EUROTRAC-2
(A EUREKA Environmental Project)

GLOREAM

Global and Regional Atmospheric Modelling

Annual Report 2000

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International Scientific Secretariat
GSF-Forschungszentrum für Umwelt und Gesundheit GmbH
Munich, Germany
September 2001

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1. Report on the work of the subproject

1.1 Summary

Also in 2000 the subproject GLOREAM was active and lively. The following items can be mentioned:

- A growing number of models which address the modeling of aerosols, including PM10 and PM2.5,
- a model intercomparison between 5 modelling groups performing real-time ozone forecasting,
- continuous progress in the area of data assimilation, inverse modeling, nesting and parallel techniques,
- model validation studies, mostly focused on measuring campaigns like Berlioz and Loop campaigns,
- an increase in the number of global modeling studies,
- a continuation of the cooperation with SATURN and GENEMIS, and an increase in cooperation with TOR-2 and CMD.

The fourth GLOREAM workshop was held in September 2000 in Cottbus, Germany. Nearly all PI's attended the workshop, 29 talks were held.

At the beginning of 2000 GLOREAM had 37 principal investigators. One new project was included during 2000, 2 projects were finalized. So by the end of 2000 GLOREAM had 36 principal investigators.

The funding situation of GLOREAM was reasonable, but mostly exclusively based on national funding, and no funding from the 5th Framework program.

An overview of the models used in GLOREAM is given at the end of this chapter in tabulated form. The table includes the capabilities of the models, and their current and future applications.

The mid term review ranked the scientific quality of GLOREAM as good, the policy relevance as high. It was recommended to put more emphasis on aerosol modeling, more synthesis is needed and the cooperation with other subprojects for validation data should be intensified.

1.2 The aims of the period's work

The general aim of GLOREAM is to investigate by means of advanced and integrated modeling the processes and phenomena which determine the chemical composition of the troposphere over Europe and on a global scale.

The aims for 2000 were:

- Further improvements in aerosol modeling
- Performance of model calculations in relation to the EU-directives
- Improvements in data assimilation
- Intensified interactions with global modeling
- Modeling of real time ozone forecasting

- Further model validation against field campaigns
- Improvement of cooperation with TOR-2 and CMD

In fact, all these items have been addressed in 2000. The closer interaction with global modeling as performed in Hamburg (Mozart model, ECHAM-5) and in Cambridge (TOMCAT model) is very welcome. Also the closer relation with TOR-2 and CMD should be mentioned. Representatives of these 2 subprojects gave presentations at the Cottbus workshop.

GLOREAM is divided in five working groups, the results of these working groups are presented below.

1.3 Model investigation and improvement

1.3.1 Activities during the year

Work has been done with emphasis on

- improvement of input data: landuse data, biogenic and anthropogenic emissions including the development of new tools based on geographic information systems (GIS),
- use of satellite data to improve cloud parameters in CTMs,
- parameterization and investigation of chemical and dynamical processes (e.g. aerosols and clouds),
- investigation of the performance and sensitivity of operationally used air pollution modeling systems,
- long-term simulations including aerosol and photochemical modeling; chemical composition of aerosols,
- development and testing tools for application to EU directives 96/62; 99/30,
- development of advanced numerical schemes for comprehensive air pollution models, e.g. for data assimilation on the basis of adjoint modeling and Kalman filtering.

Incorporation of improved input data, sensitivity studies

Emission data used in the modeling systems participating in GLOREAM are in a permanent process of improvement by intensive interaction with the EUROTRAC subproject GENEMIS. Emission data has been used for source category dependent emission scenarios (Jonson *et al.*: investigation of the impact of emissions due to ships with the EMEP model with regard to the „International Convention for the Prevention of Pollution from Ships“; MARPOL) or modeling of field experiments (e.g. BERLIOZ; Memmesheimer *et al.*). Some specific efforts have been undertaken to generate emission input data for the DMU-ATMI-THOR and REGINA model. Data from EMEP and GENEMIS are combined with local data sets for Denmark (Brandt *et al.*). Emission data for primary TSP, PM10 and PM2.5 has been generated by the TNO for the European scale. This data has been used by several groups within GLOREAM.

Satellite data is used to improve cloud parameters in air quality modeling (Östreich *et al.*, NOAA-AVHRR; Meteosat).

A Geographic Information System (GIS) has been used to prepare land use data and emissions for modeler’s use together with a Relational Databank Management System (RDMBS). The GIS/RDMBS has been used to improve the interface to air pollution models and to investigate the effect of spatial resolution and different numerical methods used to generate land use data for model applications. The uncertainty of biogenic emissions based on landuse data has been investigated. Methods developed are used in the MCCM (Smiatek). The boundary and/or initial values for chemical transport models have been generated by data assimilation (Kalman-Filter; Builtjes *et al*., 4DVar, Elbern) or results from global models (Jonson *et al*.). Data assimilation has also been applied focusing on emission input.

CO_2-emissions based on the EDGAR system (1°x1°) for the further development of the Danish Eulerian Hemispheric Model (DEHM) to handle CO_2 are implemented in hemispheric modeling.

Improvement and investigation of process parameterization

Considerable efforts have been undertaken to include atmospheric particle modeling and multiphase interactions (gas-phase, aerosol-phase and cloud modeling). Highly sophisticated modules have been developed which can be used for air pollution planning e.g. with respect to EU air pollution directive 96/62 and its daughter directive 99/30. The new aerosol modules have been implemented into 3D models and applied within long-term simulations on the time scale of a year (EURAD-MADE-system for 6 months in 1995; Ackermann *et al*.; LOTOS-model for 1994 and parts of 1997 and 1998; Builtjes *et al*.; EUROS-System for 1994; Matthijsen *et al*.). The MADE module allows for the treatment of secondary organic aerosols, sedimentation and cloud-aerosol interactions. The chemical composition of atmospheric particles can be considered for different seasons. Aerosol modeling within EURAD-MADE includes the nesting option and has been applied from the European scale to North-Rhine-Westphalia (Memmesheimer *et al*., interaction with subproject AEROSOL).

Calculations and analysis of dynamical and chemical processes on the basis of episodic model simulations have been done with the EURAD model, in particular for BERLIOZ and the alpine region (VOTALP) (Memmesheimer *et al*.).

The coupling of atmospheric chemistry with a convective boundary layer model is used to investigate the interaction of chemistry and dynamics. It has been applied to field data taken over the Gulf of Mexico (Lüken *et al*.).

Turn over of sea-salt particles and formation of sodium nitrate particles has been included in the EMEP photochemical model (Jonson *et al*.).

Model hierarchy, linking of different scales; operational use

Models in GLOREAM now usually use the nesting technique to consider regional and local scales (Europe -> urban). Application of the nesting technique has been used to simulate episodes where field campaigns have been done (e.g. EURAD with respect to BERLIOZ, PIPAPO and VOTALP). Nesting techniques are able now to handle the modeling of particulate matter in the troposphere and their interactions with clouds. This has been tested for the region of North-Rhine-Westphalia.

Some models have been applied to seasonal/annual time scale or coupled to global modeling systems, e.g. the EMEP Eulerian photochemistry model (Jonson *et al*.). In general, the models are used operationally, e.g. the DMU-ATMI-THOR air pollution forecast system (Brandt), REM3 (Östreich *et al*.) and the model system of the German Weather Service (LM coupled with EURAD-CTM and the emission model ECM; Tilmes/Zimmermann/Jacobsen). The

Danish system DMU-ATMI-THOR also has been coupled to a street canyon model (OSPM). Parts of the prognostic results have been made available to the public using the World Wide Web. A comparison of different models used operationally has been done (Tilmes *et al.*, Brandt *et al.*).

Data assimilation and numerical techniques

Considerable work has been undertaken in the further development of data assimilation techniques including adjoint modeling and Kalman filtering (Builtjes *et al.*; Elbern and Schmitt, 1999). Data assimilation techniques have been used to improve the initial values of models and, with regard to 4Dvar, for the investigation of the sensitivity of air pollution models. Some of these activities are related also to the recently formed EUROTRAC project TROPOSAT.

Several advection schemes have been investigated within the framework of the Bulgarian Air Pollution modeling system with strong emphasis on the application to non-homogeneous grids (TRAP scheme; Syrakov). New advective schemes for horizontal transport have been implemented within the REGINA model (Acurate Space Derivative (ASD), Brandt *et al.*).

1.3.2 Principal results

Incorporation of improved input data, sensitivity studies

Improved input data, in particular emission data from GENEMIS provided by the IER, has been used for the simulation of field experiments and evaluation purposes (e.g. and BERLIOZ, Memmesheimer *et al.*; Nester *et al.*).

The impact of emissions by international shipping has been investigated by the EMEP model (Jonson *et al.*). It could be shown that ship emissions have a marked effect on European pollution levels.

TNO emissions for particulate matter (PM_{10} and $PM_{2.5}$) have been used by several models, in particular for long-term simulations.

Combinations of different emission data sets for Europe (EMEP, GENEMIS) and of local importance (Denmark) have been used within the operationally used DMU-ATMIS-THOR system.

There are still considerable gaps with respect to appropriate emission data for atmospheric particles with respect to chemical composition. Such data is needed to investigate the sources of atmospheric particles and their fate in the atmosphere. It is also important to have such data for air quality planning in Europe, in particular with regard to the recent EU directives.

Improvement and investigation of process parameterization

Improvement of aerosol modeling within regional air pollution models leads to the following result (MADE/EURAD, Ackermann *et al.*): The long-term simulations with EURAD/MADE for 1995 show the importance of secondary particles generated in the atmosphere.

The analysis of different contributions to the mass budget of ozone performed with the EURAD model (Memmesheimer *et al.*) show clearly that the near source regions act chemically as sink for ozone, in particular in larger cities or highly populated areas as parts of

Nordrhein-Westfalen or the Benelux region. This points towards the strong importance of vertical exchange processes and horizontal transport to compensate the losses due to chemical processes in urban areas and to explain the temporal development of near-surface observations. Chemical net production calculated with the model could be shown to be in a good agreement with observations during the BERLIOZ experiment.

Sea salt concentations obtained with the EMEP model are compared to measurements for several sites in Norway. At most sites calculated concentrations are overpredicted but mostly within a factor of 2. Over the European continent the calculations indicate that only a small fraction of the total nitrate is in the form of sodium nitrate in winter. However, in summer a significant fraction of total nitrate is calculated as sodium nitrate.

Model hierarchy, linking of different scales; operational use

Nesting techniques have been used to couple different scales from Europe to local scales. Applications involve the BERLIOZ field campaign to investigate ozone formation in the plume of Berlin and particulate matter in Nordrhein-Westfalen (EURAD, Memmesheimer *et al.*). Typical grid sizes are 1-2 km. Process analysis on the basis of modeling show the impact of dynamical and chemical processes for ozone formation. Ozone formation in urban areas seems to be most effective in an altitude region of several hundred meters.

The EMEP model has been linked to the global scale by the use of boundary values, the Danish model (DMU-ATMI THOR) includes street canyon modeling and might serve as a link to SATURN. Results from the Danish model are available via internet/www on an operational basis (3 days air pollution forecast) from hemispheric scale (REGINA model) down to street canyons (OSPM model)(Brandt *et al.*). The OSPM model is able to handle ten different streets in the centre of Aalborg. Special emphasis is laid on the high resolution modeling of coastal areas to account for land-sea interactions. On the basis of the Danish Eulerian Hemispheric Model (DEHM), which was initially designed for studying the transport of sulfur into the Arctic, further development took place to handle CO_2.

Cloud parameters used to improve CTM-modeling in REM III are also used for climate modeling studies with REMO.

The EMAP model used in Bulgaria (Syrakov) has been used to calculate transboundary transport between Greece and Bulgaria.

Data assimilation and numerical techniques

An episode in August 1997 has been selected to test a 4D variational data assimilation scheme with the aim to optimize the initial concentrations used in CTM simulations (EURAD, Elbern and Schmitt, 1999). It could be shown that the initial values obtained by the data assimilation scheme leads to an improved subsequent forecast. Sensitivity runs have been carried out with different data assimilation windows and radii of influence to find an optimal configuration for the application of the data assimilation scheme. 4D-Var data assimilation has also been used to improve emission data used for modeling.

1.3.3 Main conclusions

Considerable progress has been achieved within 2000 in the handling, quality and uncertainty analysis of input data, development, implementation and investigation of parametrizations for atmospheric models, in the extension of episodic modeling to seasonal or annual applications and the use of complex atmospheric models for operational forecast. Most of the results from operational models are available in the world wide web.

An interface between air pollution modeling systems and biogenic emissions generated within a Geographical Information System has been developed and tested for complex regional model applications. Emission scenarios for different reference years (e.g. 2010) have been developed and are planned to be used with respect to EU directive 96/62 and 99/30.

Process analysis allows for the investigation of the contribution of chemical and dynamical processes to the temporal evolution of atmospheric trace gas fields. It can be used to explore the mechanisms for the exchange of atmospheric constituents between the planetary boundary layer and the free troposphere (e.g. vertical turbulent exchange, role of mountains, cloud transport, frontal systems). This leads to direct links to the subproject TOR-2, task 2.

Aerosols have been included in episodic and long-term regional modeling. This offers the opportunity to study the source contributions to aerosol load in the atmosphere which is an important and valuable extension of pure photochemical modeling - also with respect to the planned EU directives. The models are now able to perform long-term simulations on a seasonal and annual basis with regard to governmental regulations. On the other hand the modeling systems can be used for the planning and analysis of field experiments as planned within the subproject AEROSOL. Composition of atmospheric particles can be calculated on a seasonal or annual basis. This will help to understand the importance of different sources of atmospheric particles.

Numerical techniques (advection schemes, data assimilation) have been improved considerably. In particular, advanced data assimilation techniques of CTMs offers the possiblity to merge observations and model results. The use of satellite data is of specific interest in the application of data assimilation. Data assimilation is also used for critical checks on emission data used in the model.

Operational use of CTMs and long term runs are new fields of application of atmospheric models. This offers new possibilities for science and environmental planning due to the large amount of model results generated. But it also points towards the need that new methods have to be developed to analyse the data and to evaluate the results of long-term runs of models with measurements taken on a routine basis for a seasonal or annual scale. The coupling of street canyon models to regional models,which is an additional step in complexity of air quality models, gives a link to the EUROTRAC subroject SATURN.

1.3.4 Policy relevant results

In general, the considerable development achieved with respect to input data and progress in process modeling (e.g. aerosols) leads to further improvement of complex models as a tool for the planning of air pollution abatement strategies. In particular, the possibility to perform long term runs provides the possibility to estimate AOT40 or AOT60 values as well as the aerosol load of the troposphere and the response on emission reduction scenarios. The recently developed modules for the simulation of atmospheric particulates can be used with respect to the planned EU directives by environmental agencies and industry.

The operational use of the models, including the availability of the results in the world wide web offers the opportunity to the public to get relevant and actual information on air quality over Europe by using the internet.

1.3.4 Aims for the following year

Aims for the future are a further improvement of input data (landuse, plant species categories, emission factors; anthropogenic emissions) and the investigation of the impact of uncertainties in input data on air quality model results. This will be achieved by stronger interaction with GENEMIS. In particular more information is needed with respect to particle emissions (size distribution, composition) with the aim to get more reliable results, in particular for the decision makers.

Chemical process modeling will be continued and supported by sensitivity studies. Interaction with CMD is planned to improve gas phase chemistry as well as heterogeneous chemistry.

Dynamical processes in the planetary boundary layer are planned to be incorporated in a more sophisticated way into the air quality models used in GLOREAM.

Emission data sets that differentiate between source categories will be used to evaluate source type contributions to the tropospheric aerosol loading and to quantify the contributions of primary and secondary particles to this load.

Process analysis and budget studies will be performed to investigate the importance of different terms in the mass continuity equation for the concentration fields over Europe and polluted subregions.

Some models will be extended from regional to local scales and even coupled with street canyon modeling. Interaction with SATURN will be enhanced in the next years with respect to these activities.

Chemistry-transport models will be optimized for operational use to perform trace gas and atmospheric aerosol forecasts. Further, long term runs including emission scenarios for 2010 are planned with respect to EU-directives including atmospheric particles and the nesting option to zoom out certain regions of interest. Models will be used to fill the gaps between the observational sites.

1.4 Global modeling

1.4.1 Activities during the year

During the year 2000 global chemistry models have been improved and evaluated and have been applied in numerous studies.

In the chemical transport model (CTM) MOGUNTIA an algorithm was developed to identify contributions of specific sources or processes. Using this technique allows to split a species into an arbitrary number of tracers – a kind of “numerical isotopes” – maintaining mass conservation. This technique has been used to study the impact of NO_x from lightning on ozone concentrations.

The CTM TOMCAT has been improved by updating the photolysis rates and introducing a new interpolation scheme which reproduces the diurnal cycle of photolysis rates better.

Simulations of ozone concentrations have been performed in order to diagnose the contributions from the stratosphere and from polluted continents to the tropospheric ozone budget. Results of these simulations have been compared to observations from two measurement campaigns, ACTO and EXPORT. Moreover, future simulations based on IPCC emission scenarios have been performed.

The CTM MOZART developed at NCAR is used at MPI Hamburg. The Hamburg version of the chemistry model is driven by ECMWF analysis. Calculated ozone concentrations have been compared to measurements over Europe focusing on mountainous sites.

The general circulation model ECHAM5 was extended by a simple CO-chemistry scheme and applied to perform real-time forecasts during the measurement campaign TRACE-P.

TOMCAT and MOZART both participate in a model intercomparison organised within the framework of the EC project POET.

1.4.2 Principal results

The MOGUNTIA simulation shows maximum lightning and with that NO_x production in low latitudes over the continents with a clear summer maximum. Whereas ozone formed due to NO_x from lightning is much more widespread and covers the globe from 75° N to 75° S. Ozone from lightning undergoes stronger mixing and has a longer atmospheric residence time compared to ozone due to anthropogenic activities.

The TOMCAT IPCC scenario experiment shows strong increases of near-surface ozone concentrations over the industrialised Far East, Europe and North America, for the 2100 emissions.

The origin of CO over Europe was studied for the year 1990 using the ECHAM5 GCM. This study shows not unexpected no influence on near-surface CO from southern hemispheric sources but not expected a relatively strong influence from biomass burning (~10%) and anthropogenic emissions (~5-10%) in South-East Asia. Contributions from these sources peak in spring and are lowest in summer. Anthropogenic sources in North America contribute less than 5% in summer and 5-10% in winter. Biomass burning in boreal forests, in Siberia and Canada, has a minor influence on the near-surface CO concentrations in Europe.

1.4.3 Main conclusions

Progress has been achieved within 2000 to improve the chemistry codes in global transport models with a particular focus on the tropospheric chemistry of ozone.

Global model studies show that local concentrations of species with a life time in the order of weeks like CO and O_3 are to some extent controlled by long-range transport. Bauer could demonstrate in her Ph.D. thesis (see also Bauer and Langmann in this report) that ozone calculations performed with a hierarchy of local (horizontal resolution 4 km) and regional models (20 and 50 km mesh size) during a persistent high pressure episode cannot fully explain observed ozone concentrations at surface and in the free troposphere as well. In particular, in the free troposphere ozone was seriously underestimated. Trajectory analysis has shown that ozone from outside of Europe caused the high concentrations observed. This emphasizes the need to provide regional modellers with global fields of chemical species concentrations to be able to prescribe realistic lateral boundary conditions. Global chemistry models have improved and have reached a degree of complexity which makes output of such models useful for the air pollution modeller community.

1.4.4 Policy relevant results

Numerical models are the only tools to study the effects of emission reduction strategies on the atmospheric composition. The considerable development achieved in modelling chemistry on the global scale allows to use results as input for mesoscale models. It has been shown that more realistic lateral boundary conditions as provided by global model simulations improve considerably mesoscale air pollution simulations.

1.4.5 Aim for the coming year

Improvement of the model components, evaluation and sensitivity studies will still be a main topic in the next year. Efforts are under way to incorporate the chemistry schemes into global general circulation models, in Cambridge and in Hamburg as well. This so-called on-line model versions enable a more realistic treatment of aqueous phase chemistry and wet removal of gases and particles. Moreover, feedback processes between meteorological processes and atmospheric composition can be taken into account. MPI Hamburg will continue to use the on-line model to do real-time forecasts during the measurement campaign MINOS which takes place in the eastern Mediterranean in July and August 2001.

1.5 Model application and assessment studies

1.5.1 Activities during the year

Among all the research groups a major topic was the improvement of initialisation procedures of atmospheric chemical model systems, both meteorological and initial concentration in order to get better results for better support decisions on abatement strategies.

Concerning the Danish Eulerian Model, in one hand (National Environmental Research Institute contribution - NERI, Z. Zlatev) the main activities were concentrated on the 1) improvement of the chemical and physical mechanism used in the model making use of several chemical schemes and reforming the ammonia-ammonium mechanism, 2) improvement on the output results presentation, 3) validation tests over model results, 4) increase the model efficiency on high-speed computers through the incorporation of better and faster numerical algorithms, 5) long term runs for a nine years period, 6) episodic runs for the summer months between 1989-1998 and AOT40 exceedences; and 7) economical evaluations of losses of crops due to high ozone levels. On the other hand (National Environmental Research Institute contribution, J. Christensen) during the year of 2000 the work on coupling a state-of-the-art weather prediction model to the model system. More detailed calculations were possible for wind and precipitation patterns over Europe and Greenland, thanks to the finer spatial resolution. Long-term runs were performed with the weather prediction model and photochemical module for Europe. Concerning the photochemical scheme, wet and dry deposition were introduced into the model and further developments of the mercury scheme implemented.

The Institute of Meteorology of Belgrade, University of Belgrade - IMUB (conducted by Lazar Lazic) continued its work on improving the Eta Model mostly on making suitable the initialisation procedures for air pollutants long-range transport simulations, mostly on model trajectory calculations. The model system was applied to air pollution problems originated by war bombing over chemistry industry, oil refinery and fuel storage in Serbia.

VITO (L. Delobbe and C. Mensink), have conducted some developments on the EUROS model, namely on the calculation of the horizontal transport and determination of mixing height (through the calculations of friction velocity, Monin-Obukov length and surface sensible heat flux; Richardson number method applied to the ECMWF vertical profiles and LIDAR measurements). In this sense, the model has been subjected to validation of the mixing height parameterisations through comparison with observations and a multi-layer structure for the representation of the horizontal transport was implemented as well as the introduction of a spatially variable mixing height (in collaboration with RIVM, Netherlands). The Faculté Polytechnique de Mons has worked out a user-friendly interface and the preliminary studies for the development of an impact (on public health and vegetation) module were also carried out.

The Technical University of Madrid (R. San José) has contributed with a new cascade of models for air pollutant concentration prognosis. The stressed items were the impact of initial concentration (zero and after five days run) of the mesoscale air quality models and the integration of global through urban scale in future air quality modelling systems. In this sense, they have been running operationally the RSM global model, and the MM5 both coupled with the CAMx chemical model.

The mesoscale systems SAIMM-UAM-V and SAQM were applied over Switzerland and San Joaquín Valley of Central California, respectively, in order to analyse the threshold values on NO_x-VOC sensitivity ozone formation. This work has been performed by the Laboratory of Atmospheric Chemistry in the Paul Scherrer Institute (J. Keller) and during the year of 2000 its activities were focused on 1) indicator-based assessment of ozone sensitivity, 2) test and modifications of the SAIMM/UAM-V package, 3) sensitivity of meteorology and ozone mixing ratio to the input parameters of SAIMM for the Swiss topography, 4) transboundary air pollution using boundary values calculated by the LOTOS model, and 5) air quality simulations over Milan area, Italy.

Critical loads and deposition were the research topics of the following contributions, the Institute of Atmospheric Air Protection (V. Kisselev and I. Morozova), from Russia, and the University of Aveiro (Borrego *et al.*). The Institute of Atmospheric Air Protection has studied the sulphur and nitrogen deposition over the Murmansk region of Russia using the climatological stochastic model of regional pollution transport. The University of Aveiro has focused on the parameterisation value of the surface resistance into the deposition module of the UAM model based on site measurements over the Great Lisbon Area. As a side activity, the University of Aveiro has also tried to analyse a high short-term ozone episode over Madrid during the early morning of the 29^{th} of April 2000.

1.5.2 Principal results

The principal results of the work performed with the Danish Eulerian Model, and conducted by Zlatev, are the following: 1) the advanced chemical and deposition schemes tend to improve model results, especially the seasonal variation of some species; 2) visualisation improvements allow a more efficient way to analyse model results; 3) the joint verification of numerical algorithms and model results show an increase in the model confidence level, but more work is necessary on this topic; 4) the development in the model numerical algorithms allows a five times increase in the model performance, compared with 1998; 5) long term runs allow the trend study concerning air pollution levels over different European regions; 6) critical levels of 90 and 120 ppb are exceeded over the most polluted areas during summer ozone episodes during the period between 1989-1997 and over some parts of Europe the critical level for AOT40 is exceeded by a factor greater than seven; 7) the reduction of the ammonia-ammonium concentrations in Denmark between 1989-1998, although the unchangeable of emissions over Denmark, is due to emissions reductions of these compounds in Germany and The Netherlands.

The work conducted by Christensen, also with the Danish Eulerian Model, have shown a better performance of the model, by the introduction of orography of Greenland and the coupling with a state-of-the-art weather prediction model.

The work carried out with the Eta Model, by the IMUB, during bombing periods over Serbia, namely the pollutants emissions resulting from fire of a large number of industrial and military facilities, has shown a long-range transport to the east around the 700 hPa level. The predominant POP removing process was found to be the wet deposition during the period between 18-20 of April and the release, transport and deposition during the war are correlated with precipitation occurrence over Serbia.

The validation of the mixing height (MH) parameterisations within the EUROS model with the Richardson number applied to the ECMWF vertical profiles and LASER measurements. The comparison has been carried out for August 1997 and significant discrepancies were found. Most part of the days the EUROS MH is 100 m during the night and grows to 1000 m in the afternoon while MH deduced from LIDAR measurements and ECMWF exhibit a larger day-to-day variability. ECMWF and EUROS MH are comparable (both are calculated for a grid) while LIDAR values differ in a significant way from the two previous ones, maybe due to the local character.

Studies on indicator-based assessment of ozone sensitivity shows that the ratio H_2O_2/HNO_3 is a successful indicator providing a better separation of NO_X - and VOC - sensitive ranges than the others, but threshold ranges for this indicator ratio are affected by emission and meteorological perturbations. Keller and collaborators found that the UAM-V does not take into account expansion and compression of trace gases when they are transported from one level to another. On the other hand, a sensitivity test to meteorological input to the SAIMM was performed, and it was found that if the model is run with the minimum nudging (1 surface station and 1 sounding) the distribution of vertical diffusivity values is distorted and the absolute values decrease in the lower layers as well as in the vertical profiles leading to modified mixing conditions in the lower troposphere, affecting the pollutants transport. Transboundary air pollution assessments considering boundary conditions calculates by the LOTOS model indicates that boundary concentrations of NO_2 and VOC are lower than literature at night and in the west part of Switzerland, revealing concentration patterns typical from rural areas.

In the deposition topic, the Institute of Atmospheric Air Protection from Russia calculated that the sulphur deposition over the Murmansk region was due to local emissions, 68 % during the year of 1998. The contribution of sulphur emitted from other countries on the deposition was relatively small. On the contrary, the sulphur emitted by the sources on the Murmansk region was responsible for deposition of sulphur over Finland and Norway. On the other hand, the nitrogen deposition over this region is mainly due to the emissions occurring on other regions of Russia, and 11% is a contribution of Finland.

The parameterisations of the surface resistance parameter into the MAR-IV indicates that literature values for this parameter give poor model results over the Great Lisbon Area when compared with values calculated from measurements. The best agreement between model results and measurements was achieved for the minimum value measured for the Rc parameter.

1.5.3 Main conclusions

The confidence level on the Danish model results has been accomplished through the introduction of advanced chemical and deposition mechanisms and the numerical algorithms accuracy checking. Results from studies performed with this model showed that the critical levels for AOT40 (both for crops and forest trees) are exceeded in nearly whole Europe during the period between 1989-1998. Also, the diminishing ammonia-ammonium concentration over Denmark is primarily due to the emission reduction of these compounds over Germany and The Netherlands. The performance of this model was also enhanced throughout the coupling of a state-of-the-art weather prediction model.

A sensitivity study performed to the Eta Model was important to correct transport simulations of air pollutants. It was found that the transport of the air pollutants resultants from heavy industry bombing in the 17^{th} and 18^{th} of April 1999 was done at 1500 m and that the washout of persistent organic pollutants (POPs) over central and southern Serbia was considered the predominant removing process during the air pollution of 18-20 of April. It was found that the release of the air substances due to the bombings causes a precipitation increase.

The study on mixing height with the EUROS model brings a contribution to the validation of MH parameterisations used in air quality models. It has been found that the EUROS formulation tends to underestimate the MH values and the day-to-day variability. Besides, the estimate based on a Richardson number method applied on ECMWF vertical profiles is generally lower than the LIDAR estimate. This study underlines the need to test new formulations proposed in the literature. The present study has also shown that the comparison between various MH data sets is not straightforward, which makes the validation procedure quite difficult. More fundamentally, the present work has shown the limitations of the mixing layer concept and its use in air pollution models.

Both systems MM5-CAMx and RSM-CAMx were applied over Bilbao and Madrid and results have been compared with observational data. Two scenarios have been used: A) Both modelling systems have been executed for the August, 2-6, 2000, for a 120 hours period over Europe with 50 km spatial resolution. Initial concentrations have been put to default. B) The same as scenario A) but initial concentrations are coming from a previous simulation July, 28-August 1, 2000, with the RSM-CAMx modelling system. Results show that improving the quality of initial concentrations is having an important improvement on the quality of the results.

The application of both systems RSM-CAMx and MM5-CAMx indicates that the forecasted initial concentration conducts to better correlation coefficients between measured data and model systems results, concerning air quality over Madrid and Bilbao.

Indicators species and ratios are suitable for delineating VOC-insensitive and NO_x-insensitive regions, through the application of the model system SAIMM-UAM-V both for Switzerland and Milan. The commercially available version of the SAIMM/UAM-V model package is not suitable for complex topography because of an incomplete formulation of the transport schemes. An inadequate choice of the parameters controlling the vertical turbulent exchange profile may substantially affect the size of the mixing layer and the levels and shapes of the pollutant's distribution. Particular care is required if there are discrepancies between prognostic data from the forecast model and data from surface stations and soundings.

The total deposition patterns of sulphur for the territory of Murmansk region show that the maximums are located near industrial centres. Between 1994 and 1998 there was no clear temporal trend of deposition although total emissions from Murmansk regions have been diminishing.

The MAR-IV system presents good results for mesoscale circulation. The Rc parameterisation with values measured at Baldios presents a better performance for the photochemical model resulting in a correct ozone mass balance. Ozone deposition fluxes show a better agreement with measured values for this parameterisation. Nevertheless, this validation methodology should be done with more deposition data, which means more field campaigns.

1.5.4 Policy relevant results

Regional chemical simulations are very important when one wants to analyse the deposition of acidic compounds and pollutants' critical levels.

The significant policy relevant results of the work carried out by the model application and assessment studies teams indicates that the emission reduction in one country may contribute considerably in their concentration in another country, as an example the ammonia-ammonium emission reduction in Germany and The Netherlands lead to a diminishing in concentration values over Denmark. Sulphur and nitrogen deposition in Northern Europe are highly dependent on the distance to the emission source and contiguous countries contribute to each other sulphur and nitrogen deposition.

Photochemical regional modelling is a very important tool for the evaluation of transboundary pollution and useful for the initial boundary conditions calculations of air quality as input in mesoscale photochemical systems. All the improvements that can be made on parameterisations of regional and mesoscale systems allow better performances.

This kind of system can be applied on the evaluation of crop, forest and public health damages after war bombing periods and the areas affected by the resultant air pollutants from events like this.

1.5.5 Aim for the coming year

The Danish Eulerian Model will be submitted to further improvements in order to make possible the 3D version in more case studies, including refinement of space grids and the version with nested model calculations. The chemical scheme, both the photochemical and the heavy metal versions, will continue to be improved in the future.

During the next year, different sensitivity studies will be conducted into the Eta model, concerning horizontal resolution, mountains representation into the model, advection scheme selection, convection effects on transport, parameterisation of horizontal and vertical diffusion, effects of the initial and subgrid scale diffusion.

Implementation of the EUROS model for policy support with respect to tropospheric ozone in Belgium, with the following specific tasks: validation of EUROS for Belgium, training of the potential users of EUROS, operational use of EUROS for policy support in Belgium, determination of optimal strategies for parallellisation of the EUROS model - design and implementation.

The Technical University of Madrid is intending to progress on the use of continental and global air quality models (meteorological and dispersion) by going all the way from street level to global scale and using recognized high quality meteorological and dispersion models. The focus will be made on the CMAQ model together with MM5 and link it to the OPANA air quality model.

The working group conducted by Johannes Keller intends to put special emphasis on the modelling of the air quality in the Milan area focusing on the NO_x/VOC sensitivity. Due to the inadequacy of the UAM-V for complex topography, it is an intention to replace it by the recently issued Comprehensive Air Quality Model with Extensions (CAMx). This model contains modules to simulate aerosol distributions and is more flexible in terms of chemistry mechanisms. The results of a preliminary test are encouraging. Regarding transboundary pollution and seasonal modelling, data from the LOTOS model and the new version of the Swiss prognostic model for the full year 2001 will be procured and used as input for the photo-chemistry model. Concerning dry deposition parameterisation, analysis of the performance of the meteorological model and the concentration and deposition fields obtained with the simulations performed with the model system MAR-IV with the best value encountered for the parameter surface resistance applied to the dry deposition module. Also, a map of critical loads for Portugal will be presented based on an empirical approach.

1.6 Computational aspects

1.6.1 Why are the computational difficulties permanently increasing?

In 1984 A. Jaffe considered, in a very general manner, the relationship between the quickly increasing power of the computer technology and the increasing desire of the scientists and engineers for developing better products. The conclusion was formulated (see Jaffe [3]) as follows:

"Although the fastest computer can execute millions of operations in one second, they are always too slow. This may seem a paradox, but the hearth of the matter is: the bigger and better computers become, the larger are the problems scientists and engineers want to solve".

Many things changed after 1984. The most important of the changes being the fact that, although the computers stopped to grow bigger and bigger, they are much faster from what A. Jaffe could anticipate 16 years ago. Many big problems can now be handled on workstations and PCs. However, the conclusion made by A. Jaffe remains still true. The scientists and engineers do need faster computers (for some of their tasks, at least). This is also true in the field of large-scale air pollution modelling. There are several reasons for this:

1. New and or more advanced modules are needed in the efforts to describe in a more adequate way the physical and chemical processes studied by the models. Such modules have to be incorporated in the models (as, for example, modules for handling data assimilation, aerosols, cloud chemistry, etc.). This leads nearly always to an increase of the computational complexity of the model. In many cases the increase is very considerable.
2. The ultimate purpose when an air pollution model is used is to apply the model in a practical evaluation of possible damaging effects due to high pollution levels (as, for

example, losses of crops due to high ozone levels; see Zlatev *et al*., [4]). This leads to a requirement to perform a long series of runs with different scenarios, which also increase the computational complexity.

3. Long-term computations are often needed in order to study the tendencies in the development of high pollution levels due to reductions of emissions (in the last 10-15 years the emissions in Europe as whole have been reduced; some of the reductions are rather considerable). The requirement for long-term runs is also contributing to an increase of the amount of computations. Long-term computations are also appropriate when the influence of high pollution levels on the climatic changes is studied.
4. Sometimes more detailed information about the pollution levels is needed. Such information can be achieved by using fine resolution models. The use of such models may lead to an increase of the number of computations by a factor of several hundreds.
5. Inverse problems are to be treated in the solution of certain tasks. The computational difficulties are enormous when inverse problems are to be formulated and handled on computers. These are very challenging tasks, both computationally and numerically. There are a lot of unresolved problems in this field.

This list can be continued, but this is not necessary because the five reasons given above show clearly that the air pollution models must continuously be improved in order to meet the requirements for achieving better (more accurate, more detailed and more reliable) results. The improvements imply increased computational complexity. Therefore, it is necessary to carry out the improvements together with attempts

a) to implement faster and sufficiently accurate numerical methods,
b) to exploit in more efficient way the great potential power of the modern high-performance computers

and

c) to visualize better the results in order to represent clearly the relationships between the investigated quantities which are normally hidden behind enormous amounts of digital information (millions and millions of numbers stored in huge output files).

This short description of the importance of the computational aspects in air pollution modelling explains why these issues were treated, also in this year, in many of the annual reports of the participants in the GLOREAM subproject of EUROTRAC-2.

1.6.2 Treating models with more advanced modules

The use of **data assimilation techniques** to existing models in an attempt to improve the results has been reported by:

- Ebel and his co-workers (the particular model to which the data assimilation techniques were applied being EURAD, developed at the University of Cologne, Germany); some recent results on this topic can be found in Elbern and Schmidt [2],

and

- Builtjes and his co-workers (the particular module was the LOTOS model developed at TNO).

Adding modules for studying **particles** in an attempt to get better results for the contribution from traffic emissions to the pollution levels has been reported by:

- Ackermann and his co-workers (the particular module is MADE; it has been used in connection with EURAD at the Aachen Forschungszentrum, Germany),
- Ebel and his co-workers (the particular model to which the data assimilation techniques were applied being EURAD, developed at the University of Cologne, Germany)
- Builtjes and his co-workers using the LOTOS-model, and applying MADE in cooperation with EURAD

and

- Matthijsen and his co-workers (the PM module of EUROS; the EUROS is developed at RIVM, the Netherlands, a Belgian version of this model is reported by Delobbe and Mensink).

Adding more advanced modules for **studying effects from biogenic emissions** has been reported by:

- Zlatev and his co-workers (this module has been applied to the Danish Eulerian Model developed at the National Environmental Research Institute, Roskilde, Denmark).

Improvement of the grid-resolution of regional models by refinement of the mesh has been reported by:

- Brandt and his co-workers (coupling different models developed at the National Environmental Research Institute in Roskilde, Denmark, in order to perform air pollution forecasts at different scales; more details can be found in Brandt *et al*. [1]),
- Ebel and his co-workers (local one way refinement on several levels performed in connection with the EURAD model, University of Cologne, Germany)
- Berkens developed improved techniques for 2-way nesting

and

- Zlatev (global refinement over the whole 4800 km x 4800 km space domain, moving from a 50 km x 50 km grid to a 10 km x 10 km grid, carried out in connection with he Danish Eulerian Model developed at the National Environmental Research Institute, Roskilde, Denmark).

Results related to the challenging problem of studying feedback mechanisms between climate change and the chemical composition of the atmosphere have been reported by:

- Feichter and Schultz (by including a very extensive hydrocarbon chemistry module in the MOZART package and running this package for the period from 1978 to 1994)

and

- Law and her co-workers (studies based on the use of the TOMCAT model as well as on the use of comparisons of the TOMCAT model with several other models).

GIS (the Geographical Information System) is still not very popular in the field of large-scale air pollution modelling.

- Smiatek has reported results obtained in the attempts to develop modules based on GIS for preparation of data for use in air pollution models. It would be interesting to see the application of his technique (or some other similar technique based on the application of GIS) to a particular air pollution model.

1.6.3 Improvement of the numerical algorithms and the computational techniques

One must improve permanently the numerical algorithms and computational techniques in the efforts to make the regional air pollution models to solve bigger tasks and more tasks. Most of the participants of the GLOREAM subproject of EUROTRAC-2 do make such improvements. The most important results have been reported in the following contributions:

- Barone and his co-workers report improvements of both numerical methods and computational techniques in the Air Pollution Model for the Campania Region developed at the Department of Chemistry, University “Federico II” of Naples, Italy. Parallel computations are applied when this model is run. Parallel computations are discussed also in the report of Knoth and his co-workers.
- Berkvens and his co-workers are reporting results obtained by different kinds of splitting. The major purpose is to identify the splitting procedure, which minimizes the error due to splitting.
- The chemical part of a large-scale air pollution model is normally the most time consuming part when the model is run on computers. Therefore, the task of finding fast and sufficiently accurate chemical mechanisms as well as fast and sufficiently accurate numerical algorithms for handling the chemical schemes on computers is very important. Some interesting results in this direction are also presented in the reports of (i) Makar and (ii) Stockwell and his co-workers.
- The treatment of some inverse problem is an important issue in air pollution modelling. Inverse problems lead to big computational tasks. These problems are normally very ill-conditioned (in the sense that small perturbations of the input data lead to big differences in the output results). Therefore, the search of efficient numerical algorithms is crucial in this field. The use of inverse dispersing modelling as a tool to derive emission data from measurements is discussed in the report of Seibert and Kromp-Kolb.
- The use of faster, but still sufficiently accurate, numerical algorithms for the advection sub-model of a large-scale air pollution model is reported by Syrakov from the Bulgarian Academy of Sciences (Sofia, Bulgaria). The model has also been used to study pollution levels in Bulgaria and surrounding areas of Bulgaria.
- Improvements of the numerical algorithms in the Danish Eulerian Model as well as applying efficient parallel techniques when this model is run on modern high-speed computers are reported by Zlatev from the National Environmental Research Institute (Roskilde, Denmark).

