INTEGRATED SYSTEMS OF MESO-METEOROLOGICAL AND CHEMICAL TRANSPORT MODELS

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ENHANCING MESOSCALE METEOROLOGICAL MODELLING CAPABILITIES FOR AIR POLLUTION AND DISPERSION APPLICATIONS

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Welcome Speech: *Peter Aakjær*

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Dear Colleagues!

I am pleased to welcome you all here at DMI to this COST728[1]/NetFAM[2] workshop on the integration of weather models and air pollution models, and I am happy to get the opportunity to address you here today.

Weather hazards, the environment and climate change are of concern to all of us. Especially, how human activities impact nature is essential to understand. And its monitoring, forecasting and research are thus of the utmost importance. Furthermore, climate change and pollution of the environment do not respect national borders, so international collaboration on these issues is indeed extremely important.

Here at DMI we have a long experience in environmental modelling and forecasting. Our activities include weather and climate modelling and modelling atmospheric dispersion, transformation and deposition of pollutants. Our weather forecasting model - DMI-HIRLAM – has been developed over many years – and is now run operationally for Denmark and Greenland in a 5 km horizontal resolution. The general quality of HIRLAM has been improved enormously over the years, and the improvements are expected to continue over the next years due to the fact that we will acquire a new High Speed Computer. We are at the moment in the ITT process hoping for at least a factor of 10-12 to the speed of our present system.

DMI has a long tradition of working on many aspects concerning air pollution, both nationally and internationally - not only research, but we are – like many meteorological institutes also the national focal point for several emergency preparedness dealing with nuclear, bio- and veterinary emergency, pollen forecasting, risk assessments, and airborne dispersion of foot-and-mouth disease virus to mention a few. And I cannot resist mentioning that we are proud that our model DERMA was one of the most successful among 28 models in the European Tracer EXperiment – ETEX - some years ago. An automatic computerized system has been developed providing real-time high-resolution forecast data derived from the DMI-HIRLAM system to ARGOS and RODOS for urban- and regional-scale atmospheric dispersion modelling. DMI led a big European project FUMAPEX, which developed a new generation Integrated Urban Air Quality Information and Forecasting System and implemented it in 6 European cities.

In the future, the increasing resolution of meteorological, climate and oceanographic models are paving the way for integrated modelling of the Earth system and the possibility to include meteorology, environment, climate, ocean, cryosphere and ecosystem interactions. DMI strongly supports this development and is currently focusing on such a project. As part of this strategy we are currently developing an on-line integrated chemical weather forecast model, ENVIRO-HIRLAM, which includes interactions between meteorology and air pollution.

I hope - and I am sure - that the discussion during this workshop will lead to synergy between the meteorological and air pollution communities and that this can be used to develop more precise models for the benefit of all of us. I wish you all a fruitful meeting and a pleasant stay in Copenhagen. The weather during your stay seems to bee on your side. Thank you for your attention.

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[1] COST Action 728: Enhancing mesoscale meteorological modelling capabilities for air pollution and dispersion applications, see: http://www.cost728.org
[2] NetFAM: Nordic Network on Fine-scale Atmosperic Modelling (NetFAM), see: http://netfam.fmi.fi

Integrated Systems: On-line and Off-line Coupling of Meteorological and Air Quality Models, Advantages and Disadvantages : *Alexander Baklanov*

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Abstract
The strategy for developing new-generation integrated Meso-Meteorological (MetM) and Atmospheric Chemical Transport Model (ACTM) systems is discussed and an overview of the European COST728 (http://www.cost728.org) integrated systems is given. Advantages and disadvantages of on-line integration in comparison with, the more common, off-line coupling of MetMs and ACTMs are analysed.

