroutine for estimating the total infrared radiative cooling/heating in the ro-vibrational bands of atmospheric gases in the mesosphere and lower thermosphere (MLT) was developed at MPI-M. It accounts for vibrational non-LTE and absorption and transformation of the near-infrared solar radiation. Analyzing the pressure/temperature variations of the "opacity distribution functions" (ODF) for a number of molecular bands a new approach was formulated which allows treating each ro-vibrational band as a single line of a special shape. These shape functions were tabulated and parameterized in respect to the pressure and temperature variations. Compared to the standard line-by-line algorithms this method dramatically reduces computational time by a factor that approximately equals the number of lines in the band (about 100 for CO_2, about 1000 for O_3). Opposite to various parameteriza-

tions available the new routine treats precisely the exchange of radiation between various atmospheric layers and all variety of collisional energy transfers. As a result it accurately reproduces the non-LTE line-by-line radiative cooling/heating reference calculations both for day and night conditions even for non-smooth profiles of atmospheric parameters when best parameterizations fail (Figure 7). A more detailed description of the new method is given by GUSEV AND KUTEPOV (2003).

Conclusions

The selection of results presented above shows that large progress was made in both analyzing observations and modeling of dynamics, radiation and chemistry in the mesosphere. New insight has been gained concerning the state of the mesosphere as well as the processes acting and interacting to produce this state. However, some of the "mesospheric mysteries" remain, and the developed tools as well as the new observations and their analyses will probably be of great value also in future mesospheric research.
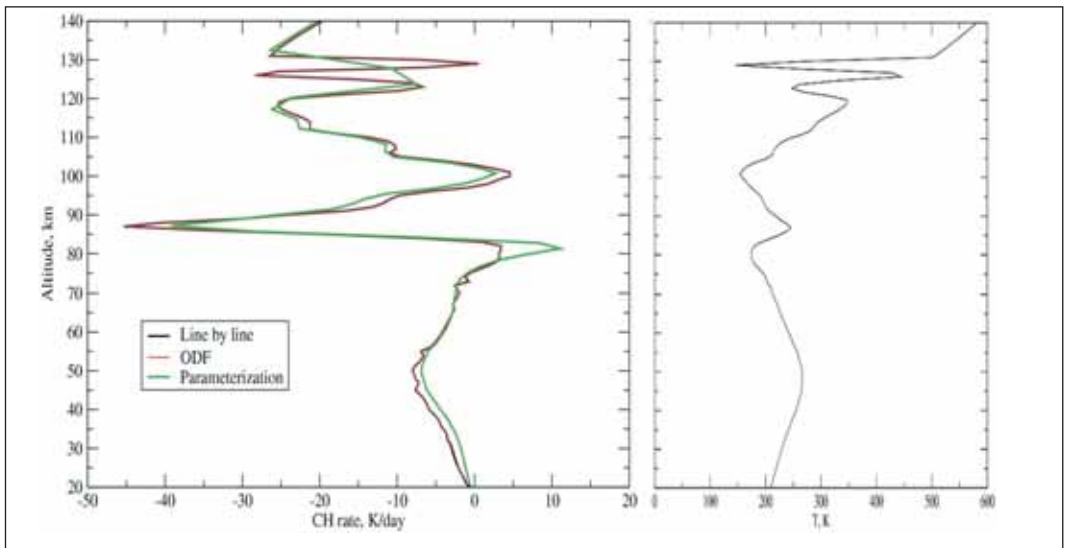
Figure 7: Cooling/heating (K/day) in CO₂ infrared bands calculated with the exact line-by-line technique (black), the parameterization of Fomichev et al. (1998, green) and the new "ODF" approach (red) for a particular temperature profile measured with the CRISTA instrument (right figure).

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On the physics of small scale structures in the upper atmosphere (OPOSSUM)

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Since some years it has been recognized that the mesosphere is probably the most sensitive region in the entire atmosphere to detect climate change. Temperature observations based on direct and indirect methods give evidence for a strong cooling of the lower mesosphere due to anthropogenic increase of greenhouse gases. At mid latitudes and at altitudes between 60 and 70 km a large negative trend of –5 K/decade is observed which is much larger compared to tropospheric trends and also much larger than expected from models. This has led to the idea that the mesosphere could be an 'early warning systeme' for climate change.

Are there any indications that such a trend may also exist at high latitudes? In summertime, layers of icy particles form in the mesopause region at altitudes between 82 and 92 km and pole-ward of 50° latitude. These ice particles cause phenomena known as noctilucent clouds (NLC) and polar mesospheric summer echoes (PMSE). The spatial and seasonal occurrence of NLC and PMSE are very similar although the physical processes involved are very different. Noctilucent clouds are the highest visible ice clouds in the Earth's atmosphere, whereas PMSE are strong radar echoes which were not explained for almost 30 years after their first detection in the 1970's. Ice clouds leading to NLC and PMSE exist due to extremely low temperatures around the polar summer mesopause (~130 K). In the frame of the OPOSSUM project we have explored the morphology of ice clouds leading to NLC and PMSE by combining modeling and observational studies.

We have investigated the major physical processes involved, in particular the micro-physical genesis of ice particles, the interaction of these particles with the background water vapor, the sensitivity of ice formation and cloud brightness to temperature variations, the role of turbulence processes in creating PMSE, and the influence of horizontal and vertical transport.

Within OPOSSUM we developed for the first time a consistent theory of PMSE which explains in detail how these strong echoes are linked to charged ice parti-cles. The theory explains why radar echoes are observed in the entire ice cloud whereas the NLC is observed in the lower part only (see Figure 1). In addition, we studied the formation of NLC/PMSE by means of a three-dimensional general circulation

model of the middle atmosphere (COMMA/IAP). In the model we filled the summer mesopause region with 20 million condensation nuclei and studied their 3-d time-dependent Lagrangian transport including ice formation, growth, and sublimation. In Figure 2 the trajectories of 1000 randomly chosen ice particles are shown for a period of 24 hours. In agreement with observations the model ice cloud appears at altitudes between 82 and 92 km and at latitudes north of ~60°N. The 'large' particles form near the lower edge of the layer (82-85 km altitude) where they are observed as NLC. Smaller ice particles extend to higher altitudes still be able to create PMSE.

The results from OPOSSUM allow for a deep insight into the physics involved in NLC and PMSE. We are now able to investigate the major physical processes influencing ice clouds in the high latitude summer mesopause region. This provides us with an assessment of the role of ice clouds for climate change detection.

Figure 1: Simultaneous observation of NLC and PMSE at Spitsbergen (78°N) on August 5/6, 2001. The NLC (black contours) was observed by the Potassium lidar of the IAP while a PMSE (coloured contours) was detected by the VHF-radar of the Max-Planck Institute in Katlenburg-Lindau. The lower edges of the two layers coincide nicely whereas the upper edge of PMSE extends to larger heights compared to NLC.

Figure 2: Trajectories of 1000 randomly chosen particles at 5-6 days after initialisation. Ice particles are shown in colour and smoke particles remaining after ice particles have evaporated are shown in gray.
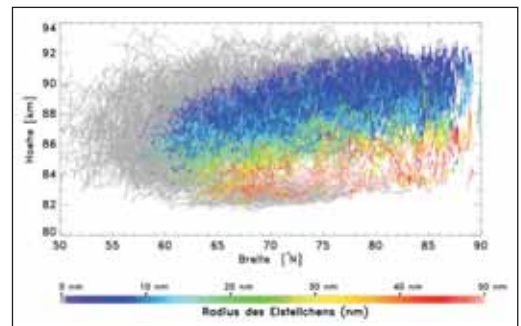
Results of Theme Group 2.2

Chemistry, Dynamics, Radiation and their Interaction: Troposphere

Photochemistry of the tropical troposphere: effects of emissions from developing Asian nations

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The tropics play a crucial role in the chemistry and climate of the global atmosphere, due to high levels of solar insolation driving rapid photochemical reactions and deep cumulus convection, and due to vast emissions, for instance from the rapidly growing populations of Asia. The AFO2000 junior research group (*Nachwuchsgruppe*) project "SAPHIRE" (Southern Asian Photochemisty and Impacts of the Redistribution of Emissions) has been examining several aspects of tropical tropospheric chemistry using two 3D models: a global model, MATCH-MPIC (Model of Atmospheric Transport and Chemistry - Max Planck Institute for Chemistry version), and a cloud-resolving model, WRF (Weather Research and Forecast Model). Here an overview of the results from SAPHIRE is given, focusing particularly on two central activities of the group: 1) the outflow of pollutants from southern Asia and the factors controlling pollution over the Indian Ocean; and 2) the role of deep convection in tropospheric chemistry, especially in the tropics, and its treatment in cloud-resolving and global models.

Southern Asian meteorology is characterized by a monsoon circulation. During the winter (or northeast) monsoon, strong, steady winds transport pollutants from the continent out over the ocean to the intertropical convergence zone (ITCZ), which is at about 5-10°S during this season. During the summer (or southwest) monsoon, the ITCZ moves north over the Indian subcontinent, resulting in a strong

onshore flow and the well-known monsoon rains. Much of our work on the pollution outflow characteristics has been highlighted in a recent review (LAWRENCE, 2004). This work began with our analyses of the observations made during the Indian Ocean Experiment (INDOEX, January-April, 1999; LELIEVELD et al., 2001). High levels of pollutants like CO at the surface in the northern hemisphere (NH) can be contrasted with the relatively pristine southern hemisphere (SH) (Fig. 1). MATCH-MPIC was able to compute and even forecast CO and O_3 mixing ratios in good agreement with observations made on research ships during INDOEX, particularly in terms

Figure 1: *The surface CO distribution predicted by MATCH-MPIC for March 4th, 1999, used to help plan a flight during INDOEX to the southern hemisphere starting from the Maldives (marked with "X").*
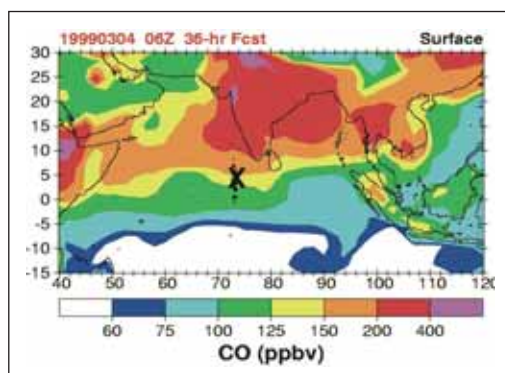
of the sharp contrast between clean SH and polluted NH air near the ITCZ (LAWRENCE et al., 2003a). The model could also reproduce very high levels of marine boundary layer (MBL) O3 observed near the Indian west coast, which exceed those observed near major coastal cities, and appear to indicate substantial photochemical O_3 production in the plumes as they are transported away from the O_3-suppressing higher-NO_x urban environments (LAL AND LAWRENCE, 2001). We hypothesized that such O_3 production would also occur over rural regions in India, which has since been supported by observations on the west coast of India (CHAND et al., 2004).

During the summer monsoon, deep convection lofts insoluble surface pollutants to the upper tropo-sphere (UT), where they are transported westward in the tropical easterly jet (Fig. 2). This plume is highly variable but occurs in every year we have simulated (1993-2002). Observing this outflow was one of the major objectives of MINOS (the Mediterranean Intensive Oxidants Study, LELIEVELD et al., 2002), based out of Crete, and was accomplished on three successful flights which observed substantial enhancements in CO in the UT associated with the Asian emissions. These flights were guided by global "chemical weather forecasts" computed with MATCH-MPIC (LAWRENCE et al., 2003a). A prototype of this forecast system was developed during INDOEX, the first time that global chemical weather forecasts were employed in field campaign planning. Routine fore-

Figure 2: Southern Asian CO (25 nmol/mol isosurface) computed by MATCH-MPIC for August 1st, 2001, showing upward transport by monsoon convection and westward transport to the Mediterranean.
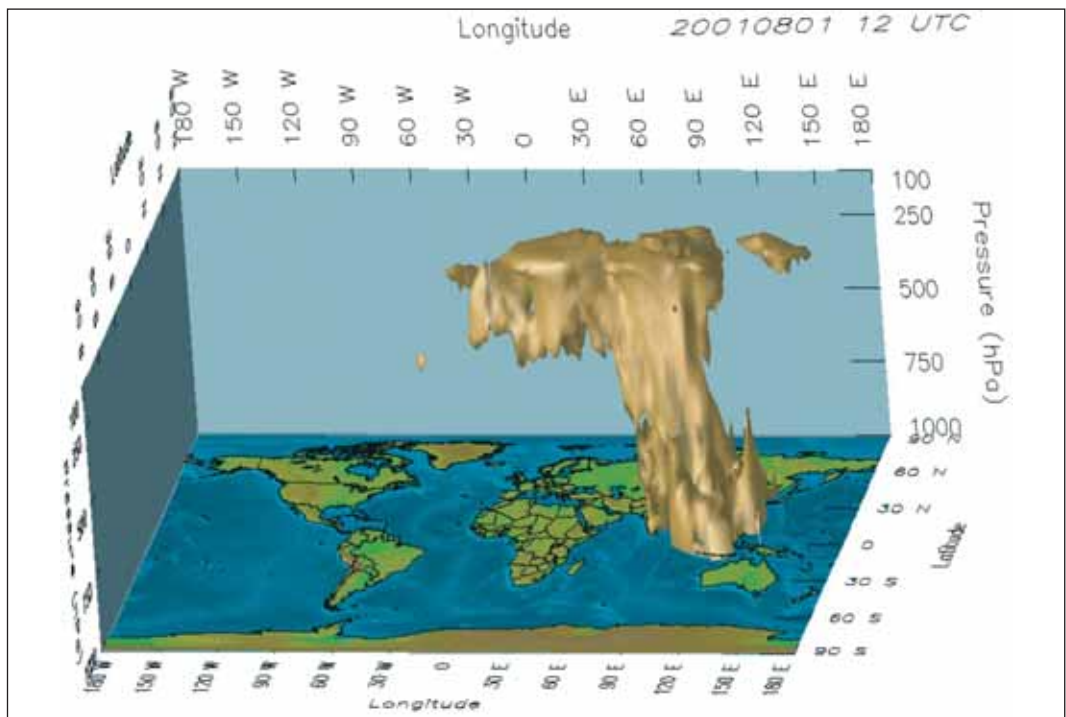
casts have been made with MATCH-MPIC since June, 2001, in support of numerous field campaigns, most notably the two field phases of the AFO2000 program CONTRACE (November, 2001, and July, 2003; PI: H. Huntrieser, DLR, Oberpfaffenhofen).

Between the summer and winter monsoons are the "monsoon transition periods", during which the ITCZ migrates north or south, the meridional winds weaken, and zonal trade winds become strong across the Indian Ocean. We have found (KUNHIKRISHNAN et al., 2004) that during these periods (around May and September), strong plumes develop across the central Indian Ocean, originating from Africa and Indonesia, primarily from biomass burning. These plumes are visible in satellite retrievals of NO_2 (Fig. 3), and are simulated by MATCH-MPIC, which shows that the transport mostly occurs in the mid- to UT (above 700 hPa). Thus, the central Indian Ocean is not always as pristine as found during the winter monsoon; in particular, during the monsoon transition periods, pollution levels are even greater south of 10°S than in the northern Indian Ocean, the opposite of the situation encountered during INDOEX.

An overall picture of the emissions which influence nitrogen oxides (NO_x) over the Indian Ocean has been developed in KUNHIKRISHNAN AND LAWRENCE (2004). Indian emissions influence the central Indian Ocean only during the winter monsoon period, and the influence is nevertheless quite weak, in contrast to the strong effect of the Indian outflow on aerosols and long-lived gases like CO. This is due to the short lifetime of NO_x and to feedbacks in the atmospheric chemical system, which we have shown can result in downwind regions often being highly insensitive to upwind emissions. Generally, the strongest direct influence on central Indian Ocean NO_x is from southeast Asia. African and Chinese emissions are important especially in the UT. The most polluted region is the Bay of Bengal, with the pollution originating mainly from India and southeast Asia during most of the year and China during part of the year.

One of the most complex and important processes influencing pollutant outflow from southern Asia and other regions is vertical transport in deep convective clouds. The net effect of deep convection on tropospheric O_3 has been debated since over a decade. We used MATCH-MPIC to carefully examine the role of convective transport in the O_3 budget (LAWRENCE et al., 2003b), and showed that convective transport of O_3 itself results in a substan-

Figure 3: Observed tropospheric NO_2 column (in 10^{14} molec/cm^2) from the GOME satellite instrument, averaged over September, 1997.
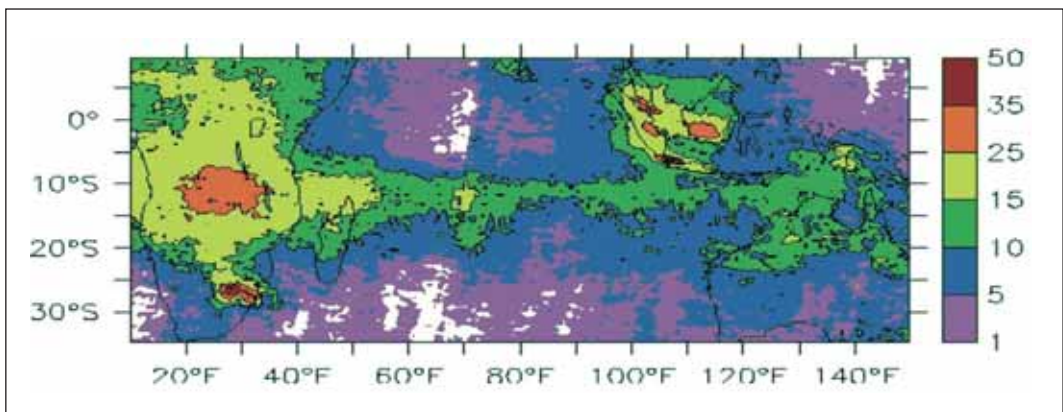
tial reduction in the tropospheric O_3 lifetime and column (Fig. 4a). However, the increase in O_3 production which results from convective mixing of O_3 precursors such as NO_x far outweighs the reduction due to the transport of O_3 itself, so that the net effect of convection on global O_3 is a substantial increase in the tropospheric O_3 column nearly everywhere globally (Fig. 4b).

Deep convection is, however, extremely difficult to simulate accurately in global chemistry-transport models (CTMs), since most convection occurs on horizontal scales of the order of a km, whereas the horizontal resolution of contemporary global CTMs is at best 100 km x 100 km. Thus, parameterizations are used to represent the interactions between sub-grid-scale processes and resolved scales, which introduces an uncertainty into our results with MATCH-MPIC and those of all other global CTMs. In order to better understand the details of convective transport of

Figure 4: Changes in the annual mean tropospheric O_3 column computed by MATCH-MPIC due to a) convective transport of only O_3, and b) convective transport of all gases.

trace gases and to evaluate and improve our parameterizations in MATCH-MPIC, our group is working with the cloud-resolving model WRF. The gas phase chemistry from MATCH-MPIC has been implemented in WRF and is being used to study the role of deep convection in O_3-related chemistry, focusing first on the Pacific warm pool region (TOGA-COARE, 1992-1993). Multi-day atmospheric chemistry simulations in a limited-area model like WRF requires careful treatment of horizontal boundary conditions and large scale vertical advection tendencies; a first major accomplishment with WRF has been developing an appropriate framework for such simulations (SALZMANN et al., 2004), which has not been available in the past. In parallel to this work, we have been examining the details of the representation of convective transport of tracers in plume ensemble parameterizations (Lawrence and Rasch, 2004), and have developed a new algorithm for computing vertical transport of tracers by deep convection, which will be a foundation for future improvements in MATCH-MPIC based on what we are learning from WRF.

The group has also been active on many other projects related to tropical tropospheric chemistry. A particularly important effort has been model evaluation. The overall global performance of the model has been extensively evaluated in comparison with O_3 sondes and composites of field campaign data (von KUHLMANN, 2001; von KUHLMANN et al., 2003a,b). In some regions, however, such as southern Asia, such observations are sparse, and we have been making use of satellite data, in particular NO_2 observations from the GOME (Global Ozone Monitoring Experiment) satellite instrument (KUNHIKRISHNAN et al., 2004a). We have also begun a detailed evaluation of the hydrological cycle in MATCH, and have developed a stronger framework for comparing modeled global mean OH and tropospheric oxidizing efficiencies (LAWRENCE et al., 2001).

Non-methane hydrocarbons (NMHCs) play a major role in tropical tropospheric chemistry. We have implemented a simplified mechanism for the
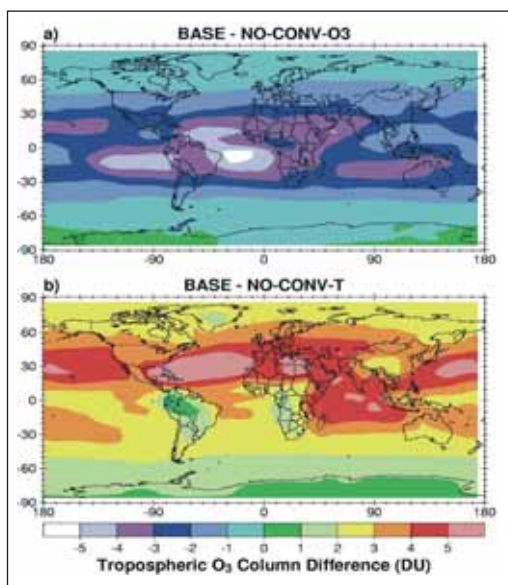
oxidation of isoprene (C_5H_8) and other NMHCs in MATCH-MPIC, and examined their effect on tropospheric O_3 (von Kuhlmann et al., 2003a,b, 2004). A scheme for the oxidation of monoterpenes ($C_{10}H_{16}$) has been implemented in collaboration with the AFO2000 project VALCHEM (P.I.: R. Forkel, IMK-IFU, Karlsruhe). The scheme is described and evaluated in Bonn et al. (2004b), and has been used to show (Bonn et al., 2004a) that hydroperoxides formed during monoterpene oxidation are likely to play a dominant role in the formation of secondary organic aerosols, although their contribution has been generally neglected in the past.

Finally, we have been delving into two further aspects of NOx and the tropospheric nitrogen cycle.

One of these is an overall analysis of the nitrogen cycle and nitrogen deposition, along with the predicted changes in the next century, as part of the international SANTAFE project. The other aspect is the production of NO_x by lightning, which is the most uncertain of all major NO_x sources. We have been particularly examining the sensitivity to the vertical distribution of lightning NO_x production (Labrador et al., 2004b), and have also found (Labrador et al., 2004a) that lightning NO_x has a very important impact on the tropospheric oxidizing efficiency, comparable to the relative effect on tropospheric O_3, which provides an additional incentive for ongoing studies of lightning and its production of NO_x, particularly in the tropics.

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Climatology of ascending airstreams and their relation to long-range transport of trace substances in the atmosphere (CARLOTTA)

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Overview

In the last decade studies have lead to the recognition that air pollution is not only of regional concern, but in fact is a truly global problem. Due to the faster wind speeds, long-range transport of air pollution occurs most effectively in the upper troposphere, which first requires ascending airstreams to lift up air pollution from the boundary layer. Within the CARLOTTA project we studied the global transport pathways of air pollutants. An important part in this project was to develop climatologies of both ascending airstreams and pollutant transport in order to uncover relationships between the two. In the following we present the main results obtained from those climatologies.

Variability of Transport to the Arctic

In the Northern Hemisphere, especially during the winter months, the most prominent and recurrent pattern of atmospheric variability is the North Atlantic Oscillation (NAO), a redistribution of atmospheric mass between the Arctic and the subtropical Atlantic. We found a significant correlation between the NAO and air pollution transport towards the Arctic (ECKHARDT et al., 2003). Two dispersion movies for a European pollution tracer were created for the winters with the highest and lowest phases of the NAO, respectively, during the 15-year period (http://www.forst.tu-muenchen.de/EXT/LST/METEO/arcticpollution/).

During high phases of the NAO (NAO[+]), European emissions take a north easterly path, crossing Scandinavia and travelling to the Arctic region. During low phases (NAO[-]), the transport is slower, and as the westerlies and the Icelandic low are weaker, less north-eastward transport occurs on the east side of the low.

To compare our results with NO_2 observations obtained from the GOME satellite instrument, we derived monthly averages of concentration fields for a very short-lived tracer from the age classes from the FLEXPART model run and derived residuals for NAO[+]-minus-NAO[-] conditions (Figure 1). The positive anomaly over Scandinavia indicates higher concentrations during NAO[+] phases and the negative one over the British Isles indicates lower concentrations during NAO[+] phases. The same method was applied to monthly concentration fields of NO_2 from GOME and, again, the same dipole structure can be seen as in the FLEXPART results. Further confirmation of our results was found in CO measurement data from Arctic stations.

A 15-Year Warm Conveyor Belt Climatology (WCB)

In contrast to the previously described low level transport, transport over the oceans mostly occurs in the middle and upper troposphere, for which a lifting mechanism is required. Numerous case studies describe lifting and the subsequent transport
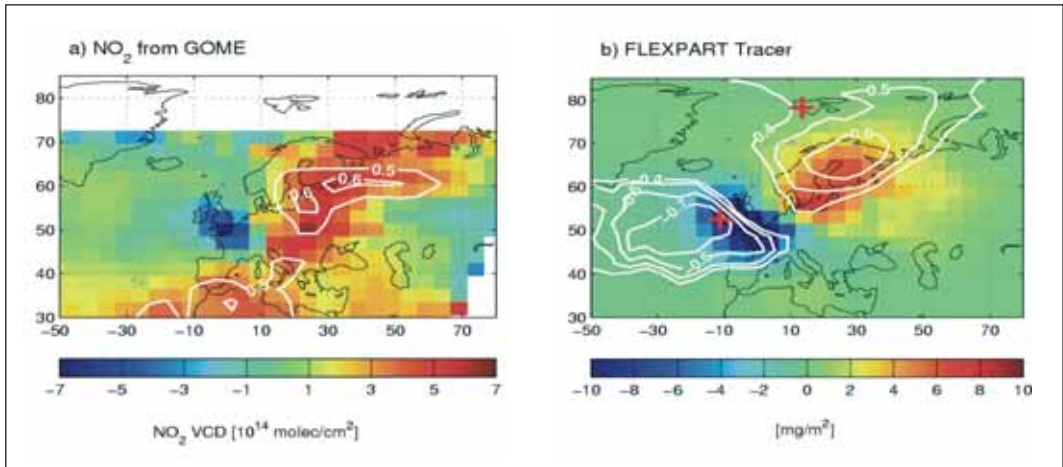
Figure 1: Comparison of observed and simulated NAO signal in pollution transport from Europe. Map of the residual NO_2 vertical columns [10^{14} molecules cm^{-2}] retrieved from GOME satellite observations (Beirle, Wagner and Platt, Inst. Env. Physics, Heidelberg Univ.) for NAO^+ minus NAO^- composites during seven (1996–2002) winters (a). Residuals of vertical columns of the European 1-day-lifetime tracer during 15 winters (1979–1993) (b). In both (a) and (b), the colours show the column residuals [mgm^{-2}]. Superimposed as white lines are the correlation coefficients between the NAO indices and the NO_2 columns (in a) or the tracer columns (in b) for the full periods with available data.

of pollutants in WCBs. We established a global climatology of these airstreams in order to find the regions with their highest frequency, get information about their meteorological properties and variability. The climatology is based on a 3-dimensional forward trajectory data set, with a resolution of 1° x 1°. Trajectories were started daily over the 15 years. The trajectory of a WCB is very characteristic, experiencing a fast ascent south of a cyclone's centre and moving east- and polewards over a typical timescale of 48 hours. The data set was checked for such characteristic trajectories and their frequency was mapped. Figure 2. shows the percentage of trajectories which were identified as WCB trajectories (15-year annual average). It turned out that WCBs are most frequent at the eastern seaboard of Asia and North America, which explains their importance for lifting emissions from these regions to the upper troposphere, and subjecting them to fast intercontinental transport (ECKHARDT et al., 2004).

Acknowledgements

Figure 2: Mean spatial distribution of WCB starting points, averaged over 15-years. Depicted is the fraction (in percent) of all trajectories that fulfil the WCB criterion.
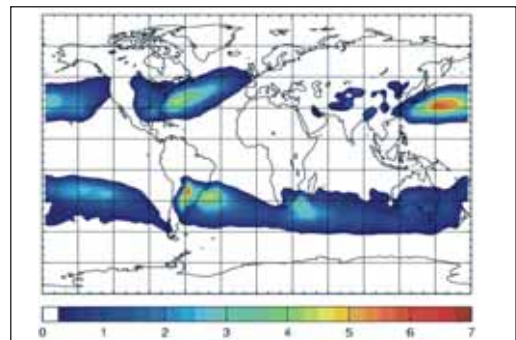
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Determination of NO$_x$ sources by combining GOME image sequences with atmospheric transport model (NOXTRAM)

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Introduction

Tropospheric nitrogen oxides (NO$_x$) are key substances in tropospheric photo-chemistry. We combine satellite data with the particle dispersion model FLEXPART to determine sources and emission strenghts of tropospheric NO$_x$, its lifetime and its transport. NOXTRAM is still in progress, therefore the finished work is presented first and a short outlook to future activities is given at the end.

Transport of NO$_x$ emissions

NO$_x$ emissions are not only a local disturbance factor. Under specific metereological conditions, if lifted to the upper troposphere, long range and even intercontinental transport can be observed. This is demonstrated by various studies carried out in the framework of NOXTRAM: Canadian forest fire emissions are transported towards Europe in August 1998 (Fig. 1, SPICHTINGER et al., 2001). In May 2003 Siberian fire plumes could be followed around the whole northern hemisphere (DAMOAH et al., 2004) In this period GOME detected enhanced NO$_2$ columns near the source regions as well. A climatological investigation of the boreal burning seasons of 1998 and 1997 points out the strong interannual variability of fire emissions (SPICHTINGER AND FORSTER, 2004, SPICHTINGER et al., 2004). Indeed, the source strength of strong fire events is comparable to anthropogenic emissions but underlies strong spatial and temporal variations. These results are important with respect to more detailed consideration of boreal biomass burning in climate models in the future.

    Anthropogenic emissions are also involved in intercontinental transport processes if they are affected by special meteorological conditions. An industrial NO$_x$ plume could be observed while travel-
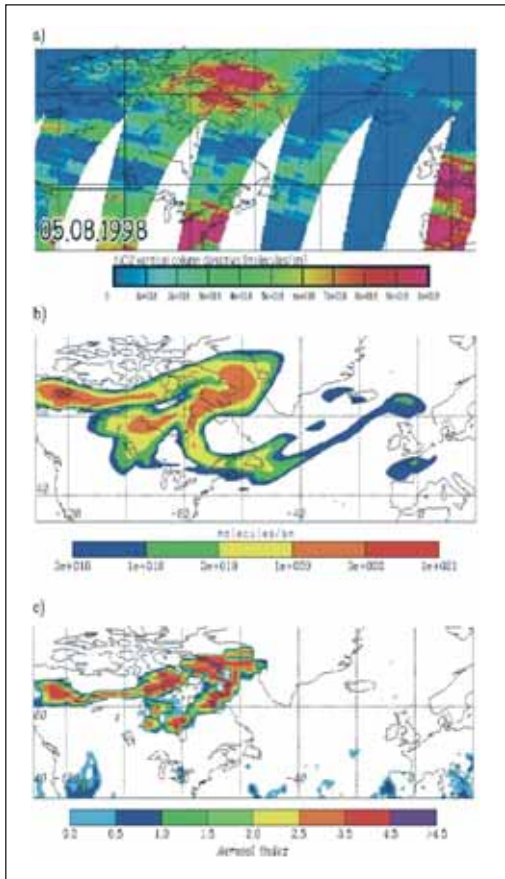
Fig. 1: a) GOME NO$_2$, b) FLEXPART NO$_x$ tracer,
c) TOMS aerosol index for August 5, 1998.

ling from the Southafrican Highveld towards Australia (Fig. 2, WENIG et al., 2003) and anthropogenic emissions transported rapidly from North America to Europe within a meteorological "bomb" (STOHL et al, 2003). Besides, transport of emissions is influenced by the North Atlantic Oscillation (ECKHARDT et al., 2003). All the studies show good agreement between GOME images and FLEXPART simulations, e.g Figs. 1, 2, and thus confirm both.

Lightning NO$_x$

Lightning activity is, besides biomass burning, a main natural source of NO$_x$, however it is the most uncertain source in magnitude. In BEIRLE et al. (2004a), we analyzed the correlation of lightning activity and NO$_2$ VCDs from GOME (Fig. 3) for Central Australia (where other sources of NO$_x$ can be neglected) and estimate a worldwide NO$_x$ production of approx. 2.8 (0.8-14) Tg N/yr, in good agreement with literature values.

SCIAMACHY

The **SC**anning **I**maging **A**bsorption SpectroMeter for **A**tmospheric **CH**artograph**Y SCIAMACHY** was launched in March 2002 onboard the ESA satellite ENVISAT. It allows the retrieval of tropospheric vertical column densities of NO$_2$ in unprecedented spatial resolution. Fig. 4 shows a mean composite of tropospheric NO$_2$ VCD. The better spatial resolution allows to identify the location of several sources. Please note that the small extent of quasi all the hot spots reflects the quite short lifetime of NO$_x$ in the boundary layer.