1.6.4 Main benefits from successful resolving of the computational problems

The computational issues are well represented in the individual annual reports of the participants in the GLOREAM subproject of EUROTRAC-2. In many of the reports it is documented that the major computational problems are successfully resolved. When this has been completed, then the following benefits were (or will be obtained in the near future) obtained:

- it is possible to improve the description of the physical and chemical processes in the models,
- it is possible to solve more tasks and bigger tasks,
- it is possible to carry out long simulations with different scenarios in order to study the response of the models to key parameters (anthropogenic and biogenic emissions, meteorological parameters, boundary conditions, etc.),
- it is possible to start to run operationally some of the models in an attempt to predict exceedance of critical levels (as, for example, ozone critical levels) in the next two to three days

and

- it is possible to start development of advanced control strategies for keeping the concentrations and/or the depositions of harmful pollutants under the prescribed critical levels.

1.6.5 Other scientific computing activities carried out by members of GLOREAM

Some members of GLOREAM participated actively in two specialized mini-workshops devoted on computational problems arising when large-scale air pollution models are treated on high-speed computers:

- Special Session on “Large-scale Computations in Air Pollution Modelling” within the Third Conference on Large-scale Scientific Computations held in Sozopol (Bulgaria), June 6-10, 2001. This session was organized by Adolf Ebel, Krassimir Georgiev and Zahari Zlatev.
- Special Session on “Large-scale Computations in Environmental Modelling” within the International Conference on Computational Science held in San Francisco (California, USA), May 28-30, 2001. This session was organized by Zahari Zlatev.

1.6.6 References

[1] Brandt, J., J.H. Christensen, L.M. Frohn, F. Palngren and Z. Zlatev; Operational air pollution forecasts from European to local scale, *Atmos. Environ.* **35** (2001) S91-S98.

[2] Elbern, H. and H. Schmidt; Ozone episode analysis by four-dimensional variational chemistry data assimilation, *J. Geophys. Res.* **106** (2001) 3569-3590.

[3] Jaffe, A.; Ordering the universe: The role of mathematics, *SIAM Review* **26** (1984) 478.

[4] Zlatev, Z., I. Dimov, T. Ostromsky, G. Geernaert, I. Tzvetanov and A. Bastrup-Birk; Calculating losses of crops in Denmark caused by high ozone levels, *Environmental Modelling and Assessment* **6** (2001) 35-55.

1.7 Model evaluation and validation

All models in use in GLOREAM, as they are depicted in the overview table, have been tested, validated in one way or an other against observations. Details can be found in the references of the different models. In 2000, a number of studies have been performed focusing on model evaluation and validation specifically.

In a study by Tilmes *et al.* five models, the THOR-model by NERI, Denmark, the EMEP 3-D model by NILU, the REM-3 model by the FU-Berlin, the SMHI-model, Sweden and the EURAD-model, Kolnhave been intercompared and tested against observations in their real-time ozone forecasting mode over the period may-september 1999. Generally, it was found that the most comprehensive models gave the best results.

Both the KAMM/DRAIS model and the EURAD-model have been tested against the observations of the BERLIOZ-campaign. The EURAD-model has also been tested against results of FLUMOB,VOTALP and PIPAPO.

The LOTOS model has been tested against ozone observations for august 1997 making use of the application of specific statistical indices and in combination with data assimilation.

1.8 Overview over policy relevant results

As part of GLOREAM models over a wide rage of scales, ranging from coarse grid global modelling to local models, are used. Most of the results presented are aiming at improving our understanding and representation of the processes going on in the atmosphere, improving the quality and thus the credibility of the policy relevant results. In this context an important initiative in GLOREAM is a recommendation for a common procedure and terminology for model quality assurance. A majority of the contributions focus on ozone, but other species such as sulphur, lead, mercury and particulate matter are also addressed. Several contributions describe systems for nesting regional and local models.

There is a limited number of contributions addressing global modelling. Model calculations using the IPCC scenario for 2100 predicts large increases in surface ozone in Europe, North America and the industrialized Far East. Furthermore, "Numerical isotopes" are used for source allocations, and thus source allocations can be made without the non-linear effects from running the model by excluding one source at a time. A substantial fraction (of the order of 10 to 20% in most of Europe)of the surface ozone is attributed to lightning sources.

Several contributions address the effect of control measures on air pollution or depositions for separate countries or parts of the countries. Regional budgets, quantifying the effects of trans-boundary advection of pollutants are also calculated. Model runs are performed in order to define strategies for meeting environmental targets for ozone, sulphur depositions etc. It is shown that significant fractions of the depositions of oxidized nitrogen and sulphur may be attributed to emissions from international shipping. Emissions from international shipping will also have a significant affect on surface ozone levels. For models covering only a limited region in Europe the sensitivity to NO_x/VOC control may be sensitive to the boundary concentrations used in the calculations. It is also shown that biogenic emissions may be the dominant local source of VOC. Regional studies have also been made of a wide range of subjects such as investigating the effects of the Balkan oil fires.

Inverse modelling is used in order to derive information on emissions from measurements on a regional scale. With this method the location of a point source in space and time can be allocated.

PM (particulate matter) is included in an increasing number of the models. For PM the response to emission reductions is strongly non-linear. Significant proportions of the PM are secondary organic particles (both anthropogenic and biogenic origin). Emissions of VOC have traditionally been lumped according to reactivity. However, a significant proportion of the organic aerosols may originate from VOCs not currently included in this lumping.

Several contributions describe the development and applications of systems for forecasting of air pollutants, mainly ozone. Such systems are intended for public warning of pollution (ozone) episodes and may also be used as a decision making tool for short term regulatory actions. Several partners to GLOREAM apply nesting from coarse to finer grid scale models.

Several forecasting systems within GLOREAM have taken part in a model intercomparison for Germany.

1.9 General aims for the coming year

The main aims for 2001, apart from just doing high level scientific research in the area of global and regional modeling, are, as also recommended by the mid-term review

- Further improvements in aerosol-modelling
- Further activities in model validation, taking validation data of other subprojects into account
- Increasing emphasis on synthesis within GLOREAM
- Emphasis on scale interaction/nesting from the global scale to the local scale.

1.10 Closing remark

The general overview of the GLOREAM annual report over 2000 has been made by the steering committee. The editing of the complete annual report has been done by GLOREAM's scientific secretary, Annette Münzenberg.

Michael Memmesheimer and Adolf Ebel were especially responsible for the chapter on model investigation and improvement. The part on global modeling has been prepared by Hans Feichter. Ana Cristina Carvalho and Carlos Borrego were especially responsible for the chapter on model applications and assessment studies. Zahari Zlatev wrote chapter on computational aspects. Jan Eiof Jonson wrote the overview over policy relevant results. The remaining text has been written by Peter Builtjes.

References to papers in this introduction and overview can be found either in the list of publications in the refereed literature, or in the references listed by individual principal investigators.

An overview over the models used in GLOREAM can be found in the two tables on the next pages.

GLOREAM models: model features

Model	horizontal resolution	vertical resolution and extent	horizontal coverage	species	remarks	PI in charge
A3UR	0,35°	variable	Europe	ozone		Toupance/Brocheton
DACFOS2	1 ... 16 km	31 layers up to 30 km	Europe	photooxidants and precursors		Gross
DEHM (Danish Eulerian Hemispheric Model)	5 ... 150 km	22 layers up to 20 km	northern hemisphere	photooxidants and precursors, sulphur, ammonia-ammonium, SO2 and sulfate, mercury and lead under development	hemispheric nested version	Christensen
DEM (Danish Eulerian Model)	10 ... 50 km	12 layers up to 7 km	Europe	photooxidants and precursors, sulphur, ammonia-ammonium, SO2 and sulfate	regional scale	Zlatev
ECHAM4-MOZART	2,8°	variable	global	SO2 and sulfate		Feichter
EMAP	20 km	6 layers, log-linear, PBL	Europe	radioactivity		Syrakov
EMEP Eulerian Photochemistry Model	50 km	20 layers up to 100 hPa	Europe	photo-oxidants and precursors, sulphur, ammonia		Jonson
Eta+Tracer	30 km	one layer trajectory model	regional to continental scale	Inert species		Lazic
EURAD/MADE System	2 ... 50 km	variable	3000 km x 3000 km ...100 km x 100 km	photo-oxidants and precursors, sulphur, aerosols, radioactive substances (Cesium)	nesting procedure	Hass, Ebel
EUROS	60 ... 7,5 km	4 layers with model top at 3 km	Europe	photo-oxidants and precursors, sulphur, aerosol, POPs		Matthijsen, Mensink
FLEXPART	10 ... 200 km	Lagrangian particle model	regional to global/Europe	passive tracers or species with prescribed decay rates (CO, NOx, NOy)		Seibert
GLOUR	up to 1 km	top layer 10 hPa, 23-32 layers tested	Europe	CBM-IV, RADM and SAPRAC-97 and 2000 chemical mechanisms	3rd generation of air quality models based on CMAQ (EPA, 2000) and MM5 models	San Jose
KAMM/DRAIS	1 ... 5 km	variable, grid size close to the ground 10 ...25 m, a few 100 m at the top, model height 4 ... 8 km	a few hundred km in both directions	photo-oxidants and precursors,sulphur, aerosols		Nester
LaMM5	up to 5 km	model top troposphere	continental ... regional	photo-oxidants and precursors, sulphur, low reactive species		Schaller
LOTOS	0,5° x 0,25°	4 layers (or more) up to 3 km (or more)	Europe	photo-oxidants and precursors, sulphur, aerosols	nested with UAM	Builtjes
MEMO-MAR IV	500 m ... 10 km (MEMO) 2 km minimum (MAR IV)	varying between 25 and 35 non-equidistant layers up to 10 km (MEMO) about 10 layers up to 3 km (MAR IV)	local to regional (MEMO) maximum 300 km x 300 km (MAR IV)	photo-oxidants and precursors, sulphur aerosols	(MEsoscale MOdel)/MARS models system MEMO coupled CSU Mesoscale model and Urban Airshed Model CB-IV MAR IV	Borrego
MATCH	1 ... 100 km	variable, depending on meteorological driver, typically 5 layers below 1 km and 20 layers below 200 hPa	regional to continental	photooxidants and precursors, sulphur, ammonia, radioactivity, aerosol bound species, base cation, PAH, arsenic		Langner
MOGUNTIA	10°x10°	100 hPa from 1000 through 100hPa	global	radioactive gases, chemical tracers, aerosol		Zimmermann
MUSCAT	1 ... 10 km	30 layers (stretched) up to 8 km	100 ... 500 km	photo-oxidants and precursors, sulphur, aerosols		Knoth

Model	horizontal resolution	vertical resolution and extent	horizontal coverage	species	remarks	PI in charge
OPANA	1 km	up to 32 layers, top layer: 6 km	maximum 500-600 km x 500-600 km	gas phase chemistry, CBM-IV	implemented in 1994, meteorological model based on MEMO, deposition model based on MEDFLUX EU project; chemistry solver based on SMVGEAR (implicit scheme); 80 % CPU for chemistry	San Jose
REMO-GESIMA	0,5° ... 4 km	10 layers	Europe	ozone, CO2, radon		Langmann
REM3, CALGRID	4 ... 30 km	3 time dependent layers (Central Europe), 14 levels (regional)	Central Europe, regional	photo-oxidants and precursors		Reimer
SPMIAC	5 km	variable, model top troposphere	southern Italy	mainly ozone		Barone
TFS network modelling system	20,8 km	36 layers in meteorological part, 15 layers in chemistry part	Central Europe	ozone		Tilmes
THOR System	10 m ... 50 km	model top 100 hPa	up to 15000 km x 15000 km	photo-oxidants and precursors, VOCs, nitrogenoxides, sulphur, aerosols, heavy metals (mercury,lead), POPs, radioactive species, CO2, ammonia/ammonium	(including: ETA, DEOM, BUM, OSPM, REGINA, ACDEP, DREAM, DEHM, MM5, ...)	Brandt
TM3-K	2,5°	19 layers	global	ozone, SO2		Kelder
TM3, TM5	1° x 1° ... 9° x 6°	19 ... 60 layers, whole atmosphere	global	photooxidants and precursors, sulphur		Berkvens
TOMCAT	2,8° x 2,8 °	31 layers up to 10 hPa	global	photooxidants and precursors		Law
TRACER	5 ... 50 km	31 layers up to 30 km	Denmark to the northern hemisphere	passive tracer		Gross
TRAP	25, 50 and 150 km	one layer trajectory model	about 500 km	sulphur and nitrogen compounds, ozone and volatile organics are included for the closure of the chemical scheme	stochastic model for meteorological variables is used	Kisselev
UAM, UAM-V	3 km, 5 km	10 layers up to 3 km	Switzerland	photo-oxidants and precursors		Keller

GLOREAM models: capabilities and applications

Model	model capabilities	examples for applications	future applications	PI in charge
A3UR	scenario runs	investigation of the impact of grid resolution on ozone formation		Toupance/Brocheton
DACFOS2	real-time ozone forecasting	verification of model results for 1998-200+, implementation of diffusion and model testing	improvement of emission uptake	Gross
DEHM (Danish Eulerian Hemispheric Model)	analysis of air pollution levels over the Arctic	14 year run for SO2 and sulfate		Christensen
DEM (Danish Eulerian Model)	damaging effects caused by exceeded critical values; economical evaluation of the losses due to high AOT40 values; biogenic emissions; application for emission reduction strategies	long term (up to 10 years) applications; sensitivity effects (various scenarios); damaging effects caused by exceeded critical levels (including health effects); economical evaluations; scenarios in connection to the CONCAWE projects (and especially with regard to some AUTO-OIL-2 data)	effects due to increased biogenic emissions; effects due to use the high resolution emissions inventories for 5 cities (London, Lyon, Milan, Cologne, Athens); increase of model resolution	Zlatev
ECHAM4-MOZART	determination of the impact of sulfate aerosols on the radiation balance	global distribution of SO2 and sulfate for the year july 1993 - june 1994	global runs for ozone and aerosols	Feichter
EMAP	ETEX (ATMES II), ETEX aims; EMEP intercomparison, check of long-term simulation abilities of participating model vs data and each other	ETEX 1st release, EMEP Cd intercomparison study	creation of interpreter for gamma background data in Bulgaria; improvement of Bulgarian Emercency Response System for nuclear accidents (3 way zooming); assessment of pollution exchange with neighbouring countries	Syrakov
EMEP Eulerian Photochemistry Model	PAUR; emission reduction studies	POLINAT, PAUR	int. shipping; emissions from international shipping; include 3D trajectory studies in the model; the model will be used in TROTREP	Jonson
Eta+Tracer	determination of the impact of intert species	Sahara dust and war time conditions of fire of oil refineries	nesting and scale interactions	Lazic
EURAD System	TRACT field experiment: model evaluation, discrepancy of meas./sim. NOx; smog episode July 94: use of monitoring network for evaluation; BERLIOZ experiment: evaluation, interpretation of measurements; RIFTOZ: vor. chem. data assimilation; emission reduction studies: AMPO: long-term scenarios (emission projections for 2005), short-term measures local in time and/or space	EUMAC wet case; EUMAC dry case episodes; TRACT episode; TOR episode; ALPEX ozone intrusion; VOTALP simulations; air traffic emission scenarios; long-term calculations; FluMOB, BERLIOZ, PIPAPO experiments; AMPO; Tschernobyl; SANA; TOASTE-C; RIFTOZ/GLOREAM episode August 1997; data assimilation	study of city plumes; of aerosol impact; long-term simulation for EC air quality directive; investigation of densely populated areas; traffic impact; use of data assimilation for applicational questions; case study analyses by advanced data assimilation techniques	Ebel
EURAD/MADE System	see EURAD (see above)	see EURAD list for episodes (see above); long-term: April 1995 - Sept 1995	application study for aerosols	Hass
EUROS	chemical dispersion modelling	short and long term -scenario- runs for national environmental outlooks and assessments; pilot to forecast ozone using data assimilation; POP dispersion linked with soil model	long-term air quality simulations; policy efficiency studies; extension to aerosols; budget analyses; POP deposition	Matthijsen
EUROS	assessment studies; emission reduction studies	specific episodes (in july and august 97), long term simulations (may -> sept 97); various impact assessment studies for the flemish government; coupling with urban air quality models; detailed emission reduction studies; various studies for Flemish Environmental Agency, policy makers at regional and federal level, environmental managers at urban level	in the framework of the BelEUROS project (supported by the Belgian authorities), implementation at the Interregional Cell for the Environment (Brussels) as a tool for policy support; design and implementation of optimal strategies for parallelisation of the EUROS code; participation in modelling intercomparison CAFE	Mensink
FLEXPART	emission reduction studies; forward and backward modeling of inert tracers and species with prescribed decay, source-receptor matrix calculation, long term and episodical application	RISKMAP study, ranking of countries according to the risk their nuclear power plants impart to any specified receptor country; application: guidance for actions to reduce the risk; ETEX; VOTALP; PIPAPO; global radionuclide transport (nuclear test ban verification)	real-time application of an Eulerian chemistry-transport model in Vienna; chemical "footprint" for background monitoring stations	Seibert
GLOUR	air quality modelling over global through urban scales based on MM5 and CMAQ models; aerosol and cloud modelling; 3rd generation of transport models	research level yet; north of Africa up to Arctic latitudes; three nesting levels up to 4 - 1 km over Madrid (tested)	building the operational version through INTERNET	San Jose
KAMM/DRAIS	real-time ozone forecasts; comparison with measurements; application in the evaluation of field experiments; emission reduction studies; mass budget simulations	participation in model evaluation for TFS; TULLA; SANA; TRACT; FLUMOB; BERLIOZ; ATHENS ...; evaluation of experiments TULLA, TRACT, SANA and Augsburg experiment	evaluation of the BERLIOZ and the Augsburg Experiments; Israel, TRACKS?	Nester
LaMM5	representativeness of surface station networks (BERLIOZ); emission reduction studies: effects and effectiveness of selective (instead of general and overall) reduction strategies	BERLIOZ/FLUMOB; emission reduction studies for "Neue Bundesländer" ("Former GDR") , scenarios with and without traffic, new roads etc.	more general studies on source-receptor relations on the mesoscale-ß (counties)	Schaller

Model	model capabilities	examples for applications	future applications	PI in charge
LOTOS	episodes; long term; evaluation studies; emission reduction studies; scenario studies for tropospheric ozone, aerosols	evaluation of ozone for August 1997 (RIFTOZ project); evaluation of long term values for O3, sulfate, nitrate; emission reduction studies: AOT40(c)(f) impact; discussion in CAFE about the model requirements in view of the EU-directives	data assimilation; aerosol modelling; ozone forecasting (in cooperation with RIVM), suited for EU directives and UN ECE calculations	Builtjes
MEMO-MAR IV	assessment and scenario studies; ozone control; episodes and specific events experiments; environment impact assessment	environment impact assessments to locate the future airport in the Lisbon metropolitan area and the implementation of a new combined cycle power plant in Carregado (NE of Lisbon city); episodes and specific experiments in the LisBex 96 campaign; study of different traffic emission scenarios on the regional level	dry deposition sensitivity studies; sensitivity studies on mixing height and implementation of mixing height parameterisations; applications on environment impact assessment; model evaluation over Aveiro's region; dynamical downscale in order to evaluate the impact of global climate change in the air quality over Aveiro's region; continuity on the impact of global climate change in air quality over Portugal; dry deposition processes; studies on production, transport and dispersion of photochemical pollutants at the Aveiro mesoscale domain and land-sea breeze strength over this domain	Borrego
MATCH	real time forecasts; scenario runs; acid deposition assessment	real time forecasts of ozone and precursors; operational real time forecasts of radioactivity; operational (annual) high resolution acid depositions assessments for Sweden; acid deposition assessments for South Asia, Africa, and South America; transport and deposition of sulfur and arsenic in central and northern Chile; assessment of environmental effects of air transportation (EU-project EIATNE), coupled to regional climate model for assessment of regional climate change on air pollution; basic transport model tested in ETEX and RTMOD	inclusion of aerosols and aerosol dynamics; high resolution applications, including forecasts; using MM5 as meteorological driver; mapping of surface ozone over Sweden for effect studies	Langner
MOGUNTIA	long term (century) global tropospheric transport/chemistry simulations on the basis of monthly mean meteorological parameters	model studies of the emission, global distribution and removal of the following species were performed and published during the EUROTRAC1 period: 1. inertial trace constituents: fluorocarbons (F11,F12); 2. radioactive gases: 85Krypton, 222Radon; 3. chemical tracers: methylchloroform (MCF); the ozone/ CH4 / CO / NOx system including clouds; 4. aerosol: mineral aerosol (desert dust), sulfate	numerical source receptor experiments; empirical relationship studies of variability vs. lifetime of tropospheric trace constituents	Zimmermann
MUSCAT	assessment studies of real situation/events (e.g. at extreme events as winter smog situations); emission reduction studies; studies on the influence of new buildings and plants on the micro-climate and the emission situation	effect of emission reduction cases in different respects (e.g. desulfurization for SO2); in respect to EU-guidelines; transport and transformation of SO2, photo-oxidants and aerosol in Saxony, OMKAS project	studies for Saxony (especially aerosols and precursors for secondary aerosol sources); BERLIOZ	Knoth
OPANA	operational air quality mesoscale model; ANA model is the research version; assessment and scenario studies; episodes and specific events; environmental impact assessment; nesting capabilities; on-line linkage between meteorological module and chemistry solver	official operational model for Madrid municipality since July 2001 used in EMMA project (IST program EU, 1996-1998); used in EQUAL project (IST program EU, 1998-2001); in EMMA over Madrid community and city; in EQUAL over Bilbao (Spain) and Leicester (UK); used in APNEE project (IST program EU, 2000-2001) over Madrid city	research studies to compare with MM5-CMAQ modelling system	San Jose
REMO-GESIMA	episodic (GESIMA) and long term (REMO, 3 months and more); modelling of photooxidants and trace species, e.g. CO2, Rn	nested modelling focussing on Europe, Germany and Berlin-Brandenburg during TRACT 1992, FLUMOB 1994, BERLIOZ 1998	global to mesoscale model applications focussing on Europe (photooxidants, biomass burning, lightning, intercontinental transport), the Arctic region (Arctic haze and ozone depletion episodes), the tropics (aerosols, photooxidants, biomass burning, lightning)	Langmann
REM3, CALGRID	episodic tests on abatement strategies against ozone peaks in summertimes	long term, episodic and for experiment (FluMOB, BERLIOZ); tests on strategic emission scenarios in relation to traffic, ozone forecasts	ozone forecast system for Germany (UBA); AMPO (UBA); BERLIOZ (BMBF); AOT40-abatement-strategies (UBA); long range model runs on AOT40 and PM10 problems in relation to measurements; short range forecast of surface ozone; Miskam/Calgrid for urban modeling	Reimer
SPMIAC	operational ozone calculations		application to southern Italy; numerical improvements; acceleration of model	Barone

Model	model capabilities	examples for applications	future applications	PI in charge
TFS network modelling system	forecasting; assessment of monitoring networks	air pollution forecasting from 1999 on; BERLIOZ	model comparison within GLOREAM; TFS evaluation for BERLIOZ episode	Tilmes
THOR System	air pollution forecasting (e.g. exceedances of critical limit values); nowcasting; part of monitoring programmes (eutrophication assessment in Danish waters every year; health effect in rural and urban areas); transport into the Arctic areas; long-term scenarios on the same scales	regional, urban background and urban street forecasting, scenarios as e.g. contribution from buses, trucks, vans and pas. cars to air pollution in major Danish cities for traffic planning; Models included as part of the DK monitoring programme (urban and rural); ETEX; Chernobyl; eutrophication and acidification studies; health effects	health effects from ultra-fine particles in urban areas	Brandt
TM3-K	calculation of the impact of aircraft exhaust on tropospheric ozone	validation with Polinat 2, Sonex and Eulinex	global model runs in relation to satellite data	Kelder
TM3, TM5	atmospheric transport and chemistry: episodic and long-term scenarios	nested model runs; atmospheric transport (stratospheric [heterogeneous] chemistry, biomass burning, NOx modeling, data assimilation, assimilation of GOME O3 data, interpretation of NO2 from GOME data, global study of influence by methyl chloroform emissions on OH)	parallel computation; coupling of (split) operators (effect of resolution on subgrid model; chemical forecast; chemical data assimilation; comparison and support and assimilation of satellite observations [Sciamachy-satellite a.o.])	Berkvens
TOMCAT	(global) scenario studies	impact of stratospheric ozone on the tropospheric ozone budget	comparisons with satellite and aircaft measurements; source attribution of ozone and precursors; trace gas budgets and future concentration scenarios for ozone	Law
TRACER	mesoscale transport of air pollutants	ETEX 1st release - intercomparison study	nuclear emergency; real time ozone forecasting	Gross
TRAP	estimation of long-term characteristics of acidifying pollutant deposition in a regional scale	calculations for Kaliningrad, Leningrad and Murmansk regions, republic Karelia (Russia)	calculations for the other territories of Russia; scenario runs; formation of the national system of regional pollution control	Kisselev
UAM, UAM-V	scenario studies; emission reduction studies; short term impact assessment	simulation of the POLLUMET period in 93; NOx/VOC reduction; replacement of Swiss nuclear power plants by fossil fuel driven plants; emission reduction studies: 1) NOx/VOC-reduction -> O3 sensitivity, studies regarding Sillman parameters; 2) influence of Swiss energy system scenarios for 2010 (reduction of traffic, replacement of nuclear power plants); LOOP (1998)	NOx/VOC limitation studies for LOOP; long term calculation for Switzerland	Keller

2. Authors and titles of theses resulting from the subproject work

Brocheton, F.; Representation des emissions anthropiques dans les modeles de chimie-transport: Sensibilite a la representation spatiale des emissions et au degre de raffinement du schema chimique, PhD Thesis, Universite Paris 12 - Val de Marne (1999).

Bauer, S.E.; Photochemical smog in Berlin-Brandenburg: An investigation with the atmosphere-chemistry model GESIMA, Ph.D. thesis, Examination Report No. 81, Max-Planck-Institute for Meteorology, Hamburg (2000).

Seibert, P.; Quell-Rezeptor-Beziehungen atmosphärischer Spurenbestandteile [Source-receptor relationships of atmospheric trace substances]. Habilitationsschrift, Univ. f. Bodenkultur Wien (2000).

Valinhas, M.J.; Modelação da deposição de poluentes atmosféricos: Aplicação ao conceito de cargas críticas (Atmospheric pollutants deposition modelling: application to the critical loads concept), Dissertation presented to University of Aveiro to obtain the Master degree on Atmospheric Pollution (2000).

3. Publications in refereed literature

In the following list only the peer reviewed papers which have appeared in international journals during 2000 are given. Other relevant literature such as internal reports, conference proceedings etc. can be found in the individual reports of the principal investigators (see Section 4.).

Ambelas Skjøth, C., A. Bastrup-Birk, J. Brandt and Z. Zlatev; Studying variations of pollution levels in a given region of Europe during a long time-period, *Systems Analysis Modelling Simulation* **37** (2000) 297-311.

Barone, G., P. D'Ambra, D. di Serafino, G. Giunta and A. Riccio; Application of a parallel Air Quality model to the Campania region, *Environ. Soft. and Model.* **15** (2000) 503-511.

Bauer, S.E. and B. Langmann; Summer haze in a polluted atmosphere, *J. Aerosol Science* **31**, Suppl. 1 (2000a) 422-423.

Brandt, J., J.H. Christensen, L.M. Frohn and Z. Zlatev; Numerical Modelling of Transport, Dispersion, and Deposition - Validation against ETEX-1, ETEX-2, and Chernobyl; *Environmental Modelling and Software* **15** (2000) 521-531.

Elbern, H., H. Schmidt, Talagrand, A. Ebel; 4D-variational data assimilation with an adjoint air quality model for emission analysis, *Environmental Modeling and Software* **15** (2000) 539-548.

Friese, E., M. Memmesheimer, I.J. Ackermann, H. Hass, A. Ebel and M.J. Kerschgens, A study of aerosol-cloud interaction with a comprehensive air quality model, *J. Aerosol. Sci.* **31** (2000) 54-55.

Fuentes, J.D., M. Lerdau, R. Atkinson, D. Baldocchi, J.W. Botteneheim, P.Ciccioli, B.Lamb, C.Geron, L. Gu1, A.Guenther, T.D. Sharkey and W.R. Stockwell; Biogenic Hydrocarbons in the Atmospheric Boundary Layer: A Review, *Bull. Amer. Meteor. Soc.* **81** (2000) 1537-1575.

Grell, G.A., S. Emeis, W.R. Stockwell, T. Schoenemeyer, R. Forkel, J. Michalakes, R. Knoche and W. Seidl; Application of a Multiscale, Coupled MM5/Chemistry Model to the Complex Terrain of the VOTALP Valley Campaign, *Atmos. Environ.* **34** (2000) 1435-1453.

Hertel, O., F. Palmgren, T. Ellermann, H. Skov, K. Kemp, M.F. Hovmand and J. Brandt; IUPAC. Air Quality in Denmark. *Chemistry International* **22**, No. 5 (2000) 133-135.

Klemm, O., W.R. Stockwell, H. Schlager and M. Krautstrunk; NO_X or VOC Limitation in East German Ozone Plumes?, *J. Atmos. Chem.* **35** (2000) 1-18.

Langmann, B.; Numerical modelling of regional scale transport and photochemistry directly together with meteorological processes, *Atmos. Environ.* **34** (2000) 3585-3598.

Lazic, L.; "Initialization" using an iterative Matsuno style scheme in the Eta Model adjustment stage. *Meteorol. Atm. Phys.* **75** (2000) 121-130.

Lazic, L. and I. Tosic; Sensitivity of forecast trajectories in strong local winds to the wind data frequency. *Idojaras* **104** (2000) 91-107.

van Loon, M., P.J.H. Builtjes and A.J. Segers; Data-assimilation of ozone in the atmospheric chemistry transport model LOTOS, *Environmental Modelling and Software* **15** (2000) 603-609.

van Loon, M., I.J. Ackermann, M. Schaap and P.J.H. Builtjes; Primary and secondary aerosol simulation using LOTOS, *J. Aerosol Sci.* **31**, Suppl. 1 (2000) 52-53.

Seibert P., F. Beyrich, S.-E. Gryning, S. Joffre, A. Ramussen and P. Tercier; Review and intercomparison of operational methods for the determination of the mixing height, *Atmos. Environ.* **34** (2000) 1001-1027.

Seibert, P., H. Feldmann, B. Neininger, M. Bäumle and T. Trickl; South foehn and ozone in the Eastern Alps - case study and climatological aspects, *Atmos. Environ.* **34** (2000) 1379-1394.

Steinbrecher, R., M. Klauer, K. Hauff, W.R. Stockwell, W. Jaeschke, T. Dietrich and F. Herbert; Biogenic and Anthropogenic Fluxes of Non-Methane-Hydrocarbons Over an Urban-Impacted Forest, Frankfurter Stadtwald, Germany, *Atmos. Environ.* **34** (2000) 3779-3788.

Stockwell, W.R., J.G. Watson, N.F. Robinson, W. Steiner and W.W. Sylte; The ammonium nitrate particle equivalent of NO_x emissions for continental wintertime conditions, *Atmos. Environ.* **34** (2000) 4711-4717.

Wolke, R. and O. Knoth; Implicit-explicit Runge-Kutta methods applied to atmospheric chemistry-transport modelling, *Environ. Model. Software* **15** (2000) 711-719.

4. Reports from the principal investigators

4.1 Introduction

GLOREAM-projects are divided into the following 5 tasks:

1. Model investigation and improvement
2. Global modelling
3. Computational aspects
4. Model evaluation and validation
5. Model application and assessment studies.

The different projects relate in the following way to the specific tasks:

Project PI	Task	1	2	3	4	5
Ackermann		*			*	
Barone				*		*
Berkvens				*		
Borrego						*
Brandt		*				*
Brocheton		*	*			
Builtjes					*	*

Project PI	Task	1	2	3	4	5
Christensen		*	*			*
Davies		*	*			
Ebel		*		*		*
Feichter			*			
Gross						*
Jonson		*				*
Kelder			*			
Keller						*
Kisselev						*
Knoth				*		*
Langmann		*	*			*
Langner				*		*
Law			*			
Lazić						*
Makar				*		
Matthijsen		*				*
Mensink		*				*
Nester					*	*
Reimer		*				*
San Jose				*		*
Schaller					*	
Seibert				*		
Smiatek		*				
Stockwell		*				
Syrakov		*		*		
Tilmes		*			*	*
Zimmermann, J.					*	
Zimmermann, P.			*			
Zlatev		*		*		*

The following remarks have to be made:

- A new project has been added: “Regional modeling of particulate matter” by Jan Matthijsen.

- The project: “Lagrangian and Eulerian modeling of acidifying species over the Italian region” by Giuseppe Calori has been ended because of change in position of the PI.

- The project “Development of a data-assimilation scheme for chemical constituents suitable for operational applications” by Jürgen Rissmann has been ended due to change of position of the PI.

- Fabien Brocheton has replaced Gerard Toupance (who has retired) as PI of the project “Sensitivity studies to the representation of the anthropogenic emissions in global CTM models”.

- Viatcheslav Kisselev has replaced Vladimir Kousnetsov as PI of the project “Mesoscale model estimates of sulphur, nitrogen deposition and critical loads of soil acidity”.

- No annual report over 2000 has been provided by Davies, Gross, Kelder, Langner, Schaller, Toupance/Brocheton and J. Zimmermann. This means that of the expected 36 annual reports for GLOREAM over 2000, 29 have been written. The originally two projects of Nester and Builtjes have been combined to one project each.

4.2 Authors and titles of the individual reports

Development of a new particulate model to study the impact of traffic emissions on air quality

A contribution to subproject GLOREAM

Ingmar J. Ackermann, Benedikt Schell, Christoph Kessler and Heinz Hass

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Summary

In order to be able to quantify tropospheric particle concentrations and to investigate the transport and transformation processes of particulate matter over Europe the Modal Aerosol Dynamics model for Europe (MADE) has been developed. The model includes treatment of primary as well as secondary particles together with the relevant dynamical processes shaping the particle size distribution and is coupled to a gas-phase chemistry transport model.

Applications of this model on the regional scale have been performed for the period April – September 1995. Scenarios for the year 2010 are currently calculated.

Aim of the research

In order to achieve their scientific tasks state-of-the-art air quality models should be capable of predicting particulate matter in addition to the gas-phase concentrations. A suitable aerosol model for the application in complex regional transport models has to:

- provide sufficient information on the chemical composition as well as on the size distribution of the atmospheric particles.
- be coupled to a photochemical model to be able to represent the interactions between the gas phase and the particle phase.
- cover the size range of atmospheric particles, i.e. several orders of magnitude.
- be computationally efficient to keep the combined model system applicable.

The Modal Aerosol Dynamics Model for Europe (MADE) has been developed as such an aerosol model based on the Regional Particulate Model (RPM; Binkowski and Shankar, 1995) and successfully applied within the EURAD model system to the simulation of tropospheric aerosols over Europe (Ackermann *et al*., 1998, 1999; Schell *et al*., 1999, 2000).

Activities during the year

The major task was to perform long-term simulations, i.e. covering the growing season rather than a short episode, in order to investigate the variations of PM over Europe with respect to the concentration levels and the source distributions. The period April - September 1995 was chosen as a reference upon which scenario simulations will be set up. Scenarios will test certain parameterizations but also predictions of future emission reductions will be used to investigate their impact on air quality with respect to particulate matter.

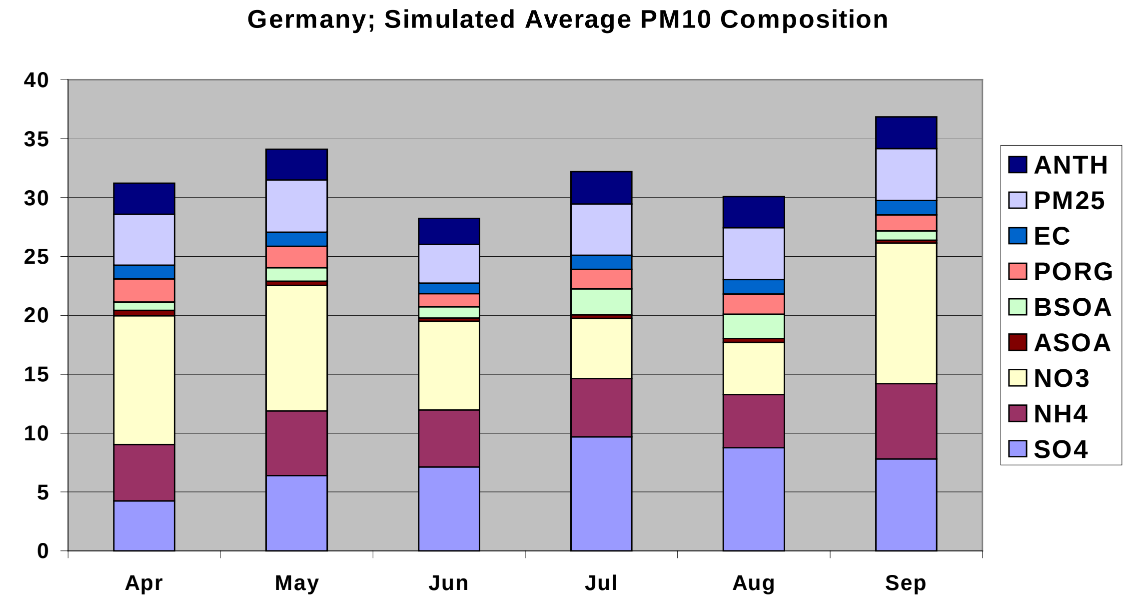

Figure 1. Monthly averaged particle load over Germany as simulated with EURAD/MADE for 1995. Particle mass is decomposed into sulfate, ammonium, nitrate, anthropogenic and biogenic secondary organic aerosol, elemental carbon, primary PM2.5 and coarse mode fraction (anth).

Principal results

- The period April – September 1995 has been simulated with EURAD/MADE for a modeling domain covering Europe with a horizontal resolution of 27 km.
- A scenario for 2010 using emission projections from the European Auto-Oil-2 has been initiated and is currently being finished.
- Preliminary analysis of the 1995 simulation shows the dominance of the secondary particles to the simulated particle loads (see Figure 1 for Germany).
- Preliminary comparison with available measurements of PM10 indicate a trend for underestimation but major events are reproduced (see Figure 2).

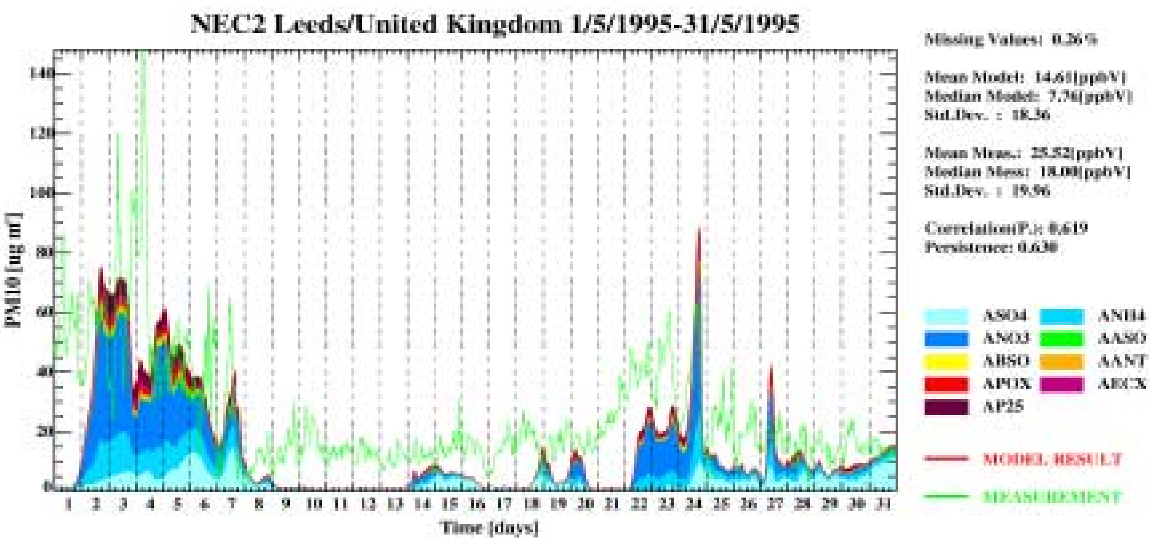

Figure 2. Comparison of model results with measurements from Leeds, UK for May 1995.

Main conclusions

First simulations – including sensitivity studies and emission reduction scenario calculations - with a full aerosol system and a modeling domain covering all Europe with a fine horizontal resolution, including primary and secondary particles throughout the whole size range of PM10, have been performed. The applications show the importance of processes which can not be neglected or simplified (sedimentation or N_2O_5 hydrolysis).

Aim for the coming year

Uncertainties in parametrisations that still exist in the model – e.g. for nucleation processes, N_2O_5 hydrolysis, etc – have to be investigated further. Measurement data from field campaigns that are becoming available will provide the opportunity to test the model performance in particular with respect to those processes that are associated with high levels of uncertainty.

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An operational model for air quality simulations over the Campania Region

A contribution to subproject GLOREAM

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Summary

Air quality models are used as the most powerful and the only scientifically relevant tools for identifying effective strategies to improve air quality. In the past, eulerian chemical transport models have been successfully used to simulate pollutant dynamics over many urban areas. This kind of studies were aimed at providing guidance to air quality management agencies; many European countries and local administrative authorities used computer simulations as basic support of their environmental policy.

In this report we describe the design of a limited-area operational air pollution model. This software has been primarily intended for policy applications on the Campania Region (in Southern Italy). It has been designed to run on heterogeneous parallel systems.