1. Introduction

Historically air pollution forecasting and numerical weather prediction (NWP) were developed separately. This was plausible in the previous decades when the resolution of NWP models was too poor for meso-scale air pollution forecasting. Due to modern NWP models approaching meso- and city-scale resolution (due to advances in computing power) and the use of land-use databases and remote sensing data with finer resolution, this situation is changing. As a result the conventional concepts of meso- and urban-scale air pollution forecasting need revision along the lines of integration of meso-scale meteorological models (MetMs) and atmospheric chemical transport models (ACTMs). For example, a new Environment Canada conception suggests to switch from weather forecasting to environment forecasting. Some European projects (e.g. FUMAPEX, see: fumapex.dmi.dk) already work in this direction and have set off on a promising path. In case of FUMAPEX it is the Urban Air Quality Information and Forecasting Systems (UAQIFS) integrating NWP models, urban air pollution (UAP) and population exposure models (*Baklanov et al., 2002*), see Figure 1.
In perspective, integrated NWP-ACTM modelling may be a promising way for future atmospheric simulation systems leading to a new generation of models for improved meteorological, environmental and "chemical weather" forecasting.
Both, off-line and on-line coupling of MetMs and ACTMs are useful in different applications. Thus, a timely and innovative field of activity will be to assess their interfaces, and to establish a basis for their harmonization and benchmarking. It will consider methods for the aggregation of episodic results, model down-scaling as well as nesting. The activity will also address the requirements of meso-scale meteorological models suitable as input to air pollution models.
The current COST728 Action (http://www.cost728.org) addresses key issues concerning the development of meso-scale modelling capabilities for air pollution and dispersion applications and, in particular, it encourages the advancement of science in terms of integration methodologies and strategies in Europe. The final integration strategy will not be focused around any particular model, instead it will be possible to consider an open integrated system with fixed architecture (module interface structure) and with a possibility of incorporating different MetMs/NWP models and ACTMs. Such a strategy may only be realised through jointly agreed specifications of module structure for easy-to-use interfacing and integration.
The overall aim of working group 2 (WG2), 'Integrated systems of MetM and ACTM: strategy, interfaces and module unification', is to identify the requirements for the unification of MetM and ACTM modules and to propose recommendations for a European strategy for integrated

meso-scale modelling capabilities. The first report of WG2 (*Baklanov et al., 2007*) compiles existing state-of-the-art methodologies, approaches, models and practices for building integrated (off-line and on-line) meso-scale systems in different, mainly European, countries. The report also includes an overview and a summary of existing integrated models and their characteristics as they are presently used. The model contributions were compiled using COST member contributions, each focussing on national model systems.

2. Methodology for Model Integration

The modern strategy for integrating MetMs and ACTMs is suggested to consider air quality modelling as a combination of (at least) the following factors: air pollution, regional/urban climate/meteorological conditions and population exposure. This combination is reasonable due to the following facts: meteorology is the main source of uncertainty in air pollution and emergency preparedness models, meteorological and pollution components have complex and combined effects on human health (e.g., hot spots in Paris, July 2003), pollutants, especially aerosols, influence climate forcing and meteorological events (precipitation, thunderstorms, etc.). In this context, several levels of MetM and ACTM coupling/integration can be considered:

<u>off-line</u>:

- separate ACTMs driven by meteorological input data from meteo-preprocessors, measurements or diagnostic models,
- separate ACTMs driven by analysed or forecasted meteodata from NWP archives or datasets,
- separate ACTMs reading output-files from operational NWP models or specific MetMs at limited time intervals (e.g. 1, 3, 6 hours).

<u>on-line</u>:

- on-line access models, when meteodata are available at each time-step (possibly via a model interface as well),
- on-line integration of ACTM into MetM, where feedbacks may be considered. We will use this definition for on-line coupled/integrated modelling.

The main advantages of the On-line coupled modelling approach comprise:

- Only one grid; No interpolation in space,
- No time interpolation,
- Physical parametrizations and numerical schemes (e.g. for advection) are the same; No inconsistencies,
- All 3D meteorological variables are available at the right time (each time step),
- No restriction in variability of meteorological fields,
- Possibility to consider feedback mechanisms, e.g. aerosol forcing,
- No need in meteo- pre/post-processors.