Lifetime

One major goal of the ongoing project NOXTRAM is the estimation of the lifetime of NO$_x$ in the troposphere. GOME data have been used in different ways to derive mean lifetimes for different regions (LEUE et al., 2001; BEIRLE et al., 2004a; BEIRLE et al., 2004b; BEIRLE et al., 2004c). We extent our lifetime studies by comparing the spatial distribution of NO$_2$, as derived from GOME and SCIAMACHY, with model results from FLEXPART for different lifetimes, to find a best match.
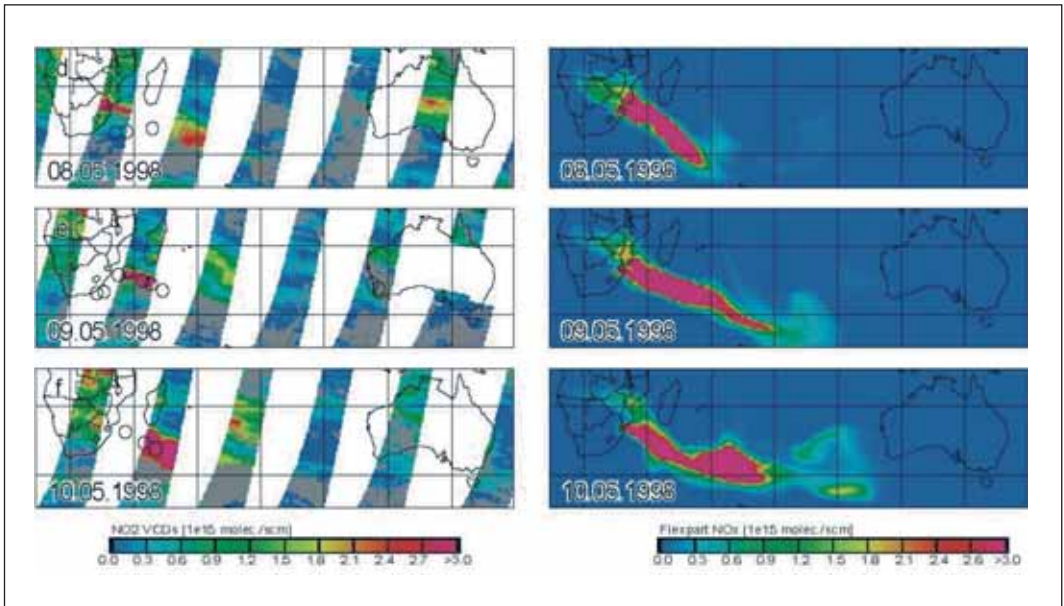
Fig. 2: Sequence of distributions of tropospheric NO$_2$ columns from the GOME instrument east of SA in May 1998 (left column). Areas with high cloud cover (>50%) are masked in grey and areas with high lightning activity (> 50 flashes per grid cell) are marked by circles. Corresponding NOx tracer column densities including emissions from lightning (right column) obtained with FLEXPART.

Fig. 3: a) Lightning activity (flashes/day/km^2, LIS data) and b) GOME NO$_2$ VCDs over Australia for different months in 1999 (taken from BEIRLE *et al., 2004a).*
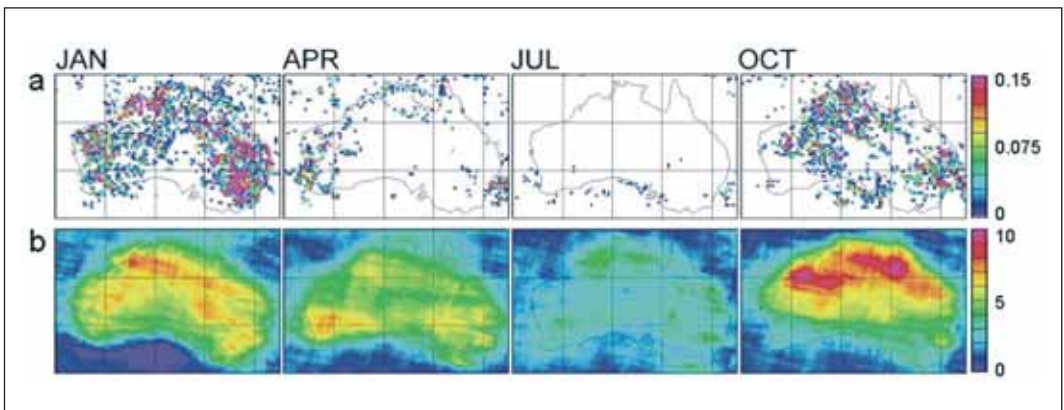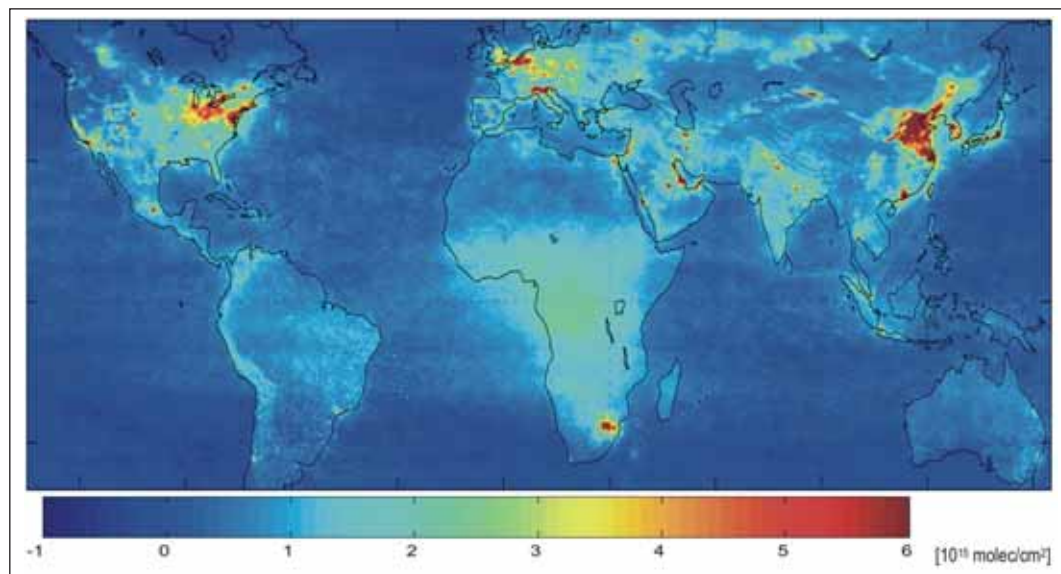
Fig. 4: Global mean of tropospheric NO$_2$ VCD from SCIAMACHY (August-December 2003)..

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Atmospheric long-range transport and its impact on the trace-gas concentrations in the free troposphere over Central Europe (ATMOFAST)

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ATMOFAST has emerged from the EU projects VOTALP (Vertical Ozone Transport in the Alps) and STACCATO (Influence of Stratosphere-Troposphere Exchange in a Changing Climate on Atmospheric Transport and Oxidation Capacity). Lidar sounding of O_3, aerosol and, starting in summer 2004, water vapour, as well as measurements of O_3, [7]Be, humidity and CO at the Zugspitze summit (2962 m a.s.l.) are combined with numerical simulations with the FLEXTRA, FLEXPART and EURAD models to evaluate the major transport pathways, the principal source regions and the chemical transformation of the air masses imported from outside Europe. In the focus of the activities are stratospheric air intrusions, intercontinental transport from North America and East Asia, Saharan dust, fire plumes and advection of low-O_3 air from the remote Atlantic. One year before the end of the project its results have already entered a total of nine reviewed publications, with several more to follow. ATMOFAST contributed to the EURO-TRAC-2 subprojects TOR 2 and GLOREAM.

Atmospheric Sounding

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Stratospheric air intrusions have been characterized in some detail with the lidar measurements at Garmisch-Partenkirchen and model calculation by the partner institutes. Since 1996, vertical sounding has been performed during more than 60 intrusion periods. Within ATMOFAST, uniquely dense measurements were achieved between February and August 2001: Each of the 17 forecasted intrusions were captured with the lidar, in sounding series extended over one to four days. In co-operation with STACCATO partners a model validation was carried out for a particularly complex case (June 20-21, 2001; Zanis et al., 2003; Roelofs et al., 2003; see Fig. 1). It was shown by FLEXPART modelling (lower panel in Fig. 1) that the ECMWF data allow the observed structures to be reproduced in great detail. The discrepancies seen for quite a few other model simulations must, therefore, be ascribed to the limited spacial resolution chosen and specific problems of the individual models. Fluxes obtained from studies with these models may, still, be rather questionable.

Filtering the Zugspitze data back to 1990 has yielded the strongest positive ozone trend at 3000 m for air masses descending from the upper troposphere and lower stratosphere (Scheel, 2002). This positive
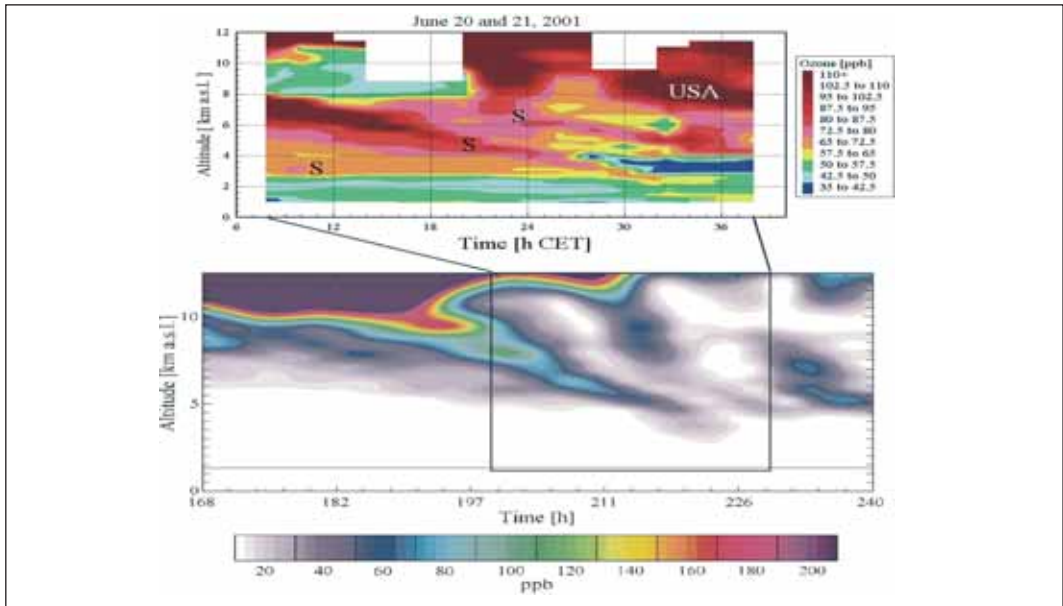
Figure 1: Complex intrusion episode on June 20th and 21th, 2001: lidar measurements (top) and FLEXPART results; S...stratospheric air, USA...air from the United States; the strange downward step at about 18 and 30 CET are both nicely reproduced by the model.

trend, which goes in line with the increase of the [7]Be values since the mid-seventies, seems to mask a downward trend in lower-tropospheric ozone expected due to the reduction in European precursor emissions since 1990, perhaps together with a growing background of East Asian origin. In fact, the overall ozone trend at the Zugspitze summit between 1990 and 2002 is even slightly positive, whereas that for the nearby Wank station (1780 m a.s.l.) is approximately constant. This is in some agreement with the observation that more than twice as many stratospheric intrusions reach 3000 m than 1800 m. The only sign of the shrinking European emissions is a decreasing amplitude of the seasonal cycle in the Wank ozone data.

Anthropogenic ozone from North America influences the Central European boundary layer only in rare cases. These contributions (80-110 ppb) pass over our area typically between 4 and 11 km a.s.l. (e.g.,

TRICKL et al., 2003). This is ascribed to upward transport of the polluted air masses in the warm conveyor belt of frontal systems both over the United States and/or the Atlantic Ocean, followed by rapid transport in the jet stream.

Some puzzling very dry layers with up to 150 ppb of ozone, in part as thick as 6 km, were recently attributed to mixed input from East Asia, the U.S. and most likely the stratosphere over the Pacific Ocean in a rapid air stream (JAMES et al., 2004; see Fig. 2). The details of the mixing of the different contributions are not yet fully analysed.

Plumes of North-American wild fires were observed with the aerosol lidar systems at IMK-IFU during the strong 2000 and 2002 fire seasons. However, the aerosol backscatter coefficients were very low. The lower-tropospheric aerosol distribution at our rural site has been dominated by Saharan dust events in recent years. Furthermore, a nine-year free-

tropospheric aerosol climatology was derived from the routine measurement of our stratospheric aerosol lidar.

Lidar and in-situ measurements were also carried out during the CONTRACE (AFO 2000) and the 2004 ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) field campaigns.

Transport Modelling

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Within ATMOFAST, the Lagrangian models FLEXTRA and FLEXPART have been used to investigate the origin of air masses sampled by the lidars and at the Zugspitze mountain site. Forward simulations of anthropogenic pollution tracers (with emissions taken from the global EDGAR emission inventory) and a stratospheric ozone tracer were carried out for a large number of episodes. Time-height sections of these tracers were produced for the lidar locations for direct comparison with and interpretation of the measurement data (Fig. 1). The forward simulations were, furthermore, compared with satellite measurements (both conventional satellite images and NO_2 data from GOME) in order to study the transport processes (convection, warm conveyor belts, dry intrusions) that were responsible for the structures seen in the measurement data (e.g., STOHL et al., 2003; TRICKL et al., 2003). Furthermore, the climatology of the pathways of intercontinental pollution transport was investigated in a model study (STOHL et al., 2002).

Another focus was on the development of alternatives to simple backward trajectory calculations (STOHL et al., 2002). The model FLEXPART was extended for backward simulations including turbu-

Figure 2: Lidar measurements on July 21-24, 2001 (left) and results from 15-day integrated FLEXPART runs for three different height levels (right); the start window of the simulations is set around label 3 in the left panel. The result for 250 m is emission-weighted and shows the most important source regions in the boundary layer.
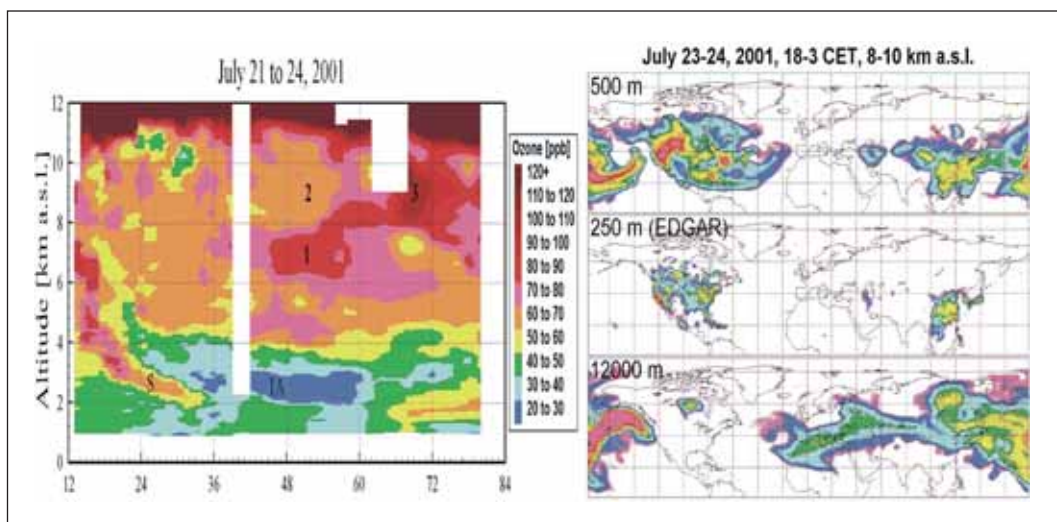
lence and convection parameterizations (Fig. 2). These runs were started in a measurement time-height bin of interest and allow a detailed account on the origin of the sampled air mass and, using an emission inventory, a quantitative estimate on how much pollution the air mass has received. Model runs were carried out backward in time from the Zugspitze position at intervals of three hours for a period of one year and, by combining the model results with carbon monoxide measurement data, allowed a detailed account on where the carbon monoxide measured at Zugspitze did originate from. The backward model options developed in the framework of AFO 2000 are now also very helpful in the analysis of the recent ICARTT field campaign.

Stohl et al. (2004) pointed out some principal difficulties in modelling the mixing between different air masses using meteorological analysis data. Deviating from the ATMOFAST proposal, James et al. (2004) used the Lagrangian model tools to investigate the origin of the water vapour that led to severe precipitation and flooding event in August 2002. These studies represent a significant spin-off from the project.

Hemispheric-scale Chemistry-transport Simulations

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The EURAD model, in contrast to the Lagrangian FLEXTRA and FLEXPART models also applied within ATMOFAST, uses an Eulerian approach to study the origin and pathways of stratospheric air intrusions and intercontinental transport events. Emphasis is on the chemical transformation of the air masses which cannot be treated by the other models.

During the project, EURAD has been extended from a regional to a hemispheric model, based on the mesoscale meteorological model MM5 (Grell et al., 1994), the EURAD-CTM (Hass et al., 1993) and the EURAD emission model (Memmesheimer et al., 1991) using the EDGAR inventory. The model has been adapted to the tropopause and free tropospheric conditions (Kowol-Santen et al., 2000). The horizontal grid distance is roughly 100 km; the vertical grid has 29 layers from the surface up to 10 hPa, with high resolution in the tropopause region. Single episodes and longer periods have been studied with EURAD.

A well-documented event with import from North America to Europe took place in May/June 1997 (Stohl and Trickl, 2001). The EURAD simulations show enhanced near-surface ozone concentrations in the south-eastern USA prior to the intercontinental transport event. These polluted air masses were lifted to the upper troposphere by a warm conveyor belt and transport towards Europe in the vicinity of the jet stream within a few days. The ozone profiles calculated for Garmisch-Partenkirchen agree well with the lidar observations there. The analysis of the model calculations indicates an efficient chemical net ozone production within the plumes after the initial lifting with decreasing rates during next days. Over Europe the North American plume is nearly neutral with regard to the ozone-chemistry. Most important for the high ozone concentrations – observed within the plume – are therefore the near-surface concentrations and emissions in the source region.
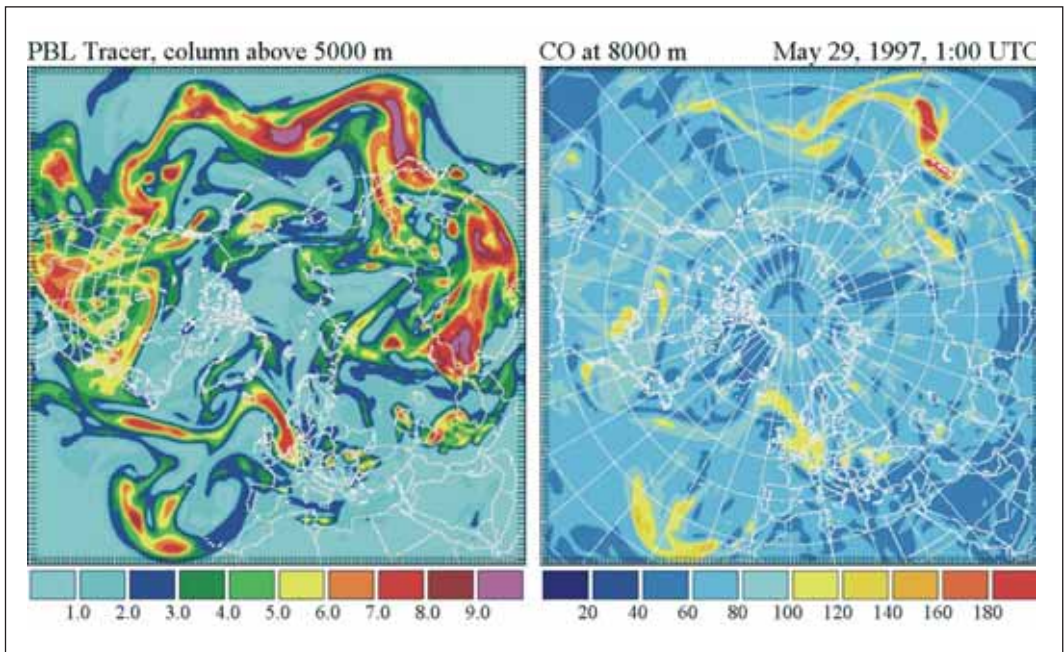
Figure 3: Calculated PBL tracer column above 5000 m and CO at 8000 m (both May 29, 1997, 1:00 UTC); the very intense trans-Pacific plume is less pronounced at 8000 m whereas the North-American export reaches Europe also in the upper troposphere, as known from the lidar measurements at Garmisch-Partenkirchen (Stohl and Trickl, 1999).

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CONTRACE -
Convective transport of trace gases into the middle and upper troposphere over Europe: Budget and impact on chemistry

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Objectives

The main objective of the CONTRACE project was to investigate the upward transport of polluted airmasses from the boundary layer to the mid (MT) and upper troposphere (UT), and to study their impact on the trace gas budget over Europe. Both the rapid convective transport in thunderstorms as well as the more slow and widespread uplift in warm conveyor belts (WCBs) ahead of cold fronts were studied. The long-range pollution transport from North America to Europe and its impact on European trace gas composition was a major aim. CONTRACE was a joint cooperation between DLR-Oberpfaffenhofen, MPI-K Heidelberg, IMK-IFU Garmisch, and TUM-Freising. The main objectives of the groups were:

+ to investigate the importance of frontal systems and long-range advection in transporting polluted airmasses to the MT and UT over Europe, and to study the impact on the ozone budget (DLR);

+ to quantify the convective transport and production by lightning of nitrogen oxides in thunderstorms and to compare it to aircraft emissions in the UT (DLR);

+ to investigate the ion-induced aerosol formation (MPI-K);

+ to investigate the impact of condensable trace gases (mainly acetone and sulphur dioxide) on the aerosol formation (MPI-K);

+ to investigate the convective transport of hydrogen peroxide and formaldehyde, and their role as radical precursors in the UT as suggested by KLEINMANN [1991] (IMK-IFU);

+ to estimate the chemical age of polluted airmasses by using VOC measurements (IMK-IFU);

+ to investigate the relative importance of frontal systems and long-range advection for the transport of polluted airmasses by using trajectories (TUM);

+ to investigate the origin of polluted airmasses observed in the MT and UT over Europe by using a CO tracer model and backward simulations (TUM).

Performance of airborne field campaigns

Two airborne field experiments were performed and coordinated by the DLR in November 2001 and July 2003. Different seasons (summer and winter) were chosen to investigate the impact of convective transport and frontal uplift. The research aircraft *Falcon* was equipped with chemical and particle instrumentation by DLR in Oberpfaffenhofen (CO, CO_2, O_3, NO, NO_y, J_{NO2}, and CN), by IMK-IFU in Garmisch (H_2O_2, CH_2O, JO^1D, VOCs), and by MPI-K in Heidelberg (atmospheric ions, $(CH_3)_2CO$, CH_3OH, and SO_2). The flight planning was supported by new developed chemical tracer forecasts provided by TUM in Freising (Lagrangian particle dispersion model FLEXPART) and MPI-C in Mainz (global chemistry transport model MATCH-MPIC). The transport of two passive tracers representing CO emissions from North America (NA) and Europe (EU) was predicted. Furthermore, forecasts of meteorological fields as well as trajectories were calculated by TUM with the FLEXTRA model.

Results from the November 2001 field campaign

The main results from the airborne field campaign carried out in November 2001 were:

+ **For the first time CO tracer forecasts were used to predict the NA pollution events and to direct a research aircraft very precisely into these polluted layers above Europe [LAWRENCE et al., 2003; STOHL et al., 2003a; FORSTER et al., 2004].**
+ **FLEXTRA trajectories and FLEXPART CO and NO_x tracers were used to analyse the airborne measurement. WCBs were identified as a key mechanism that can transport airmasses from NA to EU on a time scale of usually 4-6 days [STOHL et al., 2002b; STOHL et al., 2003a; ECKHARDT et al., 2004]. One special episode was observed when pollution was transported in a meteoro-**

logical bomb from NA to EU within only one day [STOHL et al., 2003b]. It was found that such fast transport episodes can affect the NO_x budget and thus the O_3 chemistry over remote regions.
+ **FLEXPART backward simulations were introduced as a new method to perform a detailed source analysis of the trace gases measured along the flight tracks [STOHL et al., 2002a; STOHL et al., 2003a]. In one selected case the main contribution to an observed CO maximum over Oslo (Fig. 1) came from the region in and around the city of New York (Fig. 2 right). The residence times in the lowest 300m, where anthropogenic emissions mainly are taken up, had their maximum at the East coast of North America (Fig. 2 left). Multiplying these residence times with the emission strengths given in an emission inventory (here EDGAR 1995 by OLIVIER AND BERDOWSKY, 2001) gave the detailed source contribution per grid box to the measured CO maximum.**
+ **It succeeded to measure the trace gas composition in several NA pollution plumes over Europe**

Figure 1: Measured vertical CO profile during the Falcon descent close to Oslo (Norway) on November 19th, 2001. Superimposed (upper right corner) are O_3-CO correlations in the free troposphere (FT, only regression line) and in the North American (NA) pollution plume. [Huntrieser et al., 2004]. (Copyright 2004, American Geophysical Union. Reproduced by permission of American Geophysical Union).
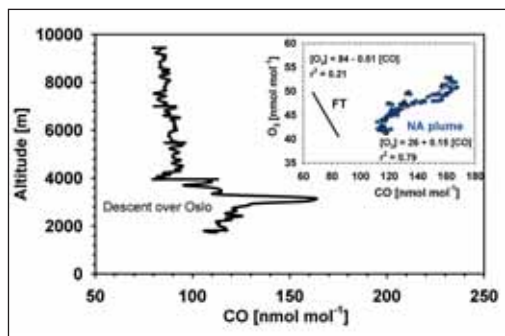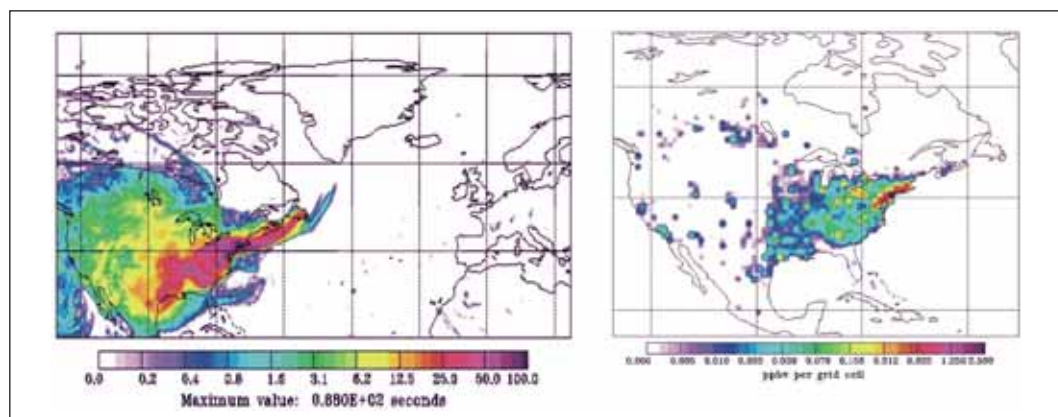
Figure 2: Left: Residence times in the lowest 300m of the particles started at 11:53 UTC on November 19th 2001, when a CO maximum was detected over Scandinavia. The times are given in percentages of the maximum residence time given below the panel. Right: Average source contribution per 0.5° grid box to the total CO mixing ratio measured along the flight track on November 19th 2001 at 11:53 UTC. [Stohl et al., 2003a]. (Copyright 2003, American Geophysical Union. Reproduced by permission of American Geophysical Union).

[HUNTRIESER AND SCHLAGER, 2004; HUNTRIESER et al., 2004]. In the most pronounced NA plume (observed in the lower-mid troposphere over Oslo) elevated CO (170), O_3 (53), NO_y (1.1), acetone (5.0), and SO_2 (2.6) mixing ratios (nmol mol^{-1}) were measured (Fig. 1).

+   In general, O_3 was elevated by ~10 nmol mol^{-1} in these NA pollution plumes in comparison to the typical background and a positive O_3-CO correlation was observed (Figs. 1, 3d). Observations indicate that the enhanced levels of O_3 were already produced near the source region over the eastern U.S. and not during the transit. Occasionally these polluted NA airmasses even descended to ground level over the Alpine region and affected the air quality considerable [HUNTRIESER AND SCHLAGER, 2004; HUNTRIESER et al., 2004].

+   In contrast, O_3 decreased by ~ 20 nmol mol^{-1} in uplifted EU pollution plumes in comparison to the typical background and a negative O_3-CO correlation was observed (Fig. 3d). Occasionally NA pollution plumes were layered above EU pollution plumes as shown in Figs. 3a-d. The

NO_y-CO correlation also showed distinct differences between NA and EU pollution plumes and was used to estimate the time since the emission of the pollutants (as suggested by STOHL et al., 2002). In the uplifted fresh EU pollution the NO_y/CO slope was much steeper than in the aged NA plume [HUNTRIESER et al., 2004].

+   For the first time large positive and negative ions with mass numbers exceeding 600 could be detected in the UT. These represent fingerprints of ion-induced aerosol formation proceeding via INU (ion-induced nucleation) [EICHKORN et al., 2002]. Furthermore, the large-ion data allow to infer the total concentration of condensable atmospheric trace gases. Laboratory data strongly suggest that the large ions were composed mostly of H_2SO_4 and that INU of H_2SO_4/H_2O took place in the upper tropospheric airmasses intercepted [WILHELM, 2003; WILHELM et al., 2004]. In relatively unpolluted airmasses INU was more efficient than HONU (homogeneous nucleation).

+   In polluted airmasses the condensable trace gases were elevated by up to 5.0 nmol mol^{-1}

acetone, 2.5 nmol mol^{-1} methanol, and 3.0 nmol mol^{-1} SO$_2$. In relatively clean airmasses in the free troposphere SO$_2$ mole fractions ranged between 0.020 and 0.050 nmol mol^{-1}. It was found that even these low values allow sufficient production of the condensable gas H$_2$SO$_4$

and thereby promote formation and growth of aerosol particles in the UT [Aufmhoff, 2004].

+ In the aged NA pollution plumes the observed formaldehyde mixing ratios were in agreement with model calculations using the RACM2 box model. Formaldehyde is rapidly decomposed by

Figure 3: FLEXPART 15-hour forecasts of (a) a North American CO tracer at 5000 m a.s.l. and (b) a European CO tracer at 3000 m a.s.l. for 22nd November 2001 at 1500 UTC. Superimposed are isolines of the geopotential height at 500 and 700 hPa, respectively. A North American pollution plume (a) reached Europe and moved rapidly from the west (British Isles) to the east (Poland). European pollution (b) was uplifted over Germany. In (c) the vertical cross section at 10 deg longitude through the FLEXPART European (EU) and North American (NA) CO tracer fields (forecast) for 22nd November 2001, 1500 UTC along 10°E is shown. In (d) the measured vertical CO profile during the descent close to Oberpfaffenhofen (48°N, 11°E) on 22nd November 2001 is presented. Superimposed (upper right corner) are O^3-CO correlations in the European (EU) and the North American (NA) pollution plume. [Huntrieser et al., 2004]. (Copyright 2004, American Geophysical Union. Reproduced by permission of American Geophysical Union).

photolysis and never exceeded values of 0.5 ppb in the aged airmasses. Typical for the UT were mixing ratios of about 0.1-0.2 nmol mol^{-1}. H_2O_2 was also found to agree with model calculations for pure gas phase reactions during transport times of more than 5 days.
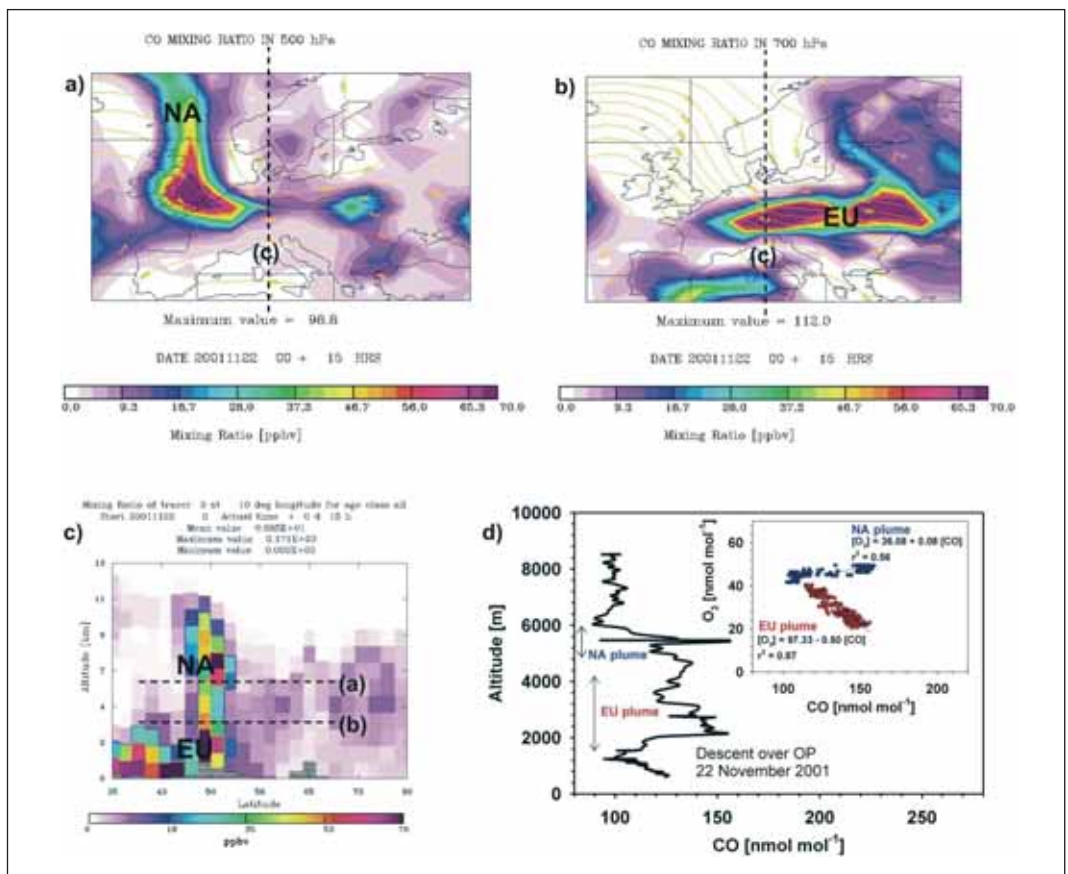
Results from the July 2003 field campaign

The main results from the airborne field campaign carried out in July 2003 were:

+ **Trace gas measurements were successfully performed in the convective outflow from European thunderstorms. An algorithm was developed to estimate the convective mass flux based on METEOSAT data [WIMMER, 2002]. The outflow from an average CONTRACE thunderstorm in the UT contained 1.2 ± 0.4 nmol mol^{-1} NO_x and up to 170 nmol mol^{-1} CO. On average NO_x produced by lightning dominated in the investigated thunderstorm anvils in comparison to the contribution from the boundary layer (60 and 40 %, respectively). The results indicate that the annual mean NO_x budget in the UT over Europe is dominated by emissions from aircraft (0.1 TgN yr^{-1}) in comparison to lightning production (0.06 TgN yr^{-1}).**

+ **One special event was observed where the outflow from a convective frontal system (active over Germany) was advected to the north out over the North Sea and then transported around a low pressure system located over the British Islands. After 5 days of circulation over the ocean the aged EU pollution plume entered Europe again at the French west coast. FLEXPART backward simulations were used to estimate the source region of this aged EU plume and it was found that the strongest industrial center in Germany (Ruhr area) mainly contributed to this pollution plume. CO and H_2O_2 mixing ratios were enhanced in the plume, however, no pronounced change in ozone was observed.**

+ **During thunderstorm flights in the UT formaldehyde mixing ratios were only occasionally elevated (>2 nmol mol^{-1}), which is close to planetary boundary layer values, while peroxide was only enhanced in clearly aged air. In most cases no significant enhancement of both trace gases was found in the UT indicating that washout processes within the convective cells significantly reduce the mixing ratios of these water soluble substances. The role of these trace gases as radical precursors in the UT seems to be minor in convection.**

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Sources, distribution and effect of reactive halogen species in the troposphere (ReHaTrop)

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Reactive halogen species have been increasingly recognised during recent years as an important factor in tropospheric chemistry. They influence the oxidation capacity of the troposphere as well as the budget of ozone and many other trace species. Here we present selected results of a joint project investigating sources, distribution and effect of Reactive Halogen Species (RHS) in the troposphere by coordinated field, laboratory (including smog chambers), and modeling studies.