Aim of the research

During the last years, many national and local environmental protection agencies have been developing and testing increasingly complex models to address air pollution problems. The aim of this work is usually to assess, through the use of such models, the cost-effectiveness of environmental regulatory acts. Following that trend, the Office of Scientific Research and the Environment Office of the Campania Regional Board (Campania is one of the Italian's regions) recently funded a research program for developing an operational model. This model covers some aspects of air pollution modeling application, namely scenario simulations, episodic studies, monitoring and forecasting on a short time scale. Moreover, since recent advances in computer technologies allow the redesigning of air pollution models, there was a strong emphasis by the Regional Offices to encourage the use of advanced parallel computers. Our software has been targeted for a Beowulf parallel computer.

This activity has been carried out at the Center for Research on Parallel Computing and Supercomputers (CPS-CNR), in Naples, by an interdisciplinary team of atmospheric chemists and computational mathematicians.

Activities during the year

The system consists of three main, uncoupled, subsystems: the meteorological driver, and emission pre-processing model and a chemical transport model, respectively. All the subsystems have been embedded in a computational framework, whose main function providing integration among the different components and a user-friendly interface.

The meteorological subsystem is the public domain software MM5-V3 (the fifth-generation Mesoscale Model, version 3, Dudhia *et al*., 2001). It has been widely used by many research and institutional centers as an operational tool for weather forecasting. MM5 provides the three-dimensional fields of the wind velocity vector, temperature, pressure, friction velocity, etc. The wind velocity vector is used to predict the mean transport of chemical pollutants in the modeling domain; temperature and pressure provide quantitative information on the stability of the atmosphere and allows one to estimate mixing heights, turbulent diffusion coefficients and the kinetic rates of chemical pollutants.

Initial and boundary conditions are generated from data of ECMWF center (European Center for Meteorological Weather Forecasting) using objective techniques.

A commercial Geographical Information System (ArcView™) and the public domain Vis5D (Vis5d, 1999) software is employed for emission data processing and visualization (either of input and output data).

An emission inventory has been historically defined as a compilation of emission data and descriptive information for sources of air pollution over the Campania region. With few exceptions, emission inventories developed for regional air quality studies are based on an emission model. The activity level or throughout, can be defined as a measure of production rate, fuel consumption, or another indication of source activity level (e.g., tons of fuel burned). An emission factor is an emission rate used with an activity level or throughput rate to estimate emissions. Emission factors are determined by measuring emissions, usually from more than one source within a source category, under varying conditions. For the present study we used the Corinair suggested emission factors. The emission model comprises numerous sub-models for estimating emissions for each source category, and for spatially disaggregation, temporally allocation, chemically speciation, and size-classification of emitted pollutants.

In the present work emissions are estimated for the Campania region during 1995. Estimates are specified using activity levels and emission factors for the following sectors:

1. Road traffic
2. Biogenic emissions

Currently we are at work to estimate emissions from residential heating and industrial combustion and processes. The sub models account for emissions of total organic compounds (VOCs), NO_x and CO.

The chemical transport model solves the continuity equations, defining the time evolution for each chemical compound (42 compounds). The continuity equation is expressed in the following form:

$$\frac{\partial C_i}{\partial t} = -u \cdot \nabla C_i + \nabla \cdot \left(K \cdot \nabla C \right) + E_i + D_i + R_i \qquad i = 1, N$$

This equation takes into account the primary emissions, the dry deposition, the effect of transport and dilution due to wind and the photochemistry. We have not taken into account cloud/fog physics, aerosol physics and chemistry, aqueous phase chemistry, wet deposition. The total duration of a simulation is of the order of some days (the typical duration of a photochemical smog event). Nesting capabilities have been added to capture steep gradients over predefined areas (e.g. urban areas).

Since the identification of the most effective reduction strategy may encompass a large number of simulation, computers equipped with parallel architecture are used. Parallelism is introduced by a two-dimensional grid partitioning, cutting the computational domain along the *x*- and *y*-directions. The application of a dynamic load balancing strategy, where a load balancing algorithm is executed periodically to determine a new and more balanced data partitioning among available processors, allows to get efficiency and scalability. The RSL (Runtime System Library, Michalakes, 1997) is employed for domain decomposition and refinement, local address space computation, distributed I/O and interprocessor communication. RSL is the interface between PNAM and the low-level message-passing library MPI (Gropp *et al*., 1999).

The system has been embedded into a computational framework which consists of an interface between the user and the system, as well as a set of low-levels modules for managing data exchange among the various components. Major functions will be embedded in three specialized components: the *data manager*, the *strategy manager* and the *system monitor*.

The *data manager* provides the interface between the various subsystems. It will typically perform internal data exchange, internal import and export, and file format conversions. Moreover, it will allow access to experimental data gathered by the Environmental Office at the Campania Regional Board.

The *strategy manager* is designed for supporting non-specialist end-users in describing, designing, executing and analyzing an air quality simulation. Using this tool, the user will be able to perform a “what if ….?” analysis, by changing landuse parameters, emission factors, initial and boundary conditions, photolysis rates, etc. The strategy manager is also able to store and retrieve large data collection of simulations from the system database. The extent and flexibility and effectiveness in the analysis of input and in the interactive exploration of simulation results is a critical issue in the design of the strategy manager component.

The *system monitor* provides supervising and state tracking capabilities on the execution of different software components.

Preliminary results have been published elsewhere (Barone *et al*., 1999, 2000).

Main conclusions

During the last year, activities have been devoted to the implementation of a modeling framework to study air pollution problems over the Campania region. Our strategy has been based on the integration of well-known software for the simulation of meteorological scenarios, and data analysis and visualization. The design of the modeling system includes a meteorological driver (MM5), an emission model and a chemical transport model. A graphical user interface allows data display and analysis and a rapid “what if” analysis.

The system has been designed to run on advanced, low cost, easy to use, parallel computer, namely a cluster of PC’s, that can be reasonable implemented and operated even at a small, local environmental office.

Aim for the coming years

We have followed a rather conservative approach in designing this system. Many advanced features will not be included in this system, mainly in the science behind the chemical transport model and the emission model, for instance aerosol chemistry, wet deposition, higher-order closure for turbulent transport, etc. The very reason for such a choice is that we are aware that there is a serious lack of data for the Campania Region. This prevents a reliable simulation or parameterization of many chemical and physical processes. In the coming years our aim is provide the local environmental authority and the scientific community with an improved operational tool.

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Advanced Numerical Simulation for Photochemical Dispersion Models

A contribution to subproject GLOREAM

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Summary

Replacing operator splitting with coupled operators combined with approximate matrix factorization leads to a remarkable reduction of the numerical errors without increasing the computational costs. The combination of two-way nesting with domain decomposition and parallelization is expected to lead to considerable savings in terms of both computer time and wall-clock time.

Aim of the research

The CWI/NWO-project `Advanced Numerical Simulation for Photochemical Dispersion Models' is aimed at developing efficient techniques for air-pollution models in general, and in particular for the TM3-model (URL http://www.phys.uu.nl/~peters/TM3/TM3S.html, URL http://www.cwi.nl/cwi/projects/TM3/) (and its successor TM5) used at IMAU and KNMI. This aim is pursued along two directions. One is the development and implementation of innovative numerical algorithms, the other is the development and implementation of new software tools.

Activities during the year

Because the numerical time integration of all the processes at the same time with standard numerical methods is too expensive (in terms of CPU time), operator splitting is widely used to solve the processes one by one within a timestep. However, this splitting introduces splitting errors which may be considerable. For this reason we have done research into algorithms which avoid operator splitting as much as possible and which are still cheap enough. Several algorithms have been implemented for the processes of vertical mixing (cumulus convection and turbulence) and convective wet deposition, (photo)chemistry, emissions, and dry and large-scale wet depositions. From numerical experiments on a realistic and challenging 1-D transport-chemistry scenario, we observed that an algorithm with fully coupled operators clearly outperforms the algorithms with operator splitting. Furthermore, theoretical considerations and numerical experiments on linear test cases indicate that further accuracy improvement or cost reduction may be possible. About this research we have written two CWI reports and several (submitted) publications, as well given presentations on various conferences.

A detailed new program structure and new data structures were designed for parallelized versions of the chemical transport model codes TM3 and TM5. In TM5 a method for two-way nesting is combined with domain decomposition and parallelization. Implementation of the enhanced codes has started.

Principal results

Several algorithms with various degrees of operator splitting have been applied to the combined processes of vertical mixing and convective wet deposition (T), (photo)chemistry

(R), emissions (E), and dry and large-scale wet depositions (D) in a 1-D vertical setting. They are briefly named and characterized as follows:

- ORI: 1st-order splitting E-R-D-T-E-R-D-T-E-R-D; 3-hour steps for T, 2-hour steps for E and D, 2-hour steps divided into 1 to 30-minute substeps for R;
- FIR: 1st-order splitting T-P, where P=R+D+E; half-hour steps for T and P;
- SYM: 2nd-order splitting P-T-T-P; half-hour steps for T and P;
- SRC: 1st-order splitting P+S(T), S(T) denotes the effect of T alone as a source; half-hour steps for T and P;
- CPL: no splitting (so fully coupled) P+T; half-hour steps for P+T; splitting at the linear algebra level (approximate matrix factorization).

In the above, the processes T, E, R, D, and P are updated with the second-order Runge-Kutta method ROS2 (Verwer *et al.*, 1999), except for ORI where T, E, and D were updated with first-order methods. Roughly speaking, the degree of splitting in the methods decreases in the given order of the methods. The methods FIR, SYM, SRC and CPL use the same computer time and memory. The costs for ORI, the original algorithm from TM3, are approximately the same for a system with 19 cells in the vertical column and 38 chemical species.

From a challenging 1-month case study with strong emissions and photochemistry, we observed the following. ORI gave unacceptably large errors. Changing the step sizes to half an hour for all processes led to considerable error reduction and gave acceptable results (species-averaged relative errors under 10 except at a few isolated points in time) for FIR, SYM, and SRC. Coupling all the operators again caused a substantial reduction of the errors for CPL (Berkvens *et al.*, 2000).

Theoretical considerations have led to two variants of approximate matrix factorization which are expected to enhance the method CPL. The first one reduces the costs and is expected to be still accurate enough, the second is expected to give more accurate results with equal costs (Botchev and Verwer, 2000).

A program structure and various data structures have been designed for domain decomposition and parallelization (DDP) of the code for the chemical transport model TM3, and for its successor TM5 which is enhanced with two-way nesting (TWN) (Berkvens *et al.*, 1999). DDP involves the distribution of the computational domain (i.e. the grid describing the atmosphere) over a set of parallel processors and the communication of I/O and results among these processors. TWN adds extra communication among the processors, which is related to the exchange of data between nested regions. For given resolution requirements, TWN is expected to reduce the necessary computer time, and DDP is expected to reduce the necessary wall-clock time. Based on a computation and communication model we expect reasonable to good speed-ups on a parallel machine.

From a model for the computational and communicational aspects of the TM5 code with two-way nesting, we expect reasonable to good speed-ups after the implementation of domain decomposition and parallelization.

Main conclusions

For the 1-D problem of vertical mixing, chemistry, emission, and deposition, the numerical errors of the original algorithm can be significantly reduced by using improved algorithms with the same costs. Reducing timestep sizes to half an hour (rather than 3-hour steps for one process) gives a significant reduction of the numerical errors, even when (1st-order) operator splitting is still used. Coupling all the operators (and instead using approximate matrix factorization) gives yet another strong reduction of the numerical errors.

Aim for the coming year

On the software level, the effort will be directed at finishing the implementation of an efficient parallel version of the TM3-code and its successor TM5. On the algorithmical level, the effort will then be directed toward further efficient coupling (unsplitting) of vertical mixing and chemistry on the one hand and advection on the other. We also plan to implement a technique which keeps the chemistry solution both positive and mass conservative.

Acknowledgement

This work has been done within the NWO project no. 613-302-040.

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Sensitivity Analysis of the Surface Resistance Parameter

A contibution to subproject GLOREAM

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Summary

During the year of 2000 the dry deposition module of the mesoscale model system MAR IV was under validation and sensitivity analysis studies. The parameterisation of surface resistance was of principal concern. The model system was applied to a domain containing the Great Lisbon Area (GLA) with four different values for the surface deposition parameter, based on measured values and also values found in the literature. Results show a better model performance for a surface resistance value of 1041 $h.m^{-1}$.

Also, the University of Aveiro have analysed an ozone episode over Madrid, in response to a question from the Technical University of Madrid.

Aim of research

The principal objective as a contribution to the subproject was the validation of the dry deposition module into the mesoscale model system MAR IV, described in the last year annual report (Barros, 1999), more specifically the surface resistance (*Rc*) parameterisation. The region under simulation was the GLA (area of simulation of 200 x 200 km^2, with a grid of 4 km x 4 km in resolution) and the available data for validation were measured during the field campaign LisbEx 97 (Borrego *et al.*, 1999). A significant number of meteorological and air quality stations acquired data for further analysis and model validation exercises.

Concerning the air pollutants fluxes between the atmosphere and surface two methodologies were applied in the field measurements: the eddy correlation and concentration gradient methods (Vermeulen, 1998). The fluxes used on this work were measured at Baldios, 70 km ESE from Lisbon, above a flat terrain covered with herbaceous vegetation.

Activities during the year

As a side activity, but considered important in the context of understanding the ozone problem over Europe and the Iberian Peninsula, some results from the study of ozone episode Madrid were presented in the GLOREAM 2000 Workshop, in Cottbus, Germany. The presented results consist of vertical profiles interpolations, direction S-N, based on radiosondes taken in the following WMO identifier: 8430 (Murcia, Sp), 8221 (Barajas, Sp), 8023 (Santander, Sp), 7110 (Brest, F) and 3808 (Camborne, UK). At this moment, other vertical profiles, direction W-E, were constructed, also based on radiosonde measurements taken in the following WMO stations: 8579 (Lisbon, Pt), 8221 (Barajas, Sp), 8301 (Baleares Islands, Sp) and 16560 (Cagliari, It). The vertical profiles W-E allow better understanding of the atmospheric dynamics that possibly have brought ozone from the stratosphere down to the surface over Madrid. Due to the fact that this subject is not part of the University of Aveiro task in the GLOREAM project, results will not be shown in this annual report.

For the main GLOREAM work, simulations were performed for the domain in a typical summer synoptical circulation, initialised with a clean air condition, characterised by an extension of the Azores anticyclone over the northern part of the Iberian Peninsula and to the location of a low pressure system to the west of the British Isles. A thermal low was created at the high and arid central plateau of the peninsula, producing a weak ($3\ m\ s^{-1}$) N-NW wind over Portugal. Strong insolation promoted the formation of mesoscale circulation.

The MAR IV system was applied in the study area with a horizontal resolution of $4 \times 4\ km^2$. In the vertical direction the grid consisted of 28 non-equidistant layers until 8 km for the mesometeorological system module and 7 layers up to 3 km for the photochemical transport system module, in both simulations.

Four different *Rc* values were used in the photochemical model application. In the first one, a literature value was used. This value was obtained in a chamber specifically designed for the study of gaseous exchanges (Killus *et al.*, 1977). In the other applications were used *Rc* values measured at Baldios. It was considered the average, maximum and minimum values for the 9 of July of 1997. This parameterisation was evaluated with deposition fluxes measured at Baldios in the referred day.

Table 1. Surface resistance (*Rc*) values used in simulations.

Parameter	Value	$h\ m^{-1}$
Rc	Literature	53
	Average	1041
	Minimum	382
	Maximum	3061

Principal results

The comparison between numerical results and observed meteorological data was done by quantitative error analysis methodology (Pielke, 1984). Therefore, if ϕ_i e ϕ_{obs} are individual predictions and observations at the same grid point, respectively, ϕ_0 e $\phi_{0\ obs}$ are the average values of ϕ_i e ϕ_{obs} at a level, respectively; and #N is the number of observations, then:

$$E = \left\{ \frac{\sum_{i=1}^{\#N} (\phi_i - \phi_{iobs})^2}{\#N} \right\}^{1/2} \qquad E_{UB} = \left\{ \frac{\sum_{i=1}^{\#N} [(\phi_i - \phi_0) - (\phi_{iobs} - \phi_{0obs})]^2}{\#N} \right\}^{1/2}$$

$$S = \left\{ \frac{\sum_{i=1}^{\#N} (\phi_i - \phi_0)^2}{\#N} \right\}^{1/2} \quad \text{and} \quad S_{obs} = \left\{ \frac{\sum_{i=1}^{\#N} (\phi_{iobs} - \phi_{0obs})^2}{\#N} \right\}^{1/2}$$

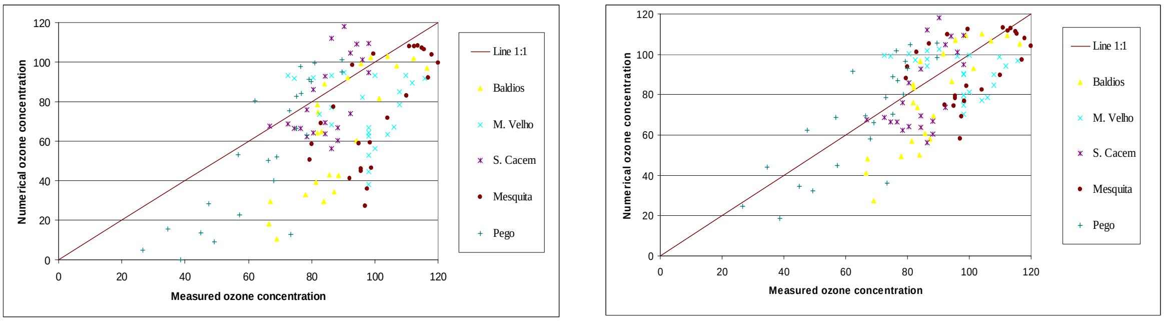
where E is the root mean square error (rmse), E_{UB} the rmse after a constant bias is removed and S e S_{obs} the standard deviations of the predictions and observations, respectively. Skill is demonstrated when: $S \approx S_{obs}$, $E < S_{obs}$, e $E_{ub} < S_{obs}$.

Table 2. Error analysis of model predicted temperatures for 12 meteorological stations.

Station	Parameter	S/Sobs	E/Sobs	Eub/Sobs
TE-Pego		1,30	1,37	1,09
Santarem		1,44	0,77	0,74
Lisboa		1,54	0,83,	0,83
Lavradio		1,75	0,98	0,97
C. Malha		1,73	1,16	0,94
Mesquita	Temperature	0,27	0,90	0,91
Setubal		0,78	0,54	0,45
Baldios		1,45	1,07	0,69
Évora		0,98	0,49	0,42
Beja		1,07	0,55	0,47
Alcacer		1,13	0,54	0,53
Sines		1,00	0,58	0,57
Average		**1,20**	**0,81**	**0,72**

A scatter analysis was made in order to assess the *Rc* parameterisation effects on the photochemical model performance. This analysis was done with literature and average *Rc* values versus measured values considering five stations in the study area.

Comparing the dispersion of observed values in Figure 1 - A and B, it is shown a better performance for the model with average *Rc* value parameterisation with a good agreement between measured and numerical values.



A B

Figure 1. Measured ozone concentration versus photochemical model results for five monitoring stations. Line 1:1 is also shown.

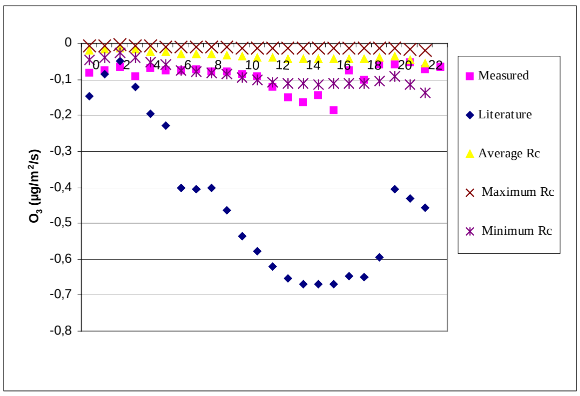


Figure 2. Ozone fluxes deposition values obtained in numerical simulations and measured at Baldios for literature value application and *Rc* parameterisation.

A point to point comparison between measured and numerical ozone deposition fluxes obtained with numerical simulations using *Rc* parameterisation is presented in Figure 2. A better model performance is shown for values resulting from numerical simulations using *Rc* parameterisation with measured values. The application using literature value overestimate the ozone deposition fluxes.

Table 3. Error analysis of model predicted ozone fluxes deposition for Baldios.

Applications	S/Sobs	E/Sobs	Eub/Sobs
Literature	5,91	11,17	5,41
Average Rc	0,30	1,95	0,92
Maximum Rc	0,10	2,52	0,97
Minimum Rc	0,84	1,00	0,98

Quantitative error analysis methodology, already explained to meteorology, was applied to deposition fluxes. Table 3 presents the results where *Rc* literature values application has poor results. From *Rc* parameterisation using measured values, the minimum value presents best results showing a good correspondence with reality.

Main conclusions

The model presents good results for mesoscale circulation. The *Rc* parameterisation with values measured at Baldios presents a better performance for the photochemical model resulting in a correct ozone masse balance. Ozone deposition fluxes show a better agreement with measured values for this parameterisation. Nevertheless, this validation methodology should be done with more deposition data, which means more field campaigns.

Aim for the coming year

Analysis of the performance of the meteorological model and the concentration and deposition fields obtained with the simulations performed with the model system MAR IV with the best value encountered for the parameter surface resistance applied to the dry deposition module. Also, a map of critical loads for Portugal will be presented based on an empirical approach.

Acknowledgements

The authors gratefully acknowledge the Portuguese Foundation for Science and Technology and Environment General Directorate for the funds given in the following projects: “Atmospheric Environment in Coastal Zones: Assessment of the Load Capacity of the Ecossystem – AMAZOC (PRAXIS 3/3.2/AMB/38/94)” and “Atmospheric Circulations and Photochemical Production over the Lisbon Region (JNICT/DGA PEAM/P/AMA/603/95)” and finally for the PhD grant of Ana Cristina Carvalho (PRAXIS XXI /BD/ 21474/99).

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Modelling of transport, dispersion and deposition; Operational air pollution forecasts on regional and urban scales

A contribution to subproject GLOREAM

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Background

The development of a new system for operational forecast modelling of transport, dispersion deposition and chemical transformation was started in 1998. The system is called the DMU-ATMI THOR air pollution forecast system. It is an integrated operational air pollution forecast system on hemispheric scale, regional scale, urban background scale and urban street scale. Currently, the system consists of a coupling of a numerical weather forecast model, Eta, the long range air pollution transport models, DEHM, covering the Northern Hemisphere and DEOM, covering the whole of Europe, an urban background model, BUM, and an operational street pollution model, OSPM. The system produces operational 3 days air pollution forecasts for the most important air pollution species, four times every day, on European scale, on urban background scale and on urban street scale.

The low-cost operational system integrates urban and regional models in a system for air pollution forecast, monitoring, scenarios, control and management, in support of decision makers and various environmental and energy policy actions. The system will support the accomplishment of the EU directives relating to air pollution limit values for human health and give the foundation for improving the quality of urban and rural life. The system will provide the necessary tool for the authorities to inform and/or warn the public and, in the future, to carry out the needed action (as e.g. restrictions on traffic) during episodes where the air pollution levels are exceeding the critical limit values. Furthermore, the system will be a part of the national monitoring programs at DMU-ATMI, both in urban and rural areas. A demonstration of parts of the system can be seen at the web-address: http://www.dmu.dk/AtmosphericEnvironment/thor.

Progress in 2000

Air pollution forecasts for the cities of Copenhagen and Aalborg, Denmark

The air pollution forecast system has been implemented and validated for the cities of Copenhagen and Aalborg, Denmark, in cooperation with the city authorities. For the city of Copenhagen, the uban background model was implemented and the operational street pollution model was implemented for a single street (Jagtvej) in Copenhagen. Operational air pollution forecasts were made available to the public in April 2000. These forecasts can be seen in the web site: http://luft.dmu.dk.

The urban background model was further developed to handle the different emission sources in the Aalborg area. Emissions were subdivided into a grid with a resolution of 1 km x 1 km. Contributions from the individual emissions are integrated along the wind direction path assuming linear dispersion with the distance to the receptor point. Horizontal dispersion is accounted for by averaging the calculated concentrations over a certain, wind speed dependent, wind direction sector, centered on the average wind direction using a Gaussian distribution. The OSPM model was extended to handle ten different streets in the center of Aalborg. The model results from the urban models have been compared to measurements from the two measurement stations in Aalborg (one at roof and one at street level). These stations are part of the Danish urban monitoring network (LMP). The system now produces operational air pollution forecasts, four times a day, for the ten streets and for the urban background.

Comparison of five Eulerian air pollution forecasting systems for the summer 1999 using the German ozone monitoring data

Eulerian state-of-the-art air pollution forecasting systems on the European scale are operated routinely by several countries in Europe. DWD and FUB, both Germany, NERI, Denmark, NILU, Norway, and SMHI, Sweden, operate some of these systems. The modeling systems are applied, e.g. for regulatory purposes according to new EU directives. An evaluation and comparison of the model systems was carried out in order to assess their reliability, see Tilmes *et al.* (2000). The model forecasts from all five systems have been compared to measurements of ground level ozone in Germany. The outstanding point in this investigation was the availability of a huge amount of data – from forecasts by the different model systems and from observations. This allowed for a thorough interpretation of the findings and assures the significance of the observed features. Data from more than 300 measurement stations for a 5-month period (May–September 1999) of the German monitoring networks was been used in the comparison. Different spatial and temporal statistical parameters were applied in the evaluation. Generally, it was found that the most comprehensive models gave the best results. However, the less comprehensive and computational cheaper models also produced good results. The extensive comparison made it possible to point out weak points in the different models and to describe the individual model behavior for a full summer period in a climatological sense. The comparison also gave valuable information for an assessment of individual measurement stations and complete monitoring networks in terms of the representativeness of the observation data.

Development of a hemispheric nested model for studying air pollution phenomena in general

A new 3-D model REGINA (REGIonal high resolutioN Air pollution model) is under development. The model is based on models developed over the last decades at NERI. The goal is to obtain a nested model capable of high resolution operation. The domain of the REGINA model is the Northern Hemisphere with several nests implemented currently covering the European and Scandinavian areas. High resolution data from nested runs with the MM5 model carried out at NERI are used for the meteorological input. The emission data used in the model are a combination of national high resolution emission data and data from the EMEP data base. Data from the GENEMIS database are to be included.

The model will be applied for studying air pollution phenomena (monitoring, forecasting and scenarios) over Denmark where there are extensive coastal areas that require a high resolution model in order to resolve the effects of e.g. land-sea interactions. It will also be applied within the Danish Background Monitoring Programme. One of the objectives of this programme is to identify the various nutrient sources giving input to the sea-areas surrounding Denmark.

The horizontal transport in the model is solved using an Accurate Space Derivative (ASD) algorithm. This method traditionally requires periodic boundary conditions, which are not applicable for nested modelling. Therefore, a new method for calculating non-periodic boundary conditions has been developed. The numerical solution to the chemistry part of the model is obtained from an implementation of a new combination of two existing numerical methods.

Extensive testing of the numerical solution of the advection and the coupling of the solution of advection and chemistry in the model has been carried out using the Molenkamp-Crowley rotation test. The same test has been applied to the model with and without nesting. The results show that the numerical methods are suitable for modelling air pollution levels on high resolution.

The further work with the development of the model includes a thorough validation with the measurements from the EMEP measurement station network.

Development of a hemispheric nested model for CO_2

As a part of the AEROCARB EU-project (Airborne European Regional Observations of the Carbon Balance), a three-dimensional Eulerian hemispheric and nested model for transport, diffusion, and surface fluxes of CO_2 has been developed. This model is a further development of the Danish Eulerian Hemispheric Model (DEHM), which was initially designed for studying transport of SO_2 and sulphur into the Arctic (Christensen, 1997). DEHM is a hemispheric model covering the Northern Hemisphere with a 150 km x 150 km spatial resolution. The model was further developed to handle CO_2, by implementing fossil emissions based on GEIA (Andres *et al.*, 1996), vegetation fluxes based on Fung *et al.* (1987) and ocean fluxes based on Takahashi (1999) and Wanninkhof (1992). Model results were tested against measurements, showing very good performance (Geels *et al.*, 2001a).

The model was then extended with a two-way nested domain covering Europe with a 50 km x 50 km spatial resolution at 60^o north. A new scheme for the time integration and a new way of treating of the boundary conditions have been implemented and tested. The predictor-corrector time integration scheme previously used in the model has been substituted with a new 3^{rd} order algorithm based on Taylor series expansions and variable time step (the latter depending on the Courant-Friedrich-Levi stability criteria). This decreased the memory requirements, which is important when applying the model with multiple nests and many species.

The spatial discretization in the model is based on Accurate Space Derivatives (ASD). This method is a precise numerical method for solving the horizontal transport. However, originally it required periodic boundary conditions, which is not applicable for nested modelling. Therefore, a new method of treating the boundary conditions in a non-periodic way was developed (Frohn *et al.*, 2000c). This new combination of the ASD method with non-periodic boundary conditions was tested using the Molenkamp-Crowley rotation test on multiple nests and showed to work very well (Frohn *et al.*, 2000b; 2000c).

Previously, the hemispheric model was run with meteorological data from ECMWF with a 2.5^o x 2.5^o resolution. Higher spatial model resolution, however, requires meteorological data with higher resolution, especially in the nested domain. Therefore the MM5 model (Grell *et al.*, 1995), developed at NCAR and Penn State University, has been implemented as a meteorological driver for the DEHM model. The MM5 model is run in a nested mode for the same domains and applied on the same projections as in the DEHM model. Furthermore, higher resolution topography and land use data (both with 30 sec resolution) from USGS were implemented in the MM5 model.

New CO_2 emissions based on Edgar (1^o x 1^o resolution) (Olivier, 1996) were implemented in the model and model results were compared to the previously used GEIA emissions. Furthermore, new monthly vegetation fluxes based on the TURC model (Ruimy, 1994) were implemented. The model results obtained using TURC fluxes was compared to results obtained when vegetation fluxes based on Fung *et al.*, (1987) were applied. Results from these sensitivity studies are presented in Geels *et al.* (2001b). New visualization programs have been developed and/or implemented for visualization of the model results from the hemispheric and European domains in 2-D as well as 3-D. In the latter case the Vis5D visualization tool was coupled to the model. The developed nested model has been applied and tested for CO_2 and Radon-222 for the two months July and December 1998. Different simulations were carried out for use in the model comparison exercise in the project.

Development of a high resolution nested meteorological model for air pollution modelling

Part of the THOR system is the weather forecast model Eta. The horizontal grid resolution currently used in the weather forecasting model is approximately 40 km x 40 km. This is a rather coarse resolution for application in air pollution models covering an area like Denmark. In a country that can be defined as mainly coastal, coastal effects in atmospheric chemistry model studies are difficult to predict using a grid resolution at 40 km. Air pollution studies for Danish conditions could therefore be improved by applying a meteorological model with finer resolution. The chosen solution is a nested version of the currently operational weather forecast model Eta. Better description of precipitation over Denmark is expected to improve the wet deposition and better description of wind fields near coastal areas could improve air pollution forecast in such areas. An example of such area is the city of Copenhagen.

Until now, different versions of the model have been tested. A version of the model with higher horizontal resolution, a version of the model where the number of layers are increased and a version with both higher resolution and increased number of layers have been tested. Some preliminary comparisons against measurements from Danish airports have been carried out. However, longer model runs and more comparisons with measurements are needed.

It is planned to change the source of land use data for a more detailed description of the surface. This change will introduce a more precise description of roughness lengths and is expected to improve the near surface winds that are important in urban air pollution forecasting.

Although the topography in Denmark is quite small compared to most other European countries, orographic precipitation over Denmark gives large differences in different parts of Denmark. This is observed in the measurements. A precise description of precipitation rates is important for e.g. the modelling of nitrogen deposition to Danish waters. The orographic precipitation over Denmark is only roughly obtained with the present resolution in the operational Eta model. By changing both vertical and horizontal resolution and improving the input data in the model, it is expected to obtain orographic effects in the distribution of precipitation.

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Modelling regional differences in tropospheric ozone and aerosols - the reliability of model results

A contribution to subproject GLOREAM and TOR-2

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Summary

The 3-D Eulerian grid model LOTOS (LOng Term Ozone Simulation) which has been used often to calculate ozone formation over Europe over longer time periods (years) on an hour-by-hour basis has been extended with the aerosol module MADE/MARS to calculate the formation of inorganic secondary aerosols. The module MADE/MARS has been received by Ford-Aachen in the framework of a joint project/cooperation. Model results for both ozone and aerosols have been analysed and compared with observations for especially august 1997, but also for other time periods including the year 1994 and 1998. The model results show large regional differences not only for ozone, but also for secondary aerosols. High nitrate concentrations are calculated over the Netherlands and surroundings, and over the Po-valley. High sulphate concentrations are found in the eastern part of Europe.
Both statistical analysis and data-assimilation has been used in relation to ozone to determine the reliability of the model results. The combination of these two methods turned out to be a useful approach for model validation. Satellite observations from Aerosol Optical Depth (AOD) over august 1997 have been combined with LOTOS calculations of AOD making use of data-assimilation. The resulting AOD-fields over Europe show large spatial differences which will be further analysed.

Aim of the research

The aim of the research is to determine and analyse the processes which result in the differences in ozone and aerosol patterns over Europe, and to make and apply a general methodology for model validation.

Activities during the year

Model calculations have been performed with LOTOS both for ozone and related species and sulfate, nitrate and ammonium aerosols. Model results have been analysed for especially august 1997, the test period of the GLOREAM subproject. A statistical analysis has been performed for modelled ozone concentrations in comparison with EMEP and UBA stations for august 1997 using fractional bias, fractional standard deviation and fraction of data within a factor 2. In general, the maximum ozone concentrations were underestimated by the model, and the spreading of simulated concentrations is smaller than observed. The statistical analysis gives information concerning both the model behaviour and the behaviour of the observations. This screening by statistical analysis has subsequently be used in the application of data-assimilation (the extended Kalman filter technique). Uncertainty (noise) has been added for all grid cells for NO_X and VOC emissions, for photolysis rate and for the surface resistance of O_3. The assumed uncertainty in observed ozone was taken as 5 ppb. In applying data-assimilation, the original residue between observations and model results over all stations was decreased from about 12 ppb to 6 ppb. The combination of statistical analysis and data-assimilation is benifical for model validation studies.

Satellite observations of AOD have been combined with the AOD of ammonium-sulfate and -nitrate as calculated with the LOTOS model for august 1997. Preliminary results show that the data-assimilation scheme is properly working also in this application, and that estimates can be made of the relative contribution of sulfate and nitrate to the AOD and their regional differences.

Principal results

- The validation of the LOTOS model for ozone for august 1997 using a combination of statistical analysis and data-assimilation.
- The combination of satellite observed AOD with by LOTOS calculated AOD by means of data-assimilation.

Main conclusion

The combination of statistical analysis and data-assimilation is a promising method for model validation. Satellite observed AOD can be combined by applying data assimilation with modelled AOD to interprete the observed AOD.

Aim of the coming year

- To improve the LOTOS-model in the calculations of aerosols, including organic and primary aerosols.
- To improve our understanding in the causes of the regional differences in ozone and aerosol concentrations over Europe.
- To improve our understanding and experience in using data-assimilation, and in performing model validation for both ozone and aerosols.

Ackowledgement

The project is partly funded by the Dutch ministry of Economical affairs via ECN, and by the Dutch Remote Sensing Board.

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Model studies of the air pollution in the Arctic by using The Danish Eulerian Hemispheric Model

A contribution to subproject GLOREAM

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Background

The Danish Eulerian Hemispheric Model is a three dimensional air pollution model, which have been under continuous development since 1990 at the National Environmental Research Institute (NERI). The model has been used to study long-range transport of pollution on the Northern Hemisphere. The model is based on set of coupled full three-dimensional advection-diffusion equations. The horizontal space of the model is defined on a regular 96x96 grid that covers most of the Northern Hemisphere with a grid-distant of 150 km at 60oN for the mother domain and 50 km for the nested domain: Greenland and Europe, and the vertical space is defined on an irregular grid with 20 grid-points up to ≈ 15 km. The present version of the model includes the long-range transport of sulfur di-oxide (SO_2), particulate sulfate (SO_4^{2-}), ozone, hydrocarbons, nitrogen-oxides, lead and mercury species. The model has been used to study the air pollution in the Arctic for a period of more than 21 years from 1979 to May 2000. The model development is a contribution to the Danish part of the International Arctic Monitoring and Assessment Programme, AMAP. The work with the heavy metal versions of the hemispheric model also contributes to the EUROTRAC subproject MEPOP (see contribution: Dynamics and chemistry of atmospheric mercury), while the work with the photochemical model contributes to the EUROTRAC subproject GLOREAM (see contribution: Modelling of transport, dispersion and deposition; Operational air pollution forecasts on regional and urban scales).

Progress in 2000

In 2000 the work with the coupling of hemispheric model with a state-of-the-art weather prediction model to the model system has continued. This has improved treatment of the physical parameterizations. It has also made it possible to do more detailed model calculations for Europe or around Greenland with a higher spatial resolution, e.g. taking into account the influence of the orography of Greenland on both the wind and precipitation pattern. The performance of the model has clearly been improved considerably. The state-of-the-art weather prediction model has been run for a 10 year period for the hemispheric domain, and for one year period for the European nested domain.

The work with the new hemispheric model with a photochemical scheme consisting of 55 species, more than 94 chemical reactions and 17 photolyse reactions have continued in 2000. The model has been coupled to meteorology, the EDGAR emissions data on a 1^ox1^o grid have been used and these emissions are redistributed to the grid used in the model, dry deposition based on the resistance method and wet deposition based on simplified scavenging has been introduced. The model has been run for several years and the validation of the model results has continued.

The work with the further development of the mercury model has continued. A state-of-the-art chemical scheme, based on the scheme from the GKSS model has been implemented. A simplified parameterization of the mercury depletion in the Arctic during the Polar Sunrise has been developed.

A special version of the hemispheric model has been developed, which is able to do nested model calculations for a smaller area with a higher resolution coupled to the state-of–the-art weather prediction model, and where there is 2 way coupling between the hemispheric area and the smaller area, f.ex. for the area around Greenland or Europe.

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Regional air quality studies using EURAD

A contribution to subproject GLOREAM

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Summary

EURAD has been applied to scientific problems and political issues in the context of air quality planning. The work has been done in cooperation within GLOREAM and other subprojects in EUROTRAC-2 (AEROSOL, TOR-2, LOOP, GENEMIS, CMD). First relations have been made to the recently established subproject TROPOSAT based on adjoint modeling. Emphasis in the model system development has been laid on the interface with emission data, application of nesting, data assimilation, aerosols and clouds, budget calculations, updates of chemical mechanisms and improvement of numerical schemes. EURAD participated in model evaluation carried out for Nordrhein-Westfalen, the Berlin area (FLUMOB, BERLIOZ) and the alpine region (VOTALP, LOOP, PIPAPO). Data assimilation methods based on 4D-Var have been developed and applied to an episode in August 1997.

Aim of the research

The general aim of the research is the better understanding of dynamical and chemical processes in the troposphere over Europe using a complex modeling system. Special emphasis is on the support of field experiments and the evaluation of the model based on quality assured observations with the aim to improve the parameterizations of the modeled processes. Model development includes the improvement of the aerosol and cloud modules, the development of advanced data assimilation methods using adjoint modeling techniques, the improvement and application of nesting techniques, deposition, gas phase chemistry and numerical methods.

Activities during the year

The EURAD modeling system has been evaluated for a summersmog episode in July 1994 based on routine observations of the environmental agency of Nordrhein-Westfalen (LUA) and data obtained during the FLUMOB and BERLIOZ episodes in the Berlin area (Ebel *et al.*, 2000a, b; Memmesheimer *et al.*, 2000). Application to the Milano area and the nearby Alps has been undertaken within the EU-project VOTALP (valley experiment, foehn episode) and for PIPAPO in close cooperation with LOOP (Feldmann *et al.*, 2000; Seibert *et al.*, 2000). The nesting capabilities of the modeling system have been extended to consider regional and local effects with horizontal resolutions from 125 km down to 1 km (e.g. Berlin, NRW) (Ebel *et al.*, 2000a, b; Memmesheimer *et al.*, 2000).

Budget calculations (Memmesheimer *et al.*, 1997) for the Berlin region have been undertaken (FLUMOB, July 1994 and BERLIOZ, July 1998) (Ebel *et al.*, 2000a, b; Weber and Memmesheimer, 2000; Memmesheimer *et al.*, 2000) and for the VOTALP campaigns (Feldmann *et al.*, 2000). The effect of the nesting on the processes involved in the budget calculations have been studied for the area of Berlin (Ebel *et al.*, 2000a, b; Weber and Memmesheimer, 2000). The budget calculations for VOTALP and FLUMOB have been used to contribute to task 2 of the TOR project (see TOR annual reports for 1999 and 2000). The modeled data has been compared to measurement for the BERLIOZ campaigns in several joint studies (Becker *et al.*, 2001; Corsmeier *et al.*, 2001).

Advanced data assimilation methods have been developed on the basis of adjoint modeling (Elbern *et al.*, 1999, Elbern and Schmidt, 2000) within the TFS and the EC-project RIFTOZ. These methods have been applied to an episode in July/August 1997. They are used to improve initialization of air quality models, chemical state analysis, for sensitivity studies and for parameter optimization such as emission rates (Elbern *et al.*, 2000). It allows for a better understanding of chemical and dynamical processes governing the chemical state of the atmosphere on the the basis of observations.