However, not always the on-line approach is the best way of the model integration. For some specific tasks (eg, for emergency preparedness, when NWP data are available) the off-line coupling is more efficient way. The main advantages of Off-line models comprise:

- Possibility of independent parametrizations,
- More suitable for ensembles activities,
- Easier to use for the inverse modelling and adjoint problem,
- Independence of atmospheric pollution model runs on meteorological model computations,
- More flexible grid construction and generation for ACTMs,

• Suitable for emission scenarios analysis and air quality management.

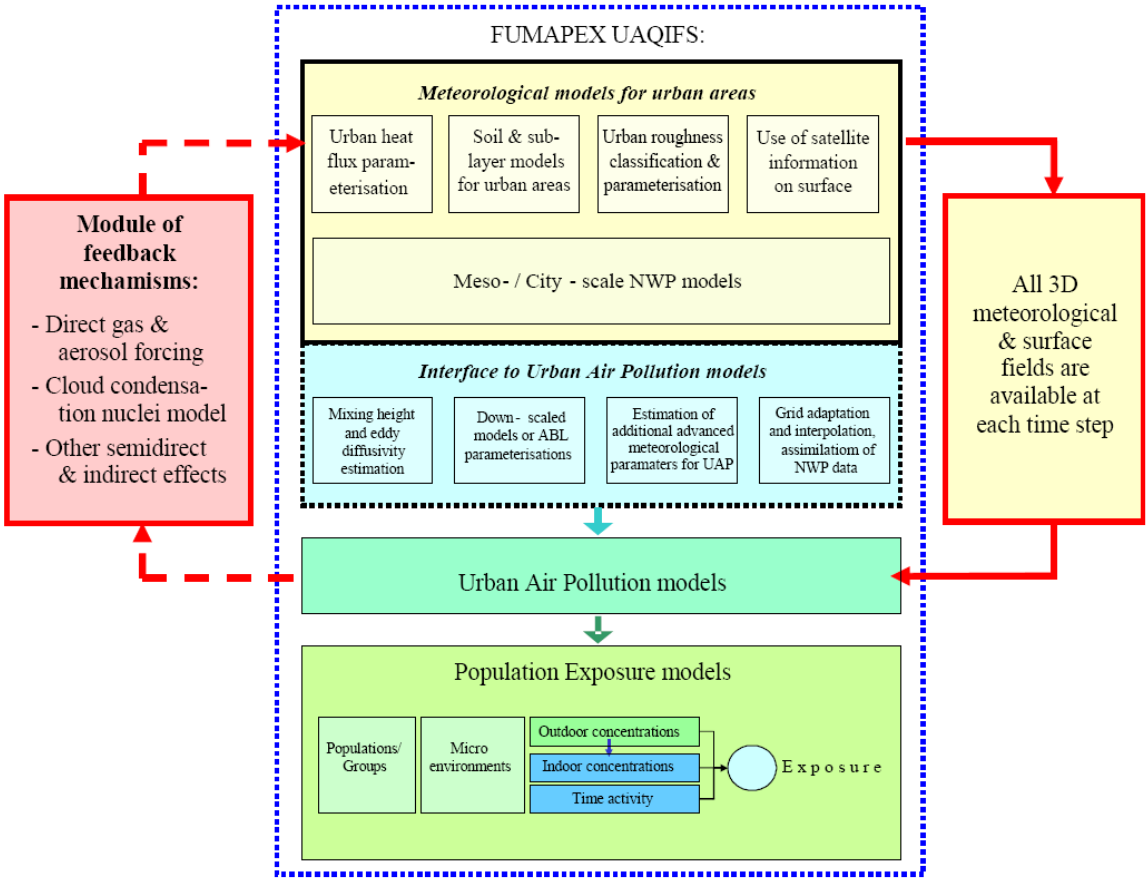

Figure 1: Extended FUMAPEX scheme of Urban Air Quality Information & Forecasting System (UAQIFS) including feedbacks. Improvements of meteorological forecasts (NWP) in urban areas, interfaces and integration with UAP and population exposure models following the off-line or on-line integration (*Baklanov, 2005*; after *EMS-FUMAPEX, 2005*).