Reactive Halogen Species in Mid-Latitude Coastal Regions

Within this part of the project, 3 field campaigns were performed to quantify sources and to study reactions and distribution of RHS in mid-latitudes, in particular in mid-latitudinal coastal regions. Two campaigns took place in a marine environment, while the third was performed in the lower Arctic. Measurements of halogen oxides (IO, BrO, OIO) as well as O_3, NO_2 and other trace gases were performed by an active Long-Path DOAS instrument, with lightpaths of up to 20km, which results in a good detection limit (e.g. 0.4ppt for IO). The site of the first campaign at the **German North Sea Coast** in spring 2002 is characterized by a moderately polluted atmosphere and slight biological activity. Halogen oxides could be detected close to and below the detection limit.

The second campaign took place in spring 2003 in Brittany at the **French Atlantic Coast**. Extended fields of macro algae lead to a higher bio-activity in comparison to the North Sea. Macro algae and phytoplankton species are sources of volatile halogenated organic compounds (VHOCs). The photolytic lifetime of some of these compounds is sufficiently short (e.g. CH_2I_2, CH_2IBr in the order of minutes and hours) to release halogen atoms locally. The halogen atoms react readily with O_3 to form halogen oxides. Very clear cycles of IO with mixing ratios of up to 13ppt were found (Figure 1). These elevated levels of IO require solar radiation and show a strong anti-correlation to tidal height. For detailed studies of the chemical mechanisms involved in the release, transformation and destruction of BrO in the polar troposphere a third campaign took place in the lower Arctic of Canada at the **Hudson Bay** in spring 2004. Ground based measurements and MAX-DOAS measurements of BrO and O_3 were performed.
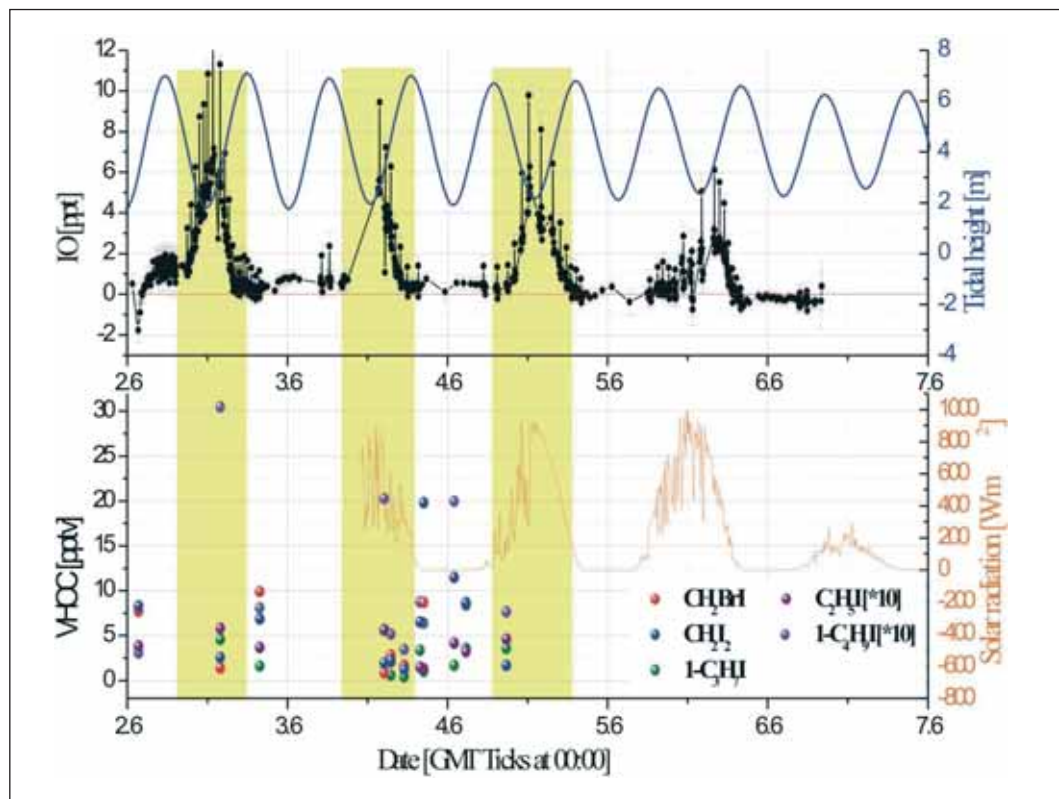
Figure 1: Four days of continuous IO measurements (June 2003) in Brittany (France). The tidal height is indicated in blue (upper panel). During low tide extended fields of macro algae are exposed to the atmosphere, leading to increased emissions of VHOCs. CH_2I_2 and CH_2IBr photolyse quickly, resulting in a clear anti-correlation to elevated IO levels at mid-day. Concentrations of 1-C_4H_9I and C_2H_5I are divided by 10 for better illustration.

Sources and Distribution of Biogenic Brominated and Iodinated Volatile Hydrocarbons in the Boundary Layer

Volatile brominated and iodinated organic compounds (VHOCs) were identified and quantified in seawater and air samples by a newly developed method coupling capillary gas chromatography with consecutive detection by an electron capture detector and an inductively coupled plasma mass spectrometer. Iodinated and brominated compounds

were measured in the coastal surface seawater and the corresponding marine atmosphere at the **German North Sea**, at the **French Atlantic Ocean**, and in the boundary layer of the **Hudson Bay**.

Average VHOC levels were always higher at the French Atlantic coast compared to the North Sea because of a higher bioactivity. At both sites CH_3I was the most abundant I-compound in seawater and the marine atmosphere but the photochemically unstable compounds CH_2BrI and CH_2I_2, were also found at significant levels (0.2-6.5/0.2-16.5 ng/L in seawater

and 0.01-9.9/0.1-13.8 pptv in the atmosphere). The highest average concentration of brominated compounds in seawater was observed for $CHBr_3$, while in the atmosphere the most abundant compound was CH_3Br. In contrast to that $CHBr_3$ and C_2H_5I showed the highest average concentrations in the air of the Hudson Bay. Good, positive correlations between the different brominated compounds were found (Figure 2) but correlation could be observed neither between the various iodinated hydrocarbons nor between the iodinated and brominated species.

Figure 2: Correlation between CH_2Br_2 and $CHBr_3$ in surface seawater samples of the North Sea at different locations with green and brown algae.
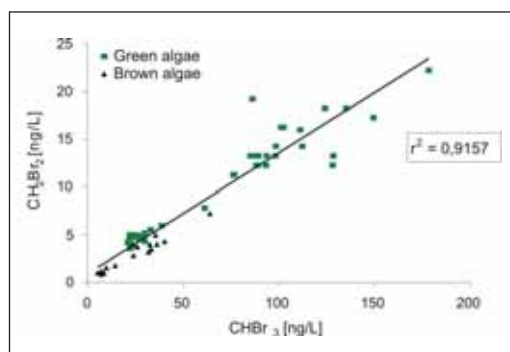
Laboratory Studies of Iodine Chemistry

IO self-reaction: The overall rate coefficient at 298 K for the IO self-reaction was determined using laser photolysis combined with transient absorption spectroscopy resulting in a pressure independent overall rate coefficient, $k(IO+IO) = 1.0 \times 10^{-10}$ cm^3 molec.$^{-1}$ s^{-1}. By adding O_3 to the reaction mixture the I atom forming channels could be suppressed, and a strong pressure dependence in $k(IO+IO)$ was observed. This allowed extraction of atmospheric pressure branching ratios of 0.6 and 0.4 to IOOI and OIO+I formation, respectively.

The IO Absorption Cross section was obtained using laser photolysis combined with transient absorption spectroscopy and three different chemical production schemes that resulted in $\sigma_{427.2nm}(IO) = (3.5\pm0.4) \times 10^{-17}$ cm^2 molecule^{-1}.

OIO Photolysis: Quantum yields of OIO photolysis were obtained using both transient absorption spectroscopy and resonance fluorescence detection methods. No evidence for loss of OIO or formation of I atoms was obtained, and upper limits to the quantum yield of I formation of 0.02 were obtained at 562.3, 567.9 and 573.8 nm. An upper limit to the overall quantum yield (all channels) of 0.15 was obtained at 532 nm. The results are summarised in Figure 3.

Reactions of $O(^3P)+RI$, $O(^3P)+I_2$, $I+NO_2$ and $I+O_3$: IO kinetic was studied by pulsed photolysis/resonance fluorescence investigation of $O(^3P)$ and I atom kinetics (see also Teruel et al. 2004) with the following results (units of $10^{-11}cm^3$ molec.$^{-1}$ s^{-1}): $k(O+CF_3I)=0.873\times exp(-216/T)$, $k(O+CH_3I)=0.988\times exp(183/T)$, $k(O+CH_2I_2)=7.36$, $k(O+C_2H_5I)=1.58\times exp(239/T)$, $k(O+1-C_3H_7I)= 1.10\times exp(367/T)$, $k(O+2-C_3H_7I)=1.84\times exp(296/T)$, $k(O+I_2)=11.8$, $k(I+O_3)=0.124$. $k(I+NO_2)$ can be parameterised (for N_2 bath gas) using $k_\infty=6.6\times10^{-11}$, $k_0=2.9\times10^{-31}$ and $F_c=0.49$.

Figure 3: Summary of wavelength dependent quantum yields (ϕ_λ) for OIO photolysis.
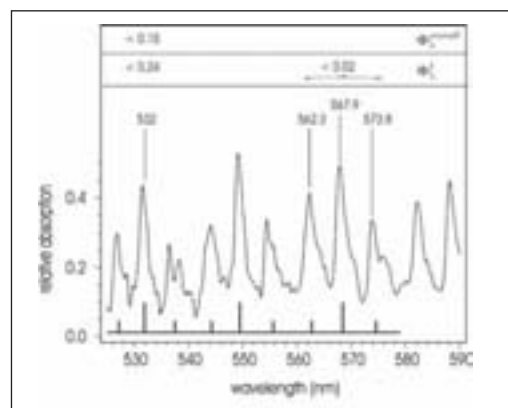
ICl uptake to sea salt: The interaction of ICl with aqueous salt solutions was investigated using the wetted wall flow tube. No release of IBr was observed due to its high solubility. An accommodation coefficient, α=0.01 was obtained for uptake of ICl to aqueous surfaces.

Effect of Halogen + VOC Reaction Products on the Tropospheric Halogen Cycle

The major aim of the project was to obtain quantitative information on the oxidation processes of organo-halogen compounds, in particular naturally occurring alkyl halides and halo-carbonyl compounds which are formed in the reactions of halogen atoms with volatile organic compounds (VOCs). Many investigations were performed in small photoreactors and the EUPHORE photoreactor facility. The following systems were investigated:

- **Kinetic and product studies of the OH and Cl radical initiated photooxidation of several chloro-** and bromoalkanes and halogenated alkenes.
- **Atmospheric photolysis rates alkyl iodides and bromides**
- **Kinetic and product studies on the reactions of O atoms with iodoalkanes**
- **Reactions of Br and BrO with CH_3OO at low temperatures.**
- **Kinetic and product studies of the $Cl+CH_3SCH_3$ and CH_3SOCH_3 including a theoretical study of the reaction between $CH_3S(OH)CH_3$ and O_2.**

It has been shown that proposed formation routes to methyl bromide (CH_3Br) such as, $CH_3OO+Br \rightarrow CH_3Br+O_2$ and $CH_3OO+BrO \rightarrow CH_3Br+O_3$, are not important at low temperatures in the atmosphere. At present a limit of $\leq 5\%$ can be placed on the molar formation yield of CH_3Br (Fig. 4).

An upper limit for the reaction BrO + HCHO of 1×10^{-14} cm^2 molec.$^{-1}$ s^{-1} was obtained rendering the reaction unimportant in the atmosphere. HOBr was the major product.

Figure 4: IR spectra showing that CH_3Br is not formed in Br/BrO + CH_3OO reactions at low temperature. Note absence of the CH_3Br band at 1300 cm^{-1} in top spectrum.
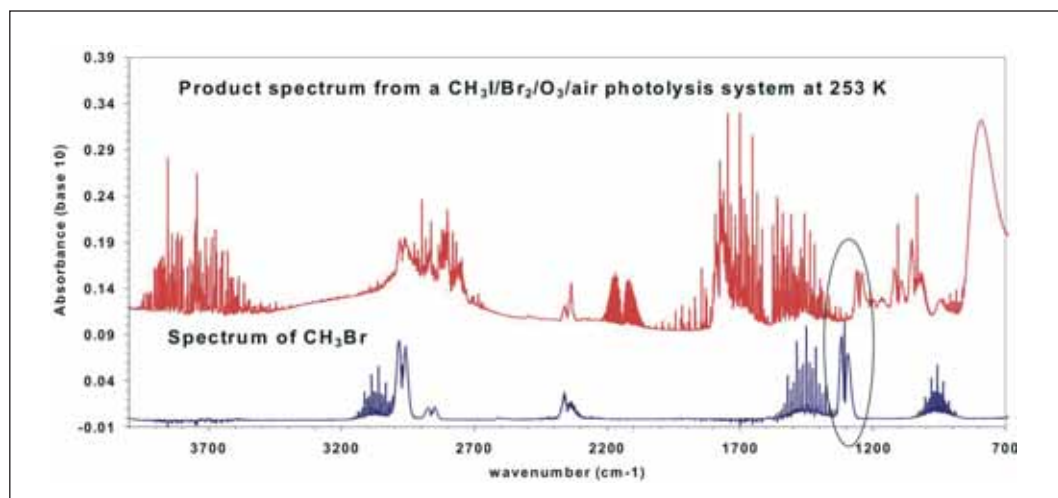
DMSO and SO_2 were identified as products of Cl+DMS. DMSO is a primary product whereas SO_2 is secondary. It was shown that the OH/Cl initiated oxidation of alkyl iodides results in large yields of reactive alkenes (≤60%). The alkene yields by alkyl bromides are much smaller (≤10%).

Project HALOCIMS

After taking over this project in Sept. 2003 our group successfully set up an apparatus (Figure 5) for detection of tropospheric ultra-trace gases via chemical ionization mass spectrometry (ULTRACIMS). Recent measurements employing the ULTRACIMS technique comprise the detection of OH, HO_2, RO_2, H_2SO_4, and MSA. Currently further development of ULTRACIMS towards HALOCIMS is in progress focusing on the detection of the atmospheric RHS BrO and IO.

The following mechanism is used to detect BrO (and analogously IO):

1. Oxidation of DMS to DMSO by bromine oxide: $BrO + DMS \rightarrow Br + DMSO$

2. Ion-molecule reaction of DMSO with ammonium ions: $DMSO + NH_4^+ \rightarrow DMSOH^+ + H_2O$

3. Detection of $DMSOH^+$ in the mass spectrometer. Tests have been made using different types of ion

Figure 5: The novel aircraft-based ULTRACIMS apparatus in the laboratory during the recent construction phase in February 2004.

sources for the production of the NH_4^+ reagent ions. In comparison with gas-discharge sources ionization by alpha particles from a radioactive [210]Po source results in a minimum production of unwanted radicals thus leading to a lower background signal. Successful long-term (6 weeks) measurements of H_2SO_4 using a 185 MBq [210]Po source were carried out in March 2004 in Heidelberg. Clear diurnal variations were observed that closely followed the solar radiation intensity with maximum mid-day concentrations approaching 1.5×10^7 cm^{-3} (0.6 ppt). The detection limit of H_2SO_4 was around 8×10^5 cm^{-3} (0.03 ppt). Very recently, first tests of an aircraft-based version of the ULTRACIMS apparatus have been successfully made in collaboration with DLR on the FALCON.

An aerosol smog chamber facility for experimental simulation of halogen activation from sea-spray and supporting experiments and modelling

Preparatory phase at Hannover: The aerosol smog chamber facilities there had to be shut down completely in October 1999 (following new directions of environmental research at Hannover) to be relocated to Bayreuth University in 2000, well before the start of the project. Unfortunately contractual details could not be solved before August 2003 resulting in a serious delay. Meanwhile, we evaluated and summarised our previous work (BEHNKE et al., 2001, 2002, ZETZSCH, 2001), did supporting experiments on concentrated and dilute salt solutions in wetted-wall flowtube and stopped-flow set-ups (BARCELLOS DA ROSA AND ZETZSCH, 2001) and performed model calculations of the gaseous uptake, solution chemistry and aerosol smog chamber chemistry. The studies include heterogeneous and homogeneous reactions of ozone with dimethyl sulphide (DMS) (BARCELLOS DA ROSA et al., 2003), chloride and bromide, the trihalide equilibria and reactions of halogens with halides, nitrite and the marine sulphur species DMS, DMSO, MSIA and MSA at various pH values, ionic strengths

and low temperatures (BARCELLOS DA ROSA AND ZETZSCH, 2002, BARCELLOS DA ROSA, 2003) and modelling of the influence of iodide on halogen activation and the uptake of HOBr on deliquescent NaCl aerosol droplets. Furthermore, a solar simulator (fluorescent lamps) and a cylindrical aerosol smog chamber (made of Teflon film) were constructed and tested.

Chamber facilities at Bayreuth: An insulated, coolable (to –28°C) laboratory room (4x5x6m) now contains two smog chambers (a glass chamber with 4 m height and 1 m diameter and the Teflon chamber). A third smog chamber (600 l) can be equipped with a White cell, foreseen for optical coupling to an FTIR instrument. Trace gases are monitored by UV-absorption and gas analysers for NO_x, O_3, SO_2, NO_2 and PAN and capillary GC. Aerosol size distributions, mass densities and chemical composition can be determined with various instruments. The reinstallation of the analytical equipment and testing and improvement of methods is ongoing, a series of aerosol smog

chamber experiments on halogen activation at low temperatures are foreseen for the next year.

Modeling reactive halogen species in the marine troposphere

We implemented our marine halogen chemistry mechanism MECCA-MBL into the global circulation model ECHAM5 (www.mpimet.mpg.de/en/ extra/models/ echam/echam5.php) using the interface MESSy (www.messy-interface.org). In contrast to prior studies (VON GLASOW et al, 2004 and YANG, 2004) we used a prognostic aerosol distribution, (the aerosol module MESSy-M7). Gas and aerosol phase chemistry were calculated using the kpp integrator (www.cs.vt.edu/~asandu/Software/KPP). We also considered several other processes (provided by MESSy submodels): dry and wet deposition (DRYDEP, SCAV), sedimentation (SEDI), emissions (EMIS, ONLEM, AEROEM), convection (CONVECT),

Figure 6: Global map of model-calculated BrO mixing ratios in the surface layer at 06:00 UTC on August 7th. Colour code gives log [BrO(mol/mol)]
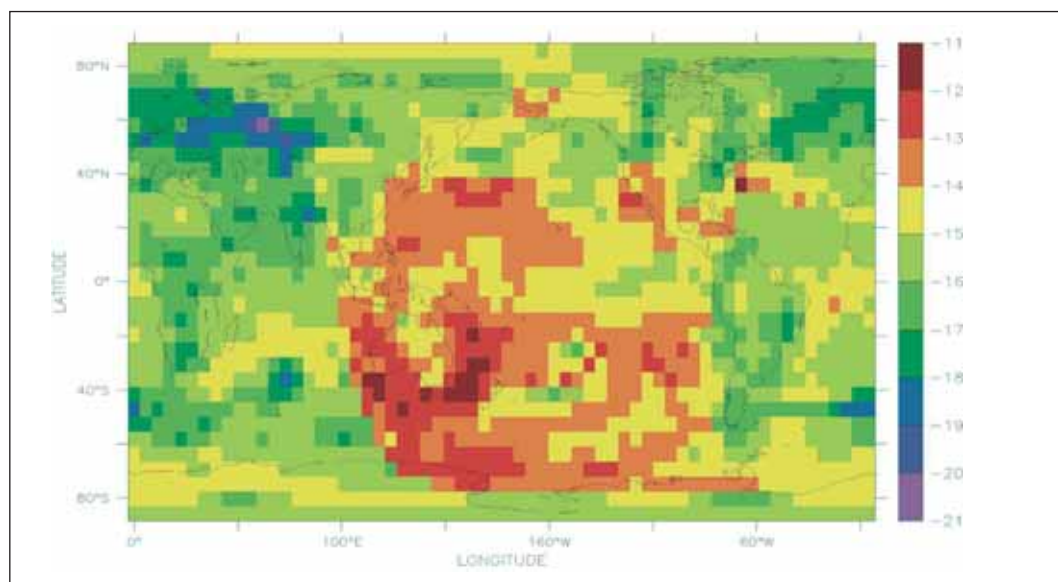
convective transport (CVTRANS), lightning NOX (LNOX), photolysis (PHOTO), and tropopause diagnostics (TROPOP). Compared to previous studies, the advanced treatment of online-calculated sea salt levels leads to higher peak values of BrO. In the northern hemispheric summer (NHS), maximum values in the MBL are a few pmol/mol, whereas average BrO levels are < 1 pmol/mol (Figure 6), in good agreement with the measurements of LESER et al. 2003. However, we model much higher peak values of BrO in the northern hemispheric winter (NHW). This results

from a simultaneously high levels of fresh sea salt (due to high wind speeds) and high acid supply. Since BrO measurements are sparse at high wind speeds, model validation is difficult. Our model results show a very pronounced annual cycle of BrO, possibly caused be the much higher acid supply in the northern hemisphere. BrO has a global effect on O_3. However, the calculated near-surface O_3 loss is smaller than predicted in previous studies. Halogen chemistry causes an O_3 reduction which is highest in the NHW, corresponding to the maximum in BrO.

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Prices:

The excellent PHD of Anastasia Schwarz was honored by the Arbeitskreises für Mikro- und Spurenanalyse der Elemente und Elementspezies (A.M.S.El.) in der Fachgruppe Analytische Chemie der GDCh in 2004.

On SOLAS open science conference, Halifax 2004 Christina Peters received the SOLAS Science 2004 prize for best poster titled: "Studies of Reactive Halogen Species in the Troposphere by Differential Optical Absorption Spectroscopy (DOAS) in the Framework of the AFO 2000 Project"

Results of Theme Group 3

Multiface Processes

Multiphase processes in the polar stratosphere: in situ measurements and simulations (POSTA)

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The POSTA cluster aimed at an improved understanding of the formation and evolution of polar stratospheric clouds (PSCs). This was achieved by a unique combination of *in situ* with remote sensing techniques, laboratory investigations, and dedicated studies with a 3D CTM including a detailed microphysical model. Salient results of the four contributing projects are briefly summarised.

Chemical Analysis of PSCs with a balloon-borne Aerosol Composition Mass Spectrometer (ACMS)

Over the last few years a balloon-borne aerosol composition mass spectrometer has been developed that provided the first chemical analysis of PSC particles [Schreiner et al., 1999].

Recent flights with additional instruments from the international aerosol community resulted in a detailed description of PSCs under leewave conditions above the Scandinavian Mountains. For the first time NAT particles were identified [Voigt et al., 2000]. Within POSTA synoptic-scale PSCs were observed with the same instrumentation on 4 Dec 2002 between 20 and 27 km again from Kiruna, Sweden. The ACMS was calibrated for H_2O, HNO_3 and HCl before and after the flight, and no change in sensitivity was observed.

New and significant results were obtained from this flight, cf. Figure 1: Within the PSC solid particles existed whenever the temperature was below the equilibrium temperature of NAT formation and liquid particles appeared when the temperature fell below an ever lower threshold about 3 K above the frost point but resulted in the co-existence of solid and liquid particles in some altitude ranges. The correlation of liquid super-cooled ternary solution aerosols with local temperature is a pronounced feature observed during this flight although the molar ratios H_2O/HNO_3 were about a factor of 2 higher than model predictions. This is an unusual result based, however, on reliable laboratory calibrations. For the first time the HCl content in liquid polar aerosols has been measured. Its abundance in particles is critical for the development of an ozone hole. The chlorine isotope signature served as a unique tool to identify unambiguously HCl. Within a narrow temperature range of about three degrees above the frost point, HCl molar ratios in liquid particles are below 1 wt-%, in fair agreement with model predictions. For a detailed discussion see Weisser et al., 2004.

Airborne lidar measurements of PSCs including mesoscale forecasts for balloon campaigning

With the airborne OLEX lidar onboard the DLR Falcon, extended PSCs were observed in the cold Arctic vortex in January 2001 and December 2002. The 6-8 km thick PSC layers were made of a

$H_2SO_4/HNO_3/H_2O$ ternary solution (STS)/nitric acid tri-hydrate (NAT) external mixture and extended along all the flight tracks inside the vortex. At the coldest regions, ice particles were embedded. In January 2001 the ice transiently formed in a gravity wave-induced temperature minimum ($T_{MM5} \approx 184$ K), in December 2002 it occurred at synoptic temperatures around the ice frost point ($T_{synop} \approx 188$ K at 30 hPa,

analysed by ECMWF) over the European Ice Sea, cf. Figure 2. As indicated by small back-scatter ratios $\gamma_{total} \approx 3$-5, particularly significant also in the perpendicular polarised channel $\gamma_{\perp}$ (not shown), and colour ratios $\beta_{532}/\beta_{1064} \approx \beta_{354}/\beta_{532} \approx 1$, NAT particles (cf. ACMS observations) covered the largest area. Liquid STS particles with depolarisation ratio $\delta=\beta_{\perp}/\beta_{\parallel} \approx 0$ and larger $\gamma_{total} \approx 10$-20 co-existed with and masked the

Figure.1: Balloon flight from Kiruna, Sweden under synoptic conditions. A particulate water, indicative of PSC layers; B H_2O/HNO_3 molar ratios, measurements and model calculations; C $H^{35}Cl$ and $H^{37}Cl$ in liquid particles, note close correlation with A; D HCl wt-%, measurements and model calculations; E, F atmospheric pressure and temperature.
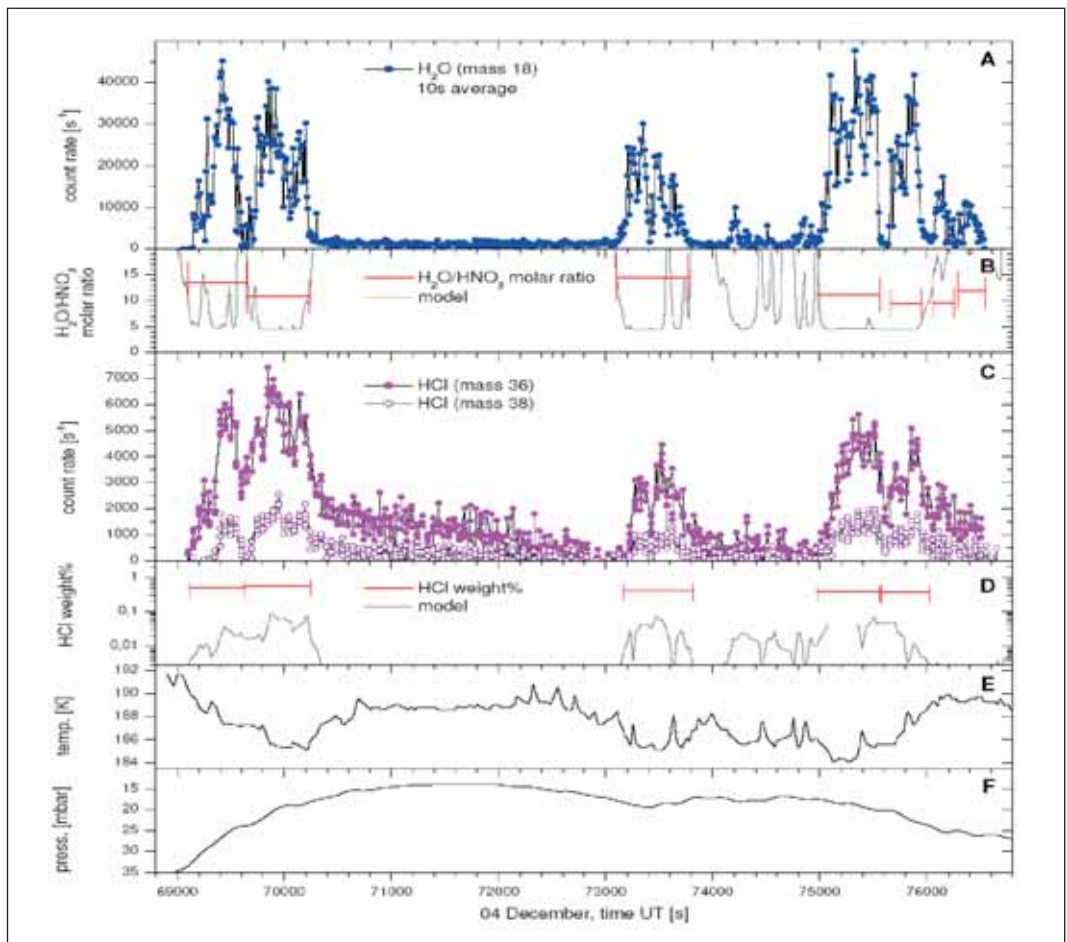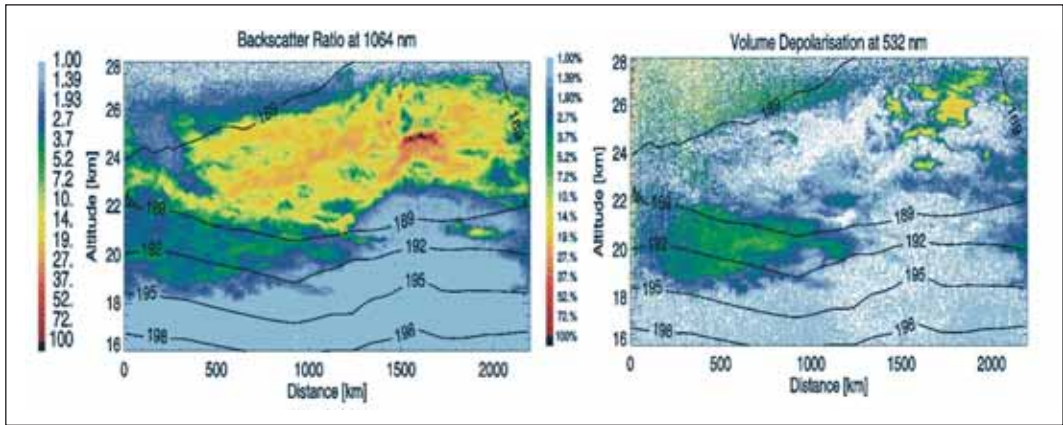
Figure 2: Backscatter and depolarisation ratio along the DLR lidar flight on 7 Dec 2002 with ECMWF temperature con-tours. Liquid STS particles with high backscatter but small depolarisation mask NAT. Patches of ice occur near a turning point at 1500 km.

NAT particles where the temperature was below T_{STS}. According to the respective colour ratios, $\beta_{532}/\beta_{1064} \approx$ 2-3 and $\beta_{354}/\beta_{532} \approx$ 2-2.3 of the liquid fraction and $\beta_{532}/\beta_{1064} \approx \beta_{354}/\beta_{532} \approx 1$ for the solid fraction indicate STS particle sizes $r_{eff} < 0.5\ \mu m$ at high concentration and NAH sizes $r_{eff} > 1\ \mu m$, occurring at low concentration. Good agreement is found with backscatter sondes launched from Sodankylä (FMI, Finland) and results from the PSC-Analysis payload launched shortly before. Larger backscatter- and depolarisation occurs due to ice particles in regions where the temperature is several K below T_{ice}.