Concerning model development major efforts have been undertaken to improve the parameterization of aerosols and clouds (Ackermann *et al.*, 1998; Friese *et al.*, 2000a, b). Relations with the subproject AEROSOL have been established to use the EURAD modeling system for the analysis of measurements and to get further ideas for model development by a close cooperation between modelers and experimentalists.

The interface to the emission data bases available within GENEMIS has been improved considerably. GENEMIS data provided by the IER, University of Stuttart, has been used to simulate the oxidant formation from the European scale to the urban scale of Berlin; landuse data genererated for Europe by the IFU, Garmisch-Partenkirchen, has been used to calculate biogenic emissions (VOCs and NO_X) for the BERLIOZ episode (Memmesheimer *et al.*, 2000).

The effect of stratosphere-troposphere exchange has been studied for several episodes and with different methods within the framework of VOTALP.

Principal results

The nesting capabilities of the EURAD model have been applied to zoom from the European Scale into highly populated, industrialized regions with high emission rates. Regions of specific interest are located in Nordrhein-Westfalen (Ebel *et al.*, 2000a, b), the Milano area (Feldmann *et al.*, 2000; see also TOR annual report 1999) and in particular Berlin (Memmesheimer *et al.*, 2000; see also Figure 1). Process analysis has been carried out for the urban area of Berlin to investigate the effect of nesting on the processes which control temporal development and spatial variations of photo-oxidants and their precursors in the Berlin plume.

The EURAD-CTM has been successfully used for the forecast of air pollutants as part of the system of the German Weather Service (Jakobs *et al.*, 2001; Tilmes *et al.*, 2001).

Aerosol dynamics and chemistry have been included into the model. First results have been obtained for a episodes in July 1994, August 1997 and January 1997 including nesting for Nordrhein-Westfalen (NRW). The nesting applications have been sucessfully extended to the nesting ratio 5 which allows to zoom from the European scale into NRW (January 1997, August 1997; more details can be found in the annual report to the subproject AEROSOL).

Data assimilation based on adjoint modeling has been tested for an episode in August 1997 (Elbern *et al.*, 1999; Elbern *et al.*, 2000; Elbern and Schmidt, 2000).

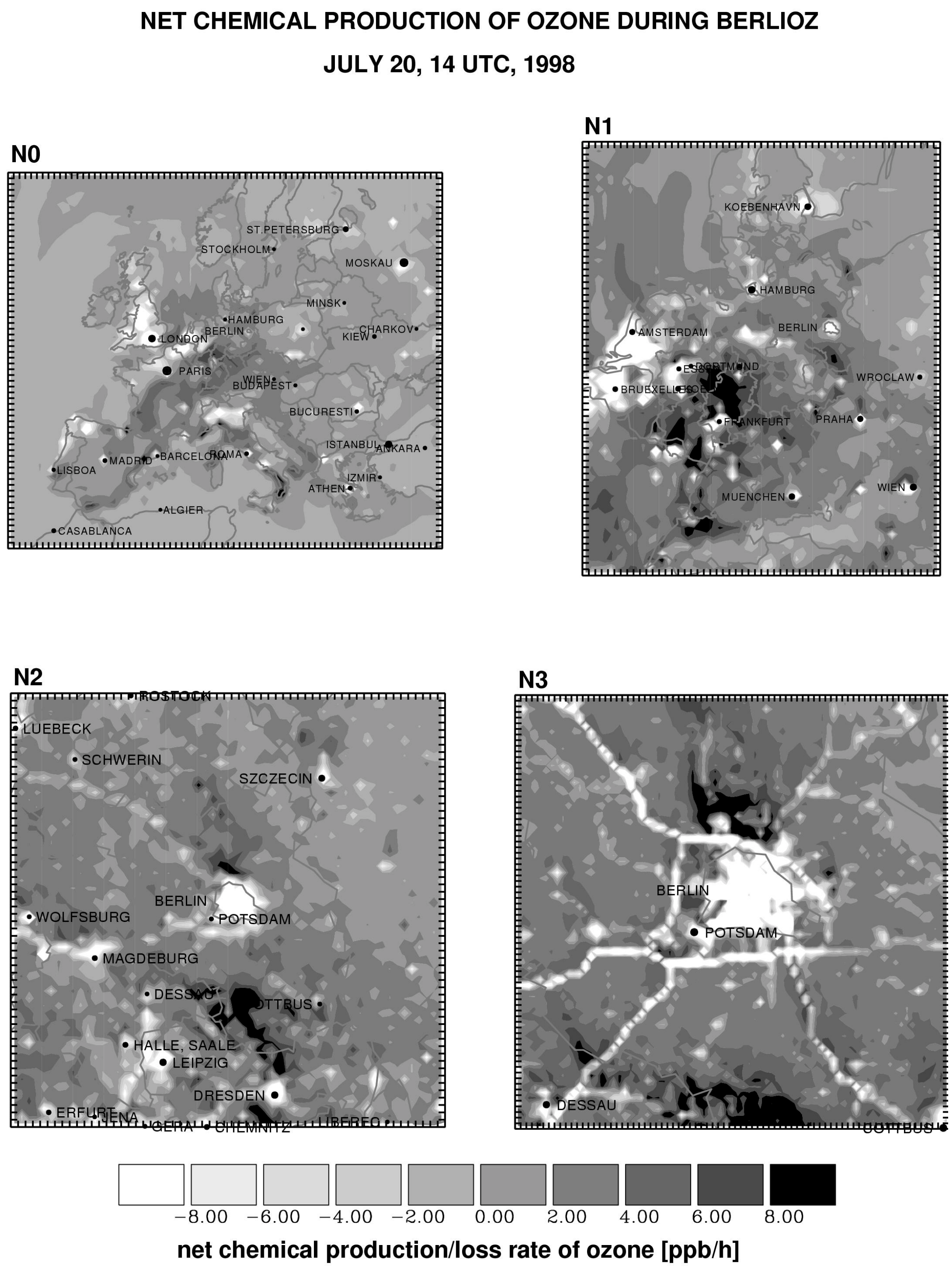

Figure 1. Net chemical production of ozone on July 20, 1998, 14 UTC for the mother domain and the corresponding nests. The results for layer 1 (about 40 m thick) are displayed. Horizontal resolution is 54 - 18 - 6 - 2 km. Regions with negative values (white) are characterized by high titration from ozone to NO_2.

Main conclusions

The EURAD modeling system has been applied successfully to various scales using its nesting capabilities. The tools available within the EURAD modeling system have been developed further to improve the understanding of dynamical and chemical processes which control the atmospheric concentration fields (graphical tools, budget analysis). Highly advanced data assimilation using adjoint modeling has been developed and sucessfully applied. The implementation of aerosol dynamics and chemistry into a 3D Eulerian modeling system allows for applications including the analysis of field experiments aiming on the characterization of aerosol patterns in different regions of Europe. EURAD is under a permanent process of evaluation which improves the knowledge of the range of uncertainty in the model results and points to possible improvements in the modeling system. The interface to GENEMIS, which has been established now allows for the calculation of emission scenarios for Europe and smaller areas of particular interest on the basis of sophisticated and permanently improved emission data. The range of applicability of EURAD has been extended and allow for the preparation and analysis of field experiments (including aerosols) as well as for the calculation of emission scenarios for air quality regulation policy on different scales.

Aim for the coming year

The EURAD modeling system will be further applied to analyse the results of the BERLIOZ episode (July/August 1998). Analysis include budget calculations, data assimilation and process oriented evaluation on the basis of observations. Additional aims are the investigation of transport of air pollutants from North-America to Europe, dynamical and chemical processes near the tropopause. Long term simulations and climatological air quality statistics based on model calculations and forecasts of the German Weather Service are planned with respect to the EU-directives. Improvement of chemical mechanisms is planned in cooperation with the Research Centre Jülich and experiments on the so-called atmospheric simulation chamber SAPHIR.

Acknowledgements

EURAD has been funded by the BMBF, Germany, within the tropospheric research programme (TFS), the aerosol research programme (AFS) and the Atmospheric Research Programme AFO2000, DG XII of the European Commission, the environmental agency of Nordrhein-Westfalen (LUA) and the German Weather Service. Close cooperation and permanent support by the Ford Research Center is gratefully acknowledged. The numerical simulations have been supported by the RRZK, University of Cologne and the Research Center Jülich (ZAM, ICG2, ICG3). Support also came from the ECMWF, DWD, UBA, EMEP, NCAR (MM5 community) and a lot of environmental agencies all over Europe. Special thanks to H. Geiß, H.-P. Dorn, T. Brauers and D. Poppe (Research Centre Jülich: ICG2, ICG3) and R. Friedrich, B. Wickert, both IER, University of Stuttgart and G. Smiatek, IFU, Garmisch-Partenkirchen, who provided important input data as emissions and landuse data.

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Study of the Feedback Mechanisms between Climate Change and the Chemical Composition of the Atmosphere

A contribution to the subproject GLOREAM

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Summary

We simulated using a global chemistry model the CO distribution for the period 1978 until 1994. In this model experiment different CO sources have been colored in order to assess the impact of different source types and source regions on the European air pollution. The seasonal behaviour of these transport processes and interannual variability will be analysed as well.

In order to improve the capability of our atmospheric global model to simulate ozone distributions correctly we implemented the chemistry package MOZART which includes a rather extensive hydrocarbon chemistry.

Aim of the research

In order to develop mitigation strategies we have to know which sources and source regions control the pollutant load over Europe. Ozone concentrations can be considerably influenced by remote sources and in particular CO undergoes due to its long life-time transcontinental transport.

Hydrocarbon chemistry has a considerable impact on ozone concentrations and has to be taken into account in a state-of-the-art global chemistry model.

Principal results

The CO tagged tracer study has been finished recently and the analyses of the budgets of different CO tracers over Europe is under way. First publications are expected until the end of the year.

The model version simulating CO has also been successfully applied to do chemical weather forecast during the TRACE-P measurement campaign. We performed real-time forecast simulations of global meteorological and tracer fields with the global 3-dimensional climate model ECHAM for two weeks prior and during the deployment of the TRACE-P field experiment. Standard maps and animated sequences have been automatically generated and made available for use in the field in order to assist the science team in detailed flight planning. One scientist from the Max-Planck-Institute has accompanied the DC-8 aircraft to all major bases of operation for this purpose. The simulations are based on ECMWF forecast fields and have been performed in T42 resolution (~ 2.8° x 2.8°) with 19 vertical levels. Additional simulations shall be performed after the mission in order to provide the science team with consistent model fields for the complete experiment time frame. We marked tracers according to their region of origin and performed a study on the budget of these tracers in the TRACE-P region. Evaluation of the simulated tracer fields shall be performed in close collaboration with the experimentalists from the science team.

Description of the chemistry model MOZART (by Horowitz)

MOZART-2 (Model of Ozone and Related Chemical Tracers, version 2) is a global chemical transport model designed to simulate the distribution of tropospheric ozone and its precursors. The model simulates the concentrations of 58 chemical species from the surface up to the lower stratosphere. The model can be driven with a variety of meteorological inputs, including data from a general circulation model or a meteorological reanalysis, such as that from NCEP or ECMWF. Meteorological parameters, including zonal and meridional winds, temperature, specific humidity, surface pressure, and surface fluxes of heat and momentum, are taken from the ECMWF reanalyses data base and are provided to MOZART every six hours. The chemical scheme used in MOZART-2 is an updated version of that used in MOZART-1 (Brasseur *et al.*, 1998). Kinetic reaction rates have been updated based on recent measurements, as compiled by Sander *et al.* (2000). The chemical mechanism includes oxidation schemes for the non-methane hydrocarbons: ethane, propane, ethene, propene, isoprene, a-pinene (as a surrogate for all terpenes), and n-butane (as a surrogate for all hydrocarbons with 4 or more carbons, excluding isoprene and terpenes). The isoprene oxidation mechanism is updated from that used by Brasseur *et al.* (1998), based on work by Brocheton (1999) and Horowitz *et al.* (1998). Heterogeneous reactions of N_2O_5 and NO_3 (as well as methylvinyl ketone and methacrolein) on sulfate aerosols are included in MOZART, with the sulfate aerosol distribution prescribed based on a sulfate aerosol mass simulation performed in MOZART-1 (Tie *et al.*, 2001). Photolysis frequencies are computed using a precalculated multivariate interpolation table, giving clear sky photolysis frequencies as a function of pressure, overhead ozone column, solar zenith angle, surface albedo, and temperature profile. Photolysis frequencies are adjusted for cloudiness by applying a cloud correction factor, as described by Brasseur *et al.* (1998). The chemical system is solved numerically using a fully implicit Euler backward method with Newton-Raphson iteration.

First results are presented in Figure 1 which shows a comparison to surface observations at two sites over Europe.

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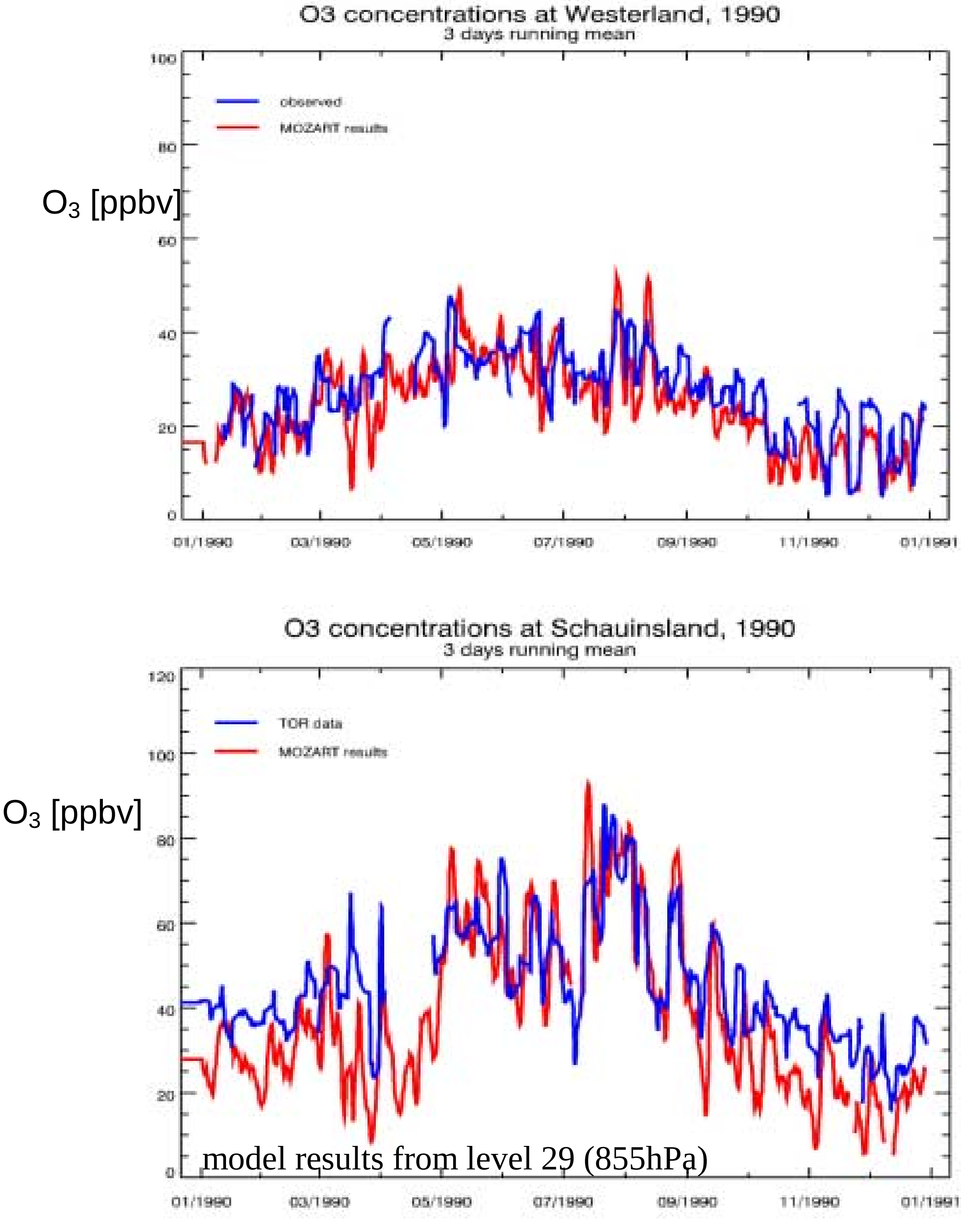

Figure 1. Observed and calculated ozone mixing-ratios in ppbv.

Status on the Development and Application of the EMEP Regional Photochemistry Model

A contribution to subproject GLOREAM.

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Summary

In 2000 model runs have been made investigation the effects of sea salt particles on the total nitrate distribution. The inclusion of sodium nitrate particles resulted in higher calculated concentrations of total nitrate (HNO_3 + particulate nitrate) at remote costal sites. Furthermore, the effects of emissions from international shipping have been studied. For several European countries these emissions contribute significantly to pollution levels.

Aim of the reaearch

The aim of the research in 2000 has been to improve the calculations by the EMEP model. Furthermore to assess the importance of emissions from international shipping relative to other sources.

Activities during the year

In 2000 extensive revisions have been made in the code. The main purpose of the revisions is to get a more flexible code with regard to chemistry scheme, chemical solver and model domain. This work will continue in 2001.

Emissions from international shipping, estimated by Lloyd's Register of Shipping, representative for 1990 for NO_x, SO_2, CO and NMVOC are now available for all sea areas surrounding Europe. The effects of these emissions on European pollution levels have been calculated with the EMEP Eulerian model.

A simple scheme describing the turnover of sea-salt particles has been included in the EMEP Eulerian photochemistry model. This scheme has also been extended to include the formation of sodium nitrate particles. Calculated concentrations of total nitrate are under-predicted by the EMEP model, in particular in remote coastal areas. Both sea salt and sodium nitrate are treated as bulk parameters with mean diameters of 6 and 3.2 μm by mass respectively. We assume that the formation of sodium nitrate is limited by the mass transfer of gaseous HNO_3 to the surface of the sea salt particles.

Principal results

Our calculations show that for European countries close to major shipping routes significant fractions of the depositions can be attributed to emissions from ships. For several countries the contribution to the depositions of sulphur and oxidized nitrogen are close to or larger than 10%, even in areas with large land-based emissions as Belgium and the Netherlands. The increase in the depositions caused by international shipping also contribute significantly to the exceedances of critical levels for acidification and nutrient nitrogen in Europe as defined in the 1999 protocol to ”Abate Acidification, Eutrophication and ground level ozone". The total annual depositions of sulphur and oxidized nitrogen and the contribution from international shipping are shown in Figures 1 and 2.

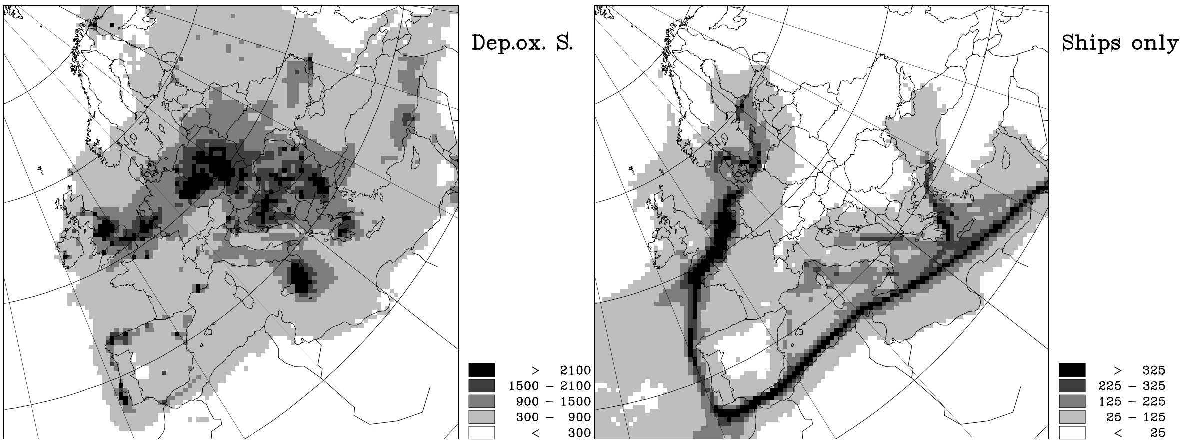

Figure 1. Annual deposition of sulphur in (mg(S)m^{-2}. Left, total deposition with all sources included. Right, the contribution from international shipping only. (Notice the difference in scale)

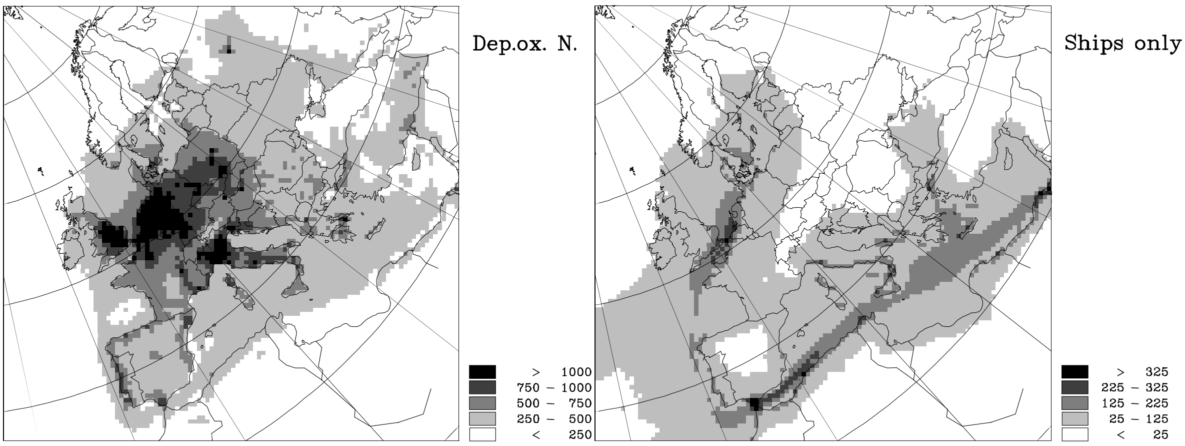

Figure 2. Annual deposition of oxidized nitrogen in (mg(N)m^{-2}. Left, total deposition with all sources included. Right, the contribution from international shipping only. (Notice the difference in scale)

There are also marked effects in ozone levels and in the exceedances of threshold values for ozone. Increases in ozone levels, and accumulated critical levels for ozone exposure, are mainly confined to the countries with a Mediterranean coastline. In and around the English Channel, the North Sea and the Baltic Sea, a combination of moderate to high emissions and low insolation, leads to decreases in mean July concentrations and accumulated critical levels for this component. This work is documented as an EMEP note (Jonson *et al.*, 2000b), and was presented at the GLOREAM workshop in Cottbus, September 2000.

Sea salt concentrations are compared to measurements for several sites in Norway. Both measured and calculated sea salt concentrations are mostly well below 1 μg m^{-3} (as Na). At most sites the calculated concentrations are over-predicted, but mostly within a factor of 2, compared to the measurements. In the North Atlantic monthly averaged concentrations are typically in the 10 to 15 μg m^{-3} range in January and the 5 to 10 μg m^{-3} range in July. In the surface layer over the ocean the calculated fraction of total nitrate in the form of sodium nitrate is often more than 90%. Over the European continent the calculations indicate that only a small fraction of the total nitrate is in the form of sodium nitrate in winter. However, in summer a significant fraction of the total nitrate is calculated as sodium nitrate. With the formation of sodium nitrate included, calculated total nitrate levels are higher in remote coastal areas where the total nitrate has been transported over the sea. Even so, calculated concentrations are in general lower than observed is these areas. This work is documented in Jonson *et al.* (2000a).

Main conclusions

Our calculations show that emissions from international shipping has a marked effect on European pollution levels. Through the 1999 protocol to ``Abate Acdification, Eutrophication and ground level ozone" control measures to reduce emissions will be, or are already implemented. As a result land-based emissions in Europe will decline in the future and the relative contribution from international shipping is likely to increase. Control measures for international shipping are the main concern for the “International Convention for the Prevention of Pollution from Ships” (MARPOL). However, this convention is likely to have a small effect only, and additional measure to reduce the emissions from international shipping in the waters surrounding Europe are under consideration.

The inclusion of sodium nitrate in the calculations results in an increase in total nitrate levels in remote coastal areas. However, compared to measurements model results are still in general underestimated in these areas.

Aims for coming year

The revision of the model code will continue in 2001. This will be followed by a careful analysis of model results obtained with the revised model version.

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Modelling of Air Quality in Switzerland and Northern Italy

A contribution to subproject GLOREAM

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Summary

Threshold values of indicator species and ratios are suitable to delineate the transition between NO_X- and VOC- sensitivity of ozone formation (Sillman, 1995). On the basis of various model simulations for two different locations (the area of Switzerland by the Urban Airshed Model, UAM, and San Joaquin Valley of Central California by SARMAP Air Quality Model, SAQM) a new approach for defining VOC-insensitive locations was examined. We found that these threshold values are not universal, but depend on the emission level and on the meteorology.

The UAM used for various studies in the past was replaced by a new software package. This package consists of the meteorological pre-processor SAIMM (Systems Applications International Mesoscale Model) and the photo-chemical dispersion model UAM-V which both have variable grid capabilities (SAI, 1995; SAI, 1999). Substantial difficulties were encountered until the program was capable of delivering reasonable results. Applied to the complex topography of Switzerland, the original version showed unexpected spatial distributions of trace gases. For instance, the mixing ratio of CO used as a quasi-inert tracer was found to increase with height. The reason was an incorrect formulation of the transport schemes.

The corrected version was used to estimate the influence of uncertainties in the meteorological input for SAIMM on the input fields of UAM-V and on the distribution of the ozone mixing ratio. In the frame of the EUROTRAC-2 subproject LOOP (Limitation of Oxidant Production) the model is currently applied to the Milan area to investigate NO_x/VOC limitation issues.

A collaboration with the LOTOS (Long Term Ozone Simulation) group at TNO Apeldoorn (NL) was established. The joint activities deal with the influence of the European air quality simulated with the LOTOS model on the trace gas mixing ratios in the Swiss model domain. In a preliminary study, the dependence of the ozone mixing ratio in Switzerland on the choice of the concentrations at the domain boundaries was estimated. For seasonally varying boundaries, a full year LOTOS data set for 1998 was made available.

Aims of the research

The objective is to simulate the spatial and temporal distribution of air pollutants for regional domains including Switzerland and to estimate their impacts on ecosystems. Particular interest is devoted to emission scenarios related to conversion processes in energy systems (e.g. power plants, vehicles, etc.). The influence of emissions in Switzerland as well as in neighbouring countries are addressed.

Activities during the year

Indicator-based assessment of ozone sensitivities

The threshold values of indicator species and ratios delineating the transition between NO_X- and VOC- sensitivity of ozone formation derived by Sillman (1995) are assumed to be universal by various investigators. However, previous studies performed on the basis of UAM simulations suggested that the threshold criteria may vary according to the locations and conditions (Andreani-Aksoyoglu and Keller, 1997). Hence, threshold criteria derived from various model simulations at two different locations (the area of Switzerland by the UAM and San Joaquin Valley of Central California by the SAQM) were examined using a new approach for defining VOC-insensitive locations. A range of boundary slopes (the ratio of O_3 reductions with NO_x control to O_3 reductions with VOC control) as indicators of NO_x- and VOC-sensitive locations was taken and the dependence of threshold values for indicators and indicator ratios such as NO_y, O_3/NO_z, $HCHO/NO_y$, and H_2O_2/HNO_3 on these boundaries was investigated. These threshold values were found to depend on the emission level and on the meteorology.

Tests and modifications of the SAIMM/UAM-V package

After having used the UAM for air quality modelling at PSI for the last few years (e.g. Andreani-Aksoyoglu and Keller, 1998), we procured the updated version UAM-V with variable grid capability. The meteorological data required by UAM-V are prepared by the pre-processor SAIMM. The model package is widely used in the U.S. in the frame of air quality programs. UAM-V creates two output data sets for each pollutant: the mixing ratio (ppb) in the lowest layer averaged over 1 hour, and the instantaneous concentration (µmol / m^3) in all layers at the end of each hour. We tested the SAIMM / UAM-V package for the complex Swiss topography. The size of the model domain was 470 km x 385 km with 5 km grid cell size and 8 atmospheric layers up to 3000 m a.g.l.

Using the long-lived CO as a tracer, we found that the instantaneous mixing ratio increased inexplicably with height. It became evident that UAM-V does not take into account expansion and compression of trace gases when they are transported from one level to another. We converted the trace gas concentrations of each grid cell to a common pressure and temperature before the mixing processes due to vertical and horizontal turbulent exchange, advection and entrainment occur. After these modifications the pressure and temperature dependence of the mixing ratios completely disappeared.

Sensitivity of meteorology and ozone mixing ratio to the input parameters of SAIMM for the Swiss topography

The SAIMM/UAM-V package requires surface data and vertical soundings as well as an initial sounding for the whole domain. A nudging procedure forces the simulated meteorological quantities towards the input fields. For simulations in the current model domain of Switzerland hourly data of the meteorological network ANETZ operated by the Swiss Meteorological Institute are used as surface inputs. The vertical soundings are taken from the hourly output of the Swiss prognostic Model (SM) used for the operational weather forecast (about 14 km x 14 km grid cell size). These data are pre-processed by SAIMM in an enlarged model domain containing 68 ANETZ stations and 29 x 20 SM grid cells. In order to estimate the influence of the number of surface stations and soundings on the simulated meteorology and the distribution of the ozone mixing ratio, we reduced the number of SM soundings and ANETZ stations. In the worst case, only 1 surface station and 1 sounding were taken.

Transboundary air pollution

In a preliminary study boundary values based on literature were compared with a dataset provided by LOTOS to investigate the effect of transboundary pollution on the ozone formation in Switzerland using UAM-V. The air quality in regional model domains may be significantly affected by the mixing ratios of the pollutants at the boundaries. Four reduction scenarios were designed to estimate the influence of NO_x and VOC emissions reductions on the ozone formation. In the view of a seasonal modeling, LOTOS data have been made available for whole 1998.

Air quality simulations in the Milan area (LOOP)

Meteorological data from various surface stations, one sounding station and one wind profiler located in the LOOP domain around Milan as well as prognostic data from the SM were used to generate reasonable SAIMM windfields for UAM-V. It turned out that the meteorological fields of the SM for the simulation period (May 11–13, 1998) differ significantly from the measured data. Hence, the SM data was omitted and the vertical wind pattern was derived mainly from the windprofiler data. The uncertainties of the model input parameters, (e.g. the soil moisture availability) caused a significant spread of the turbulent exchange coefficient leading to uncertainties in the temporal and spatial distribution of the pollutants. Most of the time was spent by creating a representative base case for the subsequent scenario simulations.

Principal results

Indicator-based assessment of ozone sensitivities

The ratio H_2O_2/HNO_3 is a successful indicator providing a better separation of NO_X - and VOC-sensitive ranges than the others. The variation of threshold criteria under two different meteorological and emission conditions shows that both perturbed cases (reduced emissions and less-stagnant meteorology) lead to similar shifts in threshold criteria towards more NO_x -sensitive chemistry. Although H_2O_2/HNO_3 provides a good separation, threshold ranges for this indicator ratio are affected by emission and meteorological perturbations (Andreani-Aksoyoglu *et al.*, 2000).

Tests and modifications of the SAIMM/UAM-V package

Using the long-lived CO as a tracer, we found that the instantaneous mixing ratio increased inexplicably with height. For instance, a test run was initialised at noon with 160 ppb CO in the layers 1 and 2 and 140 ppb in the layers 3 to 8. After 2 hours of simulation the mixing ratio increased up to 200 ppb in the upper layers. In addition, the Swiss topography was mirrored in the horizontal CO distribution (Figure 1). The concentration field (in $\mu mol/m^3$), however, did not show this effect. After numerous calculations it became evident that UAM-V does not take into account expansion and compression of trace gases when they are transported from one pressure level to another. In applications linked with air quality regulations it is usually sufficient to consider the lowest layer only. On the one hand the mixing ratios in this layer are strongly influenced by the emissions. On the other, most model domains investigated so far do not exhibit large altitude variations. Under these conditions the shortcomings mentioned above are invisible.

We converted the trace gas concentrations of each grid cell to a common pressure and temperature before the mixing processes due to vertical and horizontal turbulent exchange, advection and entrainment occur. After these modifications the pressure and temperature dependence of the mixing ratios completely disappeared (Keller *et al.*, 2000a).

Sensitivity of meteorology and ozone mixing ratio to the input parameters of SAIMM for the Swiss topography

If all surface and sounding data are included in the input data set for SAIMM (maximum case), the *wind pattern* of the model is very similar to the SM wind field because of the strong nudging effect. On the other hand, the wind field is substantially distorted and the average wind speed increases if only 1 surface and 1 sounding data are used. The inclusion of all 68 surface stations and of 1 sounding decreases the wind speed due to the lower surface velocities, but the pattern is similar. We found an optimum number of 8 x 5 SM soundings. The difference to the maximum case is not significant; this is valid for all layers.

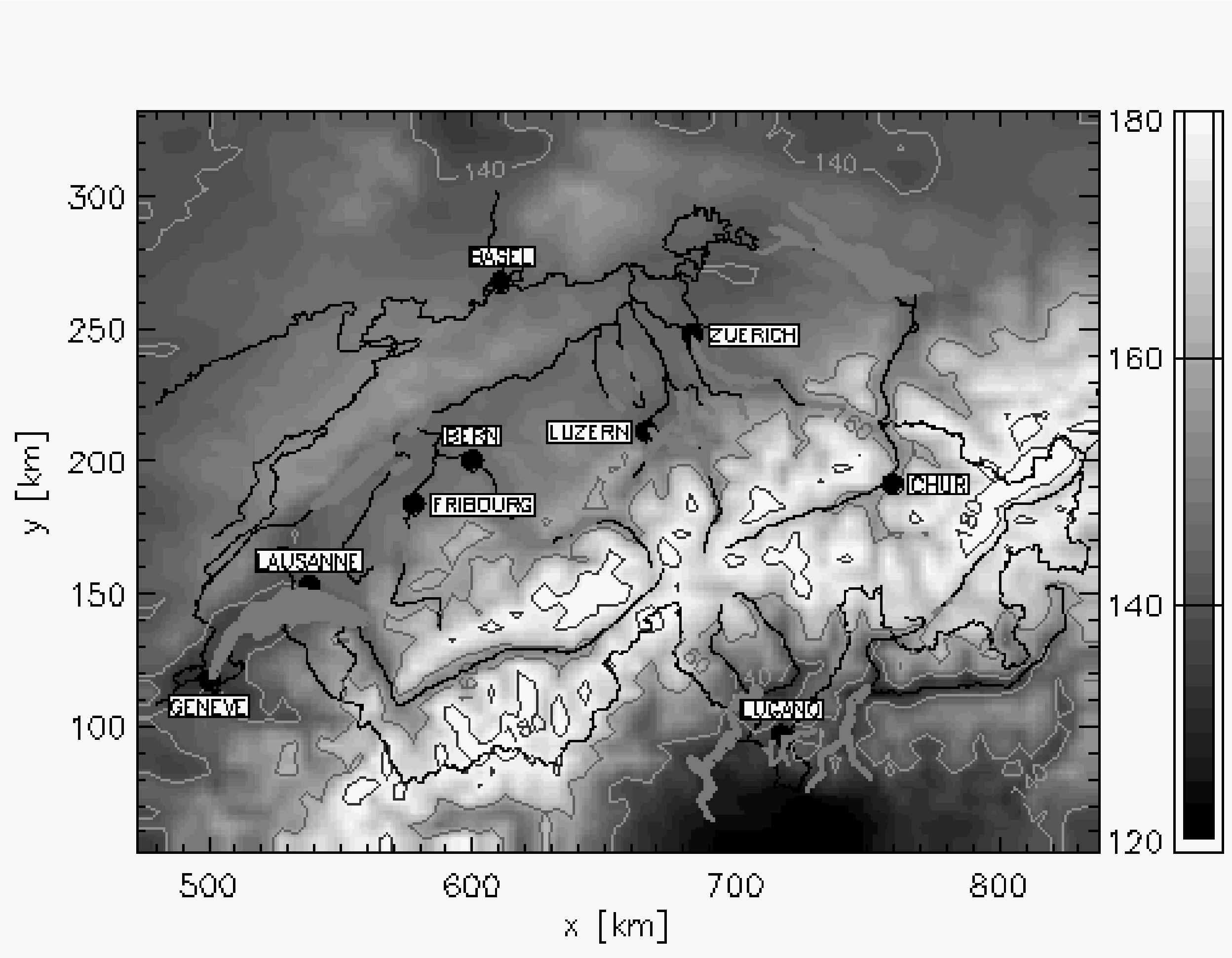

Figure 1. CO mixing ratio (ppb) of layer 7 (2000 to 2500 m a.g.l.) on July 28, 1993, 14:00 CET. The simulation started 12:00 with a constant mixing ratio of 140 ppb.

The *vertical diffusivity* k_v in the complex domain of Switzerland is highly variable. In the afternoon, we found values up to 150 m^2/s over water and >350 m^2/s over land. At that time the diffusivity is usually higher over mountain ridges than over flat terrain and water surfaces. If the model runs with minimum nudging (1 surface station, 1 sounding) the distribution is distorted and the absolute values decrease in the lower layers. The vertical profiles change as well, leading to modified mixing conditions in the lower troposphere.

The combination of distorted windfields and modified mixing layers influences the transport of pollutants. As an example, the city plumes of *ozone* are significantly distorted. The mixing ratio at a specified site may decrease or increase by up to 20 ppb if this site is located outside or inside the plume, respectively (Keller *et al.*, 2000b).

Transboundary air pollution

The boundary O_3 data from the LOTOS model shows lower concentrations at night compared to the boundary concentrations based on literature. The literature based boundary concentration for NO_2 (1-7 ppb) are significantly higher than the LOTOS data (0.3-0.8 ppb). The literature based boundary conditions reflect moderately polluted areas in Europe, while the LOTOS model data in the west of Switzerland reveal concentration patterns typical for remote rural sites. The same differences as for NO_2 can be observed for the VOCs.

A reduction of the anthropogenic emissions of NO_x and VOC lead to changes in ozone mixing rations and to the regions characterized by NO_x and VOC sensitive ozone production. The LOTOS boundaries yield slightly larger areas with VOC sensitive conditions in and downwind of the plumes and smaller regions with NO_x limited ozone production (Ritter *et al.*, 2000).

Air quality simulations in the Milan area (LOOP)

Due to the inconsistency of the experimental data and the prognostic fields of the SM it was difficult to create reliable windfields. Omitting the SM data and deriving the vertical profile from the wind profiler data lead to windfields which could be used as reasonable inputs for UAM-V. The turbulent exchange is controlled by various model parameters such as the soil moisture availability which are not clearly defined. These uncertainties contribute to the variability of the simulated mixing ratios of the pollutants and are one reason for the difficulty to match experimental and modelled data.

Main Conclusions

Indicators species and ratios are suitable for delineating VOC-insensitive an NO_x-insensitive regions. Thresholds of these quantities, however, are not universal, but depend on the emissions and the meteorology.

The commercially available version of the SAIMM/UAM-V model package is not suitable for complex topography because of an incomplete formulation of the transport schemes. This results in a unreasonable increase of mixing ratios with height. Reducing the concentrations to a common pressure and temperature level before calculating the pollutant fluxes eliminates this drawback.

On the basis of simulations with SAIMM/UAM-V for Switzerland and for the Milan area, the crucial role of realistic meteorological input data was obvious. Too few stations included in the nudging process of the mesoscale model lead to substantial distortion of the gridded windfield and the vertical exchange coefficients. An inadequate choice of the parameters controling the vertical turbulent exchange profile may substantially affect the size of the mixing layer and the levels and shapes of the pollutant's distribution. Particular care is required if there are discrepancies between prognostic data from the forecast model and data from surface stations and soundings.

Aims for the coming year

Special emphasis will be put on the modelling of the air quality in the Milan area focusing on the NO_x/VOC sensitivity. Because of the inadequacy of the UAM-V for complex topography, we intend to replace it by the recently issued Comprehensive Air Quality Model with Extensions (CAMx). This model contains modules to simulate aerosol distributions and is more flexible in terms of chemistry mechanisms. As a convenience, it uses the same input file structure as UAM-V. The results of a preliminary test are encouraging. Regarding transboundary pollution and seasonal modelling, data from the LOTOS model and the new version of the Swiss prognostic model for the full year 2001 will be procured and used as input for the photo-chemistry model.

Acknowledgements

The work was supported by the Swiss "Kommission für Technologie und Innovation (KTI)".

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Mesoscale Model Estimations of Sulphur, Nitrogen Deposition and Critical Loads of Soil Acidity

A contribution to subproject GLOREAM

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In 2000 the activity of the Institute of Atmospheric Air Protection in the field of regional pollution transport investigation was concentrated on the study of sulphur and nitrogen deposition from the atmosphere on the territory of Murmansk region of Russia, covering Kolsky peninsula and the nearby areas. Located in the northern part of Europe, close to the Scandinavian and Baltic countries, Murmansk region is subject to the influence of atmospheric transport of pollutants from abroad and in its turn influences in the same way the territories of the above countries.