The on-line integration of meso-scale meteorological models and atmospheric aerosol and chemical transport models enables the utilisation of all meteorological 3D fields in ACTMs at each time step and the consideration of feedbacks between air pollution (e.g. urban aerosols), meteorological processes and climate forcing. These integration methodologies have been realised by several of the COST action partners such as the Danish Meteorological Institute, with the DMI-ENVIRO-HIRLAM model (*Chenevez et al., 2004; Baklanov et al., 2004, 2008; Korsholm et al., 2007*) and the COSMO consortium with the Lokal Modell (*Vogel et al., 2006, Wolke et al., 2003*).

These model developments will lead to a new generation of integrated models for: climate change modelling, weather forecasting (e.g., in urban areas, severe weather events, etc.), air quality, long-term assessments of chemical composition and chemical weather forecasting (an activity of increasing importance which is due to be supported by a COST action starting in 2007).

3. Overview of European On-Line Integrated Models

The experience from other European as well as non-European union communities, will need to be integrated. On-line coupling was first employed at the Novosibirsk scientific school (*Marchuk, 1982; Penenko and Aloyan, 1985; Baklanov, 1988*), for modelling active artificial/anthropogenic impacts on atmospheric processes. Currently American, Canadian and Japanese institutions develop and use on-line coupled models operationally for air quality forecasting and for research (GATOR-MMTD: *Jacobson, 2005, 2006*; WRF-Chem: *Grell et al., 2005*; GEM-AQ: *Kaminski et al., 2005*; CFORS: *Uno et al., 2003, 2004*).
Such activities in Europe are widely dispersed and a COST Action seems to be the best approach to integrate, streamline and harmonize these national efforts towards a leap forward for new breakthroughs beneficial for a wide community of scientists and users.
Such model integration should be realized following a joint elaborated specification of module structure for potential easy interfacing and integration. It might develop into a system, e.g. similar to the USA ESMF (Earth System Modelling Framework, see e.g.: *Dickenson et al. 2002*) or European PRISM (PRogram for Integrating Earth System Modelling) specification for integrated Earth System Models: http://prism.enes.org/ (*Valcke at el., 2006*).
Community Earth System Models (COSMOS) is a major international project (http://cosmos.enes.org) involving different institutes in Europe, in the US and in Japan, for the development of complex Earth System Models (ESM). Such models are needed to understand large climate variations of the past and to predict future climate changes.
The main differences between the COST-728 integrating strategy for meso-scale models and the COSMOS integration strategy regards the spatial and temporal scales. COSMOS is focusing on climate time-scale processes, general (global and regional) atmospheric circulation models and atmosphere, ocean, cryosphere and biosphere integration, while the meso-scale integration strategy will focus on forecast time-scales of one to four days and omit the cryoshpere and the larger temporal and spatial scales in atmosphere, ocean and biosphere.
The COST728 overview (*Baklanov et al., 2007*) shows a surprisingly large (at least 10) number of on-line coupled MetM and ACTM model systems already being used in Europe (see also more information in Table 1):

- BOLCHEM (CNR ISAC, Italy),
- DMI-ENVIRO-HIRLAM (DMI, Denmark),
- LM-ART (Inst. for Meteorology and Climatology, FZ Karlsruhe, Germany),
- LM-MUSCAT (IfT Leipzig, Germany),
- MCCM (Inst. of Environmental Atmospheric Research at FZ Karlsruhe, Germany),
- MESSy: ECHAM5 (MPI-C Mainz, Germany),
- MC2-AQ (York Univ, Toronto, University of British Columbia, Canada, and Warsaw University of Technology, Poland),
- GEM/LAM-AQ (York Univ, Toronto, University of British Columbia, Canada, and Warsaw University of Technology, Poland),
- WRF-CHem: Weather Research and Forecast and Chemistry Community modelling system (NCAR and many other organisations),
- MESSy: ECHAM5-Lokalmodell LM planned at MPI-C Mainz, Univ. of Bonn, Germany.