While the distribution of STS particles is determined by the STS temperature threshold, the correlation of the solid particles' occurrence with temperature is lower, indicating that ambient temperature is a weaker indicator for NAT. In Dec 2002 the temperature history is the same in the mixed-phase and solid-phase parts of the cloud, and liquid particles are observed only where the final temperature is few K lower. As temperatures dropped below T_{NAT} only 24 h before the measurement and exceeded T_{NAT} on the previous days, the time available for formation and persistence of the solid particles was short and likely variable in space, depending on the individual trajec-

tory. This may also account for the fair correlation of the dispersion of solid particles with T_{NAT}.

MIPAS-B-profiling and 3D-modelling of trace gases in the polar vortex with a focus on denitrification

Polar Stratospheric Clouds (PSCs) trigger and enhance ozone loss by heterogeneous reactions and denitrification. Within POSTA two balloon flights with MIPAS-B and model calculations with the 3-D chemical transport model KASIMA have been performed to investigate NO_y partitioning and denitrification. In addition, detailed microphysical modelling on backward trajectories was performed in cooperation with the DMI. The balloon flights took place in January 2001 and December 2002 in close coordination with the POSTA partners. In both cases MIPAS-B detected synoptic-type PSCs as well as a large number of PSC-relevant gaseous constituents such as HNO_3 and H_2O and ozone-relevant species such as nitrogen and chlorine compounds. The wealth of data has been used to investigate PSC properties in terms of composition and size classes under various geophysical conditions as well as to determine the

uptake of HNO_3 by the PSCs and the amount of chlorine activation. Spectra measured between T_{NAT} and T_{STS} in the boundary regions of the PSC were consistent with the presence of NAT, mean particle radii 1-3 µm, in broad agreement with the in-situ und Lidar observations. Uptake of gaseous HNO_3 by the particles was found to within 1 and 7 ppbv indicating a large spatial variability. The MIPAS-B observations indicate that up to 70 % gas phase HNO_3 were taken up by particles inside the cloud, while outside the cloud large amounts of gaseous HNO_3 were found suggesting that irreversible removal of HNO_3 had not happened (yet) (Figure 3). Except REPROBUS, the 3-D models KASIMA and SLIMCAT calculate a significantly weaker uptake of HNO_3 by the cloud particles. Along with a different treatment of H_2SO_4, the positive temperature bias in these models, together with a coarser horizontal resolution are most likely responsible for this discrepancy. PSCs are modeled with a simple equilibrium treatment.

Model calculations with a dedicated microphysical model developed at the DMI Copenhagen [LARSEN et al., 2002] have been used to test scenarios for the

nucleation and growth of PSC particles (Figure 4): adopting published surface nucleation rates [TABADAZEH et al., 2002] yielded much too fast NAT formation along with excess uptake of HNO_3 (blue solid line in Figure 4). The nucleation rates had to be reduced by about two orders of magnitude in order to get the removal of gaseous HNO_3 in agreement with the MIPAS-B measurements.

Chemical analysis of artificial PSCs and studies of phase transitions in the large coolable aerosol chamber AIDA

A comprehensive FTIR study of binary and ternary solution droplets was performed under simulated PSC conditions [WAGNER et al., 2003]. It provided the basis for homogeneous nucleation experiments of PSC particles in the AIDA chamber. The focus was on NAD nucleation from supersaturated HNO_3/H_2O droplet aerosols which occurred in AIDA on time scales of several hours. NAD was unambiguously identified by ACMS, and on the basis of FTIR spectra which also yielded mass concentrations and size dis-

Figure 3: HNO_3 profiles as measured by MIPAS-B (lines with symbols) and calculated with European state-of-the-art 3-D chemistry transport models KASIMA, REPROBUS, and SLIMCAT.
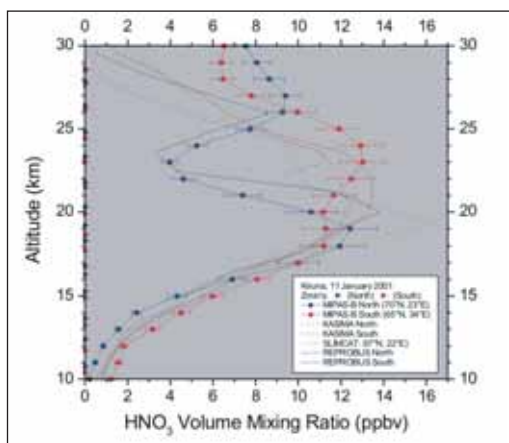
Figure 4: HNO_3 profiles as measured by MIPAS-B (lines with symbols) and calculated with the detailed micro-physical model of Larsen et al. (2002) (solid and dashed without symbols).
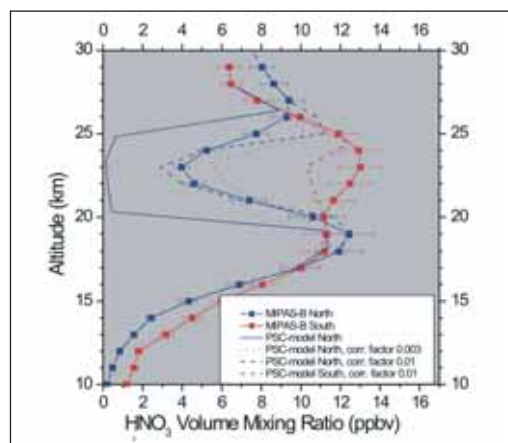
tributions and indicated that the particles were oblate with aspect ratios > 5. NAT particles were never observed, probably because the critical super-saturation threshold was not exceeded in the chamber. This contrasts with the identification of

large NAT particles in real PSCs by the balloon-borne ACMS of MPI-K and by the Falcon-borne OLEX instrument. The change in super-cooled droplet composition due to loss of water vapour to the cold chamber walls was retrieved from time series of FTIR spectra, while interstitial water vapour was measured quasi-continuously and in situ with a tuneable diode laser. This yielded NAD super-saturations as function of time. Thereby the NAD nucleation threshold could be accurately determined. The data were ideally suited to test the hypothesis of TABAZADEH et al., 2002, that homoge-neous NAD nucleation is surface-induced, in contrast to the classical assumption of volume induction [SALCEDO et al., 2001]. An example is shown in Figure 5 which compares measured size distributions with theoretical calculations assuming volume- and alternatively surface-induced nucleation. The thermodynamically more stable NAD particles grow rapidly at the expense of the evaporating droplet aerosol. Note that the size distribution measured with the optical particle counter is severely broadened due to the large aspect ratio of the NAD particles. The microphysical model calculations reproduce the onset of NAD formation and droplet evaporation only when volume-induced nucleation is assumed. In the case of surface-induced nucleation NAD formation and droplet evaporation is predicted to occur much earlier than observed.

Figure 5: Time evolution of measured (upper panel) and modelled size distributions in an experiment at ˜192 K. The NAD saturation ratio increased slowly from S = 4 until NAD nucleation started at S ˜ 10. The model calculation based on volume-induced nucleation (middle panel) captures the essential features of the experiment while NAD nucleation starts much too early when surface-induced nucleation is assumed (lower panel).

As was shown in the previous section, detailed modelling of MIPAS-B profiles inside the vortex leads to the conclusion that surface-induced nucleation of NAT is also incompatible with the retrieved HNO_3 profile.

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Carbonaceous aerosol components: Chemical composition, reactivity, and hygroscopicity (CARBAERO)

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Introduction and Motivation

The effects of aerosols on atmospheric chemistry and physics, climate, and public health are among the central topics in current environmental research. Aerosol particles can scatter or absorb radiation, influence the formation of clouds and precipitation, and affect the abundance of trace gases via heterogeneous chemical reactions and other multiphase processes. Moreover, they can cause respiratory, cardiovascular, and allergic diseases. The primary parameters which determine these effects are particle size, structure, and composition. The concentration and size distribution of aerosol particles in the troposphere is temporally and spatially highly variable, and their physicochemical properties and transformation (aging) are poorly understood.

In the lower troposphere carbonaceous aerosol components (organics and elemental carbon) account for up to ~ 50 % of fine air particulate matter. The total carbon content (TC) is usually determined by thermochemical analysis and divided into an ele-

Figure 1: Classification and molecular structure of carbonaceous aerosol components (Pöschl, 2003).
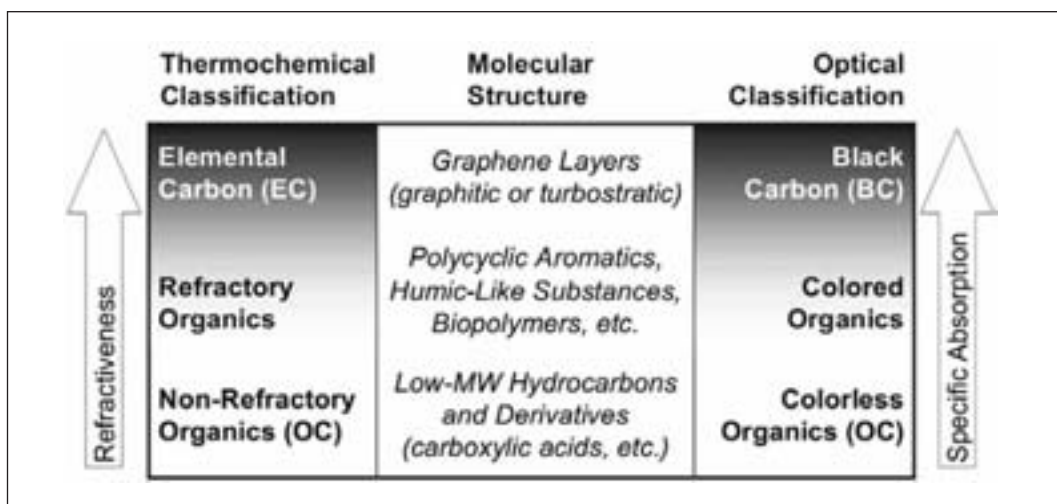
mental carbon (EC) or black carbon (BC) fraction and an organic carbon fraction (OC). As illustrated in Figure 1, however, there is a more or less gradual decrease of thermochemical refractiveness and specific optical absorption going from graphite-like structures to non-refractive and colourless organic compounds. Depending on the applied thermochemical and optical methods, EC and BC measurements include not only graphite-like material from soot and other combustion particles but also refractory or coloured organics. This can lead to substantially different results and limits the comparability and suitability of BC, EC, and OC data for the determination of mass balances and physicochemical properties of air particulate matter ("How black is black carbon?"). Besides different types of graphite-like materials there are at least two classes of organics which can contribute to the absorption of visible light by air particulate matter: polycyclic aromatics and humic-like substances. Hundreds of organic compounds have been determined but only 10-40 % of the total organic particulate matter (OPM) in air have been identified at a molecular level. Most organics can efficiently interact with atmospheric photooxidants and water, but the mechanisms and rate parameters of mass transport and chemical reactions are hardly known (Ammann et al., 2003; Pöschl, 2002; 2003; and references therein).

Therefore the project CARBAERO was aimed at the experimental investigation and mechanistic description of the chemical composition, reactive transformation, and water interaction of carbonaceous aerosol components. The investigations were focused on biopolymers, humic-like substances, polycyclic aromatic compounds, and elemental carbon.

Methods and Results

Within the project CARBAERO research activities have been pursued and scientific results have been achieved in the following areas:

1) Development and optimization of analytical methods (liquid and solid phase extraction, liquid and gas chromatography, optical spectroscopy and mass spectrometry, enzymatic and immunochemical assays, etc.) for the determination of carbonaceous aerosol components: polycyclic aromatic hydrocarbons (PAH, Bömmel et al., 2003; Schauer et al., 2003); nitrated and oxygenated PAH derivatives (Letzel et al., 2001; Schauer et al., 2004); proteins and nitrated derivatives (Franze et al., 2003a; 2004a; Walcher et al., 2003); elemental carbon (Sadezky et al., 2004); cellulose, humic-like substances, and water-soluble organic carbon (Schaller et al., 2003).

2) Aerosol field measurements at urban, rural, and high-alpine locations (Munich, Hohenpeissenberg, Schneefernerhaus/Zugspitze; measurement of particle number concentrations and size distributions; gravimetric and chemical analysis of filter and impactor samples; determination of PM2.5, TC, EC, PAH, nitro-PAH, proteins): detection of high protein concentrations (up to 7 % of PM2.5, Franze et al., 2003b); characterization of PAH filter sampling artefacts (up to 100 %; linear correlation with ambient ozone; Schauer et al., 2003); detection of nitro-PAH in a high alpine clean air environment (Schauer et al., 2004); observation of characteristic local differences and seasonal trends of aerosol physical properties and chemical composition (Franze et al., 2003b; Schauer et al., 2003; 2004; Zerrath et al., 2003).

3) Experimental investigation and mathematical modelling of the interaction of aerosol particles and components (soot/PAH, proteins) with reactive trace gases (O_3, NO_2) and water vapour: identification of previously unknown PAH nitration and oxidation products (Schauer et al., 2004); detection of efficient protein nitration by polluted air and synthetic gas mixtures (Franze et al., 2003a; Franze et al., 2004b); deconvolution of adsorption and surface reaction processes and determination of adsorption equilibrium and reaction rate parameters for O_3, NO_2, and H_2O on soot/PAH (Pöschl et al., 2001; Pöschl, 2002); development of a kinetic model framework for aerosol surface reactions and gas-particle interac-
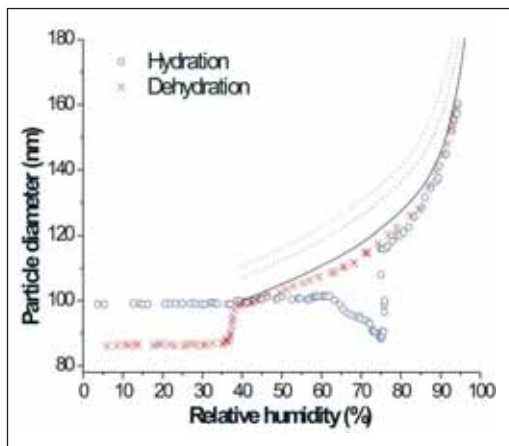
Figure 2: Microstructural rearrangement, phase transitions, and hygroscopic growth of mixed protein/salt particles (BSA/NaCl, 1:1; Mikhailov et al., 2004).

tions (Ammann et al., 2003; Pöschl et al., 2004).

4) Experimental investigation and mathematical modelling of the interaction of water vapour with aerosol particles of complex chemical composition (mixtures of salts and biopolymers, etc.): electric charge effects and microstructural rearrangements; phase transitions and hygroscopic growth; kinetic limitation of deliquescence and water uptake by protein envelopes; parameterisation of the practical osmotic coefficient for globular macromolecules (Figure 2, Mikhailov et al., 2004).

Summary and Conclusions

The abundance and molecular structures of carbonaceous aerosol components and their interaction with water and reactive trace gases have been investigated in laboratory experiments, field measurements and model calculations. New and improved analytical techniques and model formalisms have been developed and applied. Some of the scientific results and implications have already been presented and discussed in journal articles (see references), and several further publications are in preparation. While some of the investigations are still under way, the most innovative findings and conclusions achieved up to now can be summarized as follows:

1) Determination of high concentrations of biopolymers and biological particles in the fine fraction of atmospheric aerosols, which may efficiently influence cloud condensation and ice nucleation (electric and kinetic effects) and indicate that the biosphere acts as a major source not only for secondary but also for primary organic particles.

2) Discovery of efficient protein nitration by polluted air (nitrogen oxides and ozone), which may influence the chemical, optical, and microphysical properties and effects of biological particles in the atmosphere and provides a molecular rationale for the promotion of allergies by traffic related air pollution.

3) Mechanistic elucidation and quantitative description of the interaction of photooxidants with polycyclic aromatic compounds and soot particle surfaces (reversible and competitive adsorption followed by irreversible surface reaction), which implies rapid chemical aging of combustion particles in the atmosphere and upon sampling with traditional techniques (artefacts up to 100 %).

4) Development of a kinetic model framework with consistent and universally applicable terminology and rate equations for gas-particle interactions in aerosols and clouds (transport/reaction, liquid/solid, surface/bulk, reversible/irreversible, competitive/consecutive).

Acknowledgement

Impact of vegetation fires on composition and circulation of the atmosphere (EFEU)

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Introduction

Vegetation fires are a significant source for atmospheric trace gases and aerosol particles (AP) on both local and global scale. The biomass burning AP affect cloud formation as well as microphysical and chemical processes in clouds (Fig. 1). They influence the radiation budget directly and via altered cloud properties. Finally, this results in changes of the atmospheric energy budgets and circulation.

The joint research project EFEU addressed these topics with a combined experimental and numerical approach of eight different research groups.

Three series of experiments were carried out at the laboratory oven facility at MPI Mainz. Characteristic vegetation from different burning regions was investigated, e.g., Musasa (Africa), Aleppo pine (Mediterranean region), spruce (boreal) and peat (Indonesia). Trace gases and a wide range of AP parameters were measured, including size distributions as well as morphological, chemical, hygroscopic and radiative properties.

For the numerical studies of the complex impact of biomass burning emissions on the atmosphere a suite of independent models was employed. Ranging from the microscale to the regional scale they com-

plement each other in terms of spatial and temporal resolution as well as complexity of the processes described. Modelling efforts covered a detailed description of the microphysics including the ice phase, the evolution of individual biomass burning plumes, effects of radiative transport on chemistry and dynamics as well as regional atmospheric budgets of trace constituents, water and energy.

Figure 1: Clouds forming in the plume of a vegetation fire (photo: Wurzler).

Selected Results

Comparison of size distributions: A comparison was made between particle size distributions obtained with the classical SMPS/APS method and with the single particle analysis. A filter sample collected was investigated for found particle sizes. An area of 2860 μm^2 of this filter shown in Fig. 2 was analyzed and a total of 220 particles with sizes ranging from 0.2 to 3 μm were measured.

Figure 2: Filter sample for single particle analysis from Indonesian peat burnt.
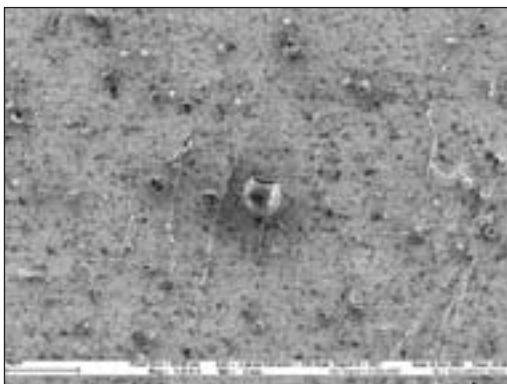
The underestimation of the smaller particles can be understood in terms of the resolution. The image taken for analysis used a magnification of 5350 and 70 nm per pixel. In order to recognize a particle some ten pixels in length are necessary.

When filter surface area and sampled air volume are taken into account, both methods correspond at sizes above 0.8 μm (Fig. 3). Single particles were studied by a scanning electron microscope (SEM) in a two step procedure. Firstly, due to resolution and the risk of destroying particles a low energy electron beam was used to investigate morphology. Secondly, elemental composition was determined using relatively high energy beams (15 keV). Chemical composition was then derived leading to a quantitative analysis which took the main elements into account.

Figure 4 depicts a particle which would be recognized as agglomerate of soot (typically 20-70 nm in diameter), if the spherules were not so large. The ones shown here are larger than 200 nm. Nevertheless, the spherules consist of carbon as is shown in the x-ray diagram. The spot analyzed is indicated by the crosshair. The presence of aluminium stems from the substrate the particle is lying on.

Optical properties of the biomass burning aerosol: Figure 5 shows 2-minute averages of the measured SSA (single scattering albedo) at 550 nm of about 9 minutes old biomass burning aerosol from an oak (dry) fire; the corresponding burning conditions are also shown as dCO/dCO_2 trace (crosses). There is an obvious positive correlation between SSA and dCO/dCO_2.

In the smouldering phase at the beginning of the experiment ($dCO/dCO_2 > 8\%$) SSA was very high with values around 0.95. With the start of the flaming combustion ($dCO/dCO_2 < 8\%$) at 11:33 SSA showed a sharp decrease down to approximately 0.7 followed

Figure 3: Particle size distributions for particles > 0.8 μm (red: SMPS/APS, blue: single particle analysis).
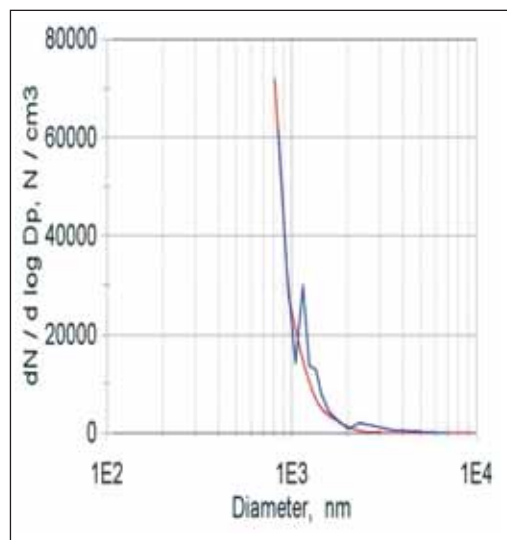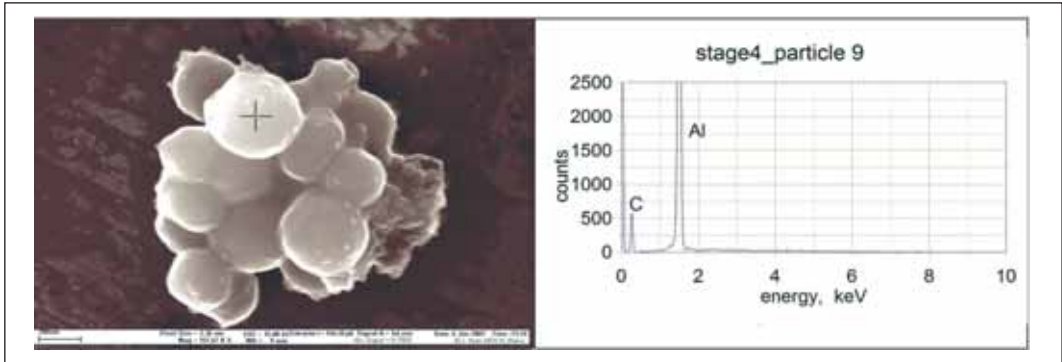
Figure 4: SEM picture and elemental analysis of an agglomerate consisting of carbon from pine burnt.

by a relatively steady increase to just above 0.9 accompanied by an increase in dCO/dCO_2.

For comparison, Figure 5 also shows Mie calculations of the SSA for three different black carbon (BC) mass fractions based on the measured number size distributions. Due to the significantly lower volatility of black carbon (BC) compared to organic carbon (OC) Mie calculations for coated spheres with a BC core ($m_{BC} = 1.75$-$0.44i$) and an OC shell ($m_{OC} = 1.53$-$0.0i$) were performed. Figure 5 shows that the modelled SSA is fairly constant for each BC mass fraction. Hence, changes in size distribution alone cannot explain the observed variations in SSA which indicates a significant role of particle chemistry and

Figure 5: Comparison between measured and modelled single scattering albedo (SSA) for a 1-hour fire with dry oak as fuel.
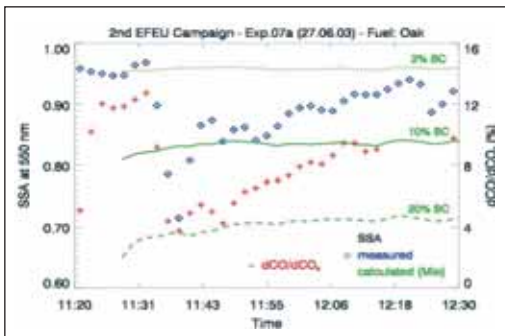
hence m (index of refraction). BC mass fractions of 2 and 20% represent the observed extreme SSA values of about 0.95 and 0.7, respectively, reasonably well. This is consistent with the expected higher BC content for more flaming fires (lower dCO/dCO_2 values) which produce higher combustion temperatures and consequently more soot. Hence, the results suggest that young biomass burning aerosol may be modelled as BC spheres coated with a variable amount of OC depending on the burning conditions.

Drop nucleation ability of AP: As a step for the understanding of the influence of biomass burning aerosol on cloud formation, potential cloud condensation nuclei (CCN) were identified by determining the CCN/CN ratio in the laboratory experiments. This was done for different dry particle sizes (50 to 200 nm) at different supersaturations from 0.24 to 1.64 %. The experimental results are compared to critical supersaturations calculated from extended Köhler theory which is also used in the detailed microphysical models applied in the project. Figure 6 shows the critical supersaturation for varying ε (soluble fraction, here ammonium sulphate) and AP sizes. CCN/CN ratios of 25, 50, 75, and 100 % are deduced from the measurements and compared to the theoretical values. The black square on the red line means, e.g., that 50 % of the 100 nm AP are activated at 0.46 % supersaturation which corresponds to a

theoretical value of $\varepsilon = 0.08$. One can see that ε generally is small ($\varepsilon < 10\,\%$ for about 50 % of the particles) as it is expected for biomass burning AP and that ε is slightly higher for smaller AP sizes (50 nm vs. 150 nm). Despite their small ε, larger particles (150, 200 nm) are activated at relatively low supersaturations that easily could be reached in clouds. Obviously, the critical supersaturations are very sensitive to $\varepsilon < 20\,\%$ especially for smaller sizes. Therefore, small changes in epsilon could highly influence the number of drops in clouds affected by biomass burning. The CCN/CN ratios are quite similar for the different wood types (spruce, pine, oak, and musasa) considered in the EFEU experiments.

Figure 6: Critical supersaturations (lines) and CCN/CN ratios (symbols) for different AP sizes and solubilities.

Figure 7 compares CCN size distributions for various supersaturations gained from the combination of SMPS and CCN/CN ratio data. Oak (left) shows the expected behaviour that the largest AP are activated first (at low supersaturations) and that the smaller AP become CCN for increasing supersaturation. In contrast to this, a certain fraction of relatively large AP from the peat smoke remains unactivated even for high supersaturations at which much smaller AP (right) activate.

Chemical processes in young smoke plumes: The chemical processes in young smoke plumes have been investigated using a chemical 0-dimensional box-dilution-model and 3-dimensional atmospheric transport simulations.

Trace gas measurements from a savanna fire in Namibia (SAFARI 2000) show photochemical production of ozone and acetone within the first 2 hours after the emission. For the investigation of the chemical processes a box-dilution model constrained to the measurements of chemically inert species was employed. The simulated ozone concentration agrees well with the observations (Fig. 8). The simulated acetone concentration was lower than the observations (not shown) pointing to an unknown photochemical source of acetone that has to be taken into account in global budgets.

In addition, the chemical processes in two young biomass-burning plumes from a boreal fire in Alaska

Figure 7: CCN size distributions for various supersaturations (left: oak, right: Northern German peat): significant reduction of large CCN in peat smoke.
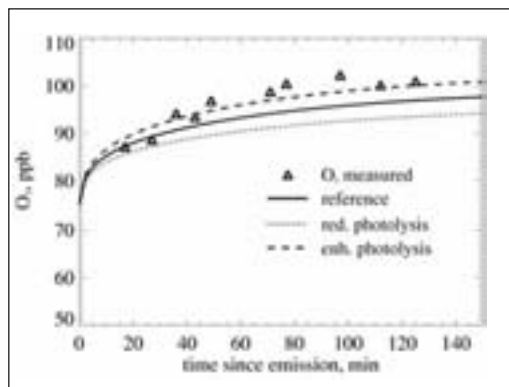
Figure 8: Measured and simulated ozone mixing ratio in the plume from a fire in Namibia.

Figure 9: Measured and simulated ozone enhancement ratios with respect to CO in two fire plumes in Alaska and Africa. Black diamonds represent observations in an Africa plume, the red triangles correspond to measurements taken in an Alaskan plume.

and an African fire in the Timbavati game reserve (SAFARI 2000) were investigated. To reproduce the observed ozone production the model had been extended by the emissions of HONO and several organic species (e.g., furan, toluene, phenol) that had recently been found in biomass-burning emissions. Figure 9 shows measurements and model results of the temporal evolution of the ozone enhancement ratio. Whereas the ozone production is reproduced for the Alaskan fire, for the African fire it is underestimated despite the HONO contribution. A comparison of the photochemical box-dilution model with a different photochemical model (Sherri Mason, SUNY, Fredonia) based on the NCAR Master Mechanism yields results that are in good agreement for both fire scenarios. The photochemical processes that lead to the rapid formation of ozone in the African plume require further investigations.

A simplified chemical mechanism has been coupled with the Active Tracer High Resolution Atmospheric Model (ATHAM), which was used to study the dynamical and chemical evolution of the smoke plume derived from a prescribed fire at the US Pacific Coast. Figure 10 shows the simulated ozone mixing ratio on an isosurface of the aerosol mass concentration. The low values close to the fire and

increasing concentrations downwind the fire are consistent with corresponding observations.

Radiative transfer modeling: Radiative transfer calculations have been performed for a fire-induced convective cloud during the Chisholm fire (Alberta, Canada). The resulting influence of the pyro-cloud

Figure 10: Isosurface of the aerosol mass concentration ($250 \ \mu g/m^3$) colored with the ozone mixing ratio from a simulation using ATHAM.
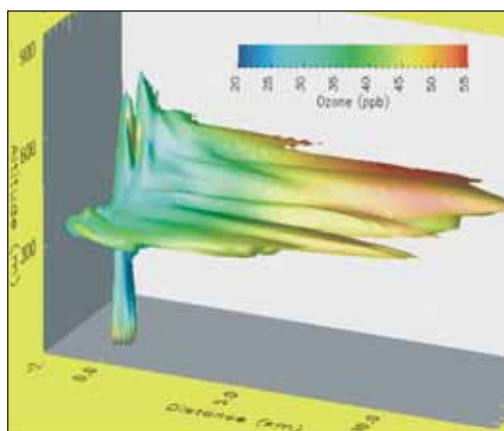
on photochemistry and radiation budget was investigated using a one-dimensional radiative transfer algorithm. The vertical cross section of the photolysis frequency for the formaldehyde photolysis in Figure 11 shows the strong reduction of photochemically relevant radiation in the uppermost part of the cloud such that nearly no photochemistry takes place inside or below the pyro-cloud.

Figure 11: Vertical cross section of the formaldehyde photolysis frequency after 30 minutes for a solar zenith angle of 30°. The contour lines show the distribution of the interstitial aerosol in µg/m³.
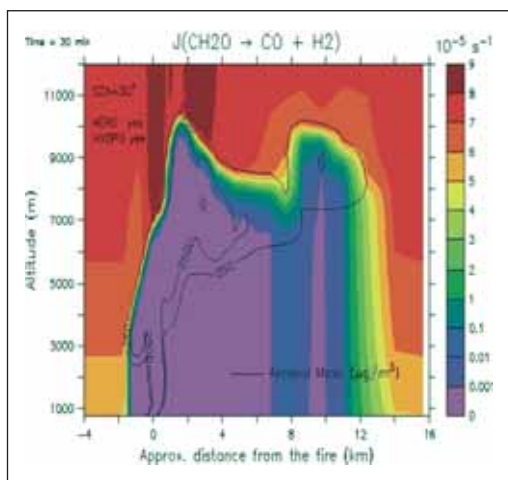
Looking at the shortwave and longwave heating rates it was found that the solar heating is concentrated to the upper part of the cloud with solar heating rates up to 12 K/h assuming a solar zenith angle of 30°. This heating is mainly caused by the absorption of the AP. In the thermal spectral range the formed hydrometeors of the pyro-cloud lead to a cooling of up to 2 K/h at the cloud top and a heating in the same order at and below cloud base whereas the AP influence is negligible. In total heating rates between –1 and 11 K/h (Fig. 12) were found, i.e. the heating rates are dominated by the warming caused by the absorption of solar radiation at the top of the

pyro-cloud. To what extent this may influence the dynamical development of the cloud has to be examined in a next step.

Figure 12: Vertical cross section of the heating rate after 30 minutes. The contour lines show the spatial distribution of the hydrometeors in g/m³.
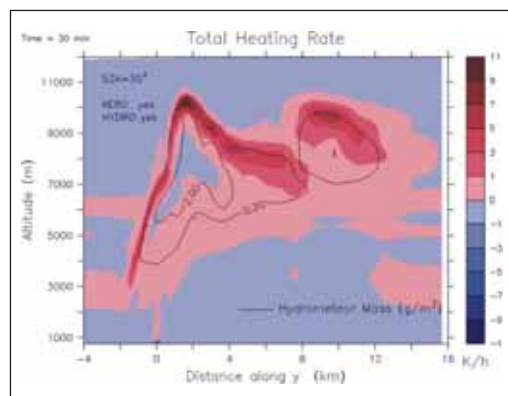
Drop freezing in biomass burning clouds: The suppression of warm rain in clouds over regions with vegetation fires raise the question if precipitation can be formed via the ice phase. Numerical simulations using two-dimensional spectral cloud microphysics in the frame of an entraining air parcel model were undertaken to investigate the effects of drop freezing. According to the high insoluble fraction of the biomass burning AP drop freezing via immersion and contact modes is very efficient. After onset of freezing glaciation of the cloud via the Bergeron-Findeisen process is effective enhancing vertical development of the cloud and leading to formation of cold precipitation. Figure 13 indicates the importance of the biogenic component of biomass burning particles. They are able to initiate freezing at rather warm temperatures compared to other ice nuclei (minerals, soot). Immersion freezing dominates for biological particles and cloud glaciation proceeds mainly by the further freezing of more and smaller drops when the temperature lowers. For mineral particles and
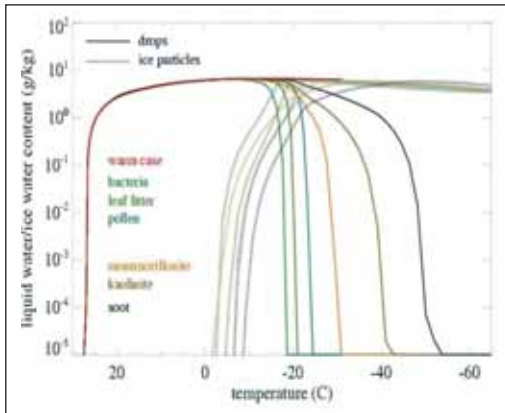
Figure 13: Liquid and ice water contents in g/kg as functions of temperature for combined immersion and contact freezing.

soot, however, contact freezing is dominant proceeding mainly by the transfer of water vapour from liquid to frozen drops forming larger ice particles.