The study was performed on the basis of the earlier developed climatological stochastic model of regional pollution transport. This model was used for the estimation of deposition of sulphur and nitrogen, emitted by the sources of Murmansk region, on the underlying surface with spatial resolution 25x25 km. For the estimation of pollutant fluxes from outside the region the data of Western Meteorological Synthesizing Centre with lower resolution were used. The high role of outside sources in some parts of Murmansk region required the development of special sliding average algorithm for integrated treatment of data with different spatial resolution.

The results of the study showed that the main part of sulphur deposition on the considered territory originated from local sources. For example, in 1998 the total deposition of sulphur on this territory was 71.72 thousands of tons, and 68% of this quantity (48.90 thousands of tons) was that emitted by the local sources. The influence of the sources of the other countries was comparatively small. In particular, the deposition of sulphur caused by emissions from Poland and Finland in the same year was correspondingly 1.4 and 1.3 thousands of tons, i.e. about 6% each. Of 191.2 thousands of tons of sulphur emitted by the sources of Murmansk region 28% deposited on its territory and 72% was exported outside. Finland and Norway were the countries subject to the maximum influence of the sources of Murmansk region. The deposition on their territories was correspondingly 11% and 4% of sulphur emitted from the considered region.

Deposition of nitrogen on the territory of Murmansk region was comparatively low and due to mainly outside sources. Only 3% of deposited nitrogen originated from local sources, and a considerable part of deposition originated from the sources of the other regions of Russia. Of the foreign countries the maximum effect was from the sources of Finland (about 11%), the role of other countries was lower than 5%.

The total deposition pattern of sulphur for the territory of Murmansk region is shown on Figure 1. It is clearly seen that the maximums of deposition field are localized in the vicinity of the industrial centres, in the other parts of the region the spatial distribution of deposition is close to homogeneous. For the considered period (1994-1998) there was no clear temporal trend of deposition, although total emissions from the sources of Russia decreased during this period. This behaviour of deposition values may be explained by high influence of local sources, the emissions of which do not demonstrate an expressed tendency to decrease.

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	275	292	276	296	299	260	274	268	265	254	254	253	243	239	213	207	233	230	258	247	214
	309	300	305	335	343	316	294	282	294	273	272	280	249	241	229	242	240	262	229	252	213
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	340	363	371	353	404	360	359	304	298	277	237	258	227	235	250	242	228	249	219	237	209
	427	420	411	417	407	373	344	341	338	297	281	274	251	247	234	234	238	243	225	233	208
	443	489	442	511	440	417	420	405	367	359	352	339	296	265	260	209	240	239	229	247	207
31	490	553	527	526	481	431	443	383	423	450	477	349	319	311	285	249	237	250	253	243	206
	581	590	606	545	660	850	571	445	495	552	516	431	359	334	322	269	249	269	259	250	205
	751	849	726	692	965	1486	833	517	689	690	695	555	419	370	299	286	276	270	276	257	204
	917	1798	1120	791	687	905	552	559	897	1889	1131	626	420	357	314	290	277	290	276	276	203
	1455	4207	1669	712	601	524	470	522	1474	3573	2217	740	435	390	340	280	248	274	286	259	202
	1777	**5160**	2111	698	603	513	445	432	486	1316	737	555	466	368	354	300	293	264	251	254	201
30	519	1673	619	533	473	486	438	352	371	426	445	552	445	335	334	308	266	265	262	252	200
	378	427	437	408	389	329	367	328	396	358	360	401	307	310	297	254	254	281	264	258	199
	308	325	344	355	320	317	333	354	487	405	338	322	323	313	311	271	263	283	273	265	198
	321	341	326	327	311	310	306	283	357	331	300	324	330	302	308	274	298	296	299	286	197
	315	345	348	350	327	313	344	292	295	316	313	309	318	301	321	291	296	301	309	299	196
	290	313	330	329	324	318	324	292	321	327	337	309	328	331	328	306	297	304	315	317	195
29	309	306	305	309	298	295	310	303	287	302	309	291	306	318	325	305	302	311	315	323	194
			16						**17**						**18**						

Figure1. Total deposition of sulphur on the territory of Murmansk region
(mg $S/m^2 * year$)

Parallelization of Chemistry-Transport Model MUSCAT

A contribution to subproject GLOREAM

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In chemical transport models, chemical reactions and the transport of species are described by very large and stiff systems of differential equations. To date, one limitation of schemes has been their inability to solve equations both quickly and with a high accuracy in multiple grid cell models. This requires the use of fast parallel computers. Multiblock grid techniques and implicit-explicit (IMEX) time integration schemes are suited to benefit from the parallel architecture (Wolke and Knoth, 2000). A parallel version of the multiscale chemistry-transport code MUSCAT is presented which is based on these techniques. The special grid structure of the model originates from dividing an equidistant horizontal grid into rectangular blocks of different size. Each block can be coarsened or refined separately. This is done on condition that the grid size of neighbouring blocks differ by one level at the most. The maximum size of the already refined or coarsened blocks is limited by a given maximum number of columns.

Our parallelization approach is based on the distribution of blocks among the processors. We consider a static partitioning where the blocks are distributed between the processors only once at the beginning of the execution of the program. Here, we use the number of horizontal cells (i.e., of columns) as measure of the work load of the respective block. Therefore, the total number of horizontal cells of each processor is to be balanced. This is achieved by the grid-partitioning tool ParMETIS (Karypis *et al.*, 1998). It optimizes both the balance of columns and in addition the ”edge cut”, i.e., it takes care of short inter-processor border lines. A distribution of 90 blocks to eight and 16 processors is shown in Figure 1.

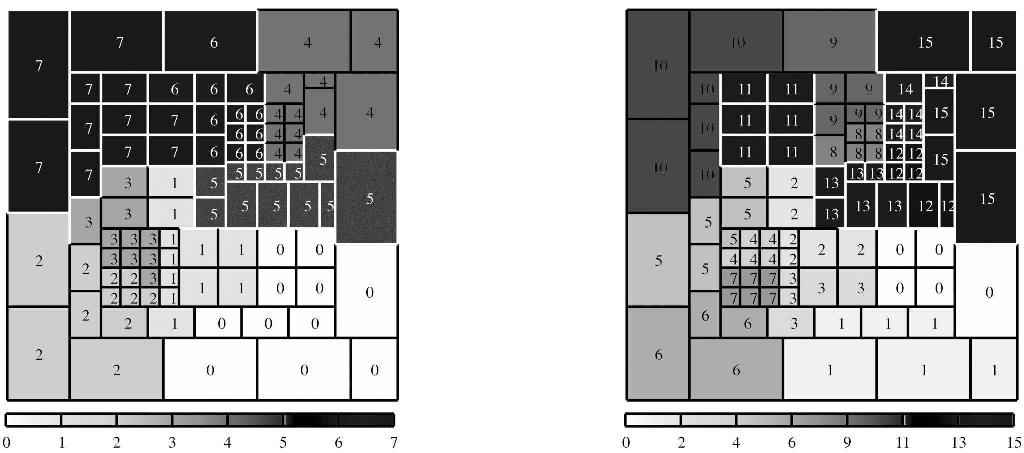

Figure 1. Distribution of the same block configuration over 8 and 16 processors.

Inter-processor communication is realized by means of the message passing interface language MPI. An exchange of data over block boundaries is necessary only once during each horizontal advection step. Each block needs the concentration values in one or two cell rows of its neighbours, according to the order of the advection scheme. The implementation of the boundary exchange is not straightforward because of the different resolutions of the blocks.

The possibilities of one cell being assigned to two neighbouring cells or of two cells receiving the same value must be taken into account. We apply the technique of ”extended arrays” where the blocks use additional boundary stripes on which incoming data of neighbouring blocks can be stored. Hence, each processor only needs memory for the data of blocks that are assigned to it.

Unfortunately the size of each block is only a crude estimate of the necessary work in the course of the integration of the chemistry-transport equations in time. These load imbalances are due to the sophisticated error control inside the used numerical algorithms. In order to improve the load balance, techniques allowing for redistribution of blocks have been implemented (Wolke *et al.*, 2000). The workload of a block work is estimated using the numbers of Jacobian N_J and function evaluations N_F applied during a past time period. They represent measures of the expense of factorizing the system matrix and solving the resulting systems. The work load of processor P is defined by

$$a_P=\Sigma_{B\in P}(N_{FB}+1.4*N_{JB})*N_{CB}$$

where the factor 1.4 balances the workload of the two parts. $B\in P$ stands for the blocks currently located on this processor, N_{CB} is the number of columns of block B. For repartitioning, we again use ParMETIS which is called if the ratio

$$\min_P a_P/\max_P a_P$$

falls below a certain critical value. According to the work loads of the blocks, ParMETIS searches for a better distribution, besides minimizing the movements of blocks. The communication required for the exchange of block data can be done by means of similar strategies as for the boundary exchange.

Figures 2c and 2d show the partitions in the beginning and the end of a twelve hour simulation using four processors. The test is performed for an ozone scenario for the Saxony area (Figure 2a) using a multiscale grid. Judging the work load, we compare the estimated work loads and the actual CPU times for runs with and without dynamic load balancing. Both values are the sum of all time steps, where in each step the ”slowest” processor is considered. The results are given for two different relative tolerances RTOL of the implicit integrator (Table 1) and have been determined on a SGI Origin 2000.

The estimated improvement in execution time through the above model is in good agreement with the actual improvement measured by the reduced CPU time. This means that the cost function is a good measure for the workload of a single block. Furthermore, Table 1 shows that for tighter tolerances RTOL dynamic load balancing leads to a larger reduction in the CPU time. The results of the 8 and 16 processors run on the CRAYT3E demonstrate the almost linear speedup of the parallel transport code MUSCAT.

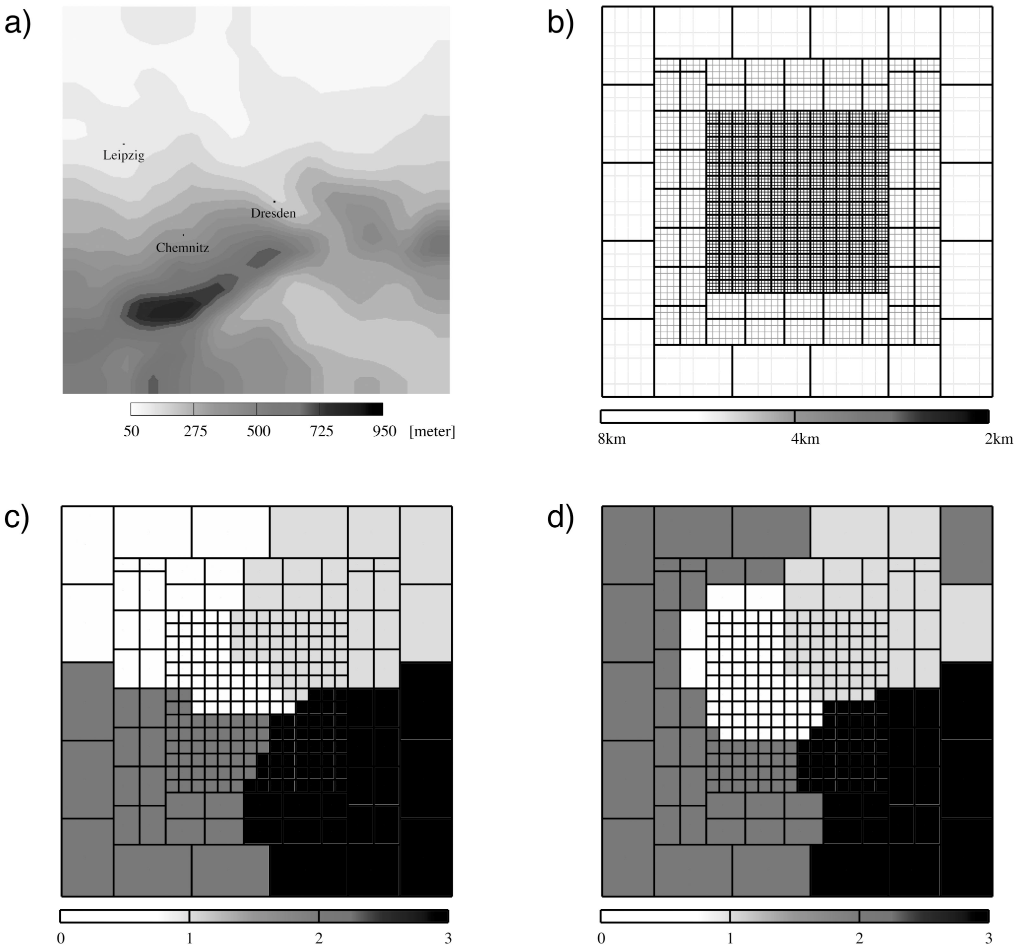

Figure 2. ”Ozone in Saxony”: Orography of the simulation area (a), multiscale 2km-4km-8km grid (b), block distribution for 4 processors in the beginning (c) and the end of a 12 hours simulation (d).

Table 1. CPU times for the ozone scenario in Saxony for different number of processors.

	CPU time		**improvement**	
RTOL	**static**	**dynamic**	**CPU**	**estimated**
	SGI (4 processors)			
1.e-2	4:17 hours	4:04 hours	94,9 %	94,2 %
1.e-3	6:57 hours	6:02 hours	86,4 %	93,6 %
	T3E (8 processors)			
1.e-2	1:18 hours	1:16 hours	98,3 %	98,7 %
1.e-3	1:48 hours	1:27 hours	80,4 %	83,3 %
	T3E (16 processors)			
1.e-2	0:45 hours	0:44 hours	97,0 %	96,9 %
1.e-3	0:58 hours	0:52 hours	89,8 %	88,7 %

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Analysis of a Summer Smog Episode in the Berlin-Brandenburg Region with a Mesoscale Atmosphere-Chemistry Model Chain

A contribution to subproject GLOREAM

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Summary

The application of the mesoscale model chain REMO(0.5^0) – REMO($1/6^0$) – GESIMA (4 km) during the FLUMOB measurement campaign in July 1994 in Berlin-Brandenburg, Germany and the analysis of the model simulations and sensitivity studies shows that

- biogenically emitted hydrocarbons dominated significantly over anthropogenically emitted ones during this specific situation,
- long range ozone transport from outside of Europe influenced ozone concentrations in the boundary layer in spite of the stagnant high pressure weather conditions during FLUMOB.

Aims of the research

- model evaluation,
- analysis of specific field campaigns,
- contribution to an improved understanding of photooxidants and other trace species distributions in the troposphere.

Activities during the year

The mesoscale model chain REMO(0.5^0) – REMO($1/6^0$) – GESIMA (4 km) has been applied to analyse extensively the FLUMOB measurement campaign in Berlin-Brandenburg in July 1994. Results are described in the submitted or already published papers of Bauer (2000), Bauer and Langmann (2000 a, b, c), Langmann and Bauer (2000) and Langmann (2000). The major results and conclusions will be presented briefly in the following sections. The BERLIOZ field experiment has also been investigated, but results will not be presented here.

Principal results

Weather conditions during the FLUMOB episode

The weather conditions during the FLUMOB measurement episode in Berlin-Brandenburg were optimal for the photochemical production of ozone and other photooxidants because of the blocking situation with high temperatures and strong solar irradiation. Due to the meteorological conditions the FLUMOB period can be divided roughly into two episodes. The first episode from July 22 until July 25, was characterised by advected air masses coming from the south-east direction. Strong vertical mixing raised the planetary boundary layer height up to about 3000 meters. During the following days the FLUMOB area was influenced by sub-tropical air masses coming from southern directions. In addition to high ozone concentrations, high amounts of precursor gases were transported to the Berlin-Brandenburg area during the second episode.

Ozone production within the Berlin-Brandenburg region

A comparison between observed and modelled concentrations (REMO and GESIMA simulation) (Bauer, 2000) close to the surface and at flight levels had demonstrated that NO_2 and urban ozone are well simulated but ozone is systematically underpredicted in the rural environment especially in the afternoon hours. As the measurements include only ozone and NO_2 observations, they are insufficient to understand and reconstruct the process of ozone formation which strongly depends on the presence of volatile organic compounds (VOC's). A theoretical study (Bauer, 2000) reveals that the Berlin-Brandenburg area is NO_x-saturated with respect to ozone production. In the morning hours the area is definitely VOC-sensitive. At noon and during the afternoon hours the system is in an intermediate state where it is VOC- as well as NO_x-sensitive.

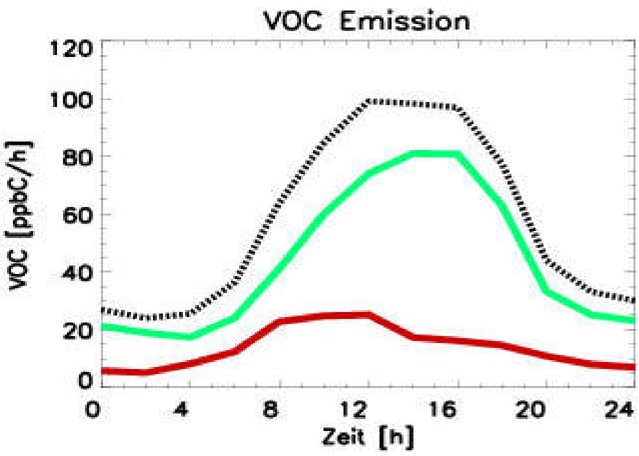

Figure 1. Total (broken line), anthropogenically (dark line) and biogenically (light line) emitted hydrocarbons as a function of time of the day (July 23, 1994) as simulated with the GESIMA model. The emissions are sampled over the whole domain of the GESIMA model covering the area of Berlin-Brandenburg.

To further characterise the VOC-composition of air masses in the area of Berlin-Brandenburg the emitted VOC's on July 23 are shown in Figure 1 subdivided into their biogenic and anthropogenic origin. The composition of hydrocarbons in Berlin-Brandenburg during the FLUMOB episode was made up by about 70-80 % of biogenically emitted hydrocarbons, i.e. monoterpenes and isoprene, and by about 20-30 % of anthropogenically emitted hydrocarbons. Thus, natural emissions released by trees in the widely forested areas of Brandenburg dominate over anthropogenic emissions. This is of special importance for the process of ozone formation in the investigated area because ozone production during FLUMOB was hydrocarbon limited so that their availability define the potential for ozone production and the peak concentrations.

Influence of large scale conditions on ozone during FLUMOB

Simulated species in limited area models, especially longer living species like ozone are sensitive to their vertical distribution at the lateral boundaries (Langmann and Bauer, 2000). Most trace species show maximum concentrations close to the earth's surface and decrease rapidly with increasing height. Contrarily, ozone concentrations are present in the whole troposphere with relatively high mixing ratios, normally increasing with height. Thus, the lifetime and distribution of ozone is also dependent on the larger scale conditions when investigating the processes of pollutant formation in Brandenburg.

Figure 2 shows the only three ozone soundings available during the FLUMOB episode at Lindenberg. Unusually high ozone concentrations were measured in the free troposphere. To explain the observed high ozone concentrations, backward trajectories, calculated by the German Weather Service, were analysed. They show that at about 700 hPa air masses from south-west Germany are advected to Lindenberg. Following that trajectory the air masses pass highly polluted areas and ozone could have accumulated day by day in the lower troposphere. Above the planetary boundary layer the observed ozone concentrations originate from the Atlantic ocean four days prior to their arrival in Lindenberg. It is assumed that the observed unusually high mixing ratios of about 100 ppbv throughout the troposphere are the result of mixing of long range transported upper level ozone and upward transported ozone from the planetary boundary layer, formed and accumulated during the summer smog event. Because of the lack of measurements it is difficult to evaluate the assumption that during this summer smog event Europe was additionally influenced by high amounts of long range transported ozone. However, it seems to be the most probable explanation for the large amounts of ozone in the free troposphere. Other ozone balloon soundings, Brussels and Hohenpeissenberg, support this theory. The shape of the European wide ozone soundings exclude also stratospheric ozone intrusion.

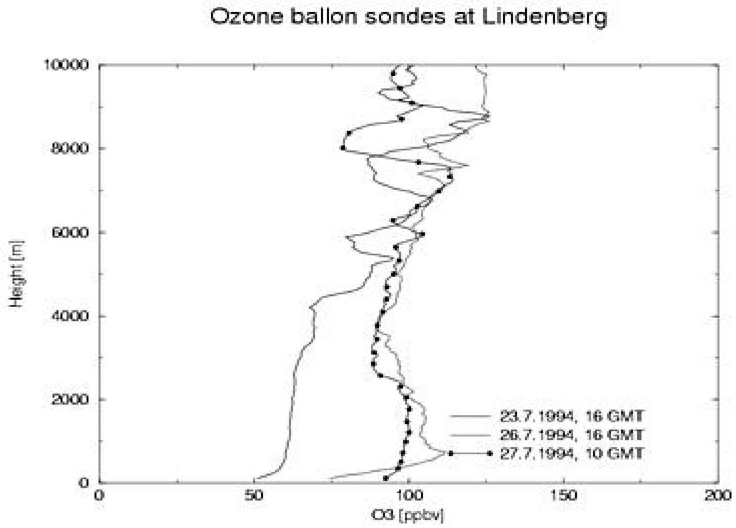

Figure 2. Ozone ballon sonde data at Lindenberg during FLUMOB.

Sensitivity of modelled species concentrations on lateral boundary information

Analysing the REMO and GESIMA model simulations and sensitivity studies (Bauer, 2000, Langmann and Bauer, 2000) reveals that

- ozone is underpredicted by the models up to 50 ppbv in the free troposphere,
- in the free troposphere, simulated ozone concentrations are strongly linked to the ozone concentrations provided at the lateral model boundaries and its initial conditions,
- in the planetary boundary layer ozone increases / decreases linearly by the amount of ozone which is added at the lateral inflow boundaries.

Main conclusions

Biogenic VOC emissions

Biogenic hydrocarbon emissions dominated significantly over anthropogenically emitted ones during July 1994 in the area of Berlin-Brandenburg where ozone production was hydrocarbon limited. With respect to ozone reduction strategies, the important role of the hydrocarbons has to be taken into account. This would possibly lead to the requirement of considerably stronger NO_x emission reduction strategies to reduce the occurrence of ozone episodes in this region than believed until now.

As this conclusion was not drawn by previous investigations of the FLUMOB project, the subsequent ozone measurement campaign in Berlin-Brandenburg, BERLIOZ carried out in summer 1998, did not focus on biogenic hydrocarbons. However, the measurements made during BERLIOZ confirm our conclusions concerning the composition of hydrocarbons in the Berlin-Brandenburg area during summer.

Long range ozone transport and lateral boundary information

In spite of the stagnant high pressure weather conditions during FLUMOB, Brandenburg was influenced by long range transported ozone concentrations. In contrast to previous analyses, this investigation showed that the high ozone concentrations in the free troposphere must originate from outside of Europe. Due to vertical mixing processes these high concentrations influenced the near surface concentrations during the second phase of FLUMOB.

Ozone was dramatically underestimated by the model system in the free troposphere. Sensitivity studies figured out that above the planetary boundary layer the amount of simulated ozone is almost completely dependent on the concentrations given to the lateral boundaries of the model domain. Even the European wide simulation with REMO was not able to rebuild this situation because climatological concentrations were assumed at the lateral boundaries. Therefore, global information about the three dimensional distribution of longer living chemical trace species and their temporal variability are necessary for mesoscale modelling systems (Langmann and Bauer, 2000).

Aims for the coming year

The FLUMOB case will be investigated again with global chemical information at the boundaries of the mesoscale model system to study the combination of large local ozone production due to the summer smog event and the advection of long range transported ozone concentration. The first step, forcing the European wide REMO simulation with global model results for trace species at the lateral boundaries look promising!

Acknowledgements

The authors acknowledge the helpful support by Ralf Podzun. This work was mainly founded by the Tropospheric Research Program (TFS) of the German Ministry for Education, Science, Research and Technology (BMBF). We also wish to thank Burckhard Wickert, IER Stuttgart, for providing the emission data sets.

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Study of photochemical oxidant budget variability in relation to dynamics, chemistry and climate change

A contribution to subproject GLOREAM

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The Cambridge chemistry transport model, TOMCAT, is an off-line grid point model using winds, temperatures, and humidity from the European Centre for Medium-Range Weather Forecasts (ECMWF) meteorological analyses. It uses an accurate non-diffusive advection scheme together with a mass flux scheme for the convective transport of trace gases. Atmospheric boundary layer (ABL) processes are treated using a realistic non-local vertical mixing scheme based on the scheme developed for the National Center for Atmospheric Research (NCAR) Community Climate Model (CCM2). A more extensive description of the TOMCAT model dynamics can be found in Stockwell *et al.* (1998) and Stockwell and Chipperfield (1999). The model also includes full treatment of tropospheric chemistry emissions, wet/dry deposition, boundary layer mixing and stratospheric fluxes. The model includes 50 chemical species and, in addition, tracers of stratospheric O_3 and NO_y.

In the last year much work has gone into further developing and testing the model. This has resulted in an updating of the photolysis rates used in the model using data from JPL 2000 (Sander *et al.*, 2000) and a new interpolation scheme which gives more realistic diurnal cycles of the photolysis rates. In addition improvements have been made to the convection parameterisation and the code has generally been made more robust and simple to use.

Chemical constituent measurements taken onboard the UKMO C-130 aircraft during May and August 2000 as part of the ACTO (Atmospheric Chemistry and Transport of Ozone) and EXPORT flying campaigns sampled many layers of different composition in the upper troposphere over Western and Central Europe. The layers sampled included stratospheric air, uplifted clean marine boundary layer air and polluted continental boundary layer air. These will be used to validate the TOMCAT model and the model will then be used to diagnose different components of the ozone budget, namely stratospheric flux, photochemistry and dry deposition over Europe. In particular, first estimates of the contribution of clean/polluted boundary layer air to the ozone budget of the upper troposphere over Europe and downwind will be determined.

An intercomparison with 3 other chemical transport models is also well under way as part of the POET program. Model runs have been performed with 2 inert tracers in TOMCAT, MOZART, IMAGES and the Oslo CTM. The tracers used were radon and a tracer of stratospheric air. Preliminary results from this intercomparison exercise showed excellent agreement between the models. This will soon be followed by full chemistry integrations with all models using the same ECMWF meteorological fields and the same emissions inventories. Also as part of the POET program work has begun to compare GOME satellite measurements of ozone, NO_2, and HCHO with TOMCAT results.

An ozone climatology of the UT/LS region was compiled using MOZAIC data for the years from 1995 to 2000. Variability of ozone in the UT/LS region was investigated on various time scales. The UT/LS region is well covered by MOZAIC ozone measurements during all seasons. The interseasonal and interannual variations of ozone mixing ratios were observed on five isentropic surfaces from 320~K to 365~K. The 340~K isentropic surface shows the lowest values of standard deviations and the ozone distribution on this level is represented most accurately by the MOZAIC measurements compared with the other four levels. In future studies, the measurements will be compared with results from chemical transport integrations.

We have incorporated a comprehensive tropospheric chemical module into the UK Met Office Unified Model (UM) to look at the climate/chemistry coupling associated with simulated changes in ozone. The detailed NO_x/hydrocarbon chemistry comprises 46 species and 186 chemical reactions. Seasonally varying surface emissions of NO_x and hydrocarbons are included. Climatological background concentrations are prescribed in the stratosphere for ozone, NO_y and CH_4. Two simulations were carried out representing present and future climate based on the IPCC scenarios for 2000 and 2100 emissions. Present day simulations are compared to observations, where there is good agreement for ozone and longer-lived gases. The model predicts very large increases in ozone concentrations, especially at the surface, over the industrialised Far East, Europe and North America, for the 2100 emissions.

Funding Acknowledgements

Funding was provided by the UK Natural Environmental Research Council (NERC), the UK Universities Global Atmospheric Modelling Programme (UGAMP) and by the EU-funded projects POET, MOZAIC and EXPORT.

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Application of Synoptic/Meso Scale Eta Model in Long Range Transport Processes

A contribution to subproject GLOREAM

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Summary

The work of the Institute of Meteorology, University of Belgrade (IMUB) group in 2000 consisted of efforts in two main directions. One was addressed to the continued improvement the Eta Model and its initialization, suitable for application to problems of the long-range transport of air pollutants. The second main direction was application of the resulting model on the cases of the air pollution originating from the greatest bombardment of chemical industry, oil refinery and fuel storage in Serbia under war conditions in Yugoslavia during 1999.

Aims of the research

Aims of the research were, in the first place, development of the meteorological part of the transport model in terms of the implementation of trajectory calculation based on the Eta Model. Studies of the requirements needed, in terms of sophistication of the representation of various atmospheric processes (mountain representation, boundary layer, land-surface processes, moist processes) needed for high-quality results in impact study situations (transport simulations with time-scale of 2-3 days). The second principal aim was the application and verification of the resulting code on the various sensitivity studies as well as to studies of chosen European long-range transport of air pollutant problems.

Activities during the year

Regarding the IMUB meteorological module (Eta Model) (Mesinger *et al.*, 1988; Janjic, 1990, 1994), work on the model data initialization, as candidate for important contribution to realistic simulation of constituent transport, has finished (Lazic, 2000). Considerable attention was given to study the model trajectory calculations. Work on a sensitivity of forecast trajectories to wind data inputs during the strong local wind conditions has finished also (Lazic and Tosic, 2000). The model has applied for various studies of chosen European long-range transport of air pollutant problems especially in the air pollution originating from fire after the greatest bombardment of chemical industry, oil refinery and fuel storage in Serbia under war conditions in Yugoslavia during 1999 (Vukmirovic *et al.*, 2000a; 2001).

Principal results

The main work was on regional air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia in the period of 24 March to 10 June 1999 (Vukmirovic *et al.*, 2000a, b, c; 2001). During the 78-day period of air-strike, a large number of industrial and military facilities have been destroyed in this country. Of those facilities the most notable and of importance as to their effects in the environment. The most severe environmental episodes in the area are expected to result from emissions that took place in the first and third weeks of April (4-7 and 12-19). This is because during the 3-day period 4-7 April targets of oil storage, refineries and other plants were hit at a large number of cities.

On April 18 at 1:00 a.m. the installations for vinyl chloride monomer (VCM) and polyvinyl chloride (PVC) production in Pancevo (20°40'E, 44°53'N), near Belgrade, were hit. A spherical reservoir with 1200 tons of VCM was destroyed and 6 train cisterns of 30 tons of VCM each. All VCM contents in the reservoir burned out for several hours. Practically simultaneous release of smoke plumes from the oil refineries in Pancevo and Novi Sad (19°50'E, 45°20'N) was occurred with total burning rate of 2,000 t h^{-1} during the first 12 h after bombing at midnight between 17 and 18 April 1999. Using the same methodology applied in the case of the Kuwait oil smoke plume (Johnson *et al.*, 1991), an average emission of carbon particles in overlapping plumes (Melas *et al.*, 2000) is estimated as 65 t h^{-1}. During the Pancevo incident large amounts of airborne toxic gases and aerosols are estimated to have been transported and deposited in Romania, Bulgaria, Moldavia and Ukraine (Vukmirovic *et. al.,* 2000a; 2001).

Forward trajectories during the Pancevo and Novi Sad oil-refinery fires episodes based on the 72 h Eta Model forecast with a horizontal grid resolution of 0.5° x 0.5° were calculated. Numerical simulation by the Eta model is initialized at 00 UTC 18 April using ECMWF (European Center for Medium-Range Weather Forecasts) analysis as the initial. Starting points in the vertical were from the middle of the eta layers, with approximate heights of 434 m, 1023 m, 1505 m, 2370 m, 3416 m, 4664 m and 6142 m. According to the trajectories, the pollutant puff was picked up over the area of oil-refinery fires and moved eastward over Romania, Bulgaria, Moldavia, Ukraine and the Black Sea. This long-range transport occurred around level 700 hPa. The regional transport of polycyclic aromatic hydrocarbons (PAHs) and of dioxins and furans as burning products of PVC and VCM from Pancevo occurred at about 1500 m to Xanthi in Greece (25°E, 41°9'N) on 18/19 April. Transport of PAHs from Novi Sad has been performed above 2000 m at Xanthi in the first day. This analysis corroborated measurements. The lower level trajectories from Pancevo indicate pollutant transport in short-regional and local scales towards the Belgrade area in the first day. Under oxidizing conditions in plume, a significant fraction of Hg(II) from the petrochemical plant in Pancevo might be adsorbed to elemental carbon particles. Relatively high concentrations of soot and polycyclic aromatic hydrocarbons (PAHs) are predicted in the air and compared with the available measurements in Novi Sad, Pancevo and Belgrade.

The next work addresses an analysis of pollution during above mentioned warfare episode and its properties as reflected on precipitation measured in Serbia. The washout of persistent organic pollutants (POPs) in central and southern Serbia is considered in the pollution episode of 18-20 April as the predominant process of removing POPs from the atmosphere. Maximum POPs wet deposition was found in central Serbia and along the 1500 m Eta Model trajectory towards south-eastern Serbia and the Bulgarian border (Unkasevic *et al.*, 2000, Vukmirovic *et al.,* 2001).

The last of this year works has shown theoretical, observed and experimental evidence of pollutants released, transported and deposited during the warfare and their impacts on precipitation in Serbia. The greatest bombardment of chemical industry, oil refinery and fuel storage in Serbia occurred during April, resulted in releases of many hazardous, toxic and cancerogenic substances, what it caused precipitation increase. The April's precipitation in 1999 are compared to the precipitation in the period from 1961 to 1990 registered at thirty meteorological stations in Serbia and especially at the Belgrade-Observatory station in the period from 1888 to 1995. In comparison with the mean precipitation sums in April, calculated from 1961 to 1990, all places in Serbia had enhanced precipitation except at the north-western part of Serbia. The maximums of days with precipitation greater or equal to 0.1 mm were at the wider Belgrade area and at the central and south-western parts of Serbia during April 1999. This results is confirmed by using the Eta trajectory analysis (Unkasevic *et al.*, 2001).

Main conclusions

- The meteorological synoptic/mesoscale Eta Model used by the IMUB group including initialization, trajectory calculations, constituent transport along trajectories, diffusion and wet and dry deposition code, has been shown to perform satisfactorily;
- A sensitivity study of forecast trajectories to wind data inputs in the flow over complex terrain has shown it to be very important for correct simulations of the transport and diffusion of tracers, and possibly also for long range transport of constituents;
- The regional air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia in the period of 24 March to 10 June 1999, was studied.
- Using the Eta model trajectory analysis, the regional pollutant transport at about 1500 m from almost simultaneously bombed industrial sites at midnight between 17 and 18 April 1999 in Northern Serbia (Novi Sad) and in the Belgrade vicinity (Pancevo) respectively, corroborated measurements at Xanthi in Greece. At the same time the pollutant puff was picked up at about 3000 m and transported to Bulgaria, Romania, Ukraine, Moldova and the Black Sea.
- The lower level trajectories from Pancevo indicate pollutant transport in short-regional and local scales towards the Belgrade area in the first day. Relatively high concentrations of soot and polycyclic aromatic hydrocarbons (PAHs) are predicted in the air and compared with the available measurements in Novi Sad, Pancevo and Belgrade.
- The washout of persistent organic pollutants (POPs) in central and southern Serbia is considered in the pollution episode of 18-20 April as the predominant process of removing POPs from the atmosphere. Maximum POPs wet deposition was found in central Serbia and along the 850 hPa Eta Model trajectory towards south-eastern Serbia and the Bulgarian border.
- The greatest bombardment of chemical industry, oil refinery and fuel storage in Serbia occurred during April, resulted in releases of many hazardous, toxic and cancerogenic substances, what it caused precipitation increase. In comparison with the mean precipitation sums in April, calculated from 1961 to 1990, all places in Serbia had enhanced precipitation except at the north-western part of Serbia.

Aims for the coming year

- Sensitivity tests will be performed on the air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia during 1999 in order to assess the effects of various modelling options on the transport/deposition results. Examples of options to be considered are the choice of horizontal resolution; representation of mountains (terrain-following vs. the step-mountain system); choice of the constituent advection scheme; effects of transports by convection; and parameterization of horizontal and vertical diffusion.

- The effects of the initial subgrid scale diffusion on the overall quality of the long range constituent transport simulations will be studied. This sensitivity study will be based on results for the air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia during 1999.
- Work on other cases will be initiated.
- Further improvement of the Eta Model and model verification will be continued.

Acknowledgements

This contribution is not being financed by any scientific foundation.

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A brief paragraph explaining results from the EUROTRAC-2 work

Initial attention of this contribution was given to include initialization to the Eta Model (Lazic, 2000) and sensitivity study of forecast trajectories to wind data inputs (Lazic and Tosic, 2000). The main work was on regional air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia in the period of 24 March to 10 June 1999 (Vukmirovic *et al.,* 2000; Vukmirovic *et al.,* 2001). Using the Eta model trajectory analysis, the regional pollutant transport at about 1500 m from almost simultaneously bombed industrial sites at midnight between 17 and 18 April 1999 in Northern Serbia (Novi Sad) and in the Belgrade vicinity (Pancevo) respectively, corroborated measurements at Xanthi in Greece. At the same time the pollutant puff was picked up at about 3000 m and transported to Bulgaria, Romania, Ukraine, Moldova and the Black Sea (Vukmirovic *et al.,* 2000; Vukmirovic *et al.,* 2001). The lower level trajectories from Pancevo indicate pollutant transport in short-regional and local scales towards the Belgrade area in the first day. Relatively high concentrations of soot and polycyclic aromatic hydrocarbons (PAHs) are predicted in the air and compared with the available measurements in Novi Sad, Pancevo and Belgrade. The greatest bombardment of chemical industry, oil refinery and fuel storage in Serbia occurred during April, resulted in releases of many hazardous, toxic and cancerogenic substances, what it caused precipitation increase. In comparison with the mean precipitation sums in April, calculated from 1961 to 1990, all places in Serbia had enhanced precipitation except at the north-western part of Serbia (Unkasevic *et al.*, 2001).

A list of theses which have resulted from the EUROTRAC-2 work

- The continued improvement the Eta Model and its initialization, suitable for application to problems of the long-range transport of air pollutants.
- The model trajectory calculations. It had included work on the implementation and testing trajectory calculations based on the Eta Model in case of complex terrain.
- Application of the resulting model on regional air pollution originating from the chemical industry, oil refinery and fuel storage fires under war conditions in Yugoslavia in the period of 24 March to 10 June 1999.
- Using the Eta model trajectory analysis, the regional pollutant transport at about 1500 m from almost simultaneously bombed industrial sites at midnight between 17 and 18 April 1999 in Northern Serbia (Novi Sad) and in the Belgrade vicinity (Pancevo) respectively, corroborated measurements at Xanthi in Greece. At the same time the pollutant puff was picked up at about 3000 m and transported to Bulgaria, Romania, Ukraine, Moldova and the Black Sea.
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Publications which have resulted from the EUROTRAC-2 work

Lazic, L.; "Initialization" using an iterative Matsuno style scheme in the Eta Model adjustment stage, *Meteorol. Atm. Phys.* **75** (2000) 121-130.

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Unkasevic, M., L. Lazic, I. Tosic and Z. Vukmirovic; Effects of Uncontrolled Particulate Matter Release on Precipitation Under Warfare Conditions. *Environ. Sci. & Pollut. Res.* (2001) Submitted for publication.

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Speciation in Chemical Mechanisms and Emissions Databases

A contribution to subproject GLOREAM

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The following report deals with the second part of this project (Part B: Lumping of Emissions for Pollution Models). Two papers describing the work in more detail have been submitted for peer review with the Journal of Geophysical Research. The following is a precis of this work.

Aim of the research

This part of the study examines the manner in which chemical lumping is performed in emissions processing systems, and the manner in which this affects the reactivity and particle-forming potential of model hydrocarbons. Past methods of speciating VOCs (Middleton *et al.*, 1990) focussed on the OH reactivity as the main factor to be preserved in creating a model VOC emission speciation. Many of the VOCs present in emissions databases have very low vapour pressures and may partition significantly to the particulate phase, but this information is lost when employing a lumping method based on reactivity alone. The current work examines the extent to which these VOCs may impact on organic particle formation.

Emissions processing systems typically make use of source profiles that split a total annual Volatile Organic Compound mass emission into hundreds of speciated VOCs. For example, the Canadian Emissions Processing System used in the current work is capable of speciating 822 different gas-phase chemicals. The gas-phase mechanisms designed for regional and global models typically have more severe restrictions as to the number of VOCs which may be retained in memory. This is due in part to the available data upon which to base reaction mechanisms, and in part to memory space and processing time requirements for regional air quality codes. These mechanisms and previous emissions processing work (e.g. Middleton *et al.*, 1990) have largely been designed with the aim of providing accurate simulations of tropospheric ozone formation. For this problem, the reactivity of the organic compounds is the key property for ensuring an accurate prediction, and hydrocarbon lumping in both mechanisms and emissions processing has been designed to preserve the net reactivity of the detailed, speciated, emissions.

More recently, particulate matter has also become an issue of importance in terms of impacts on human health, and many efforts are underway to improve regional models to allow for the prediction of particulate mass and composition. For organic compounds, other factors in addition to reactivity come into play when particulate matter formation is of interest. The structure has a greater role, and the species' vapour pressures and solubilities become of key importance. Emissions processing speciations that preserve reactivity instead of these other properties may therefore lose crucial information for particulate forming potential.

Activities during the year

An emissions speciation designed and processed with the Canadian Emissions Processing System (see GLOREAM Annual Report 1999) was used to estimate RO_2 production and particulate matter formation for a grid covering much of eastern North America, for typical summer and winter days. The emissions speciation of 81 hydrocarbons is designed for use with the Canadian AURAMS model, and will hereafter be referred to as the AURAMS81.