However, it is necessary to mention, that many of the above on-line models were not build for the meso-meteorological scale, and several of them (GME, ECMWF GEMS, MESSy) are

global-scale modelling systems, originating from the climate modelling community. Besides, at the current stage most of the on-line coupled models do not consider feedback mechanisms or include only simple direct effects of aerosols on meteorological processes (COSMO LM-ART and MCCM). Only two meso-scale on-line integrated modelling systems (WRF-Chem and DMI-ENVIRO-HIRLAM) consider feedbacks with indirect effects of aerosols.

Table 1: On-line coupled MetM - ACTMs (*Baklanov et al., 2007*)

Model name	On-line coupled chemistry	Time step for coupling	Feedback
BOLCHEM	Ozone as prognostic chemically active tracer		None
ENVIRO-HIRLAM	Gas phase, aerosol and heterogeneous chemistry	Each HIRLAM time step	Yes
WRF-Chem	RADM+Carbon Bond, Madronich+Fast-J photolysis, modal+sectional aerosol	Each model time step	Yes
COSMO LM-ART	Gas phase chem (58 variables), aerosol physics (102 variables), pollen grains	each LM time step	Yes (*
COSMO LM-MUSCAT (**	Several gas phase mechanisms, aerosol physics	Each time step or time step multiple	None
MCCM	RADM and RACM, photolysis (Madronich), modal aerosol	Each model time step	(Yes) (***
MESSy: ECHAM5	Gases and aerosols		Yes
MESSy: ECHAM5-COSMO LM (planned)	Gases and aerosols		Yes
MC2-AQ	Gas phase: 47 species, 98 chemical reactions and 16 photolysis reactions	each model time step	None
GEM/LAM-AQ	Gas phase, aerosol and heterogeneous chemistry	Set up by user – in most cases every time step	None
Operational ECMWF model (IFS)	Prog. stratos passive O3 tracer	Each model time ste	
ECMWF GEMS modelling	GEMS chemistry	Each model time step	Yes
GME	Progn. stratos passive O3 tracer	Each model time step	
OPANA=MEMO+CBMIV		Each model time step	

*) Direct effects only; **) On-line access model; ***) Only via photolysis

4. Feedback Mechanisms, Aerosol Forcing in Meso-Meteorological Models

In a general sense air quality and ACTM modelling is a natural part of the climate change and MetM/NWP modelling. The role of greenhouse gases (such as water vapour, CO_2, O_3 and CH_4) and aerosols in climate change has been highlighted as a key area of future research (*Watson et al 1997, IPCC 2001, AIRES 2001*). Uncertainties in emission projections of gaseous pollutants and aerosols (especially secondary organic components) need to be addressed urgently to

advance our understanding of climate forcing (*Semazzi, 2003*). In relation to aerosols, their diverse sources, complex physicochemical characteristics and large spatial gradients make their role in climate forcing particularly challenging to quantify. In addition to primary emissions, secondary particles, such as, nitrates, sulphates and organic compounds, also result from chemical reactions involving precursor gases such as SO_x, DMS, NO_x, volatile organic compounds and oxidising agents including ozone. One consequence of the diverse nature of aerosols is that they exhibit negative (eg sulphates) as well as positive (eg black carbon) radiative forcing characteristics *(IPCC 2001*, *Jacobson 2001*). Although much effort has been directed towards gaseous species, considerable uncertainties remain in size dependent aerosol compositional data, physical properties as well as processes controlling their transport and transformation, all of which affect the composition of the atmosphere (*Penner et al 1998, Shine 2000, IPCC200*1). Probably one of the most important sources of uncertainty relates to the indirect effect of aerosols as they also contribute to multiphase and microphysical cloud processes, which are of considerable importance to the global radiative balance (*Semazzi 2003*).