Modelling results from REMO: Based on emission factors and peat area burned the atmospheric particulate matter burden over Indonesia could be simulated successfully with regional model REMO. This was shown qualitatively by a comparison of REMO results with the TOMS Aerosol Index.

To account for the impacts of smoke on cloud microphysics, the influence of AP and drop numbers on coalescence was introduced in the model. In the presence of smoke this leads to a strong reduction of convective precipitation in the interaction run compared to the control run (Fig. 14). However, grid scale precipitation events with low rain rates increase so that a redistribution of precipitation from higher to lower rain rates was observed.

Summary and Conclusions

Results from the EFEU laboratory campaigns indicate that hygroscopic properties and drop nucleating abilities are rather similar for AP from burns of different types of hard wood but different to AP from other burning material such as peat where large CCN are

reduced significantly. Generally, the soluble fraction of the AP is quite small and their BC content fairly high. Radiative properties (SSA) are well correlated with the burn conditions (flaming/smouldering). For radiation calculations, young biomass burning AP can be regarded as OC coated BC spheres. The spherical shape is supported by SEM pictures.

Investigations of photochemical processes in young biomass-burning plumes showed that the temporal evolution of the ozone concentration could be well represented for a boreal and a mid-latitude fire. However, the rapid production of ozone observed in one African smoke plume requires further investigations. For the case of a pyro-cloud forming above an intense boreal fire a strong reduction of photochemically relevant radiation due to

Figure 14: REMO model results: convective precipitation [mm/6h] over Indonesia, March 1st 1998, 6 UTC: suppression of convective precipitation by smoke in the interaction run.
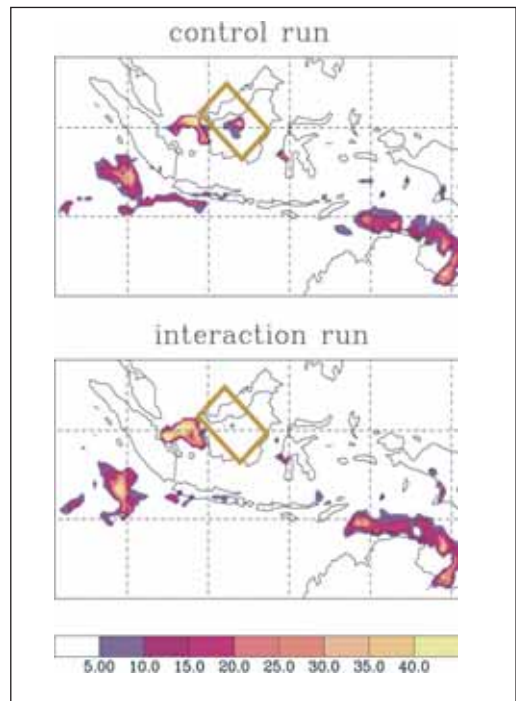
absorption by the smoke aerosol and hydrometeors was found in the uppermost part of the cloud. This absorption also dominates the heating rates. A reduction of convective precipitation and a redistribution of rainfall was observed in a case study over Indonesia in the vicinity of smoke. Emissions of trace gases and particulate matter were derived from peat area burned using the corresponding emission factors. Sensitivity studies concerning drop freezing induced by different particle types being available in smoke plumes indicate enhanced ice nucleation potentially initiating precipitation via the ice phase.

A coupled field and modelling study on aerosol - cloud interaction (AFO 2000 projects FEBUKO and MODMEP)

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Introduction

The FEBUKO and MODMEP projects within AFO 2000 will improve the understanding of tropospheric multiphase processes and especially the interaction of aerosols and clouds with an emphasis on organic particle constituents. Field experiments on aerosol and cloud chemistry and physics, model development and model application are combined to investigate chemical and physical transformation of particles within a cloud passage.

A complex experimental data set was provided by two field campaigns. In MODMEP the development is directed towards a cloud module which combines a complex multiphase chemistry with detailed microphysics. The description of both is given with high size resolution of the drop spectrum. The influence of simplifications within single components

and the kind of their coupling on the simulation results is investigated for different tropospheric situations. Furthermore, techniques are provided and tested which allow an effective implementation of multiphase processes in multidimensional cloud and chemistry-transport models.

The FEBUKO experiments at the three research sites in the Thüringer Wald (Goldlauter, Schmücke and Gehlberg) were carried out in the autumn of 2001 and 2002, respectively, to characterize the aerosol and cloud water with respect to their chemical composition and their physical properties (Figure 1). The speciation of organic components was one of the most important tasks for the different size classes of the particle phase and in cloud water. The air masses encountered in the experimental region are of anthropogenically influenced origin and have then

been exposed to biogenic emissions on their way from the Rhein-Main area to the Thüringer Wald.

Experimental

At the two valley measurement sites (Goldlauter (GL) – upwind site, 605 m a.s.l., and Gehlberg (GB) – downwind site, 732 m a.s.l.) and at the mountain site (Schmücke (SM), 937 m a.s.l.) a complex set of meteorological parameters, trace gas components, cloud parameters and particle characteristics was determined. The sampling of particles using filter samplers and impactors for the chemical constitution measurements was carried out at the valley stations together with measurements of particle number concentrations, size distributions and hygroscopic properties. On top of a 20 m research tower a variety of different

Figure 1:Schematic of the FEBUKO experiments.
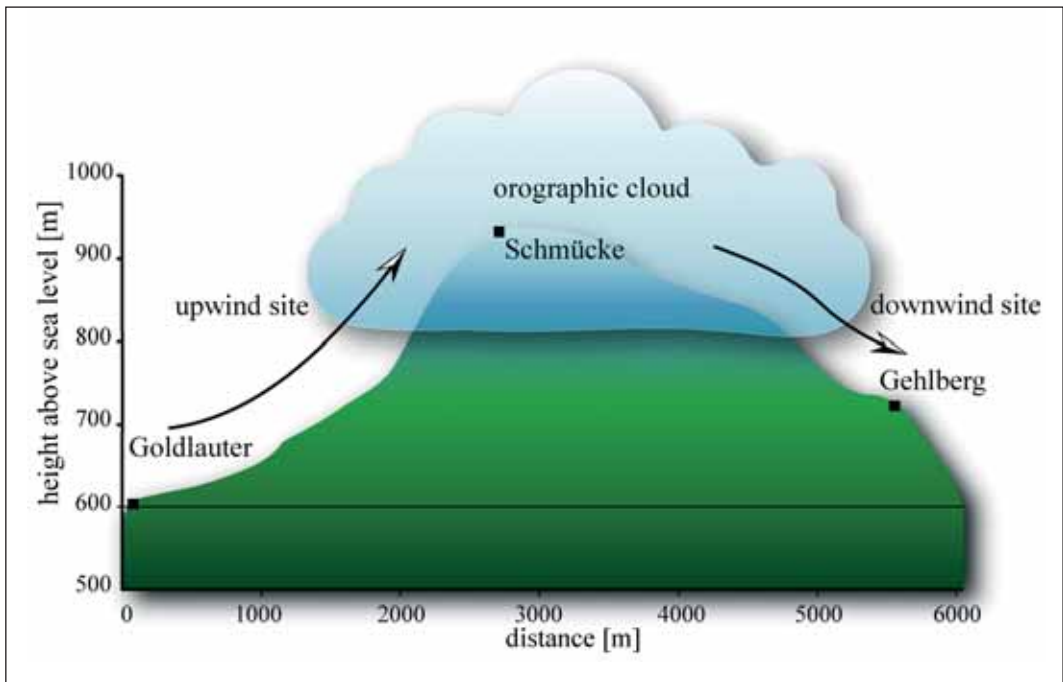
cloud water samplers were operated on the Schmücke. The interstitial aerosol and the cloud droplet residues were collected and physically characterized by mean of a counterflow virtual impactor (CVI) and an inlet to interstitial particles (INT) as well.

The flow connection between the three stations is an important prerequisite for the whole experiment. Hence, special tracer experiments, measurements of number and size distribution of particles as well as trace gas concentrations were performed. In addition, the synoptic experimental conditions were carefully investigated and flow simulations for the complex terrain area are currently in progress.

Results and discussion

In both campaigns in 2001 and 2002 a total set of 14 cloud events was identified for intensive measurements. The most important conditions of (i) a connected flow, (ii) an orographic cloud and (iii) good data coverage were best fulfilled during three events. The results within the present contribution refer to a 15 hours measurement period on 26/27 October 2001 (22:00 to 13:00 UTC).

On the modelling side, at the time of writing of this contribution simulations were made with an air parcel travelling from Goldlauter to the top of the

Figure 2: Particle number size distribution at Goldlauter (GL), Gehlberg (GB) and Schmücke (SM) on Oct. 27, 2001. The black and green curves show the mean size distribution at GL and GB, respectively, whereas the red and light blue curves represent the interstitial (INT) and CVI size distributions. The size distributions of SM INT and GB have been normalized the level of GL by a factor of 1.13. Finally the dark blue curve shows the deviation between the GL and SM INT size distribution.
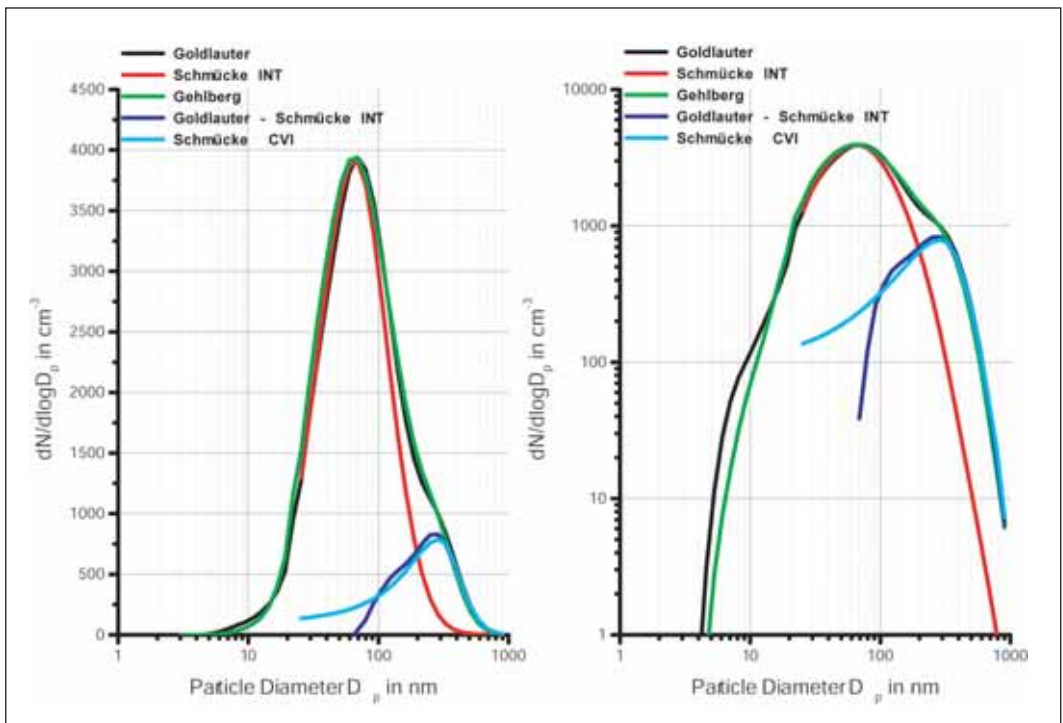
mountain into the orographic cloud. The goal of the simulations was to investigate the multiphase chemistry occurring in orographic clouds and to understand the interaction of particle phase and gas phase of atmospheric trace constituents due to phase transfer and chemical transformation. Simulation results have been compared with cloud water measurements in order to interpret the experimental data and for validation of the multiphase chemistry model.

Physical aerosol characterization

Particle number concentration

Particle number size distributions were measured in GL and GB using a Differential Mobility Particle Sizer (DMPS) system. These measurements are complementary to the interstitial (INT) and Counter Flow Virtual Impactor (CVI) number size distribution measurements on the mountain station Schmücke (SM). In Figure 2, mean number size distributions at all sites are plotted for October 27th, 2001, 0:00 to 13:00 UTC. The black and green curves show the number size distribution measured in GL and GB while the red one represents the interstitial aerosol at the top of Schmücke in cloud. The peak number concentrations of the Aitken modes at GB and Schmücke are adjusted by a factor of 1.13 to the peak concentration at GL to recognize also small deviations. By this adjustment, the rising edge in the small particle size range between 25 and 60 nm of both curves are becoming identical not only qualitatively but also quantitatively. The mean number size distributions between GL and GB do not show a significant difference.

Mean number size distributions of the droplet residues (light blue) and interstitial particles (red) are shown. They have been measured in-cloud at SM during the cloud event in the size range between 25 and 900 nm by two DMPS systems attached to the CVI and INT inlet system. Particles larger than a certain size are expected to become activated, which is demonstrated by the deviation of both distribu-

tions above 70 nm. Figure 2 shows this difference (dark blue) and represents the indirect determination of the CCN number size distribution. This curve agrees well with the direct CVI measurement, which confirms that the phase partitioning due to the droplet activation is successfully observed during the connected flow between GL and SM.

Chemical aerosol characterization

In order to gain a deeper insight into budgets and conversions of specific organic compounds in tropospheric clouds and to have a realistic data set for the model initialisation, one of the aims of FEBUKO is a comprehensive organic speciation at all three measuring sites in addition to standard particle and cloud-water chemical analyses. Organics are usually trace compounds in different atmospheric phases which make their determination a challenging task. A variety of sampling devices was applied, some of them exclusively for organic speciation. With impactors, filters and spray collectors aerosol particles were sampled. Sorbent-filled cartridges were used for gas-phase measurements and four different types of cloud water samplers collected the liquid phase on the mountain. Most of the instruments give time-resolved information about organic concentration levels with a sampling time interval of 2 hours. Complementary to this the impactors allow insights into the size distribution of condensed organic material before and after cloud processing. After the campaign the samples were analysed using analytical equipment like gas chromatography/mass spectrometry (GC/MS), high performance liquid chromatography/mass spectrometry (HPLC/MS) or capillary electrophoresis (CE). With this field measurement equipment it was possible to create an extensive pool of data for dozens of organic substances in their respective most relevant atmospheric phases. Aliphatic and aromatic aldehydes and ketones as well as other volatile organic compounds (VOCs) like short-chain alkanes, alkenes, and volatile aromatics could be

identified and quantified in the gas phase on a time-resolved basis. Depending on their polarity some of them can be scavenged by the cloud and therefore they were measured also in the liquid phase. The possible photooxidation products of carbonyls and VOCs such as higher oxygenated species like monocarboxylic acids in the gas phase and in cloud water and dicarboxylic acids in the particle phase and in cloud water were analysed in a time-resolved manner. Size distributions before and after the cloud passage as well as the corresponding liquid-phase concentrations are as well available for lower vapour pressure compounds such as long-chain alkanes, alcohols, sugars, fatty acids, dicarboxylic acids, and several biogenic compounds such as pinonaldehyde, squalene, and others. Bulk concentrations from filters for interstitial particles allow us to calculate directly the in-cloud scavenging for some of these compounds.

The complex data set obtained in the FEBUKO field experiments is of huge importance for the MODMEP model initialisation and validation. Besides, we can interpret it by comparing, correlating and balancing concentrations of selected species for different atmospheric phases, different times and different particle sizes. In combination with the model results we may now be able to achieve a better understanding of phase partitioning and chemical multiphase processes of tropospheric organic material.

Flow Characterization of the FEBUKO measurement periods with nested non-hydrostatic atmospheric models

The meso-scale and local flow conditions during the ground-based cloud passage experiment FEBUKO were investigated in order to quantify the impact of the local topography on air flow. The main objective was to verify that the air flow over the complex terrain of the Thüringer Wald satisfied the requirements for experimental design during the 14 cloud events. The meso-scale flow conditions were analysed by means of the non-hydrostatic meteorological code LM (Local

Model) with grid spacing of 7, 2.8 and 1.1 km. The meso-scale flow modelling aids to assess if the mountain ridge was overflowed and air masses were lifted up out of valleys so that orographic clouds could be formed or if the air tended to pass around rather than over the mountain range. Furthermore it had to be verified that the chemical and physical properties determined at the different measurement sites were attributed to identical air masses. In order to investigate the connected flow conditions between the different locations ASAM, an anelastic non-hydrostatic model in conservation form, was operated with initial and boundary conditions from the LM. The results of LM were compared with calculated Froude numbers, a non-dimensional flow parameter, and measurements of the natural tracer gas ozone. Also results of tracer experiments using SF_6 were applied to validate the flow characterization by the model ASAM. It was found out that the orographic effect of the Thüringer Wald is expressed in blocking effects, channelling of air flow, especially in the valleys of Werra and Gera, and gravity wave activities. Figure 3 shows an example with air passing over the ridge very well because of high wind speeds and a weak atmospheric stratification. Due to the overflow lee waves are formed .

Figure 3: Vertical cross section of isentropes and zonal wind speed through the experimental site.
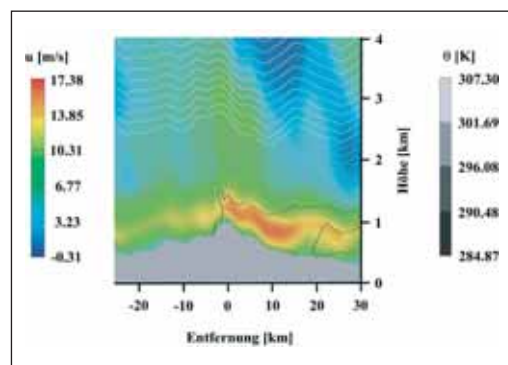
On the basis of this flow analysis some cloud episodes could be selected, which provided the most adequate conditions in terms of overflow of the mountain range as well as the connected flow. These events are recommended for further investigations.

Model initialization

The model developed within the course of the MODMEP project is initialized by means of the particle composition based on measurements with a five stage Berner impactor at the upwind site GL as described before. In the case of the cloud event on the 27th of October 2001, most of the mass was found on the third impactor stage, which collects aerosol particles with a diameter between 0.42 μm and 1.2 μm. Nitrate, sulphate, ammonium, organic and elementary carbon are the main components of the aerosol. In order to correct for losses encountered by the Berner impactor in the case of chloride and nitrate, data obtained with the Steam Jet sampler were used and distributed over the impactor stages according to the relative size distribution of each of these species derived with the Berner impactor. The alkaline and earth alkaline metal initial concentrations, i.e. sodium, magnesium, calcium and potassium, are taken from the Berner impactor. Other metal ion concentrations, i.e. manganese, iron, copper, aluminium and zinc, are initialised according to measurements made with a High-Volume-Sierra-Anderson (PM_{10}) sampler. These measure total contents are distributed over the impactor stages as the mass determined by the impactors. The initialization of the dicarboxylic acids is based on impactor and spray collector measurements. To calculate the water mass of Goldlauter, aerosol growth factors were used. The used growth factors were measured during the ACE2 campaign, and are in a good agreement with the measured growth factors by a HTDMA analyzer within the FEBUKO field campaign.

In the gas phase NO, NO_2, O_3, and SO_2 were measured every 5 seconds with commercial instruments.

For the measurement of HNO_2 and HNO_3 the wet effluent diffusion denuder technique was used. The measured organic species were implemented as stand-alone species or were lumped together into groups as used in the regional atmospheric chemistry mechanism RACM. Implemented stand-alone species are: formaldehyde, ethane, ethene, formic acid, glyoxal, methylglyoxal, and isoprene. Measured lumped gas phase species considered are aldehydes, ketones, unsaturated monoaldehydes, acetic acid and higher acids. For example 19 measured aldehydes were summed up to the lumped species 'ALD'. For some unmeasured species initial concentrations were adapted from the CAPRAM standard scenario 'urban' due to a reasonable agreement between measurements and the above-mentioned scenario.

Tracer experiments were carried out within the FEBUKO field campaign in order to verify the existence of a connected flow and to determine the transport time between the three sampling stations. Based on the experiments a realistic transport time between Goldlauter and Schmücke of 10-20 minutes resulted, depending on meteorological conditions. For the initial wind speed, data from the measurement site of the German Weather Service station located in Meiningen was used. For the cloud event on the 27th of October 2001 at 9.00 UTC an initial wind speed of 4 m×s^{-1} was applied, which led to a simulation time of about 11 minutes from the downwind site to the summit. For the simulations described here, a constant size-independent deposition velocity of 10^{-3} s^{-1} was considered.

Coupled time-integration of detailed microphysics and complex multiphase chemistry in a size resolved cloud box model

To better understand the interaction, effects and evolution of the different physico-chemical processes taking place in the atmosphere their modelling requires a detailed description of all

transformations with equal rigor. The most recently available models focus either on detailed microphysics or complex multiphase chemistry. In the framework of the joint AFO2000-project MODMEP the air parcel model SPACCIM ("SPectral Aerosol Cloud Chemistry Interaction Model") was developed for the description of cloud processes by coupling complex multiphase chemistry and detailed microphysics. The description of both process groups is given for a size-resolved particle/drop spectrum. Either the movement of the air parcel can follow a predefined trajectory (e.g., simulated by the regional model system LM-MUSCAT) or the vertical velocity is calculated for a buoyant parcel in prescribed environmental conditions. Entrainment and detrainment processes are included in a parameterized form. The model allows a detailed description of the transformation of gases and particles shortly before cloud formation, during the cloud life time and shortly after cloud evaporation.

Coupling scheme and time integration

The model variables can be grouped into microphysical variables, chemical variables and mass fluxes between different particle/droplet classes caused by microphysical exchange processes (e.g., by aggregation, break up, condensation). These mass fluxes are schematically shown in Figure 4. When two particles coagulate, for instance, their masses are added to the resulting particle class assuming internally mixed aerosol in each class. The feedback of changes in the chemical composition by gas scavenging and chemical reactions on microphysical processes (e.g., water condensation growth rates via changes in surface tension and the Raoult term) is also implemented.

The time integration scheme for solving the multiphase problem is based on the multistep implicit method (Backward Differentiation Formula). In our approach, the resulting linear sparse systems are solved exploiting the properties

of the Jacobian (sparsity, block structure, different types of coupling). An approximate matrix factorization to decouple the microphysical transport terms and the other parts of the Jacobian is possible. This is seen as an operator splitting at the linear algebra level and yields a computing gain (WOLKE AND KNOTH, 2002). The model SPACCIM is evaluated with data from the FEBUKO field campaign and other case studies from literature. It offers several parameter setups allowing sensitivity studies to reach optimal runs for each studied case.

Spatial description of clouds and their boundaries in multi-dimensional Eulerian grid models

In another MODMEP subproject, the volume-of-fluid (VOF) method (e.g., KAO et al., 2000) was utilised in order to eliminate numerical dilution and loss of clouds as well as undefined cloud boundaries. The volumetric description of the cloud phase, diagnostic reconstruction of the interface, and geometrical reinterpretation of the governing equation provide for

Figure 4: Schematic representation of mass transport between several sections due to microphysical processes. The red hatches part represents the mass fraction of one selected species in the corresponding section.
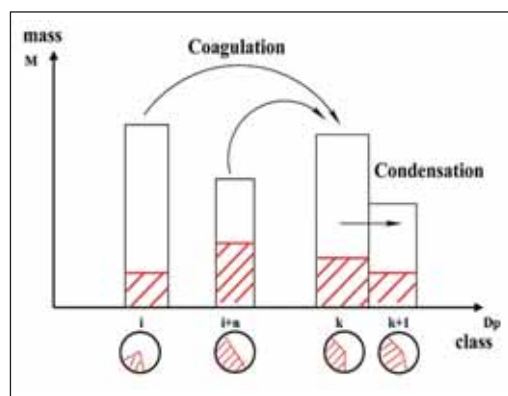
separate treatment of the cloud and its environment. The VOF method proves fast and effective in prognosing the distribution of clouds, because no sub-grid is applied and only minor auxiliary computation near the cloud boundary is needed.

Figure 5 demonstrates for the case of a rising thermal (initialised radius 80 m and height of centre 160 m), that the VOF method enables far longer lifetimes, as soon as it is applied to the advection and diffusion process together, independent of precise knowledge of the effective molecular diffusion coefficient. The variation of the grid resolution and turbulence conditions leads to following conclusions, compared to traditional modelling: (i) Exclusion of the numerical diffusion; (ii) no numerically induced phase conversion processes; (iii) phase-specific application of phase-dependent cloud processes; (iv) non-diffusive and loss-free advection of clouds; (v) only moderate cloud loss by turbulent diffusion; (vi) numerically unhindered development of intensity, extent, and lifetime of clouds; (vii) independence of the grid resolution; (viii) special suitability for long-term processes, small clouds, and coarse grid resolution.

Microphysical model results

Figure 6 shows the cloud liquid water content (LWC, blue lines) and drop number concentration (red lines) on Schmücke for the event on 26/27th of October 2001. The model calculates the LWC adiabatically which leads to an overestimation of about 25 % compared to the measurements. Drop number is overestimated as well. The time dependent structures are represented well for both parameters except for drop number after 9 UTC.

Figure 7 shows the model sensitivity versus changes in the soluble particulate fraction epsilon compared to the drop numbers shown in Figure 6. A constant offset of ±0.1 in epsilon throughout the whole AP size range leads to moderate changes in drop number of up to ±15 %. A higher soluble content (blue line) enhances drop number whereas a lower one (red line) results in less drops.

A completely different picture is obtained when the size resolved representation of epsilon is replaced by a constant average value for all AP sizes (green line). For most of the cases only minor changes occur but for the cases with relatively low LWC large positive deviations of 30-70 % show up. Low LWC corresponds to steep orography (compare Figure 6) with high vertical velocities and, therefore, higher supersaturations causing the activation of rather small AP due to overestimation of their epsilon.

This underlines the importance of a size resolved representation of epsilon and shows that the sensitivity of model results (here drop number) can highly vary for each specific set of parameters.

Figure 5: Cloud after 600 seconds for small constant turbulence coefficient (0.1 m² s⁻¹) and different degree of inclusion of the VOF method: a) without VOF; b) advection with VOF; c) advection and diffusion with VOF (maximal molecular diffusion); d) same as c (minimal molecular diffusion). Selected region: 520 m (width), 220 - 660 m (height). Grid resolution: 5 m.
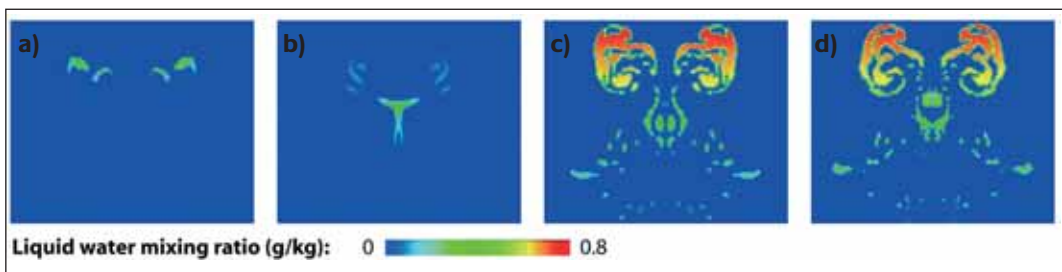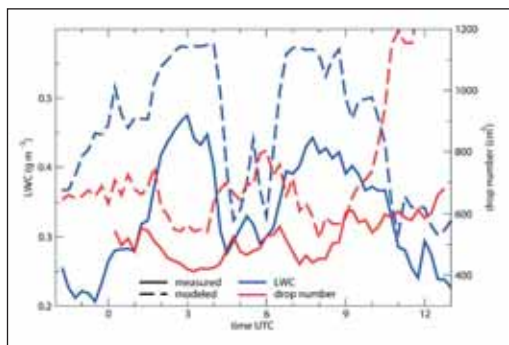
Figure 6: Liquid water content (LWC, blue lines) and drop number (red lines) from measurement (full lines) and model (dashed lines) for the 26/27th October 2001.
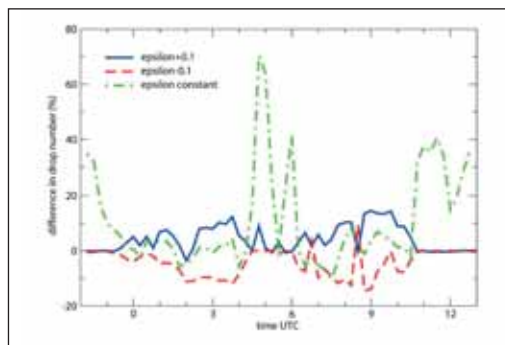
Figure 7: Drop number deviations for changes in soluble particulate fraction.

Simulations with the coupled chemistry microphysics model SPACCIM and CAPRAM

By means of SPACCIM, simulations of the hill cap cloud passage experiment taken place on 27th of October 2001 were carried out. Calculations with an air parcel following a trajectory from the upwind site to the downwind site were performed. The simulation results have been compared to experimental data from the summit and downwind site.

The applied explicit aqueous phase radical mechanisms consist of CAPRAM 2.3 (HERRMANN et al., 2000) and CAPRAM 2.4 (MODAC-mechanism, ERVENS et al., 2003). The gas phase chemistry is described by the regional atmospheric mechanism RACM (STOCKWELL et al., 1997). Phase transfer processes are treated by means of the resistance model of Schwartz considering Henry's equilibrium, gas phase diffusion and mass accommodation. For the simulations a highly resolved particle spectrum is considered. A total number of 64 size bins ($2.3 \cdot 10^{-3}$ μm $< d < 7.5 \cdot 10^{3}$ μm) is considered, where multiphase chemistry occurs in droplets where the LWC exceeds 10^{-9} g·m^{-3}. In the near future a feedback from the chemistry to the microphysics will also be implemented.

For the initial wind speed data from the measure-ment site of the German Weather Service station located in Meiningen was used. For the cloud event on 27th of October 2001 at 9:00 UTC an initial wind speed of 4 m s^{-1} was applied, which led to a simulation time of about 11 minutes from the upwind station to the summit. For the simulations a constant deposition velocity of 10^{-3} s^{-1} was considered.

Simulation results

For the respective cloud event on 27th of October 2001 the exact time for the simulations was fixed at 9:00 UTC due to a good agreement between the microphysical model and the measurement with regards to the total LWC and number concentration. At the upwind station a LWC of about 10^{-4} g·m^{-3} exists. At the summit station the total LWC will reach a value of $4.9 \cdot 10^{-1}$ g·m^{-3}. At the end of the simulation, at the top of the mountain most of the LWC will be in the droplets with a diameter between 8 and 16 μm. The maximum of the LWC distribution, as can be seen from Figure 8, occurs at droplets with a diameter of circa 12 μm. In Figure 8 also the LWC spectra measured by an FSSP (Forward Scattering Spectrometer Probe) is plotted. It can be seen that the measured spectrum of the orographic cloud is broader than the calculated spectra. This dis-

crepancy could be caused by a too broad spectra pro-
duced by the FSSP a too narrow spectrum resulted
from the microphysical model or a combination of
both. Activation of the aerosol particles will occur after
a simulation time of about 300 s.

In Figure 9 the SO_4^{2-}, HSO_4^- and the total S(VI)
concentration profile over the simulation time is rep-
resented. At the summit the cloud water concentra-
tion was measured with a cloud water bulk collector
and at the downwind site the concentration in the
particles was measured with a five stage Berner
impactor. As can be seen from the plot there is a rela-
tively good agreement between the calculated and
measured values. At the summit, in the small
droplets with less water content there will be a high
sulphate concentration. In the small interstitial
aerosol particles the sulphate concentration reaches
values of about $2\,10^{-1}$ mol·l^{-1}. In the size range where
most of the liquid water can be found a sulphate con-
centration of circa $5\,10^{-5}$ mol·l^{-1} occurs.

A further detailed analysis concerning the trans-
formation of inorganic species, e.g. nitrate, chloride,
the speciation of transition metal ions, e.g. iron,
copper, manganese, and the aqueous oxidation of
organic compounds, e.g. acetaldehyde, glyoxal,
oxalate, is ongoing.

Summary and outlook

A coupled version of detailed microphysics and a
complex multiphase chemistry in a box model is real-
ized, allowing a high flexibility concerning the use of
chemical mechanisms and the simulation of case
studies like the FEBUKO field experiment. In the sim-
ulations of the latter experiments using SPACCIM
and CAPRAM for some species there is a quite good
agreement between the measurements and model
results, with deviations below a factor of two, e.g.,
NO_3^-, SO_4^{2-}, K^+, HCOOH, CH$_3$COOH. For other species,
e.g. Fe(II)-Fe(III), HCHO, bigger differences were
encountered. For a better description of organic
chemistry, simulations with the CAPRAM 3.0 mecha-
nism will be conducted. The CAPRAM 3.0 mechanism
incorporates the CAPRAM 2.4 mechanism and con-
siders the oxidation of higher organic compounds
containing up to six carbon atoms.