Since the previous year's report, biogenic VOC emissions have been added, using the CEPS CBEIS emissions processor. The RO_2's produced by biogenics dominate in terms of the net effect across the grid (see Figure 1), but in urban centres, the anthropogenically emitted RO_2 precursors may have an equal (Atlanta and south-eastern US cities) or up to six times greater impact (Northern US and Canadian cities) than the biogenic RO_2's. The implication of this finding is that VOC controls will be less effective in southern regions in which the biogenic emissions are high, and will be more effective in northern urban regions than outside of the urban areas.

The potential for the formation of organic particulate matter from each of the VOCs was also examined. This was done in two stages. VOCs with chemical properties allowing partitioning to the condensed phase directly after emissions were used in conjunction with the absorption partitioning formulae of Pankow (1994a,b) to determine directly partitioned particle mass. The 20 VOC classes in the AURAMS81 speciation scheme considered to be potential condensable VOCs are listed in Table 1. Many of these classes were selected on the basis of observational evidence showing their presence in atmospheric aerosols. Others (methyl, ethyl, and propyl alcohol, di-and tri-alcohols, esters, and petroleum distillation spirits) were included on the basis of high aqueous solubility and boiling points higher than the typical range of ambient temperatures, characteristics that might allow the given compounds to exist in the liquid or aqueous phase under ambient conditions.

Table 1.

Species or Species Class
Water
Alkanes (C_{20} to C_{26})
Alkanes (C_{27} to C_{35})
Alkanes (C_{36} to C_{43})
High-C-Number Alkyl Phthalates
Low-Vapour-Pressure PAHs
Phenol
Cresols
C_{6+} Aldehydes
Aromatic Aldehydes
C_{6+} Ketones
Formic Acid
Acetic Acid
C_{4+} Diacids
Methyl Alcohol
Ethyl Alcohol
Propanol
C_{4+} Alcohols
Di- & Tri-Alcohols
Low-Reactivity Esters
High-Reactivity Esters
Petroleum Distillation Spirits
Furandiones

The “direct partitioning” calculation was followed by a calculation for the amount of organic aerosol mass produced by oxidation of the more volatile emitted compounds using the parameterization of Odum *et al.* (1996). The AURAMS81 VOC classes considered for VOC oxidation/SOA formation and the sources for α_i and K_i data are listed in Table 2. Note that three of these classes – Phenol, Cresol, and Aromatic Aldehydes – are also listed in Table 1. It can be seen from Table 2 that 9 of these 19 species classes have laboratory-based α_i and K_i values available; the remaining 10 have been observed to produce organic aerosols in chamber experiments, but ORGM-dependent yields are not available at the current time. For the latter species, α_i and K_i were assigned from the 9 species for which data are available – the values for these species are therefore approximations, and subject to future revision.

Table 2.

VOC Species or Class
Alpha-Pinene
Beta-Pinene
D-Limonene
D-3-Carene
End C_{9}-$_{19}$ Alkenes
Toluene
Mono-Alkyl Aromatics
Di-Alkyl Aromatics
Tri-Alkyl Aromatics
Alkene Aromatics
Cresol
C_{9-10} Alkanes
C_{11-19} Alkanes
End C_{6-8} Alkenes
Internal C_{6-8} Alkenes
Internal C_{9-19} Alkenes
Napthalene
Aromatic Aldehydes
Phenol

The results of these calculations over the model grid showed that the effect of the high-molecular mass, directly partitioning species may be as important or more important towards organic particle formation than the formation of aerosols through secondary oxidation. Figure 2 shows the net grid condensed-phase VOC mass; the main part of the mass is taken up by high molecular mass “alkane” groups (each of these are actually an amalgam of many hydrocarbons of similar structure and mass) and anthropogenic diacids. Figure 3 shows how the composition may vary between individual cities. For large urban centres (New York City, Toronto), the SOA portion of the aerosol mass is equal to the directly condensed aerosol mass.

Main conclusions

The most significant conclusion on the work to date is that a significant proportion of the organic aerosol mass may originate in VOCs which were not resolved in previous VOC emissions speciations. These VOCs make up a small fraction of the total emitted hydrocarbon mass, and have a small impact on reactivity, but may outweigh the SOA fraction of organic aerosols. Emissions inventories which lack this level of speciation will seriously underestimate the total organic aerosol mass.

Other conclusions stemming from this work:

- Biogenically emitted species were found to have a lower impact than anthropogenic species (noting that the calculations here included local effects only; no transport and hence interaction between biogenic and anthropogenic species was considered).
- The relative contribution of different VOCs towards particle production varies substantially between different urban centres.
- RO_2 production from anthropogenic emissions was shown to be approximately six times higher than biogenic RO_2 production in northern cities, but approximately equal to biogenic RO_2 production in southern cities.
- Biogenic emissions are dominated by isoprene, high-reactivity esters, and alpha-pinene on a mass basis, and by isoprene, high-reactivity terpenes, and internal C_{6-8} alkenes on a reactivity basis. Anthropogenic emissions were dominated by C_{4-5} *n*- and *iso*-alkanes, C_{6-8} *n*- and *iso*-alkanes, and toluene on a mass basis, and by internal C_{4-5} alkenes, internal C_{6-8} alkenes, ethene, and high-reactivity amines and amides on a reactivity basis. The latter group of compounds is usually not resolved in regional-air-quality-model VOC speciations, while the current work suggests that it may be the fourth greatest contributor to RO_2 production. Its inclusion in future reaction mechanisms is therefore recommended.

Aim for the coming year

Two papers have been submitted for review with the Journal of Geophysical Research on the work. The final stage of the work will be to install the new emissions speciation into the AURAMS regional air quality model, allowing these effects to be examined in the context of transported, size and composition resolved aerosols. The final stage of the current study will be an examination of the effects of reactivity lumping on ozone and particulate matter formation through 3D model simulations.

Acknowledgements

This work has been supported by the Air Quality Research Branch of Environment Canada.

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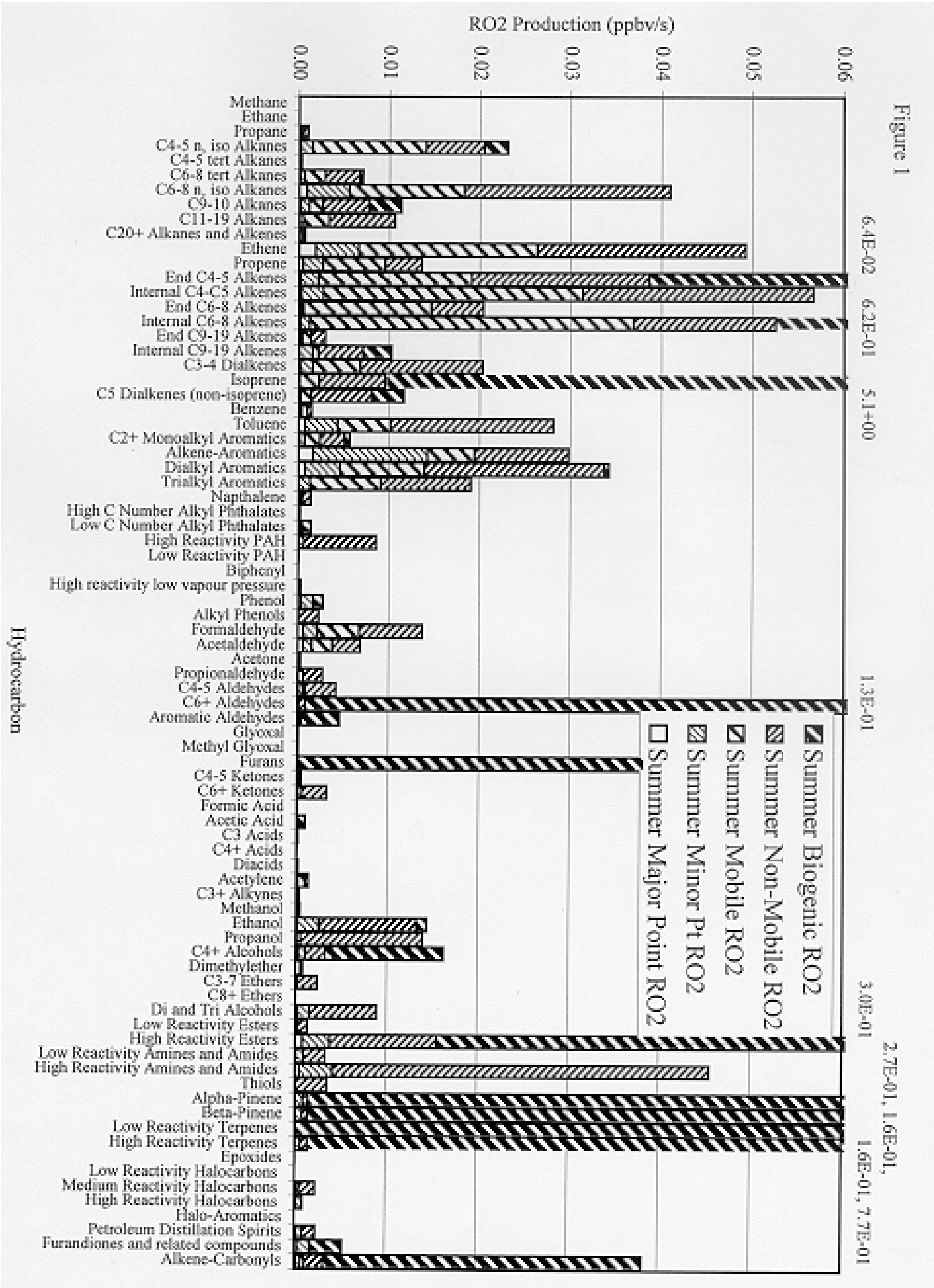
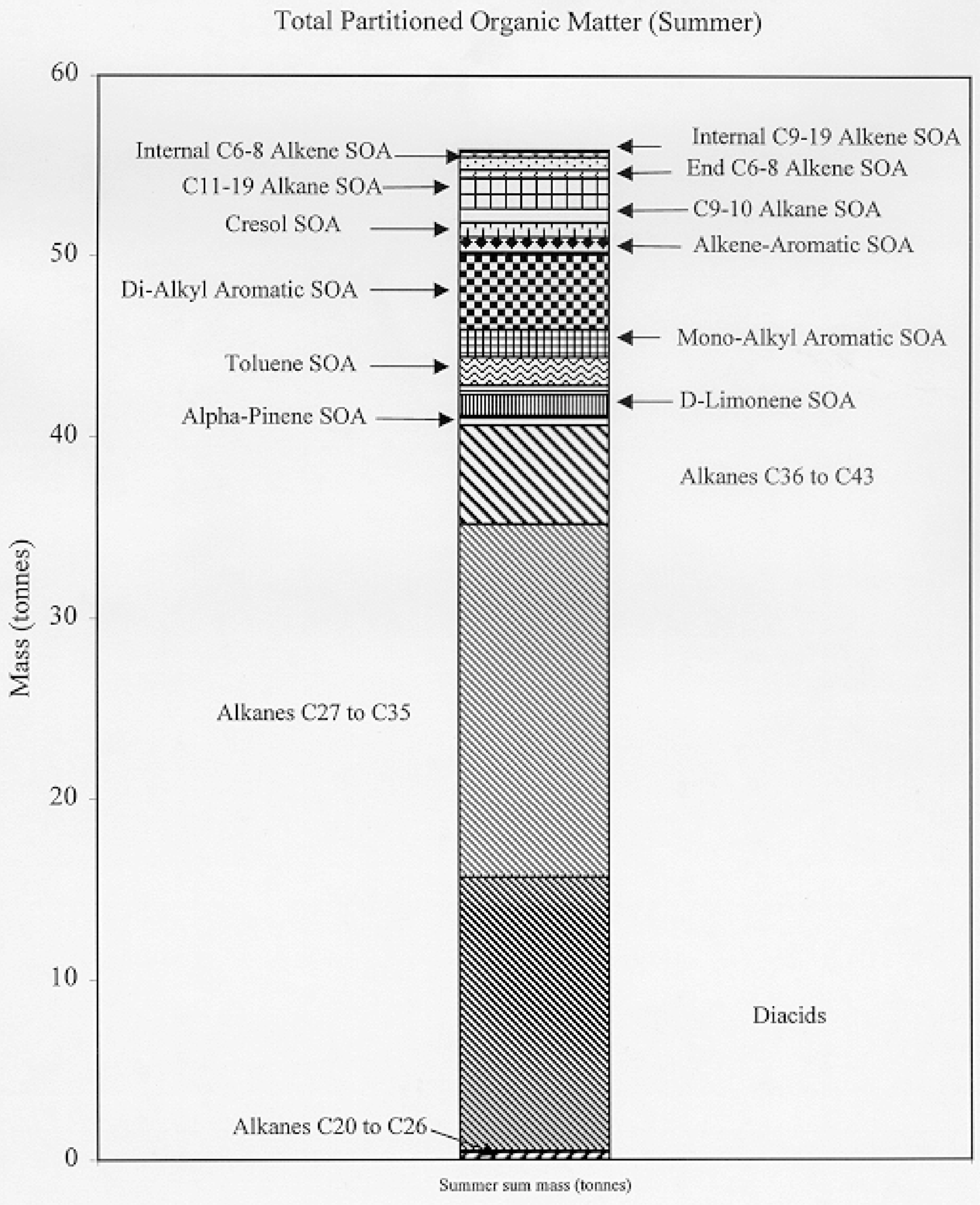

RO2 Production (ppbv/s)
0.00
0.01
0.02
0.03
0.04
0.05
0.06
Hydrocarbon
Methane
Ethane
Propane
C4-5 n, iso Alkanes
C4-5 tert Alkanes
C6-8 tert Alkanes
C6-8 n, iso Alkanes
C9-10 Alkanes
C11-19 Alkanes
C20+ Alkanes and Alkenes
Ethene
Propene
End C4-5 Alkenes
Internal C4-C5 Alkenes
End C6-8 Alkenes
Internal C6-8 Alkenes
End C9-19 Alkenes
Internal C9-19 Alkenes
C3-4 Dialkenes
Isoprene
C5 Dialkenes (non-isoprene)
Benzene
Toluene
C2+ Monoalkyl Aromatics
Alkene-Aromatics
Dialkyl Aromatics
Trialkyl Aromatics
Napthalene
High C Number Alkyl Phthalates
Low C Number Alkyl Phthalates
High Reactivity PAH
Low Reactivity PAH
Biphenyl
High reactivity low vapour pressure
Phenol
Alkyl Phenols
Formaldehyde
Acetaldehyde
Acetone
Propionaldehyde
C4-5 Aldehydes
C6+ Aldehydes
Aromatic Aldehydes
Glyoxal
Methyl Glyoxal
Furans
C4-5 Ketones
C6+ Ketones
Formic Acid
Acetic Acid
C3 Acids
C4+ Acids
Diacids
Acetylene
C3+ Alkynes
Methanol
Ethanol
Propanol
C4+ Alcohols
Dimethylether
C3-7 Ethers
C8+ Ethers
Di and Tri Alcohols
Low Reactivity Esters
High Reactivity Esters
Low Reactivity Amines and Amides
High Reactivity Amines and Amides
Thiols
Alpha-Pinene
Beta-Pinene
Low Reactivity Terpenes
High Reactivity Terpenes
Epoxides
Low Reactivity Halocarbons
Medium Reactivity Halocarbons
High Reactivity Halocarbons
Halo-Aromatics
Petroleum Distillation Spirits
Furandiones and related compounds
Alkene-Carbonyls
6.4E-02
6.2E-01
5.1+00
1.3E-01
3.0E-01
2.7E-01, 1.6E-01,
1.6E-01, 7.7E-01
Summer Biogenic RO2
Summer Non-Mobile RO2
Summer Mobile RO2
Summer Minor Pt RO2
Summer Major Point RO2

Figure 1

Figure 2.


Total Partitioned Organic Matter (Summer)
60
50
40
30
20
10
0
Mass (tonnes)
Internal C6-8 Alkene SOA
C11-19 Alkane SOA
Cresol SOA
Di-Alkyl Aromatic SOA
Toluene SOA
Alpha-Pinene SOA
Alkanes C27 to C35
Alkanes C20 to C26
Internal C9-19 Alkene SOA
End C6-8 Alkene SOA
C9-10 Alkane SOA
Alkene-Aromatic SOA
Mono-Alkyl Aromatic SOA
D-Limonene SOA
Alkanes C36 to C43
Diacids
Summer sum mass (tonnes)

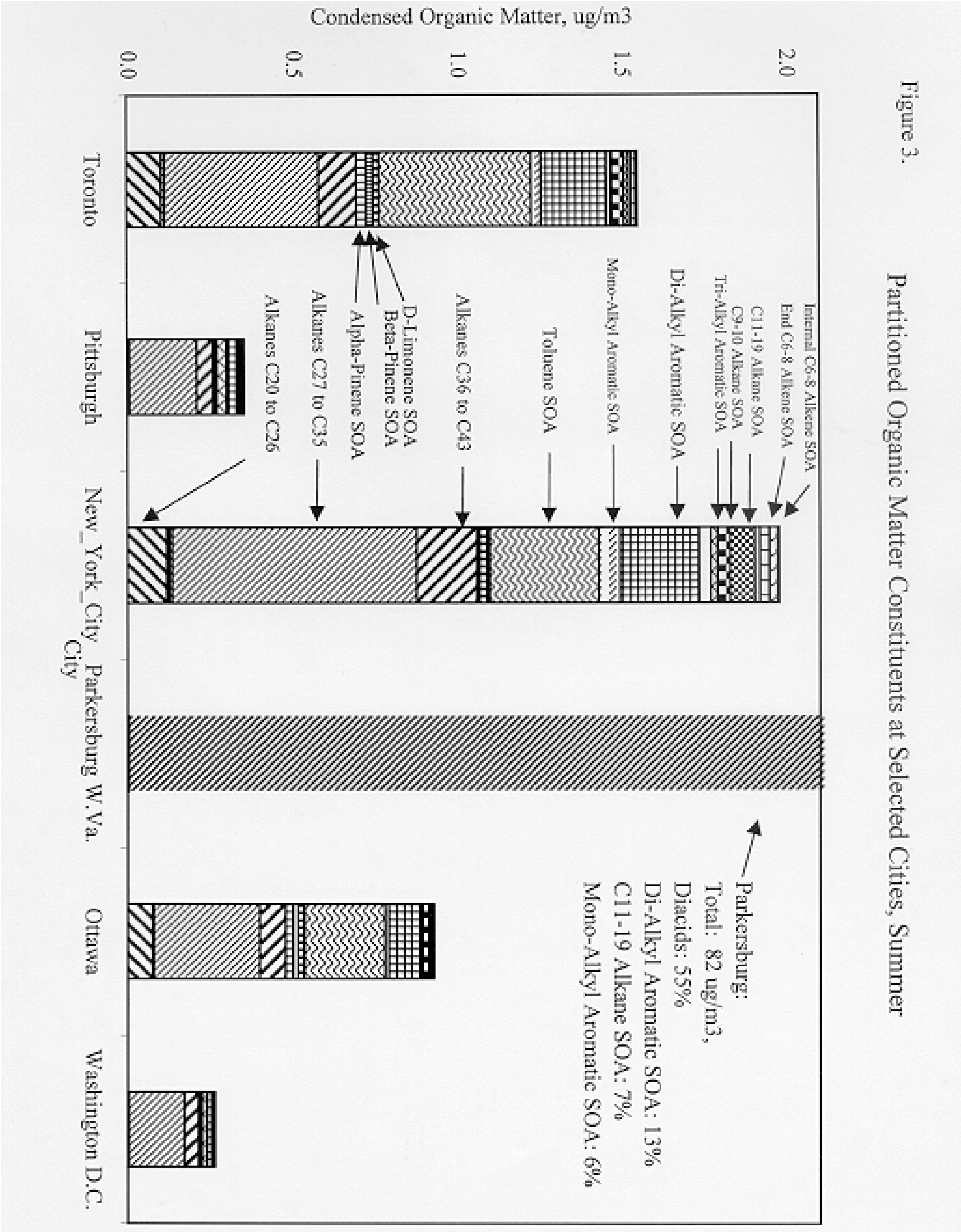


Figure 3.
Partitioned Organic Matter Constituents at Selected Cities, Summer
Condensed Organic Matter, ug/m3
0.0
0.5
1.0
1.5
2.0
Internal C6-8 Alkene SOA
End C6-8 Alkene SOA
C11-19 Alkane SOA
C9-10 Alkane SOA
Tri-Alkyl Aromatic SOA
Di-Alkyl Aromatic SOA
Mono-Alkyl Aromatic SOA
Toluene SOA
Alkanes C36 to C43
D-Limonene SOA
Beta-Pinene SOA
Alpha-Pinene SOA
Alkanes C27 to C35
Alkanes C20 to C26
Parkersburg:
Total: 82 ug/m3,
Diacids: 55%
Di-Alkyl Aromatic SOA: 13%
C11-19 Alkane SOA: 7%
Mono-Alkyl Aromatic SOA: 6%
Toronto
Pittsburgh
New_York_City
Parkersburg W.Va.
Ottawa
Washington D.C.
City

Regional modelling of Particulate Matter

A contribution to subproject GLOREAM

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Summary

The future implementation of the EU-directive [http://europa.eu.int/eur-lex] on particulate matter (PM) has implications for national emission regulations. For the assessment of these implications it is necessary to understand the processes which determine the sources and sinks of PM and precursors on a European scale. Therefore, RIVM is developing a module to describe the dispersion of PM (PM_{10}, $PM_{2.5}$) over Europe. A first version of such a module has been completed. It describes hourly concentrations of primary emitted particles and the secondary formation of sulphate and nitrate particles in the regional chemical dispersion model EUROS. EUROS is a Eulerian air-quality model, which has been developed at RIVM and is used to evaluate possible policy measures for long-range trans-boundary air pollution issues.

Aim of the research

We aim to develop a model which describes daily and yearly average surface PM concentrations for Europe. At the moment the emission data included is limited to anthropogenic sources, because our main goal is to assess possible policy measures which are directly related to (Dutch) anthropogenic sources. However, the uncertainties related to all PM aspects (sources, sinks, dispersion and health effects) are still relatively large. Therefore, it is premature to limit the description to anthropogenic sources and we aim to extend the module with other relevant PM sources and sinks allowing an improved specification of the relative importance of certain measures. This research is performed in collaboration with PM health-effect studies (Rombout *et al.*, 2000).

Activities during 2000

The first version of the PM module in the EUROS model has been set up. The dispersion of anthropogenic emitted primary particles is described distributing PM_{10} over 5 size classes. Secondary PM (sulphate and nitrate) formation is described in a condensed ozone scheme, which includes 4 production reactions for particulate sulphate and nitrate: 2 gas-phase production reactions and 2 first order reactions representing formation in cloud water and on existing aerosols. PM is removed through dry and wet deposition (at present scavenging by rain of primary PM is not applied). Yearly averaged emission input data include anthropogenic primary sources of PM_{10}, SO_2 and NO_x. The SO_2 and NO_x emissions are described with monthly, weekly and daily variation. PM10 emissions have a daily emission profile.

Principal results

First results have been produced with EUROS describing the dispersion of primary anthropogenic PM sources and secondary particulate sulphate and nitrate. Figure 1 shows as an example of the first EUROS results the distribution of PM_{10} over Europe.

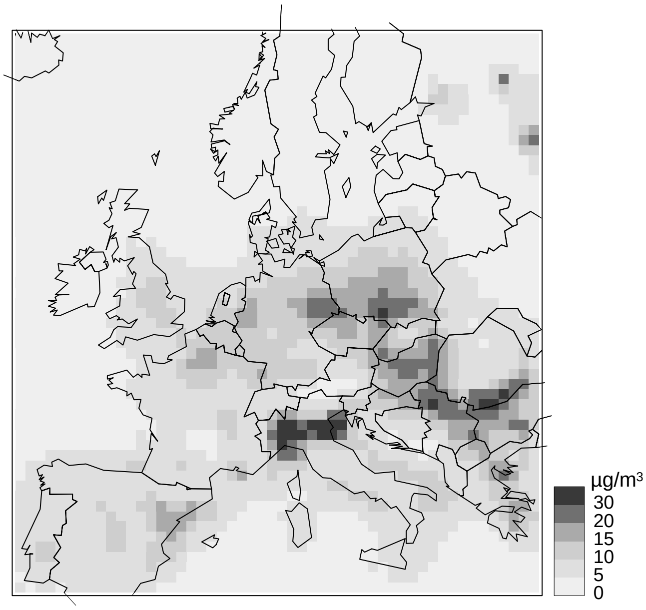

Figure 1. Yearly average PM_{10} concentrations ($\mu g/m^3$) over Europe, simulated with the EUROS model. The modelled PM_{10} consists of primary anthropogenic emitted particles and secondary sulphate and nitrate.

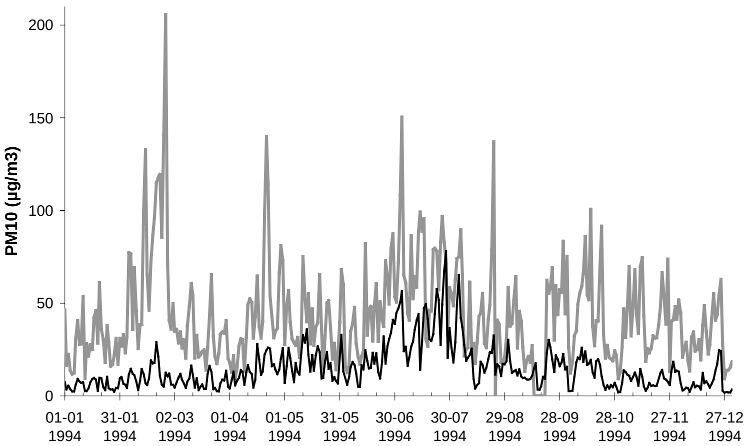

Figure 2. Daily average PM_{10} concentrations ($\mu g/m^3$) measured at the Dutch background air-quality station 'Vredepeel' (grey line) and simulated for the same location with EUROS (black line). The modelled PM_{10} consists of primary anthropogenic emitted particles and secondary sulphate and nitrate.

Main conclusions

The completed PM module results in hourly, daily and yearly averaged values for PM_{10} of anthropogenic origin. Non-episodic concentration changes from day-to-day are qualitatively well described. First results underestimate the measured PM_{10} values. Peak concentrations during for instance the episode in February 1994 (Figure 2) are not reproduced. It has been shown (Visser and Römer, 1999) that the variability in daily average PM_{10} has a strong correlation with meteorological parameters which are related to the magnitude of vertical exchange (mixing layer height, wind velocity and inversion strength). The vertical resolution and parameterisation for vertical exchange in the applied EUROS version is probably inadequate to represent such episodes.

Possible policy relevance

The research performed at RIVM is policy related since it is directed to inform the Dutch government on public health and environment issues.

Aim for the coming year

Validation and evaluation is ongoing. In addition implementation of grid refinement is planned to obtain a grid resolution of about 7.5 x 7.5 km, which is high enough to represent the PM sources and sinks of industrialised and/or urban areas in Europe. Furthermore, it is anticipated to extend the present secondary aerosol parameterisation with NH_3/NH_4^+ and include a secondary organic aerosol scheme.

Acknowledgements

The authors would like to thank the Netherlands organization for applied scientific research (TNO) and the energy research centre of the Netherlands (ECN) for their contribution to the development of the PM module in EUROS. Furthermore we thank Markus Amann of the International Institute for Applied Systems Analysis (IIASA) for discussions on PM and emissions.

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Development and implementation of the EUROS model for policy support in Belgium

A contribution to subproject GLOREAM

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Summary

The EUROS (EURopean Operational Smog) model is a regional atmospheric model developed and implemented to simulate photochemical smog situations on a long-term base. EUROS can provide policy support by modelling the impact of emission reduction strategies and emission reduction measures on ozone formation. The model has originally been developed at the National Institute of Public Health and Environmental Protection (RIVM, Bilthoven, The Netherlands) (van Loon, 1996). In the framework of the BelEUROS project, supported by the Belgian federal authorities, the EUROS model is further developed and implemented in Belgium for providing policy support with respect to tropospheric ozone.

The latest developments on the EUROS model concern the calculation of the horizontal transport and determination of the mixing height. The present report mainly describes the second aspect. The mixing height (MH) is a fundamental parameter in many air pollution models. Its practical determination is often based on simple parameterisations, which only need a few input meteorological parameters. For validation purpose, it is useful to compare the results of these parameterisations with observational data. In this study we have compared the mixing height estimates from EUROS with those obtained from LIDAR measurements and from radio soundings.

Aim of the research

The aim of the research is the implementation of the EUROS model in Belgium as an operational tool used by the Belgian decision makers for the evaluation of potential emission reduction measures on the tropospheric ozone concentrations. This includes the following tasks:

- further development of the model,
- validation of some model parameterizations (mixing height, for example),
- implementation of EUROS at the Interregional Cell for the Environment (IRCEL, Brussels) as a tool for policy support in Belgium,
- development of a user friendly user interface,
- evaluation of abatement strategies from emission scenario computations.

Activities during the year

Development of the EUROS model

- validation of mixing height parameterizations through comparison with observations,
- implementation of a multi-layer structure for the representation of the horizontal transport and the introduction of a spatially-variable mixing height.

These tasks were performed in close cooperation with RIVM in The Netherlands.

Implementation at IRCEL for policy support

- development of a user friendly input/output user interface (work carried out a the Faculté Polytechnique de Mons),
- preliminary study for the development of an impact module (impact on public health and vegetation).

Principal results

We have compared the mixing height evolution at Bilthoven (The Netherlands) for August 1997 estimated in 3 different ways: (1) from the parameterisation used in the photo-chemical smog model EUROS, developed at RIVM (van Loon, 1996), (2) as derived by VITO from the ECMWF vertical profiles using a Richardson number method, (3) from the LIDAR measurements at RIVM.

Methods used for the determination of the mixing height

1. EUROS Parameterization. In EUROS, the MH is calculated from from the friction velocity (u*), the Monin-Obukhov length (L) and the surface sensible heat flux (h_s), using simple parameterizations found in the literature (Nieuwstadt, 1981; Holtslag and Westrhenen,1989; Tennekes, 1973). Distinct parameterizations are used in stable, neutral and unstable conditions. The surface meteorological parameters u^*, L, and h_s are calculated using a software library developed at KNMI (Beljaars and Holtslag, 1990). Input parameters are wind velocity (at 10 m for example), surface air temperature (at 2 m), aerodynamic roughness length and cloud cover from synoptical observations. Input data are taken from the gridded NCAR synop observations (referred to as ODS, observational data set).

2. Richardson Number method applied on ECMWF vertical profiles. This method allows estimating the mixing height from the vertical profiles of temperature, moisture and wind. It has been used by numerous authors (see review in Seibert *et al.*, 2000). The top of mixing height is given by the top of the layer where the Bulk Richardson Number exceeds a critical value. A detailed description of the method can be found in Delobbe *et al.* (2000).

3. LIDAR measurements. The third estimate of the MH is based on the LIDAR measurements carried out at RIVM (Bilthoven, The Netherlands) (Van Pul *et al.*, 1994).

Comparison and discussion

The comparison has been carried out for August 1997. The results are illustrated in Figure 1 for the week 12-19 August 1997. Significant discrepancies are found. For most days, the EUROS MH is 100 m during the night and grows in a monotonic way up to a value around 1000 m in the late afternoon. The LIDAR and ECMWF exhibit a much larger day to day variability. In the night, ECMWF values are comparable with the EUROS estimate while the LIDAR values differ significantly.

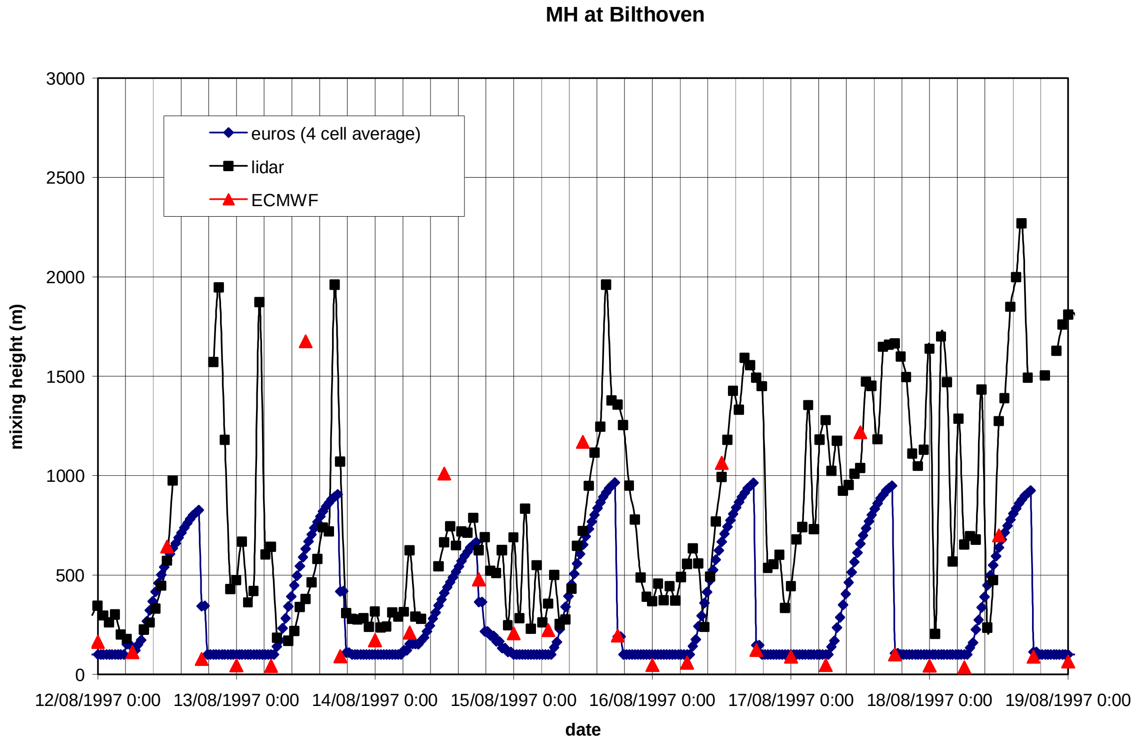

Figure 1. Mixing layer height (m) evolution at Bilthoven (The Netherlands) (1) as estimated by the EUROS parameterization (2) as derived from the LIDAR measurements (3) as derived from the ECMWF data.

During day time, the EUROS MH is usually underestimated in comparison with the ECMWF and LIDAR values. Several causes of discrepancies between the three MH estimates can be mentioned. First of all, the method used in EUROS for the calculation of the MH has its own limitations. Errors may arise from the calculation of the surface meteorological parameters (Obukhov length, friction velocity and heat flux) but also from the determination of the MH from these parameters. The meteo input used for the calculation of the surface meteo variables, for example the 2 m-temperature and the 10 m wind, may also induce inaccuracies. The meteo input used in EUROS results from a spatial interpolation from synoptic observations and a time interpolation from the 4 input times of these synoptic observations (00, 06, 12, 18 UT). This interpolation partly explains the fact that the ML diurnal cycles simulated by EUROS are much smoother than the observed diurnal cycles from the LIDAR. Another possible cause of discrepancies arises from the fact that the LIDAR observations are local while, for ECMWF and EUROS, the estimate is an average over a grid cell, which size is about 60 x 60 km^2 (for both models). The LIDAR measurements are much more sensitive to local conditions such as updraft or downdraft in convective conditions.

Concerning the estimates from ECMWF using a Richardson number method, a first source of error results from the relatively coarse vertical resolution of the ECMWF data: around 400 m in the boundary layer. The Richardson method has also its own limitations (e.g. Seibert *et al.*, 2000). In this study, the surface excess temperature has not been applied which may induce significant underestimation of the MH in convective situations. A previous study has shown the high sensitivity of the MH estimate to the surface temperature (Delobbe *et al.*, 2000). The determination of the MH from LIDAR measurements has also its limitations especially during the night (low mixing heights) and in rainy conditions.

Main conclusions

This study brings a contribution to the validation of MH parameterisations used in air quality models. It has been found that the EUROS formulation tends to underestimate the MH values and the day to day variability. Besides, the estimate based on a Richardson number method applied on ECMWF vertical profiles is generally lower than the LIDAR estimate. Our study underlines the need to test new formulations proposed in the literature. The present study has also shown that the comparison between various MH data sets is not straightforward, which makes the validation procedure quite difficult. More fundamentally, the present work has shown the limitations of the mixing layer concept and its use in air pollution models.

Aim for the coming year

Implementation of the EUROS model for policy support with respect to tropospheric ozone in Belgium, with the following specific tasks:

- validation of EUROS for Belgium,
- training of the potential users of EUROS,
- operational use of EUROS for policy support in Belgium,
- determination of optimal strategies for parallellisation of the EUROS model,
- design and implementation.

Acknowledgements

The authors would like to thank the Prime Minister’s Services Federal Office for Scientific, Technical and Cultural Affairs (OSTC) for their financial support in carrying out the projects and activities mentioned above. Furthermore, we would like to thank the Laboratorium for Air Research of the RIVM at Bilthoven (NL), the Meteorology and Air Quality Group of the University of Wageningen (NL), the Catholic University of Louvain-la-Neuve (Belgium) and the Engineering Faculty of Mons (Belgium) for their fruitful co-operation.

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Evaluation of the Model System KAMM/DRAIS

A contribution to subproject GLOREAM

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Summary

The evaluation activities in the frame of the German Tropospheric Research Programme for the Chemistry-Transport Model System KAMM/DRAIS during the last year were focused on the BERLIOZ case (Becker, 2001), a summer smog episode in July 1998 in the area of Berlin. Although a great number of species concentrations are simulated, the presented results are restricted to ozone as most important species.

The ground level ozone data have been compared especially in the time period between 11 UTC and 16 UTC, where the highest ozone concentrations occur. For the BERLIOZ episode the differences between the measured and simulated ozone concentrations in the mentioned time period are lower than 8 ppb in 50 % of the cases. This is also true, if the results of the TRACT, FLUMOB and BERLIOZ cases are combined. The combination improves the slope of the regression line and the correlation coefficient remarkably.

A comparison with measurements performed by the IBUF aircraft has also been carried out. Although the meteorological conditions are very well simulated by the model, the formation of ozone in the city plume of Berlin is underestimated in distances of about 75 km downwind. The underestimation lies in the order of 10 ppb. In the other regions around Berlin the agreement is remarkably better. Because the formation of ozone is strongly dependent on the VOC and NO_x emissions, it should be tested if a modification of the emissions provides better agreements with the measurements.

Aim of the research

The model system KAMM/DRAIS consists of the meterological model KAMM and the dispersion model DRAIS. It is designed to simulate the transport, diffusion, deposition and chemical transformation of the relevant species in a mesoscale area. The main objective of the evaluation is the estimation of the reliability of the model system KAMM/DRAIS in predicting the air pollution distribution, especially that of the ozone concentration. Up to now the model evaluation is based on data from the following episodes:

- 16 September 1992, an intensive measuring period of the TRACT experiment which took place in south-west Germany and parts of France and Switzerland,
- a summer smog episode on July 26/27, 1994 in Northrhine-Westfalia (NRW),
- a summer smog episode on July 26/27, 1994 in the area around Berlin (FLUMOB),
- a summer episode on July 20/21, 1998 in the area around Berlin (BERLIOZ).

The results of such a model evaluation shall not only indicate how realistic the simulations are but also where there are still weak points in the model approaches or in the input data.

Activities during the year and principal results

During the last year the BERLIOZ episode has been evaluated. The model domain for the simulation has an extent of 200 km x 200 km and encloses the city of Berlin, which lies nearly in the centre of the domain. The horizontal grid resolution for the simulations with the model KAMM/DRAIS was 2 km. The external forcing for the KAMM model and the initial- and boundary conditions for the species concentrations in the DRAIS model have been determined from the results of the European scale model EURAD. The grid size for this simulation was 18 km. The emission data for this episode has been provided by IER, Stuttgart.

The ozone concentration distribution near ground level at 14 UTC of July 20, 1998 shows a pronounced ozone plume downwind of the city of Berlin (Figure 1). The maximum ozone concentration in this plume is less than 70 ppb. The maximum value occurs at a distance of about 70 km.