The SPACCIM model development initiated after
MODMEP will continue to further refine the available
model framework. The CAPRAM mechanism will be
continuously updated and last, but not least, the
Schmücke hill-capped cloud experimental site is

*Figure 8 : Size and time dependent evolution of the LWC
between the upwind and downwind site, comparison between
measurement and simulation result at Schmücke,*

*Figure 9: Concentration profile of SO_4^{2-}, HSO_4^- and the total
S(VI),comparison between simulation results and measurements
at the summit (green circle) and downwind site (orange circle).*
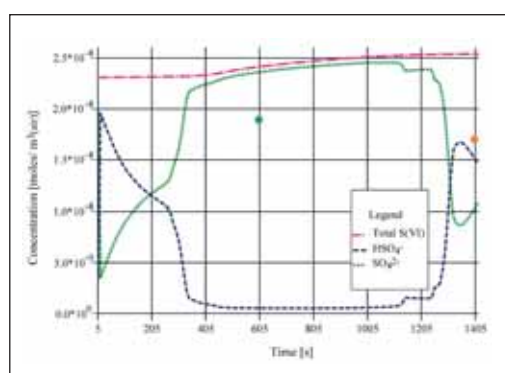
ready for further focussed ground based aerosol-cloud-interaction experiments with emphasis on organics under real conditions and hence complementary to more specialized laboratory experiments.

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Results of Theme Group 4

Atmospheric System Analysis: Models and Data

Integration of satellite observations with the chemistry transport Model MOZART for the analysis of the chemical composition of the upper troposphere (ISOTROP)

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Scientific objectives

Ozone in the upper troposphere is controlled by stratospheric-tropospheric exchange (STE) and in-situ production which depends on the concentration of precursors such as the nitrogen oxides NO_x. Currently, there is no possibility to actually measure the budget of ozone in the troposphere or to distinguish between transport and chemistry on a global scale. Models have to be used to quantify the individual budget terms of ozone. But there are large discrepancies between different models for instance in the calculated cross tropopause fluxes of ozone. Therefore, the main objectives of this project are :

+   **determination of the sensitivity of model calculated cross tropopause fluxes of ozone on meteorological and chemical boundary conditions**
+   **comparison of model calculated ozone concentrations with observations in the upper troposphere on a global scale**
+   **improvement of the estimated global emission pattern of nitrogen oxide emissions using satellite data**

Modelling studies of the global tropospheric ozone budget

Quantitative knowledge of the global tropospheric ozone budget is an important prerequisite for making reliable estimates of the future background air quality and climate forcing of non-CO_2 greenhouse gases. Tropospheric ozone is either advected from the stratosphere or formed in-situ from the precursor species NO_x, CO, and hydrocarbons. Its sinks are chemical destruction in the air or on the surface (dry deposition). Due to the various feedback processes between different chemical reaction cycles and between atmospheric chemistry and meteorology, it is difficult to delineate the contribution of individual factors to the global ozone budgets. Intensive field studies can provide snapshots of the atmospheric chemical composition and allow for reasonably well constrained estimates of the chemical ozone formation and destruction rates, but they are necessarily limited in space and time. Satellite data are very useful to identify large-scale horizontal distributions of ozone and some precursor species (e.g. NO_2, HCHO), but they lack the required vertical resolution. Ground-based measurements, on the other hand, are often influenced by local effects, and extrapolation on continental or hemispheric scales is

thus impossible. Therefore, comprehensive chemistry transport models (CTMs) provide the only way to assess the tropospheric ozone budget in a globally consistent and complete manner.

Within ISOTROP, we accomplished further development of the MOZART-2 CTM (Horowitz et al., 2003), and we investigated the interannual variability of ozone concentrations over Europe (Fig. 1). Based on a multi-year simulation driven with ECMWF winds, we found that the influence of different wind patterns between years is much larger than the effects through changes in emissions or radiation. The magnitude of stratosphere-troposphere

Figure 1: Difference in the ground level ozone concentration over Europe caused by variations in the wind fields alone between May 1993 and May 1996. The values are ozone mixing ratios in ppb from a multi-annual simulation with the MOZART-2 model.
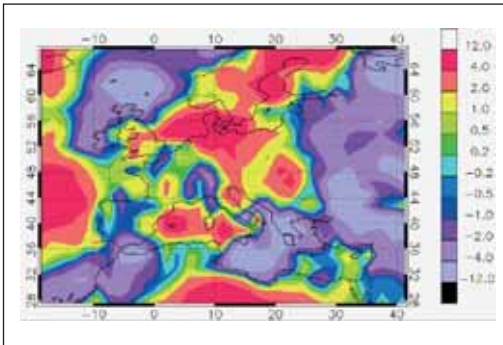
exchange in the model is highly dependent on the wind fields used (Fig. 2). ECMWF and NCEP fields yield cross-tropopause fluxes which differ by almost 20% for the same calendar year. In order to better evaluate the MOZART-2 model, we have begun to statistically compare the simulation results with data from the European MOZAIC project (Marenco et al., 1998; Fig. 3). This study reveals that the seasonal cycle is well reproduced by the model, but there is a significant bias in the lower stratospheric/upper tropospheric ozone concentrations caused by excessive vertical diffusion in the model. A minor fraction of

the bias can also be explained by sampling biases in the observational data (altitudes limited to commercial flight corridors) and the model (vertical resolution > 1 km in the lower stratosphere).

Figure 2: Comparison of MOZART-2 predictions (black) with MOZAIC data (red) for ozone in the UTLS over Europe (43°-60°N; 10°W-30°E) from Jan. 1993 to Jan. 2002. The phase of the seasonal cycle and some of the interannual variability is well reproduced by MOZART-2. The absolute concentrations, however, are overestimated by 100 ppb in the LS (T+1) and at the TP and by 50 ppb in the UT (T-1).
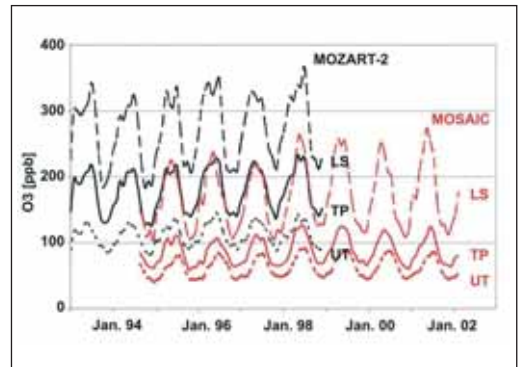
Figure 3: Sensitivity of the MOZART-2 cross-tropopause flux of ozone on different meteorological input fields.
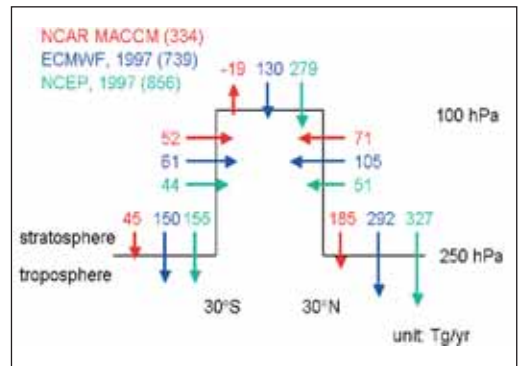
Correlation analysis of the distribution pattern of nitrogen oxides

Due to the short tropospheric lifetime of NO_x its global distribution strongly corresponds to the distribution of emissions. We want to quantify the relative contributions and the geographic distributions of individual NO_x emission sources using a variety of satellite data. Nitrogen dioxide NO_2 measured by the GOME satellite (RICHTER et al., 2002) has been compared to night-time light emissions observed from space by the DMSP OLS satellite (ELVIDGE et al., 2001; Fig. 4). The light emissions from human settlements can serve as a proxy for emissions of NO_x from fossil fuel combustion. The correlation of tropospheric NO_2 measured by GOME with light density of human settlements at the Earth's surface (R=0.75) is as good as with the estimated anthropogenic emissions in the EDGAR database (OLIVIER et al., 2001) which are widely used in global chemistry models.

To use all of these data sources for an assessment of the global distribution pattern of NO_x we developed a multi array correlation method. This method is universally valid if three or more independent sources of information are available. The multi array correlation method is able to derive pattern error estimates for each of the involved data sources and to evaluate weighting factors for the optimal combination of the available information. With data from GOME, DMSP OLS, and EDGAR, we were able to assign ranges for the pattern errors of the GOME satellite instrument of 0 to 37% for regions dominated by anthropogenic emissions. Combining all information reduced the estimated pattern error of anthropogenic emissions in the EDGAR database from 25±5% to 15±2%. The regions with the highest uncertainty and the least agreement between the instruments seem to be north-eastern China and megacities like London and New York.

Figure 4: Night-time lights of the world, DMSP Operational Linescan System; white:human settlements, red:ephemeral lights (primarily fires), green: gas flares, blue: heavily lit fishing boats, Oct 1994 - Apr 1995 (Elvidge et al., 2001)

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4D-CLOUDS: Inhomogeneous clouds

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The 4D-CLOUDS project aims at capturing the radiative influence of inhomoge-neous clouds and at implementing these influences in the modelling of transport and exchange processes in dynamical atmospheric models. Especially, the project wants to contribute to the major scientific questions around enhanced absorption, the relation between albedo and optical depth in satellite retrievals, and the relation between dynamical exchange processes and the 4D cloud structure.

The measurement component of this project consisted of two comprehensive campaigns, the Baltex Bridge Campaign (BBC) in 2001 and the BBC2 campaign in 2003 (CREWELL et al., 2004). Both were held in the Netherlands around Cabauw. Some results of the analysis of the data from these campaigns are in this paper.

In the radiation modelling part of the project, three dimensional radiative transfer models in all relevant wavelengths are performed. They will be used to develop parameterisations of radiative transfer in atmospheric models and to develop retrieval algorithms for satellites that take cloud structure into account. The 4D-cloud project runs for 5 years, thus this paper represents its mid-term status.

Cloud structure and solar radiation (IfT Leipzig)

The airborne measurements from the Baltex Bridge Cloud (BBC) campaign were used to study the impact of cloud microphysical inhomogeneities on solar spectral irradiances. The measured cloud microphysical parameters (liquid water content, LWC, and effective droplet radius, R_{eff}) were used to construct the three-dimensional (3D) microphysical cloud field, needed as input for subsequent 3D radiation calculations. Three methods were utilized: (a) A homogeneous cloud field ("hom_cloud") was generated by averaging the measured profiles of LWC and R_{eff} and distributing the average over the entire model grid. (b) For the "pdf_cloud" the LWC and R_{eff} were randomly distributed over the grid such that they were consistent with the measured two-dimensional probability density functions (PDFs) of both quantities. (c) In the "shift_cloud" model horizontal variations in the cloud parameters were introduced by shifting the average measured profiles considering a LWC time series from a horizontal in-cloud flight leg.

These three reconstructed cloud types (a) to (c) were used as input for 3D radiative transfer simula-

tions. A typical example of the results is given in Figure 1, which compares the measured and calculated PDFs of the upwelling (reflected) irradiances above cloud for a specific case. Overall the simulations fit best with the measurements for the "shift_cloud" case, which represents the most sophisticated of the three cloud reproduction methods. Both the measured mean value and the variability of F↑ are well reproduced by the calculations for the "shift_cloud". The mean value is slightly overestimated by all three schemes, though the bias is within the measurement uncertainty. The measured variability is sufficiently covered by the "shift_cloud" only. Overall different properties of the radiation field are best reproduced by different cloud reproduction schemes.

Experimental investigations and modelling of the radiation budget in the cloudy atmosphere (Free University Berlin)

In order to improve the understanding of multiple scattering and absorption, a systematic investigation of the effect of 3d cloud structure on the atmospheric radiation field was carried out. The investigations are based on radiance observations taken during the BBC campaign and on radiative transfer simulations. The combined results led to a better understanding of the processes affecting the results and enhance their reliability (SCHRÖDER, 2004a).

The input fields for the radiative transfer model were either generated with a model developed by VENEMA et al. (2004), with a LES model, or by a Fourier model developed by the authors. In case of the LES model, a new mixing scheme was introduced to account for non-adiabatic LWC profiles (CHOSSON et al., 2004). The study of input parameters for these models is based on data from CREWELL et al. (2004) and MEYWERK et al. (2002), and revealed a much stronger degree of non-stationarity for LWP compared to literature values (decrease of power spectrum with –8/3 instead of -5/3) and higher standard deviations of cloud base height than cloud top height.

The effect of gas absorption, surface albedo, and two layer clouds on the nadir radiance was investigated by applying the power spectrum analysis to the obser-vations and simulations. The outcome of the analysis of the observations is given in SCHRÖDER and BENNARTZ (2003a), SCHRÖDER et al. (2003b) and SCHRÖDER et al. (2004b). In particular,

Figure 1: Comparison of measured and simulated reflected irradiances F↑ above cloud at 500 nm wavelength for the three cloud reproductions "hom_cloud", "pdf_cloud", and "shift_cloud".
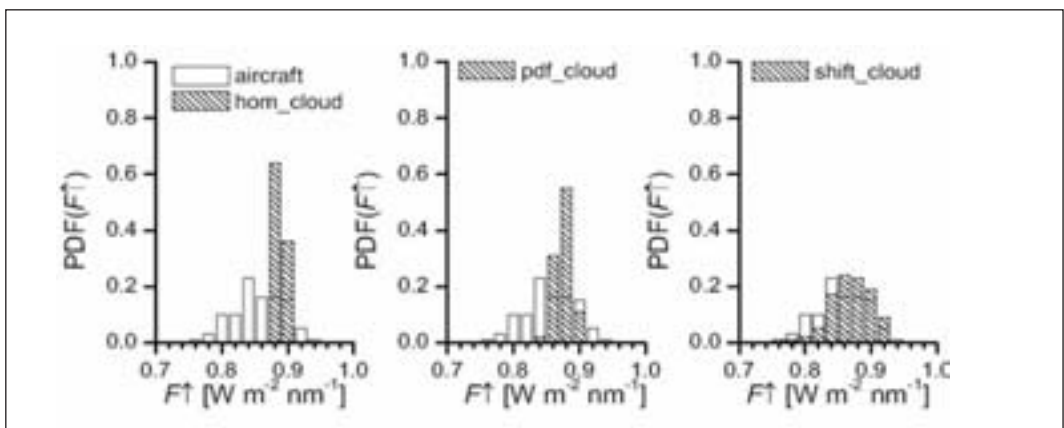
the term cloud radiative smoothing was extended to account for large and small scale effects. The results from the observations and simulations are in excellent agreement. However, the computations revealed that the occurrence of large scale cloud radiative smoothing depends on the relative distribution of the optical thicknesses for two layer cloud systems (SCHRÖDER et al., 2004c).

The effect of vertical layering and averaging was studied on the basis of realistic cloud profiles (SCHRÖDER et al, 2004d). As soon as absorption is effective, large effects on nadir radiance observations are found, and the intensity of the absorption and the volume extinction at cloud top are the dominant dependencies. The mixing scheme and the layer thickness are of minor importance. Since the vertical profile of the extinction is usually unknown, the retrieval of cloud properties should be carried out in the core of convective cells. Great care is required, if satellite observations of different resolution are compared. A simple adjustment of the resolution should be accompanied by an exact allocation of the measurements. Utilising adiabatic profiles the mean photon path length was parameterised with the optical thickness and the effective radius. The parameterisation offers a way to improve the retrieval of atmospheric properties which rely on gas absorption. The inclusion of a microphysical parameter may enhance the quality of the computation of heating rates in GCMs.

Measurements and synthesis of cloud LWC (Bonn)

For many applications it is desirable to have 3D cloud liquid water content (LWC) fields. Unfortunately, this is impossible to measure. Thus, we are working on generating 3 dimensional LWC fields that have several statistical properties of the cloud that we measured. We call these fields surrogate cloud fields. We developed a simple and fast iterative method to make surrogate cloud fields, that allows specifying

both, the power spectrum and the amplitude distribution (VENEMA et al, 2004a). It allows one to generate 3D Liquid Water Content (LWC) fields based on 2D LWC profile measurements assuming horizontal isotropy, see Figure 2. Furthermore, theoretical statistics can be used to make, e.g., monofractal surrogates with a certain slope of the log-log power spectrum and an (almost) arbitrary LWC distribution. To validate the method we have created stratocumulus and cumulus fields that have almost the same radiative properties as LES cloud fields. The LES clouds and their surrogates had almost the same radiative properties. (VENEMA et al., 2004b)

Figure 2: A 2D LWC retrieval (top) and the 3D IAAFT surrogate field, made from its statistics (bottom).
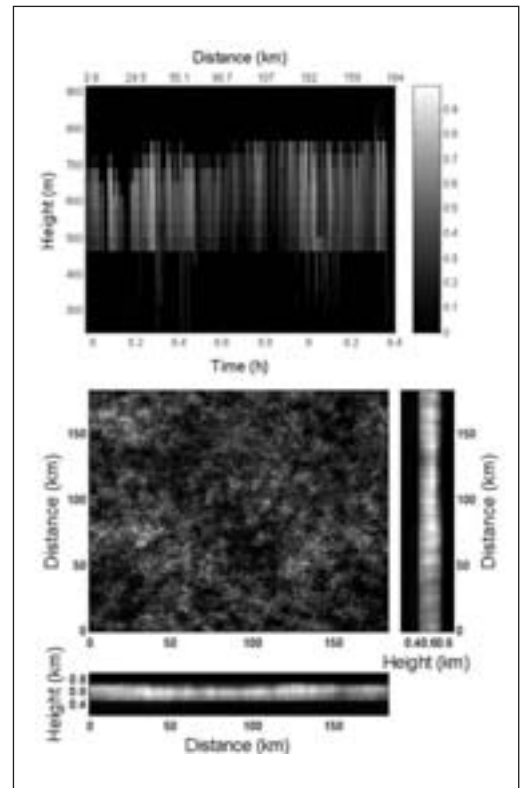
A constrained method using a search algorithm is a very flexible way to generate a cloud field (3D LWC field) with almost arbitrary measured statistical cloud properties. We have used an evolutionary search algorithm for its robustness and relatively good efficiency (VENEMA, 2003).

The algorithms can easily be used for other geophysical fields as well. See http://www.meteo.uni-bonn.de/victor/themes/surrogates/ for example programs.

Photon path length distribution (Heidelberg)

Within the AFO-2000 sponsored 4D-CLOUDS project, the Institute for Environmental Physics (IUP) at the University of Heidelberg tackled various facets of the radiative transfer under the clear and cloudy skies. The conducted research included studies on (a) the absolute measurement of the extraterrestrial solar spectrum within the 320 – 650 nm spectral range (GURLIT et al., 2004), (b) the UV/vis radiative transfer in the stratosphere (BÖSCH et al., 2000, and WEIDNER et al., 2004), (c) the spectroscopic parameters of collisional complexes, metastable and stable clusters relevant for atmospheric absorption (PFEILSTICKER et al., 2001 and 2003) (d) the large scale variation and stationarity of zenith radiance time series measured under cloudy skies (SAVIGNY et al., 2002), and (e) measurements of clear and cloudy photon path length distributions and their relation to cloudy sky parameters measured otherwise, like the cloud vertical distribution, liquid water path, and cloud optical thickness (FUNK AND PFEILSTICKER, 2003 and SCHOLL et al., 2004). While the research activities performed under (d) and (e) fall under the primary research objectives of the 4D-CLOUDS project, the research activities covered under items (a) to (c) are nevertheless regarded to be rather important to improve our understanding of the absorption of solar radiation in the Earth atmosphere, and the potential disturbances to it due to human activities.

Our research on the temporal and spatial variation of zenith sky radiances (SAVIGNY et al., 2002) indicated the existence of horizontal scale breaks (1) due to radiative smoothing for distance of the order of the cloud vertical extension, and (2) due to a transition from non-stationary to stationary behaviour at spatial scales of a few tens of kilometres. Our study also revealed the feasibility of zenith sky radiance measurements to infer cloud optical thickness, when the former are corroborated by radiative transfer models.

Further, our cloudy sky photon measurements using high-resolution oxygen A-band spectrometry (FUNK AND PFEILSTICKER, 2003 and SCHOLL et al., 2004) are clearly indicating the transition of normal to anomalous diffusive photon transport for optical cloud thick cloud covers. Generally speaking the transport of solar photons under cloudy skies obeys the physical laws of normal diffusion for simple, i.e. single layered stratiform clouds, whereas the physical laws of anomalous diffusion govern the photon transport for complex cloud covers, i.e. multi-layer, convective and broken in place cloud covers.

Future research will need to focus on the connection of our results to the clear and cloudy sky radiative transport, as currently implemented in global climate models.

Cloud liquid water profiles from sensor synergy and inter-comparison with in situ measured values (GKSS)

Among other parameters cloud liquid water content and its vertical distribution within the cloud are essential information when it comes to modelling the radiative transfer of clouds. Within the project we used a 95 GHz cloud profiling radar, together with data from a ceilometer, time series of liquid water path from a passive microwave radiometer and radio sounding information to correct the radar data for attenuation effects. All these data has been

collected simultaneously during three field phases, which had been conducted jointly with the CLIWA-NET field phases during April and May 2001 (pre-experiment), August and September 2001 (main field phase) and Mai 2003 (post experiment).

From the above mentioned sources, the time series of liquid water profiles have been derived using a method, which redistributes the liquid water path to the entire cloudy column, weighted by the radar reflectivity (FRISCH, 1998). In addition to this the radar reflectivity has been corrected in a recursive manner to account for attenuation effects due to water vapour, oxygen and the liquid water itself (modified FRISCH, MEYWERK et al, 2002, 2004). It turns out that its correction increases the liquid water content at its maximum (the upper third of the cloud) by on average 14%.

Figure 3: Time series of liquid water path (a) using Frisch (1998), (b) corrected Frisch, and (c) adiabatic LWC.
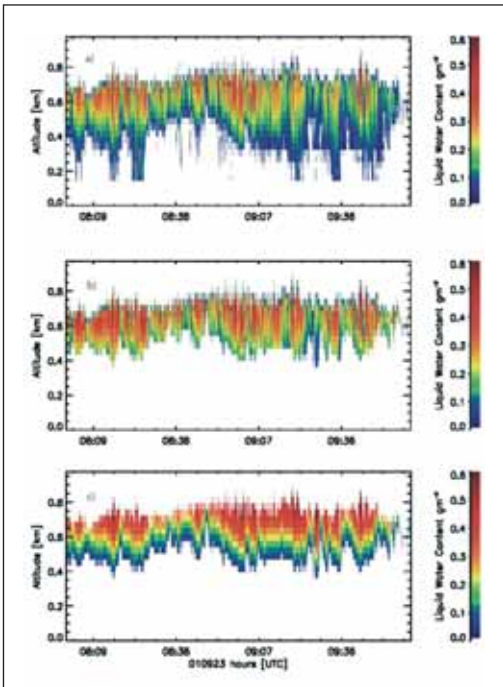
The calculated liquid water profiles have been compared to those which have been calculated with classical methods (relating liquid water to the radar reflectivities alone, ATLAS (1954), BAEDI et al. (2000), FOX AND ILLINGWORTH (1997), SAUVAGEOT AND OMAR (1987)) and in-situ measured liquid water profiles from aircraft. It comes out that the classical methods by far underestimate the liquid water profiles if compared the modified Frisch and in situ measurements. This is because for our case investigated, the air masses were of continental origin, while the classical methods have been developed for marine air masses. As a major result from this part of the project an examples of a time series of liquid water content profiles from September 23rd, 2001 in comparison to uncorrected profiles, as well as the adiabatic values are given in Figure 3. These have been widely used by project partners for radiative transfer calculations.

Radiative transfer in 3D inhomogeneous clouds and development of parameterisations (Leipzig)

To assess three-dimensional (3D) radiative effects of realistic clouds resolved 3D cloud extinction fields are required for the radiative transfer (RT) computations. Surrogate clouds generated by means of stochastic cloud generators developed in the 4D-CLOUDS joint project, which preserve the relevant statistical properties of the measured clouds, can be used as input for such calculations. Here we present results for 45 Sc clouds measured in the BBC1 campaign and for 45 pairs of surrogate Sc clouds.

Figure 4a shows how well the domain-averaged reflection for all pairs of surrogate clouds agrees with the reflection of the 45 different measured 2D clouds. The RT was calculated with a 3D Monte Carlo RT model (GIMENO GARCÍA AND TRAUTMANN, 2002) for a wavelength of 550 nm and a solar zenith angle of 60°.

As a second application to speed-up 1D RT computations we applied the so-called linear radiative perturbation theory (LPT) approach (BOX et al.,

1989) using the RT model DISORT to perform the forward and adjoint multiple base case calculations for a 3D cloud field in independent-pixel mode. LPT allows one to treat altitude dependent changes of the extinction coefficient and the scattering phase function. Hermite interpolation was employed to extend the LPT results over a considerable range of cloud optical depths. Fig. 4b presents the downwelling flux field for a partially cloudy Sc layer (the cloud extinction field was kindly provided by Dr. R. Scheirer, DLR). Using LPT the absolute error for domain-averaged transmission and reflection turned out to be below $3*10^{-3}$. With a non-optimized version of the method we obtained a speed-up factor of at least three.

Exact 3D-radiative transfer (Kiel)

Remote sensing of the cloudy atmosphere relies on the proper interpretation of the solar reflected and the thermal emitted radiances. This in turn requires a most realistic simulation of the radiative transport. In the framework of 4D-CLOUDS radiation transfer models have been developed/improved/applied and

made public available. These models are based on the Monte-Carlo method and thus allow for the consideration of arbitrary complex 3d atmospheric settings.

MC-UNIK (MACKE, A. et al., 1999) is a forward MC method with a local estimate scheme for efficient calculations of radiances at discrete directions. It has been extended to calculate the spatial weights of cloud leaving radiances. These weights are used to semi-empirically account for the 3d structure in cloud retrievals from satellite based radiance measurements.

GRIMALDI (SCHEIRER, R. AND MACKE, A., 2001) is another forward scheme to solve the radiative transfer in 3d scattering and absorbing cloudy atmospheres with direct (solar) illumination. It is based on MC-UNIK and provides monochromatic fluxes and spectral band fluxes, radiances and photon path length distributions by photon counting. In 4D-CLOUDS it is used for calculating domain average radiative fluxes which in turn are correlated with domain average cloud properties to obtain new radiative flux parameterisations.

3RAD-UNIK is a backward Monte Carlo scheme to solve radiative transfer problems in a 3d emitting

Figure 4: (a) Domain-averaged reflection for surrogate clouds and their measured counterparts. (b) Multi-base case perturbation results for the downwelling flux field of a partially cloudy Sc layer of size 20 km x 10 km.
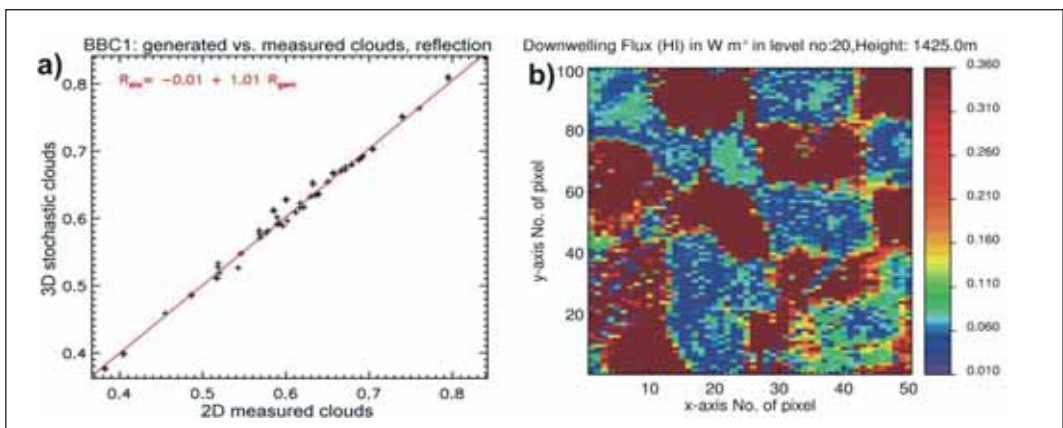
cloudy atmosphere (LIU, Q. et al., 1996). It has been extended to the thermal spectral range in order to simulate and to better understand thermal emission from 3d clouds.

The backward Monte-Carlo scheme MC-UNIK-BW has been developed from MC-UNIK to efficiently simulate photon path lengths for specific illumination and viewing conditions. Path length distributions have been simulated for 3d cloud fields and the moments of the distributions have been correlated to local and to domain averaged cloud properties. Comparisons with indirect measurements of path length distributions enable new remote sensing techniques for large scale cloud properties with full account on the 3d cloud structure.

Reconstruction of three dimensional cloud fields (Dresden)

In this ongoing part of the 4D-CLOUDS project the aim is to generate a method to reconstruct three dimensional cloud fields from 2 two-dimensional cloud property input fields.

These fields consist of one horizontal field of vertically integrated cloud parameters like liquid water path, cloud fraction and cloud top height derived from satellite data and a vertical slice located in the observed volume. The vertical slice is derived from time series of radar and microwave radiometer data, which include liquid water content profiles, cloud base and cloud top height.

Because measured three dimensional cloud fields do not exist, model data are used to develop the recon-struction method. Therefore data of the Lokalmodell of the German Weather Service, which have been operated for selected days of the Baltex Bridge measurement campaign in the Netherlands in August / September 2001 with a refined horizontal resolution of 1 km, have been combined with satellite data and ground based measurements. From these merged three dimensional cloud field, the two dimensional input fields described above have been extracted.

To reconstruct the three dimensional cloud field, information about the vertical distribution of the liquid water within the columns of the vertical slice have been used to vertically resolve the liquid water path of the columns besides the slice. Subgrid cloud fraction has been resolved by different cloud masks. Different parameterizations of the effective radius have been applied.

Three dimensional radiative transfer simulations have been performed with the reconstructed cloud field to validate the quality of the cloud field by the deviation of the simulated radiance from the radiances observed by satellite.

Problems arise from the bad correspondence of the data of the different instruments observing the same volume like liquid water path from satellite and microwave radiometer. Furthermore clouds contain variabilities of liquid water content, effective radius and cloud top height on the subscale range which are relevant for the radiation field, but are not observed. Another problem results from the missing information about the subgrid structure of the cloud field.

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Transport, chemistry and trace gas distribution in the tropopause region (TRACHT)

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Objective

The focus of TRACHT has been to study mesoscale processes affecting the trace gas distribution in the tropopause region. To achieve this aim, high resolution satellite observations from the second CRISTA flight in August 1997 (OFFERMANN et al., 1997) have been combined with global and regional scale models. The global scale model NCAR-Rose (RIESE et al., 1999) was used to assimilate the CRISTA data. Global eddy transport analyses employing the observations have been carried out by Schaeler et al. (2004). The mesoscale aspects of the trace gas

fields have been investigated using the chemistry transport model EURAD (HASS et al., 1993; EBEL et al., 1997) adapted to the tropopause region (KOWOL-SANTEN et al., 2000). Two special cases (blocking, tropical convection) have been analysed in detail (see Fig. 1). In addition, the EURAD model has been evaluated employing CRISTA and MOZAIC data. As a result it has been concluded that the model can well be used for simulating distributions of temperature, ozone, water vapour and CFC11 in the tropopause region.
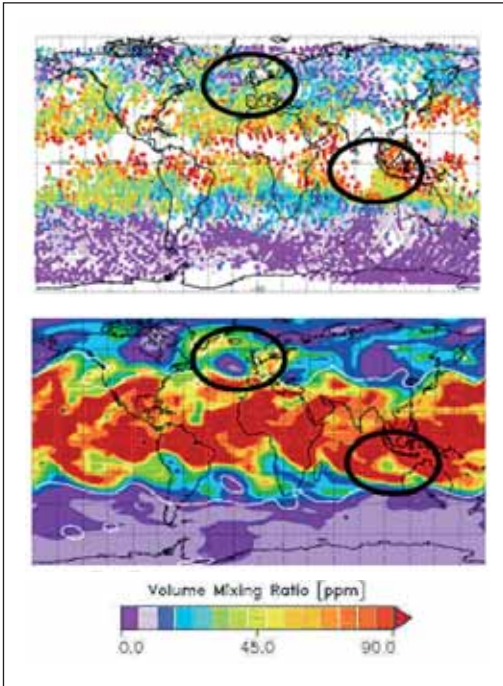
Figure 1: Global water vapour distribution as observed with CRISTA-2. Upper panel: Mixing ratios (ppmV) at the 215-hPa constant pressure surface from CRISTA-2 measurements compiled for the episode August 8th – 15th 1997. Lower panel: H_2O mixing ratios (ppmV) at 2^6 hPa on August 12th 1997, 00 UTC, from CRISTA-2 observations assimilated with the ROSE model. The intersection between the 215-hPa constant pressure and the 2.5 PVU constant potential vorticity surface is indicated by a white line. The latter surface indicates the tropopause outside the tropics. The meso-scale features analyzed in this study are encircled.

Figure 2: CFC11 [pptV] on August 12th 1997, 0 UTC from EURAD simulation. (top) horizontal cross section at 215 hPa (bottom) vertical W→E cross section through the Ω structure (cut-off low, central ridge and streamer).

Omega pattern (blocking)

The circulation over the eastern North Atlantic and a large part of North and East Europe was controlled by a blocking event nearly lasting during the whole mission period of CRISTA-2. The pressure distribution exhibited an omega pattern with its central high pressure part over the North Sea, a rather persistent cut-off low to the west over the eastern Atlantic and a variable trough to the east. The horizontal and vertical cross sections in Figure 2 indicate that CFC11 like other tracer distributions was heavily controlled by the blocking. Marked differences of horizontal and vertical air mass transport could be identified for the different parts of the blocking with net downward fluxes in the wings and net upward flux in the central ridge.
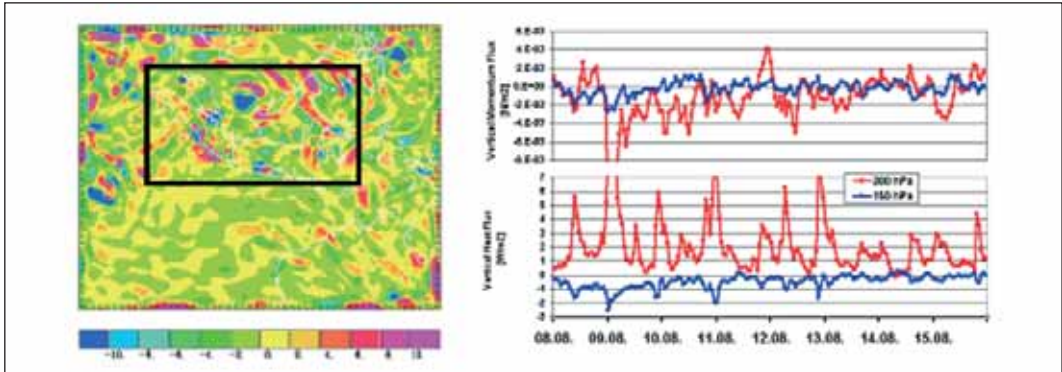
Figure 3: (left) Eddy heat flux at 200 hPa. Summing up the eddy fluxes in the area marked in the figure one obtains a quantitative esti-
mate of possible integral contributions of convectively induced disturbances to the vertical transport on the meso-scale as shown in
the right panel for heat and horizontal momentum. An interesting finding is that upward eddy fluxes of horizontal momentum may
episodically ad up to pulses with magnitudes equivalent to those found in severe tropical storms (around 0.5 N·m⁻²) near the
tropopause over a region with ongoing strong convection.

Convection over the Indonesian Archipelago

CRISTA-2 data over Indonesia are available with particularly high horizontal resolution (100x100 km, instead of typically 200x600 km). Furthermore, two peculiar features were found in this region, namely an intrusion of dry air from the southern subtropics into the humid tropical belt and increased gravity wave activity, assumed to be due to strong convection (Preusse, 2001). Simulations with EURAD confirm this assumption, showing that convection

during CRISTA-2 episode was partly controlled by the intrusion of dry air and partly by the land-sea distribution. Mesoscale vertical fluxes have been derived from the simulations for several parameters.

Examples for heat and momentum are given in Figure 3 exhibiting wavelike structures in the horizontal heat flux distribution (left) and indications of flux bursts (right) which in the case of momentum may penetrate into the lowest stratosphere. A tropical plume (Kuhnel 1990) extending from Indonesia to Australia has also been identified in the southern subtropics.

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Community modelling initiative (COMMIT)

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The past decades have led to tremendous advances in knowledge about the Earth's atmosphere and climate system. Much of the progress made has been due to the development of highly sophisticated and comprehensive numerical models describing dynamical and transport processes as well as the chemistry in the atmosphere. Such models are extremely demanding in terms of computing resources and they require great programming skills in order to ensure efficient use of resources and numerical stability under different environments. Historically, general circulation models (GCM, aka. climate models) and atmospheric chemistry transport models (CTMs) have been developed independently at several institutions in Germany, Europe, and elsewhere. While GCMs generally have a very poor representation of chemical processes (e.g. uniform fields for CO_2 and methane and constant climatologies for ozone), CTMs on the other hand, are generally less sophisticated in terms of the parameterisations of physical processes, and they do not allow for the investigation of feedbacks from chemical changes onto the atmospheric circulation. Because of the tight coupling of these processes in the real world, a new strategy has been formulated recently to develop numerical models, which include both, the circulation and the chemistry in the atmosphere in one consistent and coherent framework. In a wider perspective, the ultimate goal is to couple these models to other climate system compartments, such as the ocean (including its biogeochemistry), the terrestrial biosphere and hydrological cycle, and the cryosphere. International programmes, including the World Climate Research Programme (WCRP) and the International Geosphere-Biosphere Program (IGBP), coordinate such "Earth System Modelling" (ESM) initiatives through their WFCM and GAIM projects, respectively.

In order to make efficient use of the limited resources for Earth System model development, the COMMIT project established a collaborative effort between various German institutions for designing and implementing a joint global chemistry climate model. COMMIT therefore developed several general model interface structures (Figure 1) and tested these in different model environments and on different computer platforms. Efficient and user-friendly modules were developed, which allow for the defini-
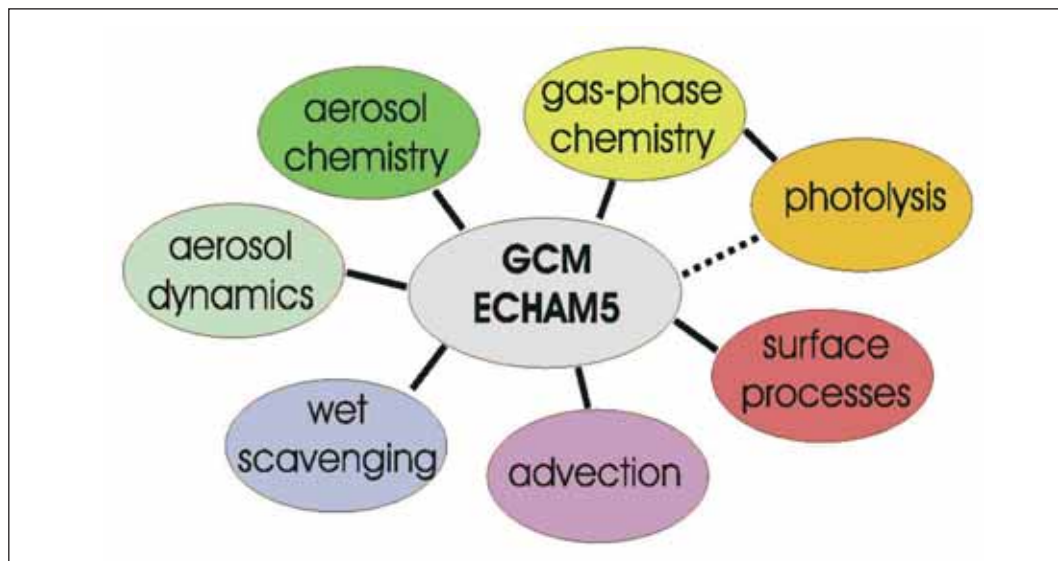
Figure 1: Schematic Overview of atmospheric chemistry related model components and their modular coupling to the parent general circulation model.

tion of transported chemical species ("tracers") and flexible output of diagnostic fields. The results from COMMIT have also influenced the European PRISM initiative and now form the basis for further community model development in the COSMOS project. Several different chemistry climate models, which use the COMMIT structures, are now under validation:

+ **MOZECH: a tropospheric chemistry climate model including 65 gas-phase species related to tropospheric ozone formation (RAST et al., 2004),**
+ **HAM: a tropospheric multi-modal aerosol model with explicit formulation of sulphate, organic carbon, black carbon, sea salt and dust particles (STIER et al., 2004),**
+ **HAMMONIA: a middle and upper atmosphere model with an upper ceiling at 250 km for studies of the influence of solar variability on atmospheric dynamics and chemistry (SCHMIDT et al., 2004).**

The COMMIT interface structures are also employed in a carbon cycle model under development at the MPI-M.

Further COMMIT activities include the development of postprocessing and visualisation tools. An interactive viewing system based on the commercially available IDL software was developed jointly at NCAR and MPI-M. The viewer contains options for plotting maps in various projections, vertical cross sections, line graphs, and animated sequences of these. Graphical output can be stored in image files of different format (e.g. ps, eps, png, jpg), images can be zoomed, and several operations can be performed on the data (e.g. differences between data sets or variables within a data set, or chemistry specific operations such as conversion from mixing ratio to concentrations). GEOV is freely available at the NCAR web site or from the Max Planck Institute for Meteorology.

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INVERT: Inversion of vertically resolved trace gas profiles from ERS-2-GOME total columns using the NCAR-ROSE chemical transport model

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The availability of longer term time series of consistent vertically resolved trace gases from ERS-2-GOME will considerably support the research work that is needed for environmental policies. It is especially valuable to improve atmospheric and climate modeling (initialization and validation) and for the evaluation of chemical transport modeling. ERS-2-GOME is in operation since April, 1995. Mainly, information on total column O_3 and NO_2 are derived on a routine, daily, basis. While derivation of vertically resolved trace gas profiles from the GOME spectra needs extensive computing time, studies using the chemistry transport model NCAR-ROSE (ROSE AND BRASSEUR, 1989) showed that it is possible to derive vertically resolved stratospheric ozone profiles with considerable accuracy in near-real time from GOME total column ozone. ROSE covers the stratosphere between 10 and 60 km altitude. It is an Eulerian model with full chemistry driven by available meteorological analysis of wind and temperature fields.

The aim of INVERT is to reprocess all historical GOME data using assimilation into ROSE in order to retrieve global 3D distributions of O_3, and, when available, also of BrO and N_2O. Therefore ROSE has been modified in terms of enhancing its resolution, updating its chemical scheme, and implementation of an optimized sequential assimilation scheme. INVERT is conducted by the German Remote Sensing Data Center at DLR (DFD) in cooperation with the Max Planck Institute for Meteorology (MPI), the Rhenish Institute for Environmental Research at the University of Cologne (RIU) and the National Center for Atmospheric Research (NCAR).

Development of a statistical profile correction scheme

The work package of the Rhenish Institute for Environmental Research at the University of Cologne aimed at the inference of improved stratospheric

ozone profiles based on total column data and additional auxiliary data. Available auxiliary data are the potential vorticity (PV) diagnostics obtained from the UK Met Office at the satellite locations. In addition, the ROSE model ozone data should be integrated in the estimation as a further auxiliary data source. The algorithmic development was performed with ozone. Different regression models were defined and applied to SAGE ozone profiles as training data sets. Among others a basic univariate regression method was implemented, establishing a linear relation between PV and ozone on an individual height level. Two statistical algorithms were then evaluated: 1. the stochastic inverse exploiting total column and PV information, and 2. the maximum a posteriori

estimator, which is able to additionally include ROSE ozone concentrations as validated against SAGE data. Both algorithms fully met expectations. In summary, significant model error reductions could be proved in relation to ROSE, which partly went beyond 30 %. With a negligible amount of compute efforts, this is a considerable benefit for operational runs. The algorithms work well throughout the stratosphere with the foreseen exception of some height levels (see figure 1): in the upper stratosphere where ozone concentration ceases and at about 30 hPa altitude, where the PV-ozone correlation changes sign leaving a low signal to noise ratio.

Improvements of the chemical-transport model ROSE

The new version of the chemistry transport model ROSE contains numerous improvements which were carried out at the Max-Planck Institute for Meteorology in cooperation with NCAR. The improved chemistry module includes an up-to-date heterogeneous chemistry scheme. Instead of the former semi-Lagrange (s-L) transport scheme a Lin-Rood (L-R) scheme can now be used alternatively. To avoid the accumulation of ozone in the lower stratosphere the model area was enlarged into the troposphere. Comparisons with observations show that in this way systematic errors in low altitudes could be considerably reduced. The antarctic ozone destruction, however, is still underestimated. Multi-year simulations with tracers and different transport schemes show deficiencies especially in the representation of the Brewer-Dobson circulation. Generally, meridional and vertical gradients of the long-lived compounds are underestimated. This points to an overestimation of vertical diffusion in the L-R transport scheme while the s-L scheme shows a too week decent of air in the polar vortex. Both effects lead to too high organic chlorine and other source gas concentrations (N_2O, CH_4) within the polar region. In the course of polar spring ozone destruction by chlorine chemistry is

Figure 1: Normalized root mean squared error scores of corrected ozone profile estimates (right panels, solid blue) for latitude belt 3S-40S. ROSE ozone skill for comparison (red dashed). Episode Jan-Mar 1996. Values smaller 1 indicate improvement over statistical mean of training data set.

therefore significantly underestimated. It could be shown that this problem can be partly compensated by an artificial transformation of the source gasses into active bromine and chlorine compounds.

Assimilation results

A sequential assimilation scheme was implemented at DLR-DFD based on the ROSE model and the statistical profile correction scheme. All observations within a 1h model time step are taken into account. For the weighting between model values and observations a fixed relative model error of 18 % is used. The corresponding GOME error value is set to 4 %. The model's total ozone first-guess is corrected by observations using optimal interpolation. Ozone is then vertically redistributed in compliance with the first-guess profile. A complete

reanalysis of ERS-2-GOME observations from 1996 until 2003 has thus been performed (Bittner and Baier, 2004). First results show that the derived data set is very valuable for scientific studies on stratospheric ozone variability (Bittner et al., 2004). The final release will be published online (http://wdc.dlr.de). It is expected to have residual mean errors in ozone mixing ratio well below 15% for stratospheric layers. As an example, figure 2 below shows zonal mean ozone for the 56 hPa model level. As an additional product the tropospheric NO_2 column can be derived by subtracting the model's stratospheric background column from GOME observations. This method was proved feasible for a November 2001 episode (Thomas et al., 2003). An extended data set including all historic GOME observations is in preparation.

Figure 2: Zonal mean ozone mixing ratios at 56 hPa derived from ERS-2-GOME ozone column observations by sequential assimilation into the ROSE chemistry transport model.
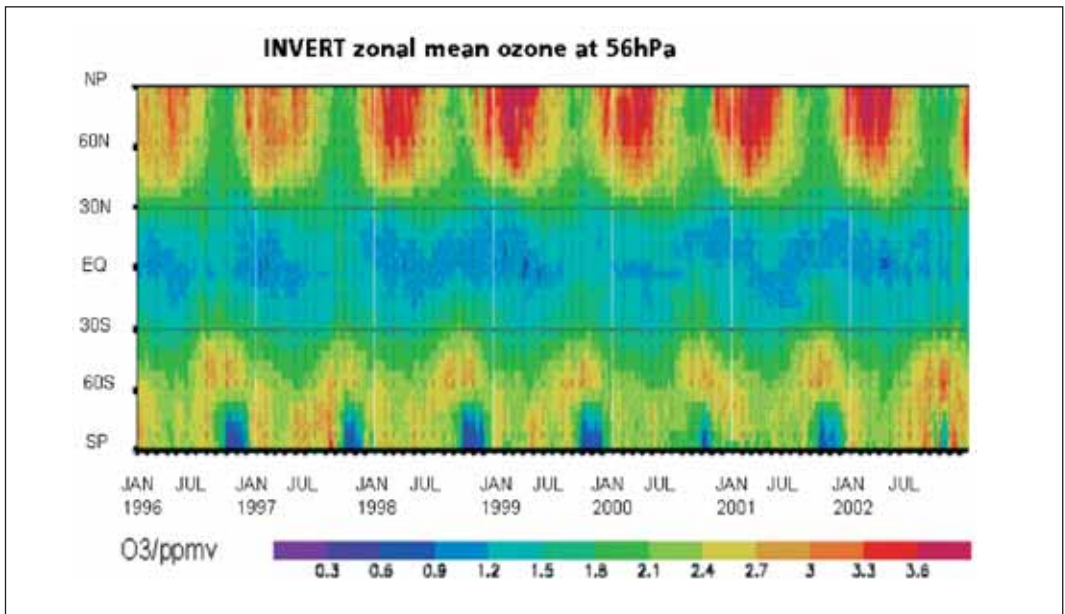
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SACADA, a new chemical data assimilation system for earth observation

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Objective

Satellite data, like many other measurement systems, provide observations scattered in time and space. SACADA (Synoptic Analyses of Chemical constituents by Advanced Data Assimilation) is designed to produce synoptic fields of constituents, thereby providing for: (i) chemical consistency, (ii) extension to the estimation of not-observed species, which are chemically coupled to observed species, (iii) cross validation of satellite data, (iv) harnessing additional parallel compute power without methodological revision of the assimilation algorithm, and (v) efficiency and stable operation of the assimilation system for routine use at DLR-DFD in near real time. Presently, ENVISAT mounted MIPAS and SCIAMACHY sensors provide observational input data.

The Assimilation System Configuration

1 Data Assimilation Method

The overall method is the four-dimensional variational data assimilation (4D-var), which is an advanced technique allowing theoretically to provide a Best Linear Unbiased Estimator (BLUE) of a dynamical system's state. This algorithm is conceptually able to satisfy the above mentioned criteria (i) to (iii) by minimising a scalar cost function which meas-

ures the discrepancy between the observation **y** and the model state at the observation location $H(\mathbf{x}(t))$ and the a priori modelled initial state $\mathbf{x}_b$.

$$J = 1/2(x_b - x_0)^T B^{-1}(x_b - x_0) + 1/2(y - H(x_0))^T R^{-1}(y - H(x_0))$$

B and **R** are the error covariance matrices of the background (= a priori) state and the observations, respectively. Key efforts implied by this method are devoted to the development of the adjoint code of the stratospheric chemistry-transport and the reduction of the compute demands, to comply with requirements (iv) and (v).

For anisotropic and inhomogeneous parameter dependent background error covariance modelling in SACADA a diffusion approach (Weaver and Courtier, 2001) has been selected and a new pseudo-diffusion method has been developed at SCAI. The method ensures positive definiteness and provides a simple way for the square root preconditioning. For this purpose a new fast, locally one dimensional, implicit, and unconditionally stable algorithm is designed for the GME mesh.

The parameterization of the background error correlation matrix requires statistical key parameters to be estimated. The basic idea is to derive characteristic correlation-length-scales for geodetic, potential vorticity (PV) and potential temperature (ϑ) coordinates plus RMS-errors for all observed species from routine simulations of the DLR-ROSE chemistry-transport model (Bittner and Baier, 2004; Thomas et al., 2003; Brasseur and Rose, 1989).

2 The SACADA General Circulation Chemistry Model

The newly developed semi-Lagrangian SACADA GCCM and its adjoint applies the Hendricks et al. (2001) stratospheric chemistry mechanism, however, with a 2nd order Rosenbrock solver. It is coupled online with the global meteorological forecast model GME of the German Weather Service, also including its icosahedral grid structure. This unique feature in chemical data assimilation ensures full dynamic consistency with special benefits for vertical transport. The icosahedral grid structure of GME is adopted for computational efficiency, allowing for a nearly isotropic resolution of 220-260 km (~2°15' ~T80, ~ ni=32). In the vertical direction 41 levels are introduced with 2 km spacing at stratospheric height levels.

In order to further reduce computing time, the GME time step is doubled in the SACADA part of the code. For semi-Lagrange trajectory computation a second order Runge-Kutta time stepping algorithm is implemented now. The vertical advection scheme of GME is also modified to ensure unconditional stability. The numerical design items are implemented by SCAI, while the adjoint modules and the composition of the assimilation system is a RIU contribution. Assimilation of 24 hours data requires about 17 hours real time on a PC cluster with 6 nodes.

3 Satellite Observations

3.1 MIPAS IMK retrievals

IMK has contributed vertical profiles of temperatures and trace gas abundances from MIPAS/ENVISAT to the SACADA Project. In addition to the standard ESA retrievals, the following species are retrieved: NO, N_2O_5, $ClONO_2$, ClO, CO, CFC-11, CFC-12, HCFC-22, SF_6, C_2H_6 und HNO_4 (Höpfner et al., 2004; Glatthor et al., 2004(a,b); Tsidu et al., 2004; Funke et al., 2004; von Clarmann et al., 2003). Figure 1 shows retrieved ClO and $ClONO_2$ mixing ratios as an example. Beyond this, for IMK data products an extensive data characterization with respect to estimated retrieval errors and averaging kernel matrices has been made available, and these products are subject to a more rigorous quality control compared to ESA retrievals.

3.2 SCIAMACHY IFE Occultation retrievals

The main objective and task of the contribution of the Institute of Remote Sensing (IFE) to SACADA is to verify and validate the chemical data assimilation
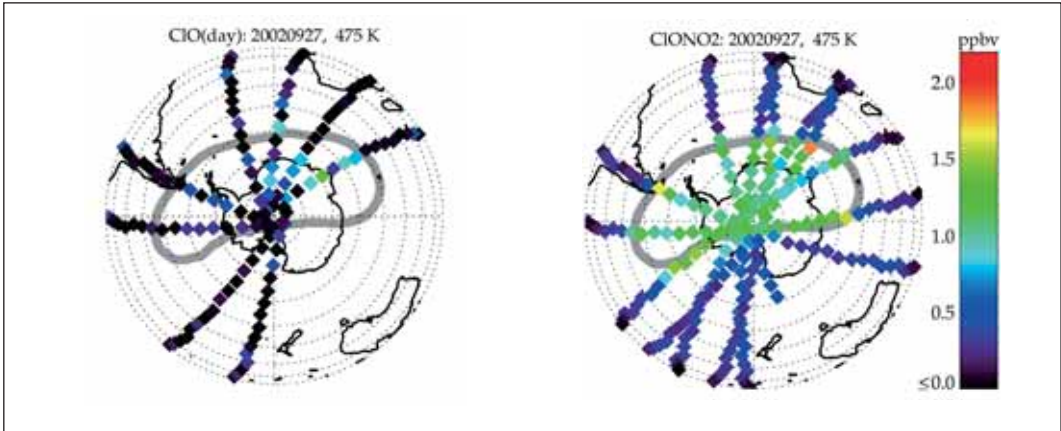
Figure 1: MIPAS/Envisat IMK-retrievals of ClO and ClONO$_2$ before the Antarctic stratospheric major warming at 475 K on 27/09/2002.

system by comparing the results of the data assimilation system with independently generated vertical trace gas profiles from SCIAMACHY solar occultation measurements.

Within this project a "standard retrieval occultation toolbox" for O$_3$ and NO$_2$ vertical profiles was adapted to the use of real data and to cope with relevant features like pointing errors, solar Fraunhofer correction etc.. The retrieval algorithm is based on the optimal estimation method applied to the solar transmission spectra and O$_3$ as well as NO$_2$ are derived simultaneously within one retrieval run. Standard retrievals have successfully been applied to a large amount of data generating scientific data

Figure 2: NO$_2$ concentration profile from SCIAMACHY solar occultation measurements: Shown is the comparison of with SAGE II data demonstrating the overall good quality of the data.
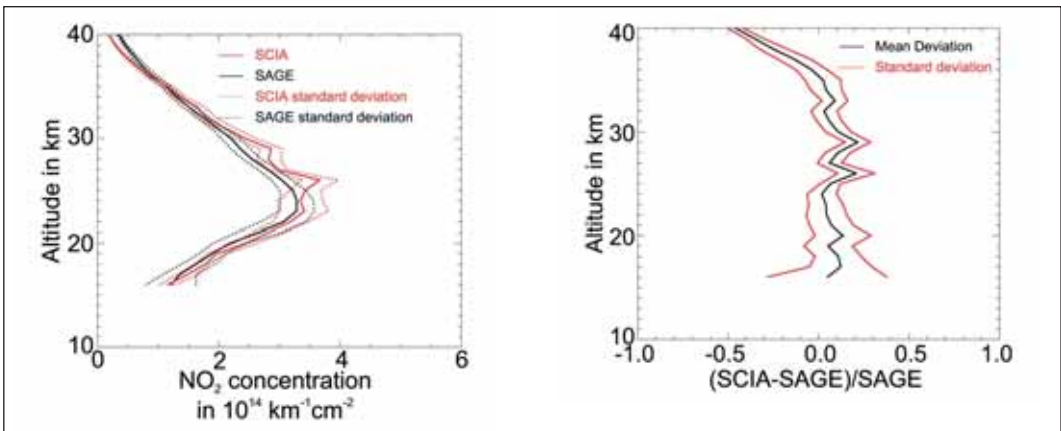
products for validation and case studies for the period September to October 2002 (SH major warming) and October to November 2003. O_3, NO_2 and BrO vertical profiles from SCIAMACHY limb measurements were also made available to the SACADA assimilation project.

The height range considered to yield a good information content from the measurements is between 15 and 40 km. Figure 2 shows first validation studies for NO_2 retrieval results in comparison to SAGE II data. Similar intercomparisons were performed for O_3. The overall agreement is already quite

Figure 3: Assimilation result versus first guess. Profiles of volume mixing ratios at 33°W, 51°N for October, 30th, 2003 and corresponding observations. The blue line is the analysis model state while the black dotted line gives the first guess.
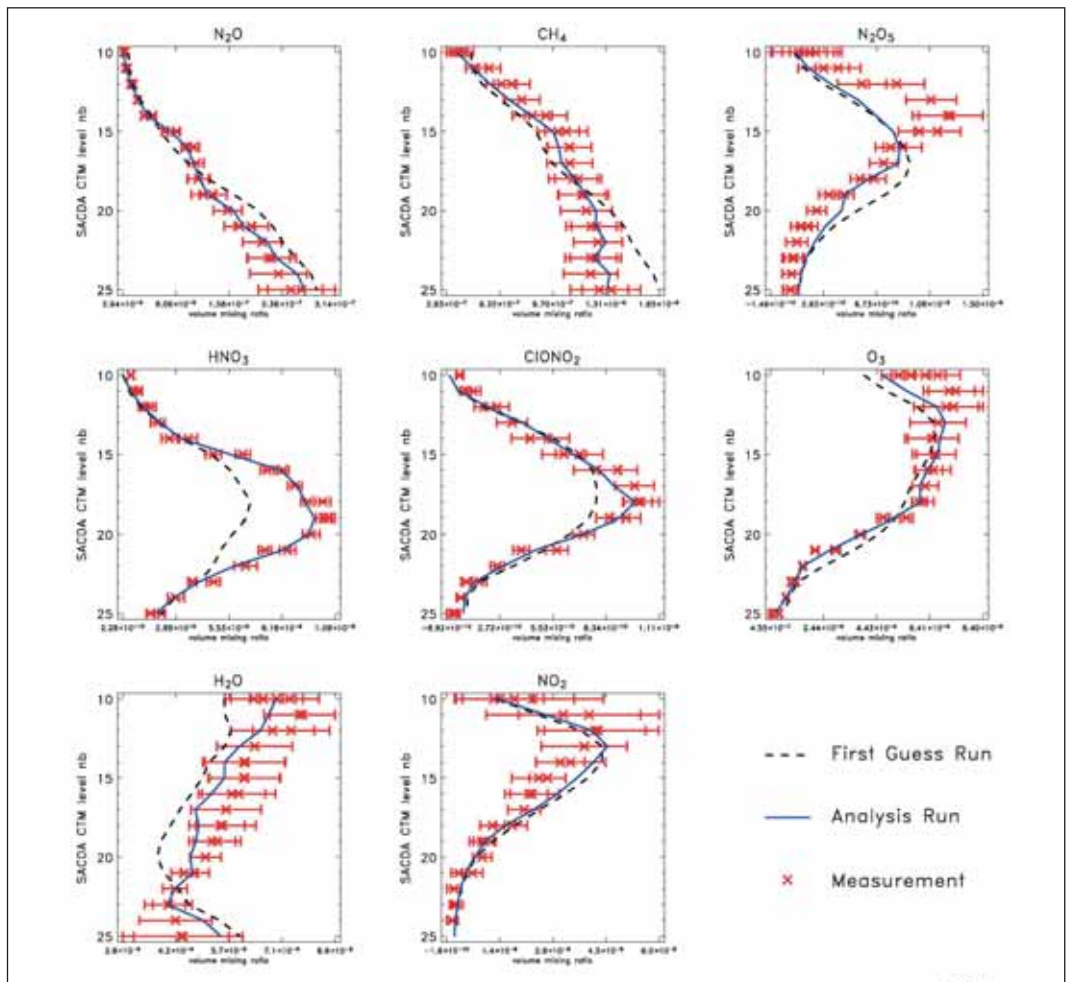
promising. Deviations are in the order of 10% in the case of ozone and about 15% for NO_2.

3.3 CRISTA and MIPAS ESA retrievals

For validation of the SACADA data assimilation system the University of Wuppertal provided height profiles with error-estimates of chemical constituents and temperature measurements, observed during two Space Shuttle missions of the Cryogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA) experiment (November 1994 and August 1997). The data sets have been successfully used at the University of Cologne for test runs of a 4D-VAR assimilation system based on the COMMA model.

DLR-DFD provides data from ENVISAT (SCIA-MACHY, MIPAS, GOMOS), meteorological background fields (UKMO, ECMWF) as well as further satellite and ground based data (MLS, HALOE, SAGE etc.) needed for validation and operation.

Figure 4: Assimilation for November, 4th, 2003, after daily application of the assimilation system for the preceeding week. Given are the results for ozone at the 7.6 hPa pressure level (~33 km). The available and assimilated ozone observations from MIPAS-IMK are shown in the upper panel. The control run without assimilation of this day is shown in the upper right panel. The final assimilation result for 12:00 UTC is displayed in the bottom right panel. Values are in volume mixing ratios.

Results

1 Assimilation

As a preparatory exercise, the ability of the 4D-var method was tested, to which extend standard ENVISAT retrievals (O_3, H_2O, N_2O, NO_2, CH_4, HNO_3) or special retrievals of MIPAS (N_2O_5, NO, $ClONO_2$, CFC-11) suffice to analyse non-observed species (observability tests). The standard data set (6 species) alone was already able to reduce estimation errors of another 18 unobserved non-CFC species.

The test suit to assess the skill of the SACADA system comprises three 6 weeks episodes, all of which are initialised with the SOCRATES 2D chemistry model, that is, with zonally symmetric constituent distributions. In the presented case, this has been accomplished for October, 29th, 2003, with consecutive daily variational assimilation until analysis November, 4th. At this early stage, covariances of the background and representativity error are based on first estimates. The assimilation procedure demonstrates the ability to reduce the model - observation discrepancy as weighted by the cost function by about one order of magnitude in this time span. Normalised with respect to the number of available observations, there is a decrease from χ^2=11.5 down to χ^2=1.5 on Nov. 4th, one week after continuous assimilation, with an ideal value of 1/2 with perfectly estimated covariances. Figure 3 presents an example of profiles of eight species observed by MIPAS as retrieved by IMK.

Figure 4 demonstrates MIPAS retrieved ozone assimilation at 7.6 hPa for November, 4th. While the a priori model state (=control run, not resting on assimilation) was not too far from observations, the observed low ozone values in the arctic region is only reflected in the assimilation result.

2 Cross validation

Cross validation of MIPAS-IMK data and SCIAMACHY occultation measurements is so far focussed on
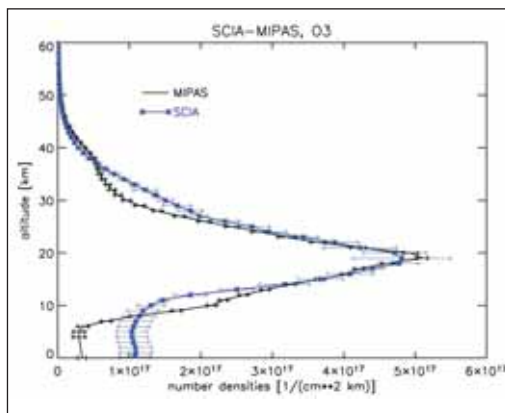
Figure 5: Comparison of ozone retrievals between MIPAS (black) and SCIAMACHY occultation mode (blue) at 68°N with 3h50min temporal difference and 550 km distance at October, 22nd, 2003.

ozone und NO_2. Ozone data compare reasonably well, as shown in Figure 5. Differences are about 10 percent in the altitude region of the ozone density maximum. For NO_2, the overlap of the sensitive altitude region of both instruments is small.

3 Flux estimates

A diagnosis and visualization toolkit for atmospheric transport processes has been developed and tested based on assimilated CRISTA observations at BUGHW. The toolkit allow for detailed analyses based on the Transformed Eulerian Mean formalism of Andrews and McIntyre (see Fig. 6). In particular, effects of the residual mean circulation, large-scale eddy fluxes, and photochemical production and loss on the temporal development of the background atmosphere can be investigated. A case study has been carried out with emphasis on the exchange of tropical and extra-tropical air by planetary wave mixing during the CRISTA-2 mission (RIESE et al., 2002). Within a second case study, CFC-11 observations of the CRISTA-1 mission have been analyzed with respect to trace gas transport in the upper troposphere and lower stratosphere region (HOFFMANN AND RIESE, 2004).
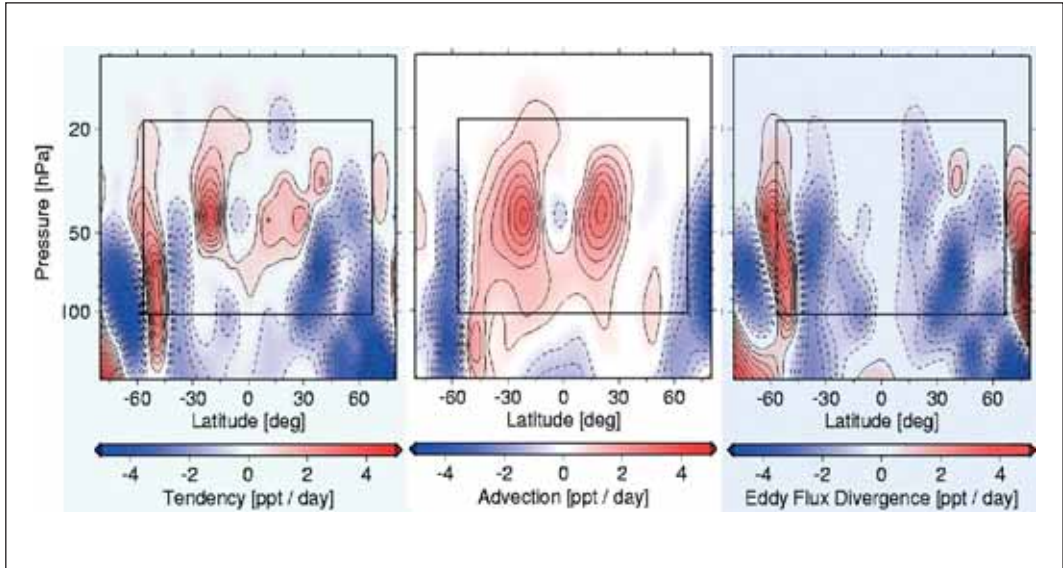
Figure 6: Transport study based on assimilated CFC-11 measurements of the CRISTA-1 mission (November 1994). The latitude-height cross-sections show the calculated tendency of the background atmosphere over a time period of four days (left) and the contributions due to the residual mean circulation (middle) and large-scale eddy-transport (right) respectively.