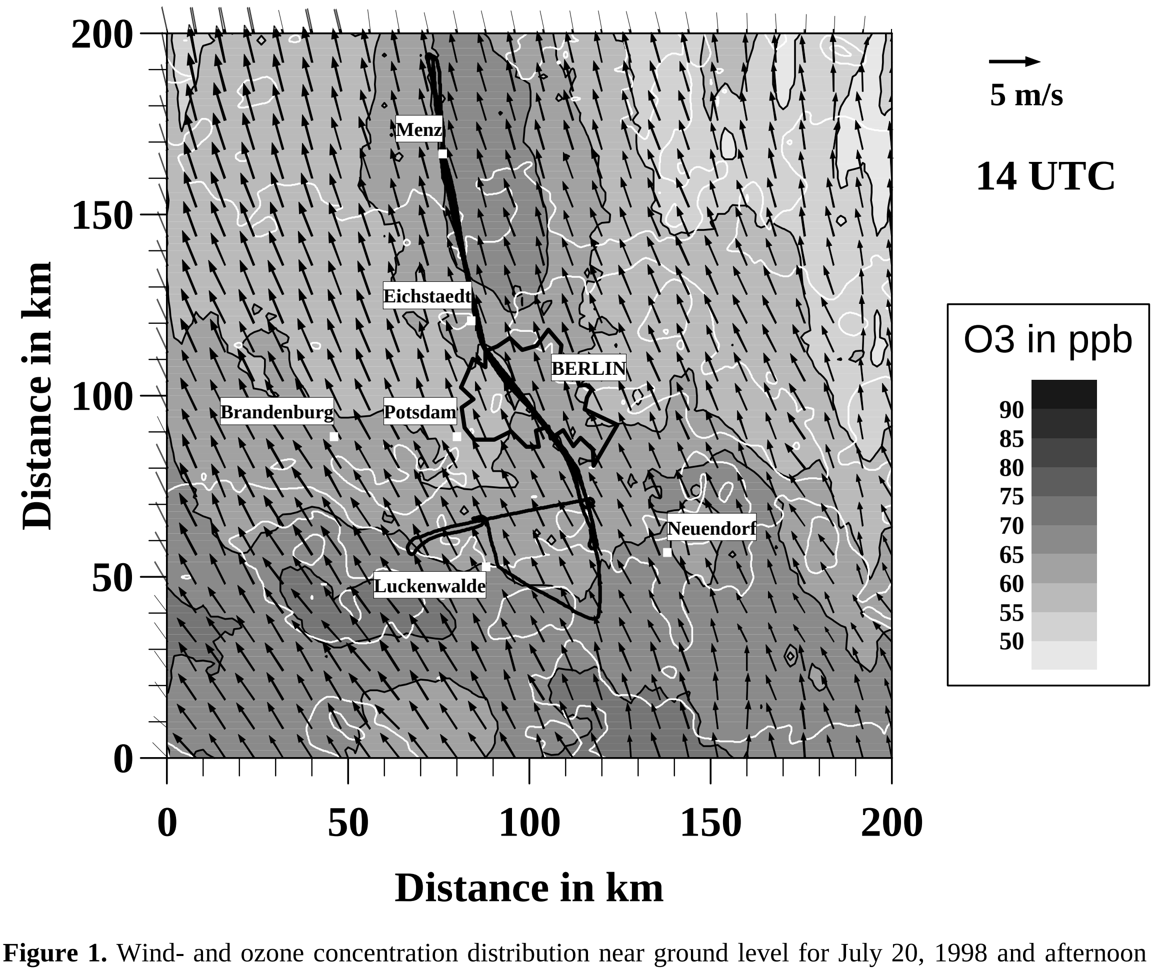
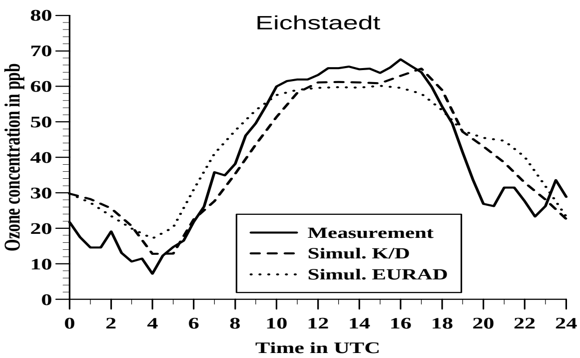


Figure 1. Wind- and ozone concentration distribution near ground level for July 20, 1998 and afternoon flight track of the aircraft IBUF.

The diurnal cycles of the ozone concentration at two stations are presented in Figure 2. At the station Eichstaedt, which is located in downwind direction at a distance of about 30 km from the city centre of Berlin, the simulated ozone concentrations fit quite well to the measurements. But at the station Menz, which is located downwind in a distance of about 70 km from the city centre, the simulated ozone concentrations are underestimated compared to the measured ones. If we compare the ozone concentrations measured along the flight track of the aircraft IBUF (see Figure 1) with the corresponding simulated concentrations a similar result is found. The main flight track begins in about 50 km on the upwind side of Berlin, crosses the city, and ends at about 100 km downwind. The aircraft flew in different levels along this flight track. The wind direction along the flight track is well simulated by the model as can be seen from Figure 3. A similar agreement is found for the wind speed. The simulated ozone concentration along the flight track agrees also quite well with the observations except of two time periods (13.8 UTC and 15.1 UTC) where the model underestimates the measured ozone values. During these two periods the aircraft flew in the area of Menz. This means, that the simulation underestimates the formation of ozone in the city plume of Berlin at larger distances by about 10 ppb although the meteorological conditions are well simulated.



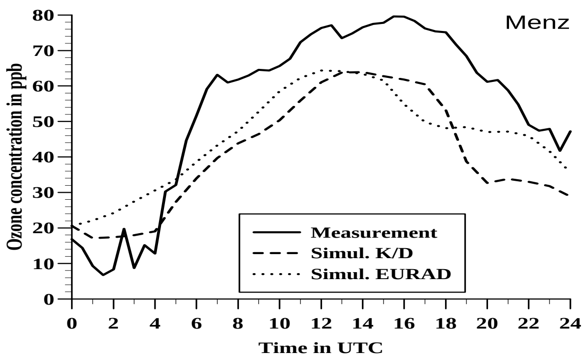


Figure 2. Diurnal cycle of the ozone concentration at ground level for the stations Eichstaedt and Menz.

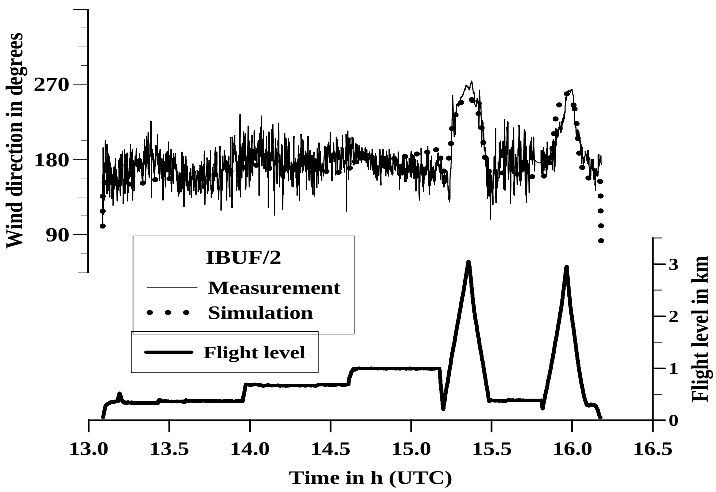


Figure 3. Wind direction comparison along the flight track of IBUF.

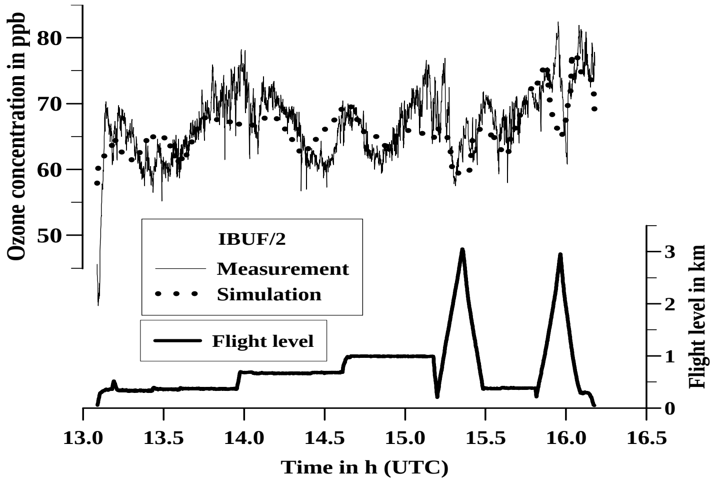


Figure 4. Ozone concentration comparison along the flight track of IBUF.

A statistical evaluation with the data from the ground level stations is also carried out in a similar way as it was done for the earlier episodes. The cumulative frequency distribution for the time period between 11 UTC and 16 UTC shows, that in 50% of the compared cases the difference between the measured and simulated ozone concentrations is less than 8 ppb. In the scatter diagram it can be seen that the data are grouped around the agreement line. The great scatter is represented in the low correlation coefficient of 0.54. On an average the higher ozone concentrations are underestimated and the lower values are overestimated by the model simulation. This is in agreement with the results of the earlier evaluated episodes.

If the data of three evaluated episodes TRACT, FLUMOB and BERLIOZ (the NRW case is not included because of the less reliable emissions) for the time period between 11 UTC and 16 UTC are combined and compared with the corresponding model results, the agreement is much better than for the individual episodes. Figure 5 shows the scatter diagram. The data are well grouped around the agreement line.The slope of the regression line and the correlation coefficient have the acceptable values of 0.76 and 0.85, respectively. The frequency distribution of the difference between measured and simulated ozone concentrations is plotted in Figure 6. This difference is lower than 8 ppb in 50% of the compared data. 8 ppb is about 12% of the average ozone concentration.

Main conclusions

At the first day of the BERLIOZ episode an ozone plume downwind of the city of Berlin develops. Although the meteorological conditions and the ozone concentrations on the upwind side of the city are well simulated by the model the observed formation of ozone in the plume is about 10 ppb more than the simulated one. Because the formation of ozone is strongly dependent on the VOC and NO_x emissions from the city of Berlin, it should be tested, if a modification of the emissions provides better agreements with the measurements.

The results of the statistical evaluations based on the simulations with the KAMM/DRAIS model system for the episodes TRACT, FLUMOB and BERLIOZ show that in about 50 % of the compared cases the difference between the measured and simulated ozone concentrations in the time period between 11 UTC and 16 UTC is lower than 8 ppb, if ground level data are considered. In this time period usually the highest ozone concentrations occur. The direct comparison of the measured and simulated ozone concentrations provides a correlation coefficient of 0.85, which is a rather good value. The slope of the regression line has a value of 0.76, which indicates that on an average the higher ozone concentrations are underestimated and the lower ones are overestimated by the model simulations.

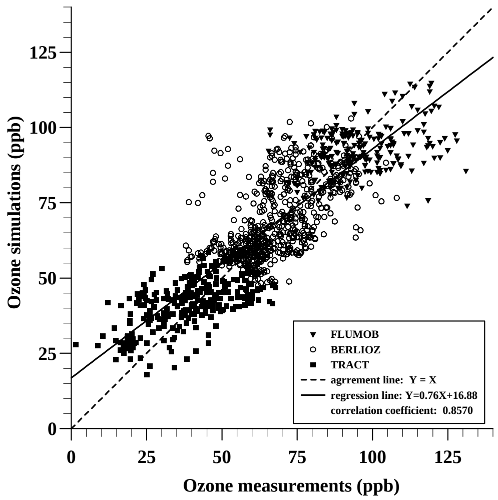


Figure 5. Comparison of measured and simulated ground level ozone concentrations (11 UTC till 16 UTC).

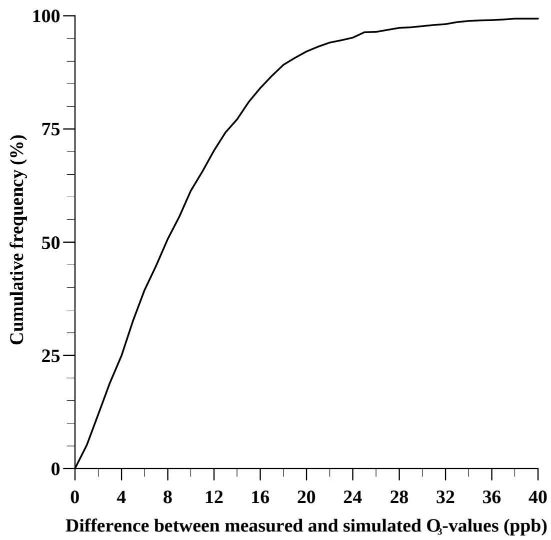


Figure 6. Cumulative frequency distribution of the ground level ozone concentration difference (11 UTC till 16 UTC)

Aims for the coming year

New simulations of the BERLIOZ episode will be carried out in order to find out which modifications of the precursor emissions provide a more realistic formation of ozone in the city plume of Berlin.

Acknowledgements

We thank the Ministry for Science and Technology for the financial support of the project. All colleagues involved in the model evaluation activities are acknowledged for the good co-operation.

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Utilisation of Cloud Parameters for Chemical Transport Model REM3

A contribution to subproject GLOREAM

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Introduction

Some studies have been carried out to improve the accuracy of cloud parameters for use in chemical transport modeling (REM3, LOTOS) by use of synoptic meteorological observations and satellite data from Meteosat and NOAA. Cloud parameters are needed for boundary layer modeling, for the determination of photolysis frequencies and for an chemical aerosol module in CTM.

In 2000 a project started within the BMBF main project GLOWA dealing with climate problems in relation to water resources and water quality in the Elbe region. Cloud parameters (cloudiness, cloud type, 3D-formation) are determined to validate the climate model REMO from MPI Hamburg and to drive the cloud module of the CTM ADOM to model deposition of nitrogene in Middle Europe.

Research Activities

A special raid system (up to 3 Tbyte) was installed at the Institut für Meteorologie to make possible the utilisation of the 10 year archive of Meteosat and NOAA satellite data for that project. Cloud parameters are analysed by a maximum likelyhood procedure developed by BERGER.

First results of the GLOWA-Elbe project concerning clouds show

- a very good accordance of cloud cover from Meteosat data with NOAA AVHRR data and a quite good agreement of satellite data with synop and model data,
- many differences between the analysis of cloud types from Meteosat and NOAA AVHRR data, caused partly by the lower horizontal resolution of Meteosat data with 6 km x 8 km in 50EN in comparison to NOAA with 1.2 x 1.2 km also compared to synoptic observations.
- a very good consistency of the cloud types from Meteosat with precipitation data from observations.

In this first step time series of precipitation and cloud parameters from observations are compared to model simulations.

A general problem for the correct analysis of the satellite data are the reflectance characteristics of the land surface which varies with changing satellite-sun-earth geometry. Complex radiative transfer models have been used to identify the essential components of plant canopy and bare soil reflectance.

A BRDF (Bidirectional Reflectance Distribution Function) model will be used to correct the albedo values from the both NOAA-AVHRR channel 1 (visible 555-680 nm) and channel 2 (near-infrared 725-1110 nm) and also for the visible channel of Meteosat (0.7-1.1 µm). The correction of albedo values by the BRDF model will result in a better determination of cloud classification, because f.e. the BRDF effect is very important to analyse the differences of optical thin cirrus or thin low clouds in relation to the cloud-free surface.

Moreover, the diurnal variations of the cloud type distribution from Meteosat can be compared to the diurnal variations of the sun azimuth.

Of special interest are weather situations showing a mix of areas with mainly optical dense clouds with sharp borders, with partly cloudy and cloud-free areas to solve for the 3D-formation by use of numerical meteorological analyses. On the other hand, cases with optical thin clouds or with partly cloudiness over large areas are of interest, to analyse the different uncertainties in the cloud parameter procedures.

For an improved analysis of precipitation data the consistency of Meteosat cloud presentation and observed precipitation from synoptic data is checked. In a first step the time series of Meteosat cloud presentation is tested to additionally adjust the numerical interpolated precipitation fields (1 and 3 hourly), which are determined by climatological interpretation of WMO actual weather classes with an adjustment by observed precipitation amounts. Also automatic hourly synoptic measurements are available for most of the observing DWD stations in Germany in addition to the usual 12 and 24 hourly sums of precipitation. The hourly precipitation values will be compared to Meteosat cloudiness series to consistently break down the 12 and 24 hourly precipitation sums to hourly gridded information.

Activities for the next years

The cloud parameters will be analysed for long time series of Meteosat and NOAA Satellite data in combination to synoptic observations to be compared to climate model runs.

For special episodes cloudiness, cloud types and 3D-formation will be analysed for diagnostic runs of the CTM REM3/Calgrid for tests on the determination of special secondary aerosol formation. For diagnostic CTModeling the 3D-cloud structure and precipitation is highly needed to localize and initialize cloud chemistry modules.

RSM-MM5-CAMx simulations over Europe

A contribution to subproject GLOREAM

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Summary

In this contribution we have used the MM5-RSM-CAMx system over the European domain with 50 km gridcell to study the impact on the ozone concentrations using different initial concentration fields. The Regional Spectral Model (RSM) (NCEP/NOAA, USA) is running over an Alpha/Compaq XP1000 machine and the MM5 model (Pennsylvania State University / NCAR, USA) over a Pentium III 1000 under LINUX OS. Both meteorological models are used to provide input meteorological information to the CAMx air pollution transport model (ENVIRON Co.). EMEP emission inventory is used to provide input emission data into the simulation system. The CBM-IV chemical mechanism (simplified version) is also used into the CAMx module.

Two different initial concentration fields are used to compare the quality of the results with observations (Madrid and Bilbao (Spain)) and also Leicester (United Kingdom). Two different scenarios are defined: a) initial concentrations are set to zero and b) initial concentrations are taken after five days of running the model.

Aim of the research

In this contribution we have focused on studying the impact of initial concentration of the mesoscale air quality models and also on the integration of global through urban scale in future air quality modelling systems. So that, use of global models (GSM) running on-line and dataset from global models transferred through Internet (MRF/AVN) and progressive nesting to urban scale together with dispersion models (off-line application) such as CAMx or UAM-V and in further developments CMAQ-Models-3 is used to study the importance and impact of long range transport of mesoscale β and γ scales in air pollution modelling.

Activities during the year

The laboratory has been running operationally the RSM and MM5 models in our web site (http://artico.lma.fi.upm.es). In addition to this capability, CAMx model (ENVIRON Co.) has been used to simulate ozone concentrations over the European domain by using RSM and MM5 meteorological input datasets.

Principal results

In Figure 1 we show the CAMx data requirements. We have used the EMEP emission inventory properly interpolated and project transformed to the CAMx projection scheme (Lambert and UTM). In addition to this, we have used the USGS 1-km NOAA/AVHRR landuse datasets for Europe to run CAMx model. The topography of the terrain is the GTOPO

30’’ Digital Elevation Model. Figure 2 shows the MM5-CAMx interface which has been applied over European domain.

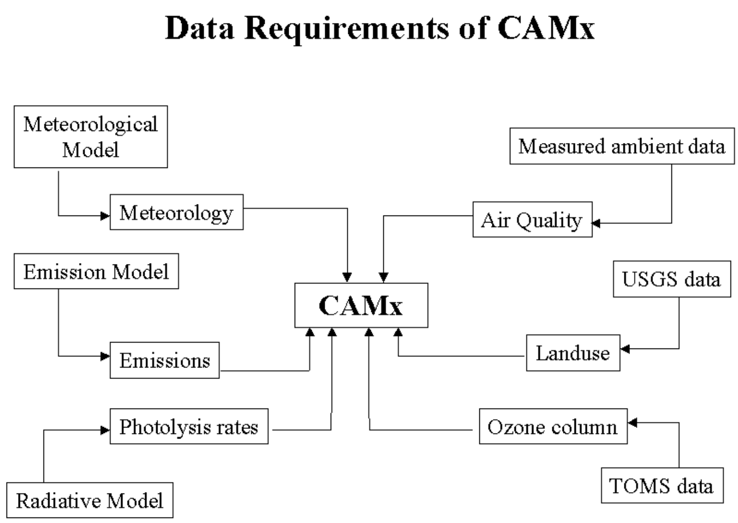

Figure 1. CAMx data requirements.

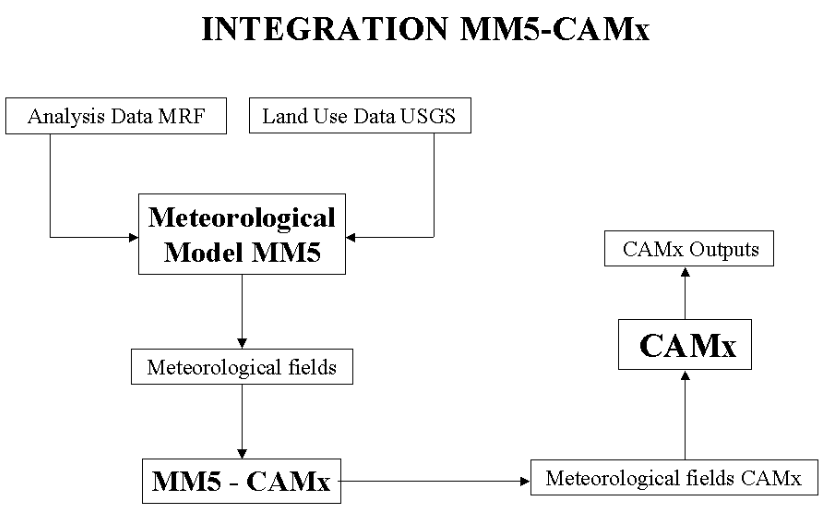

Figure 2. MM5-CAMx modelling system.

Main Conclusions

Observational data in Bilbao and Madrid have been compared with simulated data from RSM-CAMx and MM5-CAMx modelling systems. Two scenarios have been used: A) Both modelling systems have been executed for the August, 2-6, 2000, 120 hours period over

Europe with 50 km spatial resolution. Initial concentrations have been put to default. B) The same as scenario A) but initial concentrations are coming from a previous simulation July, 28-August, 1, 2000, with RSM-CAMx modelling system. Results show that improving the quality of initial concentrations is having an important improvement on the quality of the results. Tables 1-4 show some results when using the so-called “window” approach.

Table 1. Window approach comparison by using scenario A (predefined initial concentrations) and scenario B (forecasted initial concentration from previous five day simulations with RSM-CAMx modelling system).

	RSM - CAMx	
60 % window approach	Coslada	Bilbao
A (predefined con.)	100 %	60 %
B (forecasted con.)	100 %	60 %

Table 2. Window approach comparison by using scenario A (predefined initial concentrations) and scenario B (forecasted initial concentration from previous five day simulations with RSM-CAMx modelling system).

	MM5 – CAMx	
60 % window approach	Coslada	Bilbao
A (predefined con.)	100 %	60 %
B (forecasted con.)	100 %	80 %

Table 3. Correlation coefficient comparison by using scenario A (predefined initial concentrations) and scenario B (forecasted initial concentration from previous five day simulations with RSM-CAMx modelling system).

	RSM - CAMx	
Correlation coefficients	Coslada	Bilbao
A (predefined con.)	0.499	0.303
B (forecasted con.)	0.648	0.601

Table 4. Correlation coefficient comparison by using scenario A (predefined initial concentrations) and scenario B (forecasted initial concentration from previous five day simulations with RSM-CAMx modelling system).

	MM5- CAMx	
Correlation coefficients	Coslada	Bilbao
A (predefined con.)	0.171	0.412
B (forecasted con.)	0.762	0.802

Aim of the coming year

The Laboratory is intending on progressing on the use of continental and global air quality models (meteorological and dispersion) by going all the way from street level to global scale and using recognized high quality meteorological and dispersion models. We are intending to use CMAQ model together with MM5 and link it to OPANA air quality model.

Acknowledgements

We would like to thank Hann-Ming Henry Juang and Masao Kanamitsu from NOAA (USA).

Inverse Dispersion Modelling as a Tool to Derive Emission Data from Measurements

A contribution to subprojects GLOREAM and GENEMIS-2

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Summary

The new inversion method based on source-receptor matrices from backward runs of a Lagrangian particle dispersion model was further refined, especially for the quantification of point sources with simultaneous location in space and time. This method turned out to be of high relevance in the context of the verification of the Comprehensive [nuclear] Test Ban Treaty (CTBT).

Aim of research

This contribution aims at the development of inverse modelling methods to derive information on emissions from measurements in the regional scale. Such methods shall be applied to suitable data sets and results be compared with conventional emission estimates. In addition, recommendations on the optimum monitoring network design shall be made.

Activities during the year

- The inversion method was refined on the basis of the ETEX tracer experiment.
- The development of a convection parameterisation for the FLEXPART model was begun, in order to improve its usefulness as a global transport model.
- Results were presented at the EUROTRAC Symposium in Garmisch-Partenkirchen (Seibert, 2001d), the International Technical Meeting on Air Pollution Modelling and its Applications in Boulder (Seibert, 2001a), and the Informal Workshop on Meteorological Modelling in Support of CTBT Verification in Vienna (Seibert, 2001b and 2001c). In addition, talks on the subject have been given at the NOAA Aeronomy Laboratory in Boulder and at the Canadian Meteorological Centre in Montreal.
- Contacts with the Comprehensive Test Ban Treaty Organisation (Preparatory Commission) in Vienna have been continued and intensified. Project team members continued to participate in the Working Group B of national technical experts and entered in a closer co-operation with the leader of the informal working group on meteorological aspects, Mr. Michel Jean from the Canadian Meteorological Service. This co-operation eventually lead to a visit in Montreal and the organisation of an informal workshop on meteorological modelling in support of CTBT verification in Vienna by the project team (Seibert, 2000b, 2000c).
- Because of these application-oriented activities and the necessity of the project team to work on other projects, too, original aims with respect to air pollution applications in a more traditional sense were not yet reached.

Principal results

A new inverse modelling approach was developed in the previous year, based on the construction of a source-receptor matrix by running the Lagrangian particle model (LPDM)

FLEXPART (see Stohl *et al.*, 1998) backward and subsequent regularised inversion. The method has presently been applied to the first release of ETEX (Seibert, 2000, 2001a).

An accurate and efficient method was implemented for the joint location of the source in time and space, utilising the knowledge that the source is a point source. The method consists of doing a temporal inversion for a hypothetical source located just in one grid cell, and repeating this for all the grid cells (or a reasonable subset). For each of these possible locations, the misfit between observed tracer concentrations and simulated tracer concentrations using the reconstructed temporal evolution of the source is calculated. Suitable measures are the root-mean-square error (RMSE) and the correlation coefficient (see Figure 1). We see that there is a “channel” of possible locations of the source, with the best correlation near the real source location. If the temporal evolution belonging to the different potential source grid cells is looked at, the expected result (earlier release for grid cells to the west of the real location, and vice versa) is found. This technique is very efficient, because it is numerically much cheaper to solve many small linear problems than a single big one.

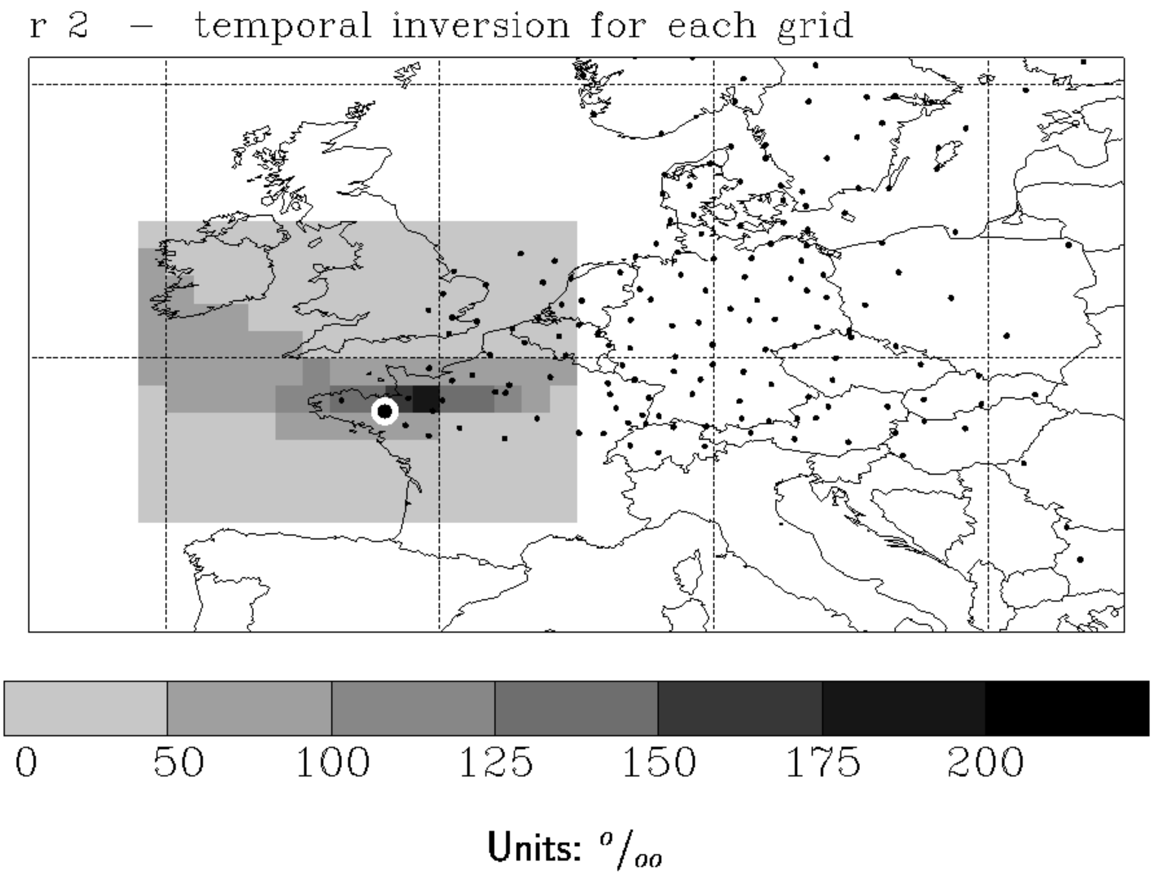

Figure 1. Application of the inverse modelling technique to the ETEX-1 release. Correlation coefficients between observed and modelled tracer concentrations (all sites, all times) for hypothetical sources placed in the respective grid cell are indicated by the grey scale, with its temporal evolution determined by inverse modelling. True source location is marked by a big dot, the small dots indicate measurements sites.

Here we see also a big advantage of the source-receptor matrix method: once this matrix has been calculated and stored, it is easy to do a number of different evaluations, to perform inversions with different cost functions, or to iterate the inversion to approximate non-linear cost functions. If the cost function is changed in a set-up with an adjoint Eulerian system which minimises the cost function directly, iterating with forward and backward (adjoint) runs, the whole procedure has to be rerun. As in the LPDM the computation time depends mainly on the number of particles and not much on the number of species, tracing a separate species for each measurement as it is necessary in our approach does not increase the computation time. It does, however, increase the memory requirements.

For more information, please check the project web page at

http://boku.ac.at/imp/envmet/invmod.html

The development of a convection scheme for use in FLEXPART is under way, as a contribution to the EU project STACCATO. This scheme redistributes particles statistically according to convective tendencies of tracers with initial distribution in form of a delta function (one level unit mixing ratio, other levels zero) calculated by the scheme of Emanuel and Zivkovic-Rothman (1999) from the given large-scale profiles of temperature and humidity. Thus, the applicability of FLEXPART in regions where convection is important (especially the tropics) and for long-time simulations as relevant for substances influencing the climate will be improved.

Main conclusions

The inverse modelling method developed in the previous year, which is based on the calculation of a source-receptor matrix with a backward running Lagrangian particle model, has been further refined for better applicability to point sources. The method continues to show its usefulness for different applications. The chosen approach combines high accuracy and efficient usage of computational resources for receptor-oriented problems. The source-receptor approach facilitates flexible usage of the output of the transport calculations.

A convection parameterisation for the Lagrangian particle model will enhance its potential for climate-oriented studies.

Aim for the coming year

- Completion of the FLEXPART convection scheme.
- Expansion of the theoretical basis of source-receptor matrix derivation from particle dispersion models to the case including sinks.
- Further refinement of the regularisation schemes.
- Participation in the ad-hoc expert group for the evaluation of atmospheric transport models test in the context of the Comprehensive Test Ban Treaty verification.
- Application to other tracer experiments (CAPTEX, ANATEX) and the release from the Chernobyl nuclear disaster 1986.
- Application to air pollution data.
- All applications will be carried out with FLEXPART and not with the box model IMPO as originally foreseen.

Acknowledgements

The funding by the Austrian "Fonds zur Förderung der wissenschaftlichen Forschung (FWF)" under grant P1295-GEO, the Austrian Federal Ministry of Education, Science and Culture (GZ 76.017/1-VIII/A/5/2000) and the European Commission (Project STACCATO, EVK2-CT-1999-00050) is gratefully acknowledged. We thank ZAMG for access to meteorological data from the ECMWF and A. Neumaier (University of Vienna) for mathematical advise.

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Results of possible policy relevance

The relevance of the methods and experiences developed in this contribution in the context of the Comprehensive Test Ban Treaty verification has been confirmed. As one consequence, a member of the project team has been invited to participate in an ad-hoc expert team tasked with the evaluation of the meteorological tools used at the CTBT Provisional Technical Secretariate's International Data Centre.

Theses

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Geographic Information System (GIS) Methods in Land Use Mapping for Air Pollution Models

A contribution to subproject GLOREAM

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Summary

Geographic Information System (GIS) make a very important contribution to meteorology and chemistry transport models. They can store, administrate and provide all necessary input data in specified formats and visualise the model output. In addition, GIS can be used to interface atmospheric models with other models, such as those used to determine critical loads and levels, biogenic VOC emissions or with integrated assessment models for development of air pollution abatement strategies.

The paper describes in broad terms the GIS support for the Meteorology-Climate-Chemistry-Model (MCCM) Information System (GSfM) developed at the Fraunhofer Institute for Atmospheric Environmental Research(IFU).

Aim of the research

The aim of the research is the development methods based on geographical information systems (GIS) and relational data base management system (RDBMS) for compilation and processing land use and plant species-specific data for use with meteorology-chemistry models**.**

Activities during the year

During the year several new features have been added to the GSfM information system. The information system has been used in several studies for provision of the input data for use with atmospherical models. Several studies have been performed using the BVOC emission module. This GIS based module proved to be an excellent tool, which combines the results of the numerical models with the GIS and RDBMS technology, in provision of BVOC emission inventories.

Principal results

GIS support for MCCM has been written in ArcInfo-AML, SQL, and Perl with additional C and Fortran programs. It consists of four major modules:

ToModel - geo data for input into numerical models

- landuse compilation for specified area
- landuse processing for specified domain and subdomain system according to MM5 requirements
- plant species specific landuse, vegetation parameters (biomass, LAI, VI) and VOC emission factors
- digital elevation models (DEM)

FromModel - visualisation of the MCCM output data

- interface to VIS5D
- read MCCM-output
- perspective view in 3D
- data export

GIS-Model

- biogenic VOC emissions
- spacial disaggregation of anthropogenic emissions

Mapping - Mapping module

The system utilises two databases: The geo database stores all spacial data in vector or raster format. Several data sets on land use, LAI, VI, DEM, soil properties for Germany and Europe are available. The relational database stores all thematic features, such as agricultural and forest statistics, emission factors, traffic counts and many others. The structure of the GSfM information system is shown in Figure 1.

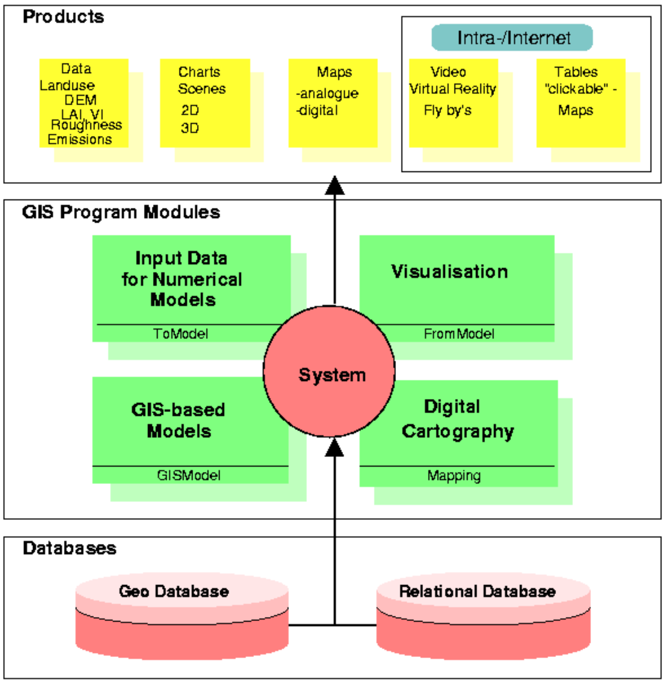

Figure 1. Structure of the GSfM information system.

Main conclusions

The GSfM information system is a comprehensive tool for managing and visualisation of both input geo data and the results of numerical models.

Aim for coming year

The major aim of the work is the further development of the anthropogenic emissions module. The module should provide MCCM tailored input emission data.

Coupling Atmospheric Chemistry with a Convective Boundary Layer Model

A contribution to subproject GLOREAM

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Summary

A highly simplified model that couples atmospheric chemistry with a convective boundary layer model was developed to be a theoretical tool to make detailed investigations of the relationship between atmospheric chemistry and boundary layer processes. The model is simple enough so that it can be used to make thousands of simulations and only about 10 minutes of computer time processing is required. Despite its simplicity the model should be able to produce reasonably realistic results. The model calculates the time evolution of the mixing height and the potential temperature of the boundary layer and its affect on the chemistry of a series of first order reactions that proceed from an emission, through an atmospheric concentration and ending in deposition. In future studies the model will be extended to more realistic chemical mechanisms and applied to the analysis of observational data.

Aim of the research

The three dominate factors that affect air quality are emissions, meteorology and atmospheric chemistry. Most chemical transport models (CTMs) incorporate as many physical and chemical processes as possible within constraints imposed by current understanding and by limited computer resources. Analysis of CTM modeling results to determine the effects of each process on air quality may be very difficult. Detailed sensitivity analysis may not be possible because large numbers of CTM simulations will require too many computer resources. In previous GLOREAM reports we have reported on the development of highly complex chemical mechanisms. Here we focus on the development of a tool for sensitivity analysis and data assimilation.

Activities during the year

A box model has been developed that simulates the development of the layer height, the horizontally averaged potential layer temperature, and the temperature jump at the upper layer boundary. The horizontal and vertical air motion, surface pressure, and time dependent surface temperature are calculated according to the parameterizations of Driedonks (1982).

The chemical mechanism consists of a simple scheme of consecutive reactions where E, k_1, k_2 and d are rate parameters for emission, reaction1 and 2, and deposition, respectively.

Emission $\rightarrow$ A	E
A $\rightarrow$ B	k_1
B $\rightarrow$ C	k_2
C $\rightarrow$ Deposition	d

An emission and/or an initial concentration of A is assumed. The species A is transformed to B and C in two reactions, and the final product, C, is deposited on the surface. In this model the chemistry is coupled to the meteorology through temperature and mixing height. If the rate parameters are temperature dependent then the temperature is calculated from the potential temperature and height of the mixed layer. The emission is treated as a volume source with a fixed mass flux. Furthermore, the layer height rise causes dilution and deposition. Therefore both the effective rate parameters for the emission of A and the deposition of C depend on the mixing height.

The model is being applied to model field data taken over the Gulf of Mexico. Observations of sea surface temperature and atmospheric temperature were made over a period of 18 hours as the research ship traveled from near New Orleans through the Gulf during the wintertime. At this location, during this time, the formation of convective boundary layers follows relatively simple physics.

This period is also interesting from a chemical point of view because here nitrogen oxides react to form ammonium nitrate particles. The simple mechanism approximates the mechanism for the formation of ammonium nitrate where $[NO]_o$ is the initial concentration, $k_1[HO_2]$ is the effective first order rate parameter of the first reaction, $k_2[HO]$ is the effective first order rate parameter of the second reaction, f is the effective yield of particles for the observed humidity and temperature and d is the deposition rate parameter for ammonium nitrate particles (Stockwell *et al.*, 2000).

Emission $\rightarrow$ NO	$[NO]_o$
$NO\ (+HO_2) \rightarrow NO_2$	$k_1[HO_2]$
$NO_2\ (+HO) \rightarrow HNO_3 \rightarrow f\ NH_4NO_3(s)$	$k_2[HO]$
$NH_4NO_3(s) \rightarrow$ Deposition	d

The meteorological boundary conditions have been obtained from measurements made within an 18 hours period. With this highly parameterized model the evolution of nitrate particle formation and deposition over this time period was simulated. The meteorology results are being compared with measurement data while the chemistry simulations can be compared with an analytical solution because of the simplicity of the reaction mechanism.

Principal results

Figure 1 shows a sample simulation with a fixed mixing height and a fixed temperature. The model reached a steady state for the concentrations of A, B and C relatively rapidly and the total deposition increases monotonically with time.

A simulation with chemistry and the convective boundary layer model are shown in Figures 2 and 3. In this simulation the convective boundary layer rises in height from a few hundred meters to about two thousand meters and the mixing layer warms several degrees, Figure 2. The changes in the meteorology cause significant changes in the chemical concentrations, Figure 3. It is interesting that species A and B reach early peak values while C does not. The final steady state concentration of C is not affected by the height of the boundary layer.

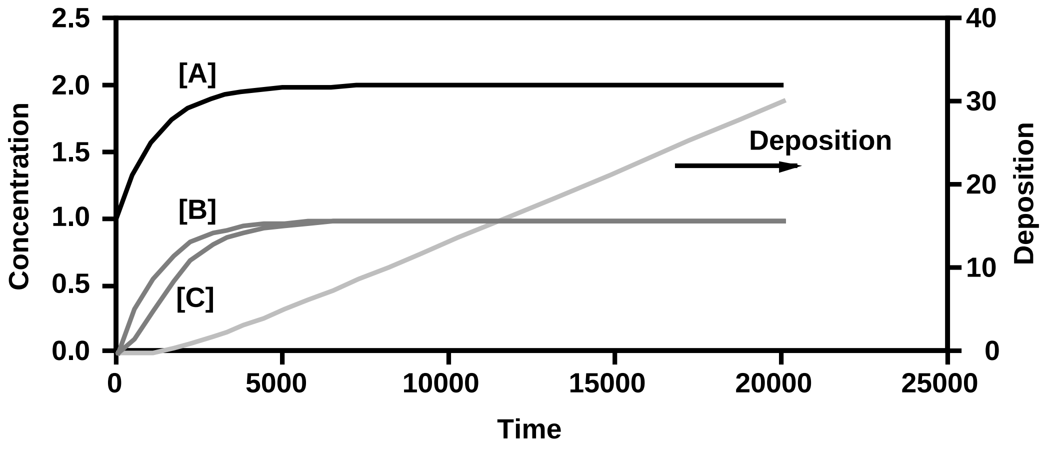

Figure 1. Plot of calculated concentrations of box model without meteorology.