Conclusions

The 4D-var method with the icosahedral grid model formulation on parallel platforms proves both powerful and efficient in providing synoptic maps of stratospheric constituents. For the envisaged routine application, the conceptual ease of ingesting heterogeneous data is demonstrated and the ability to exploit the expected plethora of future remote sensing data in an optimal way can now be taken advantage of. Scheduled routine operations will make all data products available via DLR-DFD's WDC internet portal http://wdc.dlr.de .

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Tracer analyses by mesoscale field experiments with 4D-var data assimilation

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Objectives

Cooperating with AFO2000 campaign projects, SATEC4D aims to satisfy the frequently enunciated need to combine data with models on the highest possible sophistication level. Advanced space-time data assimilation methodology is the optimal choice. In practice, the four-dimensional variational (4D-var) data assimilation algorithm, which implies the development of the adjoint models, is an ambitious, yet computationally feasible option. It satisfies the optimality-condition of a Best Linear Unbiased Estimator

(BLUE), while a consistent evolution of the chemical state of the observed and modelled system is attained. The underlying chemistry transport model (CTM) is the EURAD (EURopean Air pollution Dispersion model), a state of the art continental to local scale air quality model.

Scheduling roughly one campaign assimilation per year, the following projects are involved in the project: BERLIOZ; CONTRACE-I; SPURT; VERTIKO, and ReHaTrop, with measurements of the first three now being analysed.

Figure 1: Model domains for BERLIOZ of mother grid CG (left, 54 km resolution) and nested grids N1, N2 and N3 (right, 2 km resolution) and measurement locations.
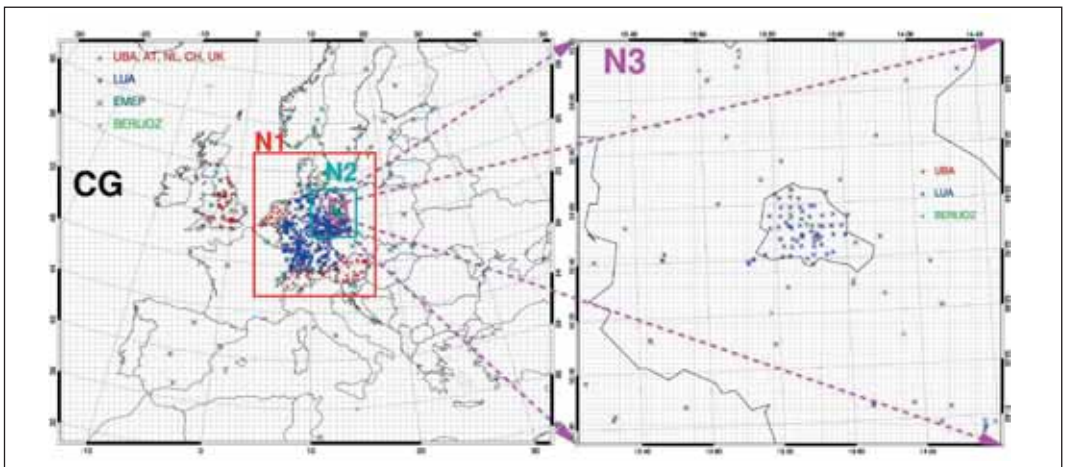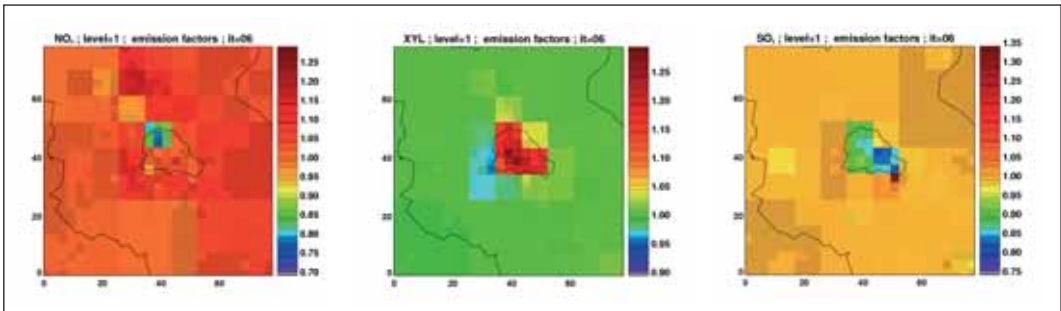
Figure 2: Optimised emission factors for nest 3 for NO$_2$, xylene, and SO$_2$ (from left to right).

Survey of assimilation system achievements

The first developed chemistry 4d-var data assimilation system in the troposphere was the starting point for SATEC4D (ELBERN AND SCHMIDT, 2001). During the project time the following principal achievements were attained: Generalisation of the optimisation parameter to emission rates, together with chemical state variables, 4D var multiple nesting with adjoint modelling, ingestion of data stemming from radio sondes and tethered balloons, LIDAR, aircraft, satellite ozone profiles and NO$_2$ tropospheric column retrievals.

Assimilation results

The spatially and timely dense observational data of the BERLIOZ campaign (BERLIn OZone experiment, July and August 1998) has been used for an episodic simulation (July 18-21, 1998) with a nested data assimilation technique. Assimilated species are O$_3$, NO, NO$_2$, CO and SO$_2$ in an assimilation window of 14 hours, from 06 UTC to 20 UTC on July 20th, 1998. The nesting procedure included a coarse grid simulation with horizontal grid size of 54 km and three nested grids with a nesting ratio of three (Figure 1).
As a unique feature, the parallel implementation of the adjoint EURAD-CTM allows to optimise initial values as well as emission rates.

Figure 2, in which emission factors for CO, NO$_2$, and SO2 on nest 3 are shown, demonstrates the mechanism: the different optimisation stages in terms of coarse grid, nests 1, 2 and 3 boxes are obvious. Each nest moderately refines the previous determined scaling factors.

Figure 3 shows time series of four observation locations measuring NO and NO$_2$, respectively. Optimising emission factors in common with initial values here leads to a strongly improved forecast skill

Figure 3: Time series for selected NOx stations (upper panel NO, lower panel NO$_2$) on nest 2. + observations, - - - no assimilation, —— N1 assimilation, —— N2 assimilation. Grey shading: assimilated observations, others forecasted.
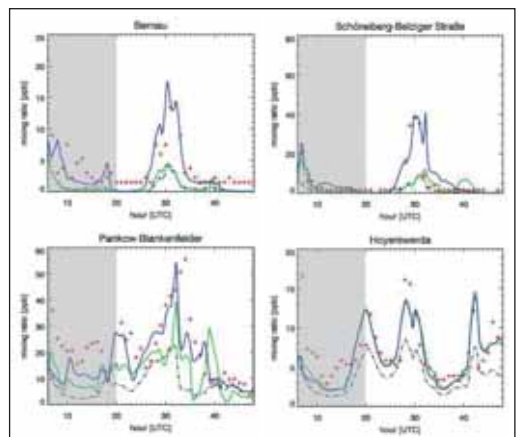
during the second day. Concentration peaks can be reproduced in a fully satisfying way.

The first CONTRACE episode with a special flight on Nov 14th, 2001 was selected for upper tropospheric assimilation, with warm conveyor belt features induced by cyclone dynamics. The vertical grid structure has been refined for that height region, having now 26 layers. Horizontal grid size is 25 km. Assimilated species aloft are O_3, NO, H_2O_2 and CO, while NO_2 and SO_2 at the surface only, Figure 4 shows forecast improvements for the data of the second flight part B during the afternoon of Nov 14th. Initial values have been optimised for mid-

night (00:00 UTC) to ensure a daily consistent chemical model state. Especially ozone and CO show very good enhancements due to assimilation while all assimilated species are improved.

The SPURT campaigns provided observational data from the UT/LS height levels, from which CO and O_3 were assimilated, giving a similar high performance as in the CONTRACE case (not shown). In summary, the chemical 4D-var method and its EURAD implementation proved to be successful in both improving the forecast skill for chemical weather applications and upper tropospheric flight campaign analyses.

Fig. 4: Time series for CONTRACE flight B on Nov 14th, 2001; measurements (••••), first guess (– • – •) and analysis (——).
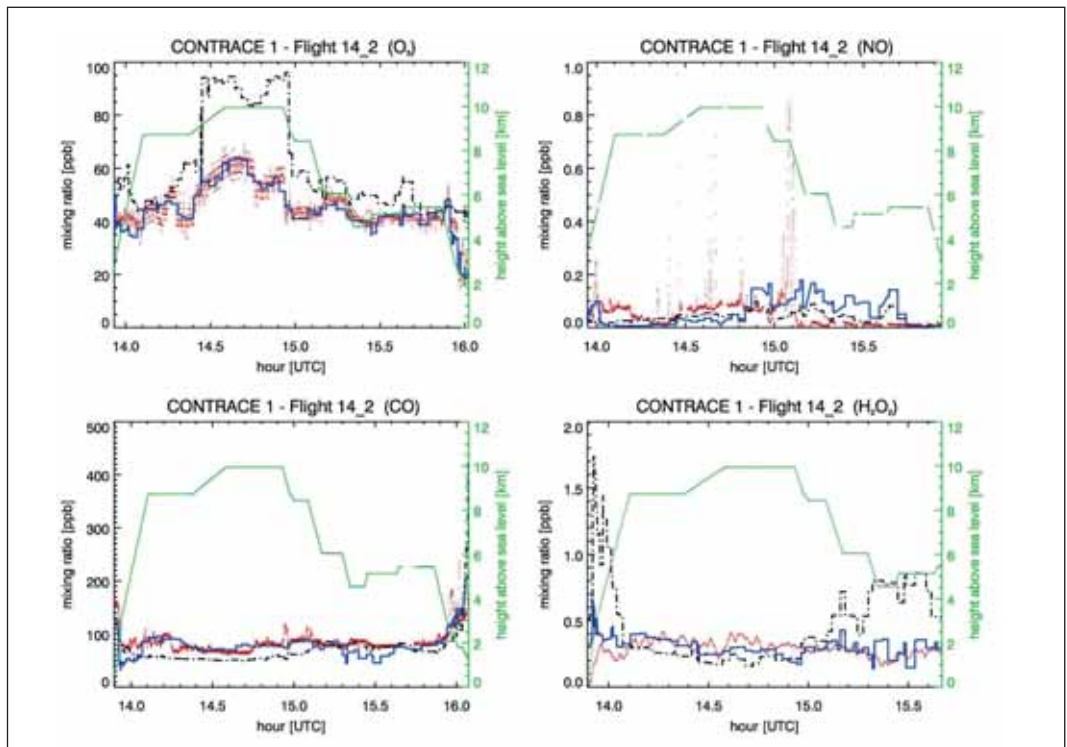
Vertical ozone distributions and stratospheric columns of NO$_2$, OClO, and BrO from GOME and SCIAMACHY nadir satellite data:
Data product optimization and scientific studies of the lower stratospheric dynamics and chemistry (GOMSTRAT)

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Overview

The main goal of this project was to derive high quality trace gas distributions, i.e. ozone profiles and stratospheric NO$_2$, BrO, and OClO columns, from long-term global nadir UV/visible spectral data measured by the GOME and SCIAMACHY satellite (1995-present). These data sets were used to study chemical and dynamical processes in the lower stratosphere, some of which are relevant to possible future climate change.

An improved and accelerated algorithm to retrieve ozone profiles from UV nadir spectra has been introduced. For deriving BrO and NO$_2$ stratospheric columns, a method to separate tropospheric and stratospheric contributions to the measured total column were introduced. The same retrieval methods can be applied to SCIAMACHY nadir observations. Both GOME and SCIAMACHY provide a unique long-term data set spanning more than a decade starting in 1995. They become increasingly important with respect to the question if ozone recovers from a long-term linear decline after stratospheric chlorine loading peaked in the late 90s.

Improved ozone retrieval from GOME

This standard Differential Optical Absorption Spectroscopy (DOAS) approach assumes that the absorber is weak and the atmosphere optically thin. Ozone in the Huggins band, however, shows significant absorption so that this basic assumption is violated (BRAMSTEDT et al. 2003). A more generalized approach, called weighting Function DOAS (WFDOAS) has been introduced for total ozone retrieval (COLDEWEY-EGBERS et al. 2004a,b). Validation with ground-based Brewer and Dobson spectrophotometer data show excellent agreement on global scale (WEBER et al., 2004).

Ozone vertical distributions can be retrieved from GOME UV/VIS spectra by application of the FURM (FUll Retrieval method) algorithm which is based upon an advanced optimal estimation (Tellmann 2004). Several improvements to the GOME retrieval have been introduced to make the long-term dataset from 1995 to 2003 usable for trend studies: first an improved a-priori ozone and temperature profile climatology including covariances were introduced (DARMAWAN, 2002, LAMSAL et al. 2004).

Spectral calibration corrections have been applied to correct for the optical degradation of GOME with time (TELLMAN et al. 2004, TELLMANN 2004). Currently the new ozone algorithms are adapted to SCIA-MACHY to continue the global time series that started in 1995.

Dynamical and chemical control of spring total ozone

Stratospheric ozone concentration is mainly maintained by the balance between photochemical production in the tropics, transport to higher latitudes, stratosphere-troposphere exchange (STE), and photochemical loss. Among these, STE and transport of

ozone are mainly controlled by the wave driven Brewer-Dobson circulation.

Figure 1 shows the compact relationship between inter-annual ozone variability (due to transport) and chlorine activation (chemical ozone loss) to winter planetary wave driving (WEBER et al. 2003). Even the Antarctic ozone hole anomaly in 2002 is not an exception from these relations (SINNHUBER et al. 2003, RICHTER et al., 2004). Any change in tropospheric wave activity that may result from the rise in greenhouse gases or from natural climate variability may influence the stratospheric circulation and hence the trace gas and ozone distribution in the near future.

Figure 1: Correlation of winter/fall total ozone ratio (left) and winter integrated maximum OClO columns (right) as a function of the winter mean eddy heat flux through the 100hPa level and between 40° and 70° latitudes : The amount of chlorine activation (and whence polar ozone loss) in a given polar winter and high latitude ozone levels is mainly determined by the planetary wave driving in each hemispheres. These compact relationships show the strong coupling between chemistry and transport and this has strong implications for the ozone-climate interaction due to potential future changes in planetary wave driving in a future warmer climate (update from WEBER et al., 2003).
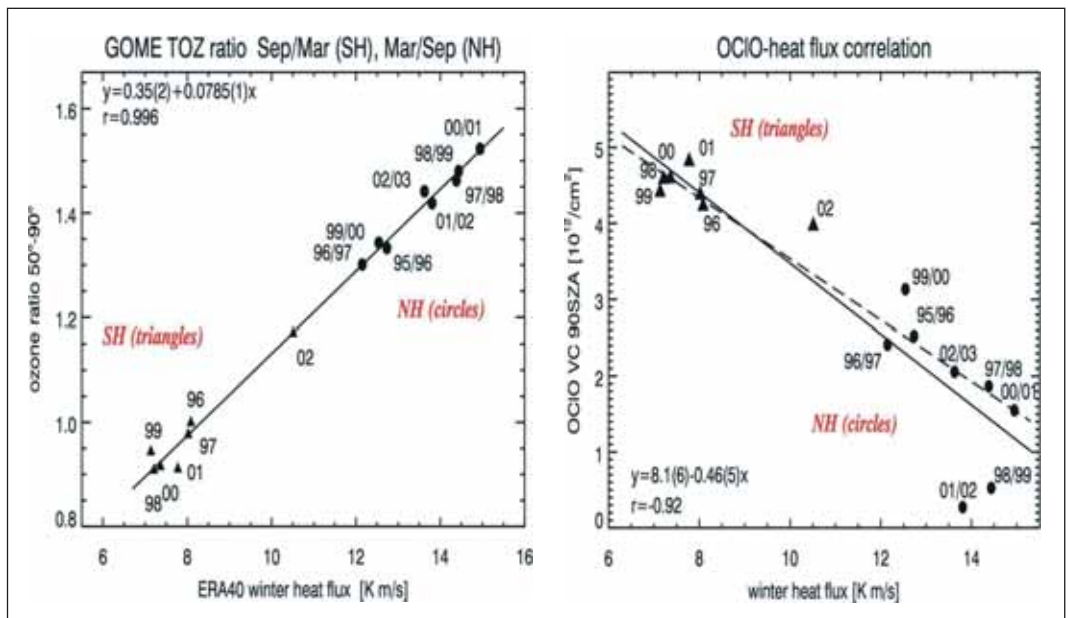
Effects of dynamical and chemical influences on the ozone layer in the changing atmosphere (DYCHO)

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Within DYCHO the understanding of the chemical and dynamical processes that regulate the abundance of ozone at northern high and mid latitudes was improved. Long term changes of these factors were identified and related to changes in climate. The model representation of the sensitivity of Arctic ozone loss on changes in climate in current 3-dimensional models was dramatically improved. This will result in more reliable predictions of the future evolution of the ozone layer over the Arctic.

The high latitude ozone layer is characterized by large interannual variability in spring, caused by the combination of two processes: variable dynamical supply of ozone with the residual circulation and variable chemical loss. We have developed approaches to quantify both contributions from observations. In DYCHO we found that the interannual variability of wintertime chemistry and dynamics both contribute about 100 DU to the total variability of the Arctic ozone column in spring. The importance of both factors for the total ozone variability is similar and a full understanding of both processes is indispensable for a reliable prediction of future ozone levels above the Arctic.

Within the project we have identified the climate parameters that drive the variability of both processes. Figure 1 shows that the interannual variability of chemical loss of ozone is nearly entirely driven by the variability of the average volume (VPSC) of air that has been below the threshold temperature for the existence of polar stratospheric

clouds (PSCs) (REX et al., 2004). Hence, V_{PSC} is the key climate parameter that drives variability of Arctic ozone loss on the year to year time scale. We found that over the past four decades maximum values of V_{PSC} reached during cold winters have increased by a factor of three (REX et al., 2004). Severe Arctic ozone losses during some winters of the nineties were the result of this climate change: If we still had the stratospheric climate of the 1960ies, Arctic ozone loss would not be of much concern nowadays, despite the

Figure 1: Ozone column loss versus V_{PSC} from observations (coloured points, black line) and from the SLIMCAT CTM (crosses; old version: gray, new version: red). The range of V_{PSC} shown here corresponds to about 5-6 K temperature variability. The sensitivity of Arctic ozone loss on changes in stratospheric temperatures is given by the slopes of fits through the data (black, gray and red lines). The observations suggest about 15 DU additional ozone loss per Kelvin cooling of the stratosphere. The old model only accounts for 5 DU per Kelvin, while the new model represents the slope and the scatter of the data relatively well (REX et al, 2004; CHIPPERFIELD et al. 2004).
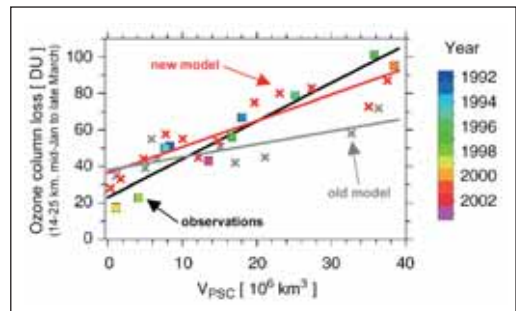
high levels of stratospheric chlorine due to anthropogenic emissions (Rex et al., 2004).

The sensitivity of chemical ozone loss on changes in climate has been quantified in DYCHO based on observational data and current 3d models. The observations show that a 15 DU additional ozone loss can be expected per Kelvin cooling of the Arctic stratosphere. Current model calculations result in only 5 DU additional ozone loss per Kelvin cooling, underestimating the sensitivity of Arctic ozone loss on climate change by a factor of three (slope of grey line in Figure 1; Rex et al., 2004).

We have shown that with the currently accepted set of kinetic data, photolysis cross sections (Sander et al., 2003) and stratospheric bromine levels, observed ozone loss rates during cold Arctic Januaries cannot be explained (Rex et al., 2003). In DYCHO the model representation of measured Arctic ozone loss rates has been largely improved by using information about reaction kinetics and bromine levels extracted from measurements during recent field campaigns in the Arctic (Frieler et al, 2004). We have also shown that coincident measurements of ozone loss rates, ClO and Cl_2O_2 abundances can only be understood with the new model (Frieler et al, 2004). We found that the same model improvements dramatically improve the representation of the climate sensitivity of Arctic ozone loss (slope of red line in Figure 1, Chipperfield et al. 2004), allowing much more reliable predictions of the future evolution of the Arctic ozone layer in a changing climate. The instantaneous dynamical coupling between the troposphere and the stratosphere has been analyzed by relating tropospheric climate indices with the stratospheric ozone distribution (Hak, 2001). We have also shown that on seasonal time scales the variability in the dynamical supply of ozone to mid- and high latitudes is driven by variability in the strength of the residual circulation and hence by the transfer of angular momentum from the troposphere into the stratosphere by vertically propagating waves (Wohltmann et al., 2004). An important result of DYCHO is the development of a new statistical model of total ozone variability that is based on a largely complete set of chemical and dynamical processes. Proxy parameters were developed for each process, including the momentum flux from the troposphere into the stratosphere, short term horizontal and vertical advection, PSC-induced ozone loss, mid-latitude ozone loss, etc. The model explains a large fraction of the day to day variability of total ozone at individual sites around the globe and allows us to develop a thorough understanding of the mechanism that drive short term and long term changes in total ozone columns in different regions of the world. For example we have shown that the long term negative trend of total ozone at northern mid-latitudes is dominated by in-situ chemical loss of ozone, with dynamical changes playing a significant role mainly in mid- to late winter (Wohltmann et al., 2004).

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Upper tropospheric humidity and ice from meteorological operational sensors (UTH-MOS)

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Introduction

Upper tropospheric humidity (UTH) is a crucial parameter for the atmosphere's energy balance [BUEHLER, VON ENGELN, BROCARD et al., 2004], but difficult to measure [BUEHLER AND COURCOUX, 2003]. The objective of the UTH-MOS project is to develop and interpret an upper tropospheric humidity climatology product from microwave data collected by the polar orbiting operational meteorological sensors of the AMSU-B type. Figure 1 shows an example of AMSU data and the derived UTH. For the correct interpretation of these data, the impact of cirrus clouds on the measurement in the microwave channels has to be well understood and taken into account in the retrieval. An auxiliary objective is to explore the potential of these data for deriving information on cloud ice particles. The research work in UTH-MOS is split between the modelling of the radiative transfer in the presence of cirrus clouds on one hand, and the actual data analysis and atmospheric research applications on the other hand.

The project will run until the end of 2005. Work so far has concentrated on radiative transfer modelling, developing the methods for the data evaluation, understanding the data properties, and validating the data by comparing to other satellite sensors and in-situ measurements.

Radiative transfer model development

The Atmospheric Radiative Transfer Simulator ARTS is an open source radiative transfer (RT) code developed jointly by the Institute of Environmental Physics, Bremen and the Department of Radio and Space Science, Chalmers University of Technology, Gothenburg. The code with documentation is freely available at: http://www.sat.uni-bremen.de/arts/. It can simulate radiances for up, limb, and down looking instruments for a wide frequency range. An overview of the clear-sky version of ARTS is given by BUEHLER, ERIKSSON, KUHN et al. [2004].

Particular attention was paid to ensure that the model correctly represents the absolute value of atmospheric absorption, not just the component that varies quickly with frequency. This requires the addition of absorption continua for water vapour, oxygen, and nitrogen [KUHN et al., 2002].
As part of the UTH-MOS project, ARTS has been extended to include the effect of ice particles in cirrus clouds, which mainly interact with the radiation by scattering. The version with scattering is described by EMDE et al. [2003], EMDE et al. [2004], and SREEREKHA et al. [2002]. The algorithm solving the radiative transfer problem uses a successive order of scattering approach in discrete ordinates. It can handle all four components of the Stokes vector in a 1D, 2D, or 3D spherical atmosphere.
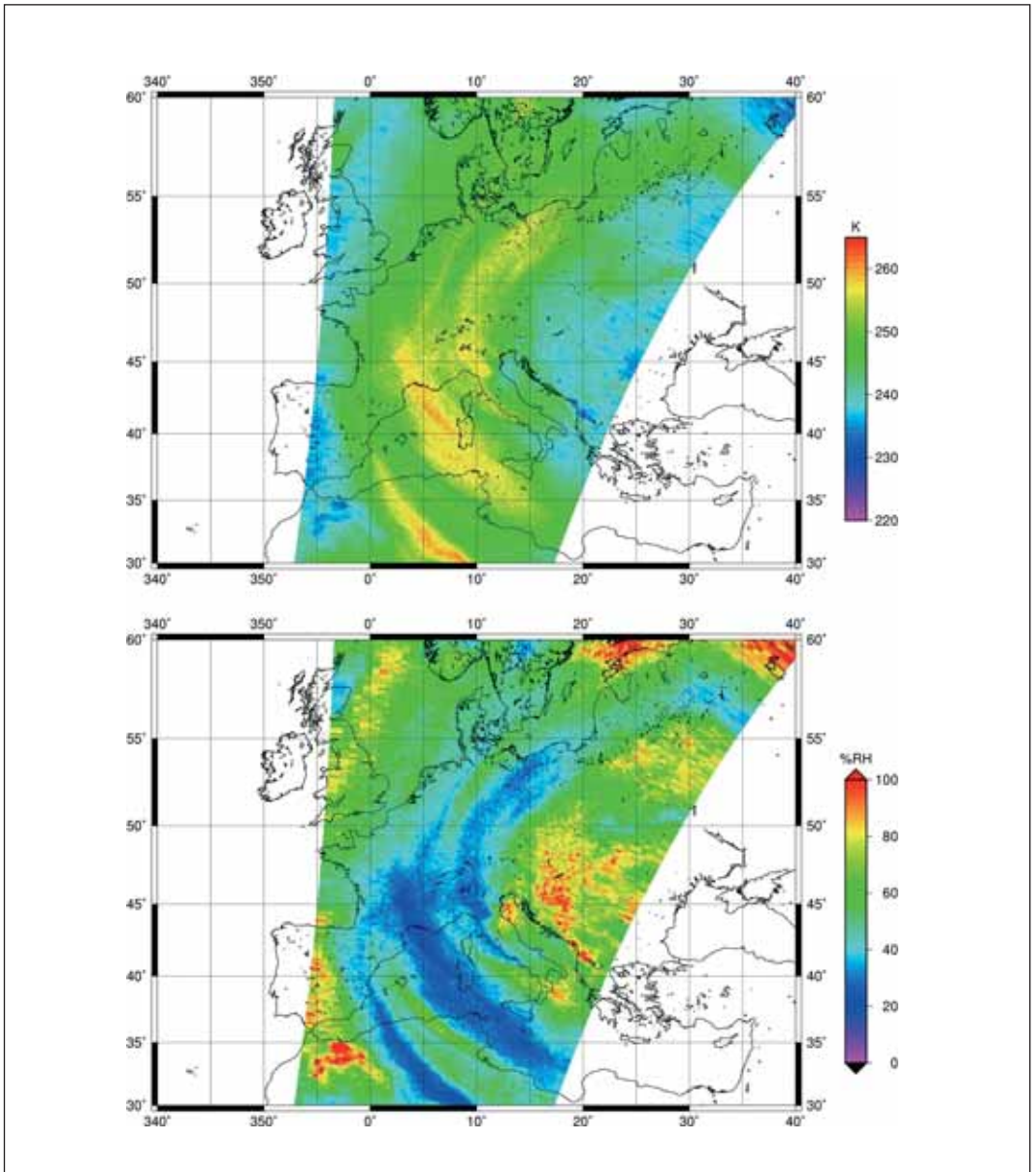
Figure 1: Top: Radiances from channel 18 of the AMSU-B instrument on the NOAA-16 satellite (June 6th, 2004). The unit is brightness temperature (BT) in Kelvin. This channel is most sensitive to the water vapour concentration at altitudes between 500 hPa (mid-latitude) and 350 hPa (tropics).
Bottom: The retrieved upper tropospheric humidity (UTH) product. The unit is relative humidity with respect to ice. On this particular day a dry air extrusion ranged from the subtropics across Europe. (Figure by O. Lemke and V. Oommen.)

Model – data comparison

A necessary condition for accurate retrieval is that the forward model, in this case the RT model ARTS, accurately predicts the measurement from a given atmospheric state, and that all errors in RT model and measurement are well understood. One way to gain confidence in the RT model is to do comparisons to other RT models, as described by Melsheimer et al. [2004]. Another way is to compare directly AMSU measurements to simulated ones, if some in-situ data are available. Such a comparison, based on radiosondes, is described by Buehler, Kuvatov, John et al. [2004]. Figure 2 shows a comparison of different German radiosonde stations performed by the same method. The general level of agreement is very satisfactory, but there are small differences between the different radiosonde stations, pointing to potential problems in the humidity data of some stations. In the context of COST Action 723 these data are currently used for a systematic European radiosonde site intercomparison.

Upper tropospheric humidity retrieval

Although there is a straightforward relationship between atmospheric humidity content and temperature on the one hand, and top-of-the-atmosphere

Figure 2: A scatter plot of AMSU radiance versus predicted radiance, based on radiosonde data. Coloured lines indicate linear fits for the different stations. (Figure by V. Oommen and M. Kuvatov.)

radiances, on the other hand, the task to invert this relationship and obtain humidity values from measured radiances is far from trivial. In principle, two approaches are possible, a variational profile retrieval [Eriksson, Jimenez, and Buehler, 2004], or a simpler statistical regression approach [Buehler and John, 2004; Jimenez et al., 2004].

The second approach is computationally cheap and thus allows the analysis of large amounts of satellite data. It requires a set of atmospheric states and matching radiances, which could in principle be obtained by collecting in-situ measurement and correlated satellite measurements. However, it is in practice difficult to get enough global data this way. Another possibility is to use a collection of atmospheric states covering the atmospheric variability, and use the radiative transfer model to generate matching radiances. This is the approach followed by us.

To give a flavour of the method's capability, one can apply the method to an arbitrary AMSU overpass. Figure 1 shows AMSU radiances and derived UTH for a pass over Europe on June 6, 2004. The top plot shows the original radiances, displayed as brightness temperatures in Kelvin, the bottom plot shows the derived UTH in relative humidity with respect to ice.

Conclusions and Outlook

Microwave data from polar orbiting meteorological sensors are well suited to study upper tropospheric humidity in climate applications. By the developed advanced radiative transfer algorithms, retrieval schemes, and validation efforts, the UTH-MOS project has paved the way for a wider use of these data. The remaining project time until the end of 2005 is planned to be used for applying the developed algorithms to the available data and for climatological studies with the derived datasets.

Below is a list of references. For those papers still in the review process or in press, preprints can be found at http://www.sat.uni-bremen.de/publications/.
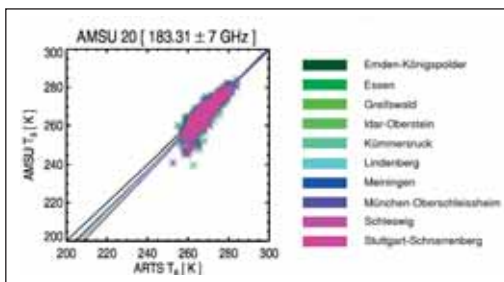
Annex

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List of Project Acronyms

Project Acronym	Theme Group	Project Coordinator	Page
4D-CLOUDS	4	Clemens Simmer	195
ATMOFAST	2.2	Thomas Trickl	145
BEWA2000	1.2	Rainer Steinbrecher	14
CARBAERO	3	Ulrich Pöschl	169
CARIBIC	2.1	Carl A.M. Brenninkmeijer	102
CARLOTTA	2.2	Andreas Stohl	139
CASH-AIC	2.1	Hermann G.J. Smit	111
COMMIT	4	Guy P. Brasseur	205
CONTRACE	2.2	Heidi Huntrieser	150
CRISCA	2.1	Dirk Offermann	114
DYCHO	4	Markus Dietrich Rex	222
ECHO	1.2	Ralf Koppmann	29
EFEU	3	Martin Simmel	172
FEBUKO	3	Hartmut Herrmann	179
GOMSTRAT	4	Mark Weber	220
HOVERT	1.1	Eberhard Reimer	62
IDEC	1.2	Hans-Peter Dorn	44
INVERT	4	Michael Bittner	207
ISOSTRAT	2.1	Thomas Röckmann	98
ISOTROP	4	Franz Rohrer	192
KODYACS	2.1	Martin Dameris	119
LEWIZ	2.1	Dieter Peters	116
MEDEC	2.1	Guy P. Brasseur	124
MODMEP	3	Ralf Wolke	179
NOXTRAM	2.2	Andreas Stohl	141
OPOSSUM	2.1	Franz-Josef Lübken	130
POSTA	3	Ottmar Möhler	164
REHATROP	2.2	Ulrich Platt	155
SACADA	4	Hendrik Elbern	210
SAPHIRE	2.2	Mark G. Lawrence	134
SATEC4D	4	Hendrik Elbern	217
SPURT	2.1	Ulrich Schmidt	107
TOM-DOAS	1.1	Irene Pundt	50
TRACHT	4	Adolf Ebel	202
TRANSVAER	1.2	Ulla Wandinger	55
UNAPRAM	1.1	Nicole Mölders	67
UTH-MOS	4	Stefan Bühler	224
VALCHEM	1.2	Renate Forkel	40
VALIUM	1.1	Michael Schatzmann	57
VERTIKATOR	1.1	Franz Fiedler	91
VERTIKO	1.1	Christian Bernhofer	68