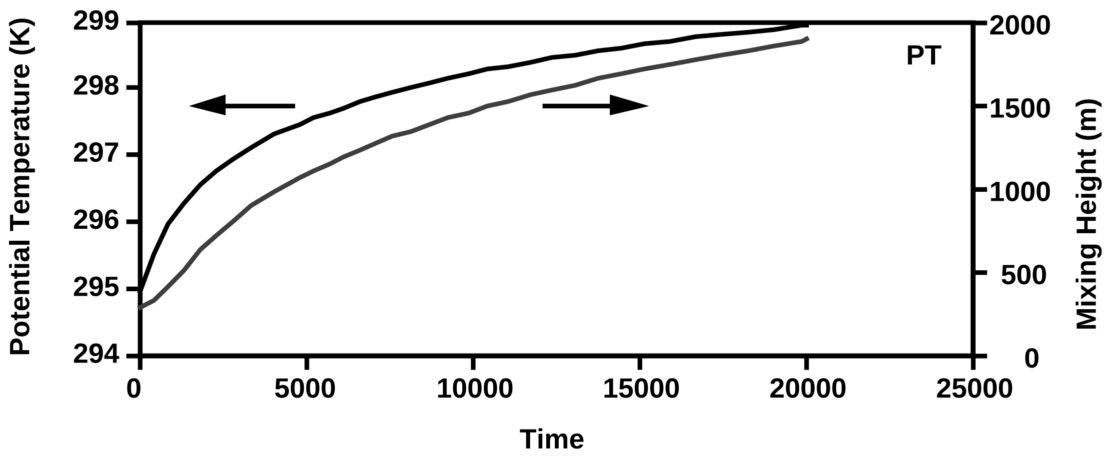

Figure 2. Plot of calculated potential temperature and mixing height for typical simulations with a rising convective mixing height.

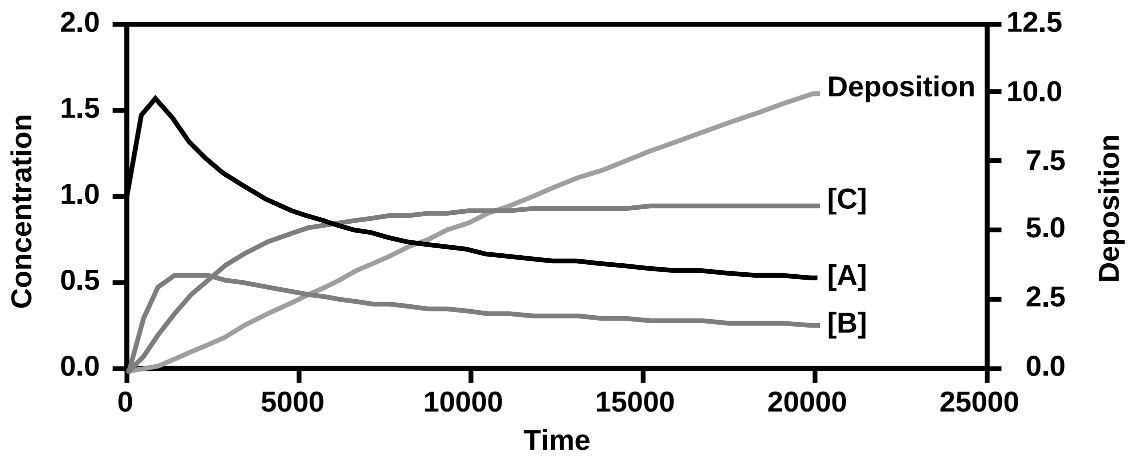

Figure 3. Plot of calculated concentrations for typical simulations with a rising convective mixing height.

Main conclusions

This simple model has been developed for the purpose of testing new methods of sensitivity analysis, including adjoint and inverse methods. Although it does not include detailed chemistry and physics it appears to make reasonable simulations of particle formation and the convective boundary layer. The system is low-order with 7 variables (3 meteorological and 4 chemical). The system is nonlinear and this leads to complex behavior that is nontrivial. The model's simplicity allows it to make several thousand simulations within a short period of time.

Aim for the coming year

The convective boundary layer model will be used for the development of new methods of adjoint and inverse methods. Furthermore we plan to develop the model to include the Regional Atmospheric Chemistry Mechanism (RACM, Stockwell *et al.*, 1997). To improve the treatment of aerosol formation, we will investigate incorporating the SCAPE aerosol mode within the model. The full model will be used to investigate the effects of ammonia, NO_x and volatile organic compound (VOC) limitations on particle formation under the influence of a rising mixing height and an increasing atmospheric temperature.

Acknowledgements

Support for this research was provided by the National Aeronautics and Space Administration, the National Atmospheric and Oceanic Administration, the Idaho Department of Health and Welfare, Division of Environmental Quality and PG&E Generating Company.

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Bulgarian Modeling: Further Development of PC-oriented Air Pollution Model for the Region of South-East Europe - Model Improvement and Applications

A contribution to subproject GLOREAM

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Summary

Two problems related with air pollution modeling are discussed in this article. The first one is adequate simulation of the transport part of the semi-empirical air dispersion equation; the second one being connected with model application for estimate of transboundary exchange of sulfur pollution between Bulgaria and Greece.

The most exploited advection scheme is Bott's one (Bott, 1989). It is explicit, positively definite, conservative, and possesses limited numerical dispersion and good transport ability. In this scheme, the Integrated Flux Concept is used when calculating mass fluxes at the cell borders. A new concept called TRAP (Syrakov, 1996) is proposed in the frame of this subproject decreasing computations to a great extent. The flux area is supposed trapezoidal and the flux is determined as a product of Courant number and a single value of the approximating polynomial, taken in the middle of the passed distance. If the same 4th order polynomial as in Bott's scheme is used and the Bott's normalization is applied, the TRAP-scheme turns out to be several times faster then this scheme displaying the same properties. The acceleration is bigger when 3rd order Bessel polynomial is used for approximating the nearest concentration values (Syrakov and Galperin, 1997). Last years, extension of this scheme to the case of non-homogeneous grid is made. In 2000, a common approach is proposed allowing the usage of all Bott-type advective schemes over non-homogeneous grids (regular and non-regular).

The 3-D Eulerian air pollution model EMAP (Syrakov, 1998) is used for estimating the sulfur deposition over South-east Europe for 1995 due to Bulgarian and Greek sources. The model region is a part of South-east Europe, taking a territory of 8x9 EMEP 150 x 150 km^2 standard grid cells with Bulgaria in the center; every cell divided to 36 25 x 25 km^2 cells. The official sources inventory for 1995 is taken from the EMEP/MSC-W 50x50 km^2 database. Additional redistribution of this data is made over the finer grid of 25 km space resolution. The meteorology input is the standard 50x50 km^2 product of the "Europa-Model" of Deutscher Wetterdienst, Germany. Annual runs with Bulgarian and Greek sources alone are made resulting concentration and deposition fields visualized and commented. As only sources over these two countries are handled the results can be considered as an estimate of Bulgarian and Greek impact in the acid pollution of the region as well as an estimate of the reciprocal pollution.

Aim of the research

The common tendency in air pollution modeling is the development of more and more complicated models oriented to high performance computers and computer networks. Usually, such models are conjugated with proper meteorological model systems producing wind and turbulence fields with high horizontal, vertical and time resolution. Such modeling is not possible for many East-european countries, so far, in spite these countries also need dispersion modeling for various applications. The aim of this contribution to GLOREAM is to develop models and model systems oriented to small computational platforms (mainly PC), working in conditions of lack of precise meteorological input. The work is directed to improvement of the Eulerian multi-level dispersion model EMAP, created in NIMH (Syrakov, 1997). It is PC-oriented and relatively simple physical and mathematical approaches are searched for as to model the various air dispersion processes. In order to perform long-term integration, special attention is paid to the time and memory optimization of the different calculation schemes.

The EMAP improvement is planned in the next points:

- Improvement of the advection scheme;
- Development of surface layer parameterization, accounting for surface sources (cases of re-emission, area sources etc.);
- Incorporation of proper chemistry block for simulation of regional scale and urban pollution events.

Activities during the year

The activities of Bulgarian group during 2000 are connected with the two above mentioned topics; namely:

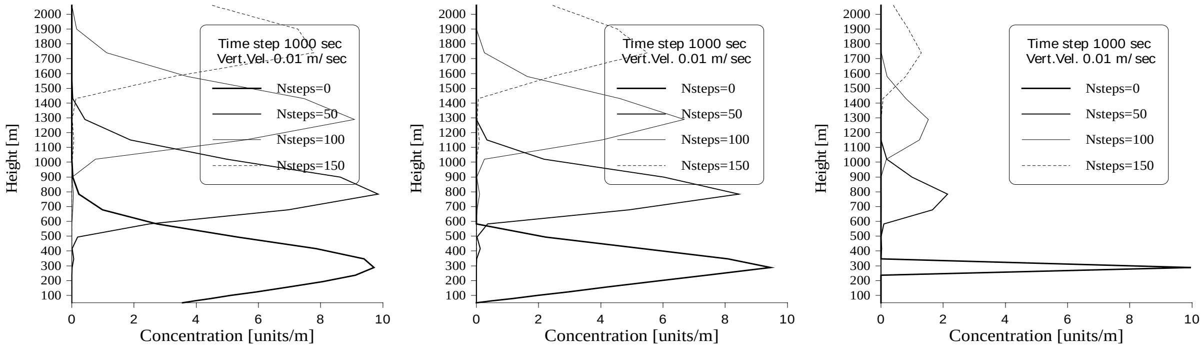
A. Improvement of the TRAP advection scheme, and
B. Application of EMAP-model for estimation of pollution exchange between Bulgaria and Greece.

The results are presented on some international forums as:

- EUROTRAC Symposium 2000, 27-31 March, Garmisch-Partenkirchen, Germany.
- GLOREAM Workshop 2000, 19-24 September 2000, Cottbus, Germany.

Principal results

A. Non-homogeneous grid versions of the TRAP advection scheme are elaborated and tested. Two kinds of non-homogeneous grids are treated and respective modifications of TRAP are made. A common approach allowing the usage of all kinds of Bott-type schemes with minimum modifications is proposed and tested. The non-homogeneous TRAP performance is compared with the homogeneous one. The TRAP performance on log-linear grid for instantaneous sources with Gaussian, triangle and point-like initial profiles is demonstrated below.



B. The air pollution model EMAP is used for estimating the sulfur pollution over south-east Europe for 1995 due to Bulgarian and Greek sources. Two species of sulfur in the air are considered - gaseous sulfur dioxide SO_2 and particulate sulfate $SO_4^{=}$ with the only transformation $SO_2 \rightarrow SO_4^{=}$. The pollutant specific model parameters used are:

	SO_2		$SO_4^{=}$	
$SO_2 \rightarrow SO_4^{=}$ rate constant α_{tr} [h^{-1}]	0.01 (winter) - 0.04 (summer)			
Wet removal constant γ [mm^{-1}],	0.3		0.2	
	Land	Sea	Land	Sea
Dry deposition velocity V_d [m/s]	0.01	0.03	0.002	0.006

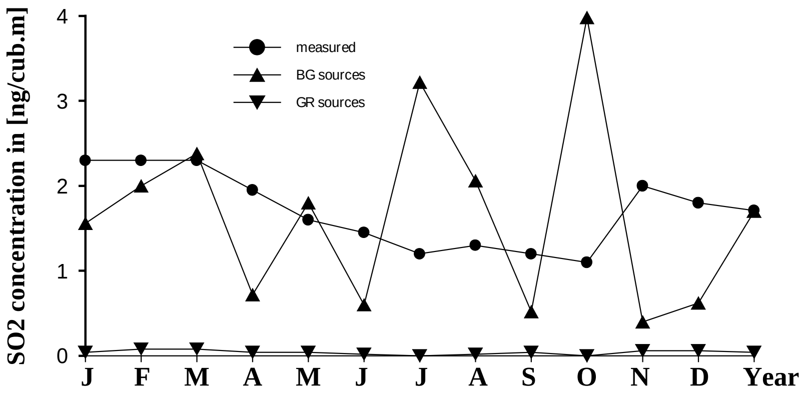
A territory of 8 x 9 EMEP's 150×150 km^2 grid cells with Bulgaria in the center is chosen in this study; every cell divided to 36 25 x 25 km^2 cells. A 5-layer log-linear vertical structure of EMAP is used with representative levels at 50, 200, 650, 1450 and 2500 m.

The sources are determined on the base of the CORINAIR methodology as reported officially to EMEP's Meteorological Synthesizing Centre – West in Oslo. Two kinds of sources exist in this inventory – Large point sources (LPS) and Area sources (AS), the last ones distributed over territories of both countries. The total amount of sulfur emission of Bulgaria for 1995 is estimated to 748.6 kt S, 651.14 kt due to LPS and 97.5 kt - to AS. The total sulfur emission of Greece for 1995 is estimated to 304.672 kt S. The amount released by large point sources is 179 kt and by area sources - 125 kt. A common height of 200 m is prescribed to LPS, 50 m - to AS.

The meteorology input has time resolution of 6 hours. It consists of sequence of analysed U_{850}, V_{850}, T_{850} and T_{surf} fields and 6-hour forecast for precipitation. The standard 50 x 50 km^2 output of the "Europa-Model" of Deutscher Wetterdienst, Germany, distributed via the Global Telecommunication System of the World Meteorological Organization is used here, data interpolated in the points of the used grid. A simple PBL model (Yordanov *et al.*, 1983), built in EMAP, uses these fields as input, producing U-, V-, W- and K_z-vertical profiles at each grid point. It provides also u* and SL universal profiles necessary in the SL parameterization. The roughness and Coriolis parameter fields are pre-set additional input to this PBL model.

Annual runs with Bulgarian and Greek sources are performed. Mean monthly and annual concentration and deposition fields for 1995 are obtained and the country-to-country pollution budget tables are calculated.

Very few measurement data are available as to validate the calculation results. There is not any EMEP station in the region. For some period of time a background station of Bulgarian Ministry of Environment used to operate in the National Astronomic Observatory "Rojen", placed on a peak with 1800 m height in the Rhodopy-mountains, situated both in Bulgaria and in Greece.



The graphical data given in "Status of the environment of Republic of Bulgaria - 1995", Bulletin of the National Center of Environment and Sustainable Development at the Ministry of Environment, page 92, and the calculated SO_2 concentrations are presented in the Figure. It can be noticed that the mean monthly values have the same order of magnitude, the differences less than factor of two. The annual values show remarkable coincidence - 1.71 ng/m^3 from the measurements and 1.74 ng/m^3 from calculations. All these show that the governing parameters are properly settled and the presented results can be considered reliable to some extent.

It is shown that about 4% of the emitted Bulgarian sulfur oxides are deposited over Greek territory. The deposited quantity is estimated to be 28 kt. Only 2% of Greece emitted sulfur compounds are deposited over Bulgaria, the quantity estimated to be 6.2 kt. It can be seen from the 10-year report of EMEP/MSC-W (Barrett and Berge, 1996) that according to their calculations the exchange of sulfur pollution between both countries is estimated right as order of magnitude, giving in the same time much more details in time and space distribution of deposited quantities.

Main conclusions

A. The TRAP scheme describes the advection of pollution forms without steep gradients pretty well. The transition to a non-homogeneous grid makes the description worse in three ways: faster decrease of profile maximum, increase of numerical dispersion and slow decrease of the movement velocity of the shape as a whole. In spite of this decrease of description quality, the TRAP scheme demonstrates a very good performance in both cases of regular and non-regular non-homogeneous grids and can be recommended for use in pollution dispersion models.

B. The results of such calculations can be used in decision-making, negotiating and contamination strategies development.

Aim for the coming year

Participation in the Mercury models inter-calibration organized by EMEP/MSC-E, Moscow.

Acknowledgements

Deep gratitude is due to the EUROTRAC Scientific Secretariat for its efforts to support my participation EUROTRAC Symposium 2000 as well as GLOREAM Workshop 2000.

Part B of this work is done in the frame of NATO Grant ENVIR.LG 973343.

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Intercomparison of Eulerian ozone prediction systems within GLOREAM for summer 1999 using the German monitoring data

A contribution to subproject GLOREAM

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Within GLOREAM numerous activities are concerned with operational forecasting of ground level ozone concentrations in Europe. Comprehensive forecasting systems based on an Eulerian approach are driven for example by NERI, Denmark, DWD and FUB, Germany, NILU, Norway, and SMHI, Sweden. To apply such modeling systems, e.g. for regulatory purposes according to new EU directives, an evaluation and comparison of the model systems is fundamental in order to assess their reliability. One step in this direction is presented in this study: The model forecasts from all five systems have been compared to measurements of ground level ozone in Germany. The outstanding point in this investigation is the availability of a huge amount of data – from forecasts by the different model systems and from observations. This allows for a thorough interpretation of the findings and assures the significance of the observed features. Data from more than 300 measurement stations for a 5-month period (May – September 1999) of the German monitoring networks have been used in this comparison. Different spatial and temporal statistical parameters were applied in the evaluation. Generally, it was found that the most comprehensive models gave the best results. However, the less comprehensive and computational cheaper models also produced good results. The extensive comparison made it possible to point out weak points in the different models and to describe the individual model behavior for a full summer period in a climatological sense. The comparison also gave valuable information for an assessment of individual measurement stations and complete monitoring networks in terms of the representativeness of the observation data.

An Application of the MOGUNTIA Numerical Isotope Concept to Tropical Thunderstorm NO_X Emissions and Ozone Formation

A contribution to subproject GLOREAM

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An inherent uncertainty in atmospheric air pollution control concerns the one to one source - receptor relationship of long lived chemical constituents, especially if nature does not provide molecular isotopes which allow to identify the contribution of particular emission sources. Chemical transport simulation models may help out of this dilemma if their algorithm tolerates the transport of subdivisions -- "numerical isotopes" -- without mass conservation deficiencies.

Ozone is the most prominent candidate to investigate in atmospheric photochemistry. In polluted air a catalytic reaction of $NO_x = \{NO \wedge NO_2\}$ inverts the sign of the net ozone production in the course of methane oxidation upside down from negative in the natural tropospheric air to positive. NO_x is released during fossil fuel combustion, e.g. in car engines or domestic heating, two major anthropogenic sources. Other sources of NO_x are soil exhalation, aircraft, biomass burning, and lightning. The two latter predominantly are situated on the tropical continents and in the air above up to the tropopause.

The NO_x molecules themselves are short lived and hardly get a chance e.g. from the tropics to reach the midlatitudes, especially the European continent in this context. However ozone, with a tropospheric lifetime of about 100 days can spread out by wind and convection all around the world. Thus background ozone contributions from local pollution sources on the large scale are generally camouflaged by considerable amounts which can be blamed to remote NO_x sources and to the natural stratospheric ozone intrusion.

In this study, the NO_x, HNO_3, and O_3 molecules from the sources mentioned above are numerically treated as isotopes of these species, a method which allows their identification within any air parcel at any place of the world with special emphasis on the troposphere over Europe.

The model parameterisation of the tropical lightning NO_x source was chosen to provide the numerical ozone isotope under investigation here. The model results are hypothetical and depend on the validity of the assumptions made in context with the lightning parameterisation presented in the following. Aim of the whole MOGUNTIA-modelling effort is to bring some more transparency into the complex interactive transport/photochemistry system.

The lightning- NO_X emission parameterization in MOGUNTIA

The principle is to derive a field of global lightning activity from statistics of deep convection events and corresponding vertical air mass fluxes using a heuristic relationship which defines flash rates in dependency of the cloud top height for the basic environments over sea and land surface.

Based on the cloud atlases by Hahn *et al.* (1986) and Warren *et al.* (1986) for continents and oceans respectively, a database was derived with locations of convection events and corresponding air mass fluxes (Feichter and Crutzen, 1990) for every 2h time step. Apparently (Figure 1) the total convective vertical mass exchange over sea by far exceeds that over land because of the larger sea surface area.

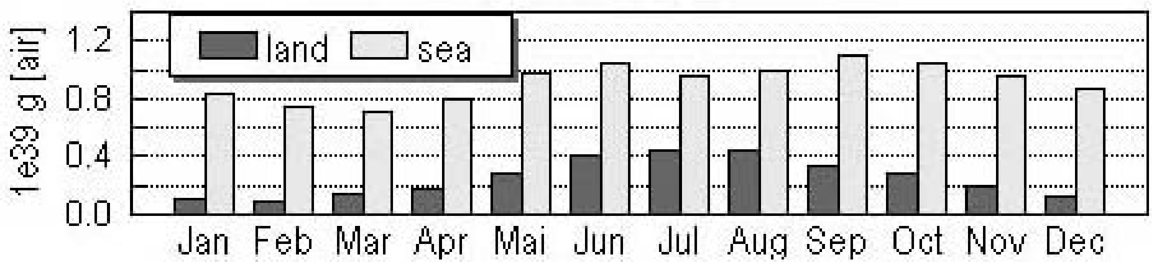

Figure 1. NH convective air mass flux.

Price and Rind (1992) derive a relationship between the cloud top height *H* and the flashrates F_{contin} (steep) and F_{marin} for both marine and continental deep convective clouds:

$$F_{cont} = 3.44\times10^{-5} H^{4.9}$$

and

$$F_{marine} = 6.4\times10^{-5} H^{1.73}$$

It is obvious from Figure 2 that for the same cloud top height the lightning activity over land is considerably higher than over the ocean. This is of essential importance with respect to the NO_X emissivity which is proportional to the flash intensity of a particular thunderstorm. In addition to the difference in flash rates, the cloud tops reach less high over the sea compared to continental convection.

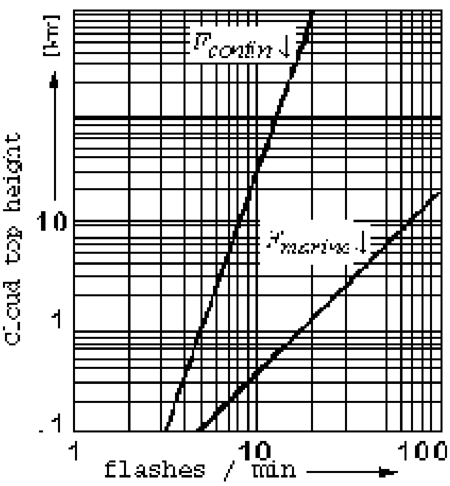

Figure 2. Flash potential of continental and marine clouds.

Monthly global distributions of lightning potential were calculated from the formulae above for every 10°×10° grid column depending on the model tropopause height and the land/sea surface distribution. The database of convection events mentioned in the first paragraph indicates the occurrence of thunderstorms, while the corresponding vertical air mass flux amounts define their intensity. A match of both fields, the land/sea - cloud top height dependent potential flash frequency with the convective air mass fluxes were normalized to result in a four dimensional NO_X emission field, density weighted in the vertical as recommended by Kovalczyk (1982). Figure 3 shows the cumulated lightning NO_X emission seasonality for the NH. Obviously the stronger continental lightning frequency leads to much higher NO_X formation over the continents with a summer maximum on the NH.

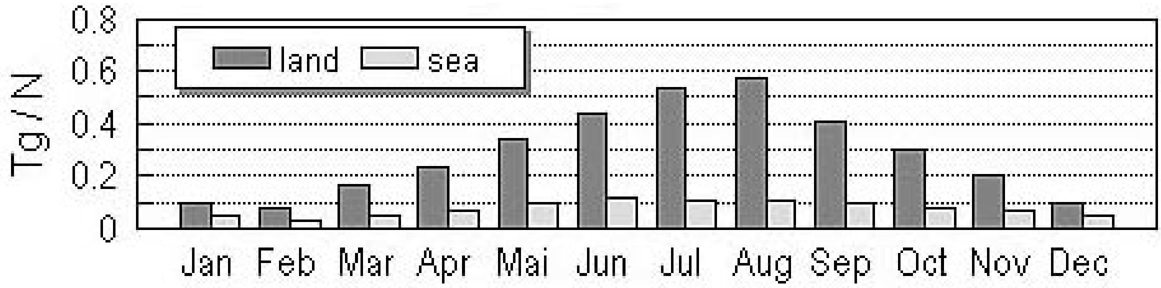

Figure 3. NH lightning NO_x emission.

A global transport / photochemistry simulation -- with an annual total lightning NO_X input of 6 Tg/N, distributed in July according the dark shading in Figure 4 -- predicts high lightning ozone productivity between the Equator and 20°N, the location of intense lightning activity (Figure 5a). The lightning ozone distribution itself (Figure 5b) is much more wide spread and covers the whole globe from 75°S through 75°N. Because of its outstanding source location, "lightning ozone" is mixed more and removed less efficiently compared to e.g. "industrial ozone". The percentage in the surface air layer over Europe is depicted in Figure 6 and reveals a local maximum of ~22% over south France.

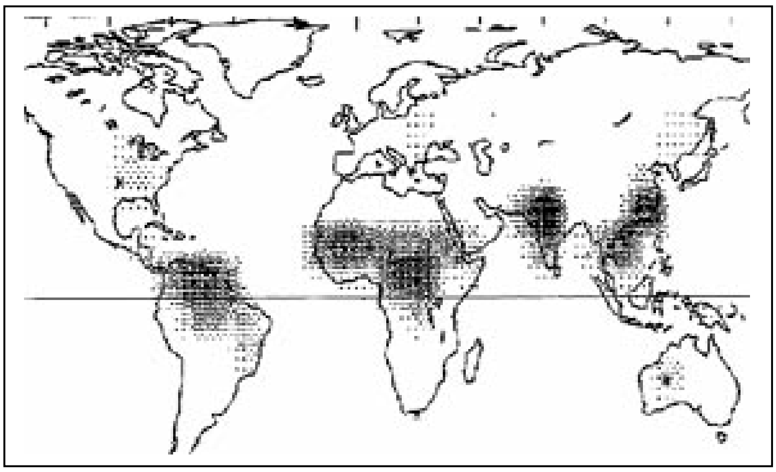

Figure 4. Calculated lightning distribution - July mean.

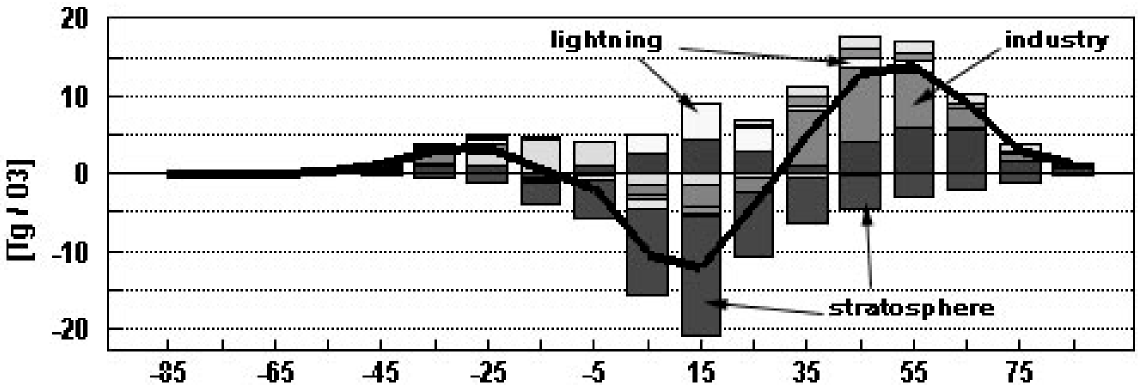

Figure 5a. Source segregated net ozone production rates per 10° latitude belt for July.

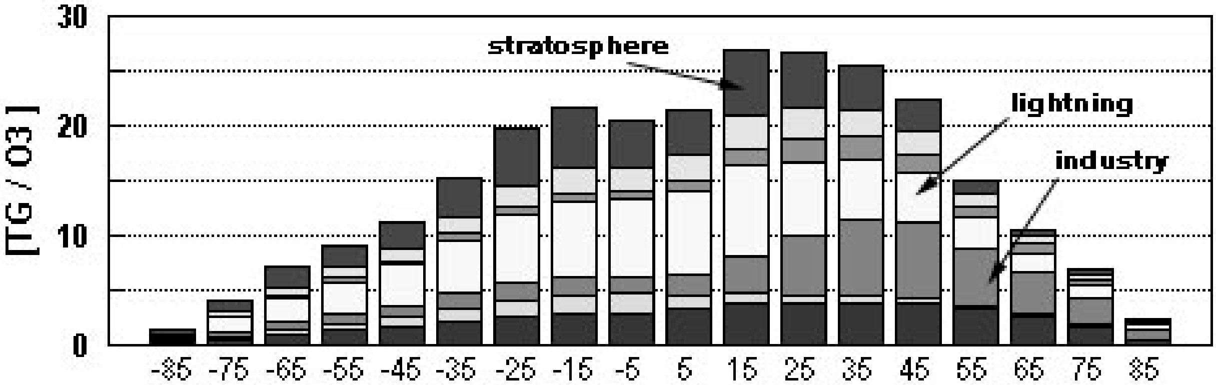

Figure 5b. Corresponding ozone distribution.

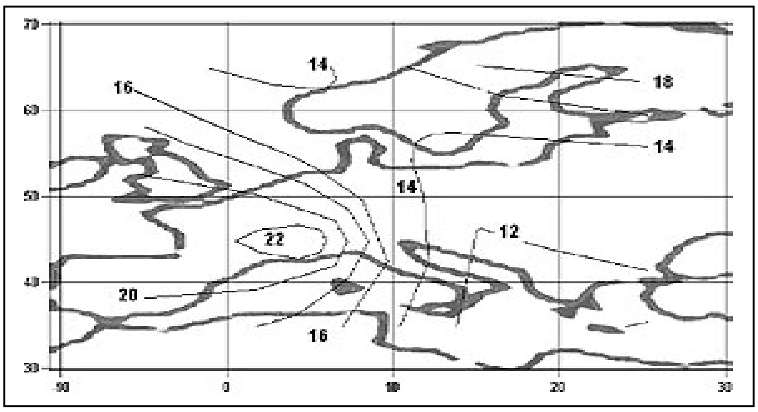

Figure 6. "Lightning ozone" at surface level over Europe.

Summary

As mentioned above the results of this model experiment strongly depend on the assumptions made in the lightning model which was developed in order to provide two hourly changing input fields of lightning- NO_X emission source distribution and strength. The validation process is phased in collaboration with the Global Ozone Monitoring Experiment "GOME" (Borrows, 2000) and will be subject of further publications. For more detail about the design of the MOGUNTIA transport and ozone chemistry please refer to Crutzen and Zimmermann (1991).

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Comprehensive Air Pollution Studies by the Danish Eulerian Model

A contribution to subproject GLOREAM

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Summary

The protection of our environment is one of the most important problems in the modern society. The importance of this problem was steadily increasing during the last two-three decades. Studying health effects as well as other damages, which are caused by high pollution levels, is a task that must be urgently resolved. Reliable and robust control strategies for keeping the pollution caused by harmful chemical compounds under certain safe levels have to be developed and used in a routine way. Large mathematical models, in which **all** important physical and chemical processes are adequately described, can successfully be used to solve this task. The use of the Danish Eulerian Model is discussed in this report.

Aim of the research

The Danish Eulerian Model is described in Alexandrov *et al.* (1997), Bastrup-Birk *et al.* (1997), Zlatev (1995), Zlatev *et al.* (1996a, 1996b). Its space domain covers the whole of Europe together with parts of Asia, Africa and the Atlantic Ocean. The domain is discretized by a (96x96x10) grid. This means that (50 km x 50 km) grid squares are used in the horizontal planes. A variable mesh is applied in the vertical direction. The CBM IV scheme (Gery *et al.*, 1989, Zlatev, 1995) with 35 chemical species is used in the chemical part of the model. The main objective is to use the model in studying relationships between emission sources and high levels of concentrations and/or depositions of certain chemical species in different parts of Europe (and first and foremost in Denmark and in the region surrounding Denmark). The model is used to study both episodic and long-term variations of the concentrations and depositions. It should be mentioned here that the two-dimensional version of the model is still used when long-term variations (over a time-period of up to ten years) are studied. It should be mentioned here that some experiments by using a finer resolution grid, (480x480x10), have also been carried out. The grid squares in the horizontal planes are (10 km x 10 km) when the finer resolution grid is used.

Activities during the year

The main activities in the work on the project *"Comprehensive Air Pollution Studies by the Danish Eulerian Model"* were concentrated in the following seven directions during 2000:

(a) Improving the chemical and physical mechanisms used in the model. Experiments with several chemical schemes have been carried out (this is a continuation of the work reported in Alexandrov *et al.*, 1997). Seasonal variations in the deposition mechanisms have been introduced. Some improvements of the ammonia-ammonium mechanisms were successfully performed.

(b) Improving the presentation of the output results. Better and faster visualization and animation routines were developed and used. The use of efficient graphical tools is very important for the validation procedures and for the dissemination of model results among interested specialists.

(c) Work on better validation of the model results. Different tests have been carried out to verify the reliability of the results computed by the numerical algorithms applied in the model. The chemistry-advection rotation test (Hov *et al.*, 1989), which is an extension of the well-known and commonly used advection rotation test (Molenkampf, 1968 and Crowley, 1968), was used to verify the algorithms used to describe the transport and the chemical reactions. Measurements, taken in EMEP stations located in different European countries, were also extensively used in the experiments.

(d) Incorporation of better and faster numerical algorithms, which will allow us to run the model efficiently on different high-speed computers. It is still rather difficult (and even impossible when the time-interval is too long) to run the three dimensional version of the Danish Eulerian Model in long-term simulations. Efforts to improve the performance of the model, when it is run on the available high-speed computers, have systematically been carried out. The final aim is to prepare a flexible version of the model based on an extensive use of object-oriented software. This aim has not been achieved in 1998, but some very promising results have been obtained. Several easily exchangable modules have been developed and attached to the model. The model was run on several high-speed computers, including parallel computers with distributed memory. A standard tool, MPI (the Message Passing Interface, Gropp *et al.*, 1994), has been used in the latter case. This will allow us to achieve easily good results on different computers with distributed memory.

(e) Running the model over a very long time period. The Danish Eulerian Model has been run over a time period of nine years (from 1989 to 1998). It was possible to perform these simulations only with the two-dimensional version of the model. This emphasizes the great importance of obtaining good results in the efforts described in the previous paragraph.

(f) Studying ozone episodes during the summer months of 1989-1998. Ozone episodes, in which several critical levels were exceeded in different European regions, have been studied. It has been shown that in many West European and Central European countries the levels of 90 ppb and 120 ppb have been exceeded in many summer days. Also the AOT40 (accumulated over a threshold of 40 ppb) values were exceeded in many parts of Europe; this being true both for the AOT40 values for crops and for AOT40 values for forest trees.

(g) Economical evaluations of losses of crops due to high ozone levels. Algorithms for calculating evaluations of losses of crops have been derived and tested. These algorithms have been used to calculate some evaluations of the losses of crops in the Danish counties for the period from 1989 to 1998; the results are discussed in detail in Zlatev *et al.* (2001).

Principal results

The major results that were obtained during 2000, in the seven major directions that are discussed in the previous section, are listed below.

(a). The introduction of more advanced chemical schemes and deposition mechanisms tends to improve the results (this is especially true for the seasonal variation of some species).

(b). The improvement of the visualization and animation techniques allows us to perform in a more efficient way different checks of the reliability of the output results; this is demonstrated (by many plots) in http://www.dmu.dk/AtmosphericEnvironment/DEM.

(c). The combined check of the accuracy of the numerical algorithms and the reliability of the output results (tested by comparisons with measurements) increased the confidence in the model, but it should be emphasized here that much more work is needed in this direction.

(d). The improvement of the numerical algorithms resulted in performance, which is five times better than the performance of the model in the beginning of 1998. This allows us to solve now much bigger tasks and much more tasks. However, much better performance is still needed. Some recent improvements are discussed in Ambelas Skøth *et al.* (2000), Georgiev and Zlatev (2001), Owcharz and Zlatev (2001a, 2001b).

(e). Runs over period of many years allowed us to study some trends concerning pollution levels in different European regions.

(f). The studies of episodes with high ozone concentrations in the summer periods of 1989-1997 indicates that the critical levels of 90 ppb and 120 ppb are exceeded in the most polluted areas in Europe. The critical levels for AOT40 values (both for crops and for forest trees are exceeded in nearly the whole of Europe (excluding the Northern and central parts of Scandinavia and Russia). In some parts of Europe the critical levels are exceeded by a factor greater than seven. Moreover, in many regions this happens every year.

(g) Some attempts to evaluate the losses of crops that are caused by high ozone levels have been carried out. The procedures used are discussed in several publications; see, for example, Dimov *et al.* (1999) and Zlatev *et al.* (2001). Furthermore, many visualizations of the obtained results are given in these references.

(h) Some studies concerning the trends in the variation of the ammonia-ammonium concentrations in Denmark were performed. The main conclusion from these studies was the following: *although the ammonia-ammonium emissions in Denmark were practically unchanged during the period 1989-1998, the averaged annual concentrations were clearly reduced in this period.* The variation of the emissions in Denmark is shown in Figure 1. The variation of the averaged model results over the whole Denmark as well as of the measurements and the model results at the three Danish stations which are in the EMEP network is shown in Figure 2. The results given in Table 1 indicate that the reduction of the ammonia-ammonium concentrations in Denmark in 1989-1998 is primarily due to reductions of the ammonia emissions in Germany and The Netherlands.

Table 1. Ammonia-ammonium concentrations in 1989 and 1998 in three European countries.

Country	**1989**	**1998**	**Reduction**
Western part of Germany	661	502	24%
The Netherlands	232	171	24%
Denmark	102	104	0%

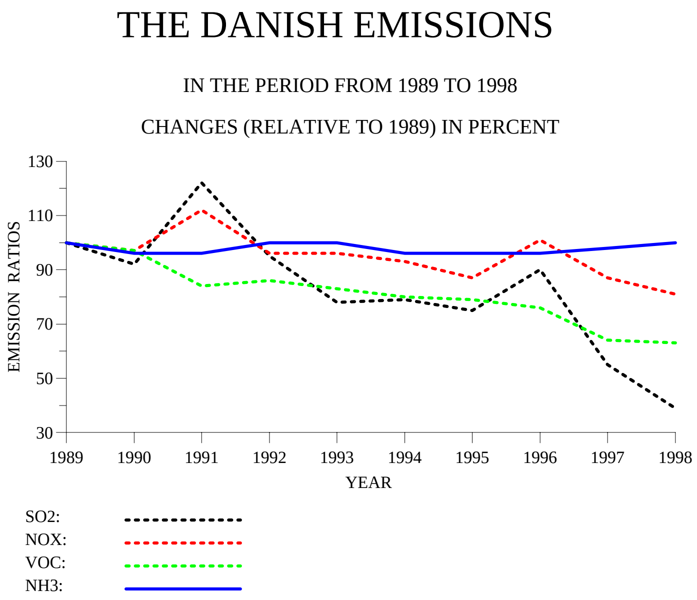

Figure 1.

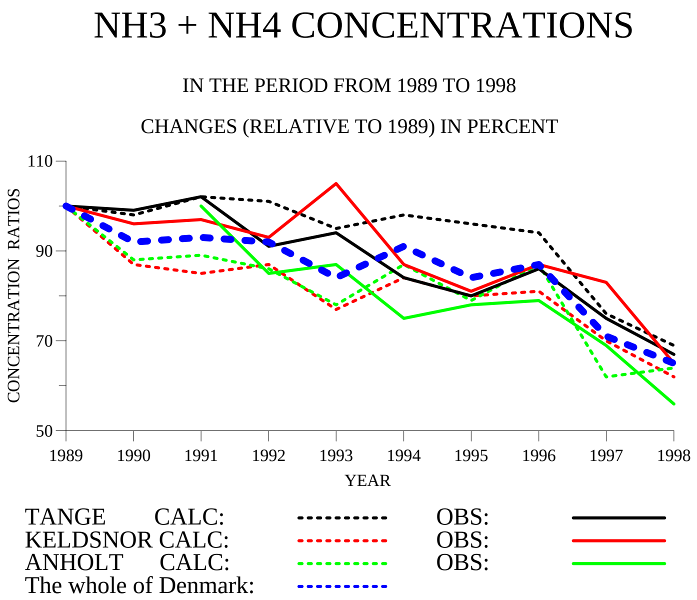

Figure 2.

Results representing different aspect of this project have been reported at ten international meetings (as invited addresses on some of these meetings). The research is documented in several papers in international journals and proceedings (some of these publications are listed below).

Aims for the coming year

There are still many open questions and many modules of the model can be further improved. Increasing the performance of the model, so that the three dimensional version can be applied in more and more cases is one of the most important tasks. This is a difficult task (see Peters *et al.*, 1995 and Zlatev, 1995), but the computers are becoming faster and faster, which indicates that some progress in this direction can be achieved. The attempts to run the model on more refined space grids will be continued. Inter-comparison with other models could gives some ideas for improving some parts of the model.

Acknowledgements

This research work on this EUROTRAC-2 project was partially supported by NATO (ENVIR.CRG.930449, OUTS.CRG.960312 and ENVIR.ARW.971731), EU (ESPRIT, #22727 and #24618) and NMR (the Nordic Council of Ministers) in a project in which teams from Denmark, Finland, Norway and Sweden are collaborating. A grant from the Danish Natural Sciences Research Council gave us access to all Danish supercomputers.

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