In addition to better parameterisations of key processes, improvements are required in regional and global scale atmospheric modelling (*IPCC 1996*, *Semazzi 2003*). Resolution of regional climate information from atmosphere-ocean general circulation models remains a limiting factor. Vertical profiles of temperature, for example, in climate and air quality models need to be better described. Such limitations hinder the prospect of reliably distinguishing between natural variability (eg due to natural forcing agents, solar irradiance and volcanic effects) and human induced changes caused by emissions of greenhouse gases and aerosols over multidecadal timescales (*Semazzi 2003, NAS 2001*). Consequently, the current predictions of the impact of air pollutants on climate, air quality and ecosystems or of extreme events are unreliable (e.g. *Watson et al 1997*). Therefore it is very important in the future research to address all the key areas of uncertainties so as provide an improved modelling capability over regional and global scales and an improved integrated assessment methodology for formulating mitigation and adaptation strategies.
In this concern one of the important tasks is to develop a modelling instrument of coupled 'Atmospheric chemistry/Aerosol' and 'Atmospheric Dynamics/Climate' models for integrated studies, which is able to consider the feedback mechanisms, e.g. aerosol forcing (direct and indirect) on the meteorological processes and climate change (see Figure 2).

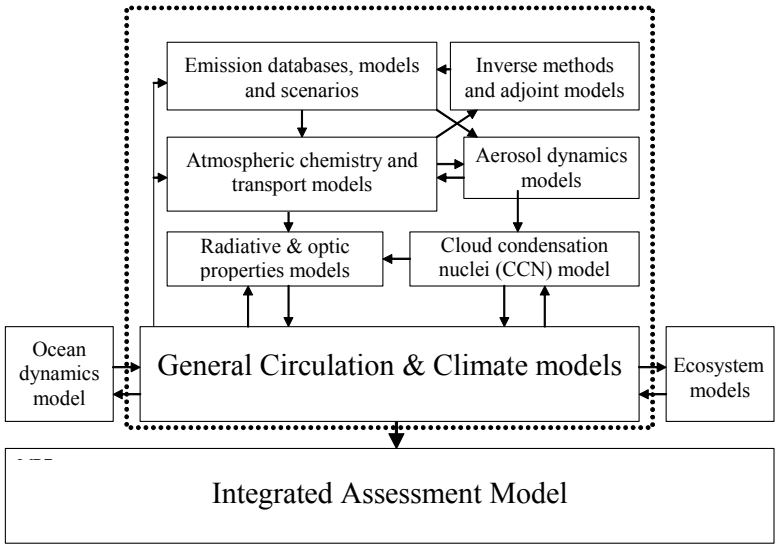

Figure 2 : The integrated system structure for studies of the meso-scale meteorology and air pollution, and their interaction.

Chemical species influencing weather and atmospheric processes include greenhouse gases which warm near-surface air and aerosols such as sea salt, dust, primary and secondary particles of anthropogenic and natural origin. Some aerosol particle components (black carbon, iron, aluminum, polycyclic and nitrated aromatic compounds) warm the air by absorbing solar and thermal-IR radiation, while others (water, sulphate, nitrate, most of organic compounds) cool the air by backscattering incident short-wave radiation to space.

It is necessary to highlight those effects of aerosols and other chemical species on meteorological parameters have many different pathways (direct, indirect, semi-direct effects, etc.) and they have to be prioritized and considered in on-line coupled modelling systems. Following *Jacobson (2002)* the following effects of aerosol particles on meteorology and climate can be distinguished:

- Self-Feedback Effect,
- Photochemistry Effect,
- Smudge-Pot Effect,
- Daytime Stability Effect,
- Particle Effect Through Surface Albedo,
- Particle Effect Through Large-Scale Meteorology,
- Indirect Effect,
- Semi-direct Effect,
- BC-Low-Cloud-Positive Feedback Loop.

Sensitivity studies are needed to understand the relative importance of different feedback mechanisms. Implementation of the feedbacks into integrated models could be realized in different ways with varying complexity. The following variants serve as examples:

One-way integration (off-line):

- The chemical composition fields from ACTMs may be used as a driver for Regional/Global Climate Models, including aerosol forcing on meteorological processes. This strategy could also be realized for NWP or MetMs.

Two-way integration:

- Driver and partly aerosol feedbacks, for ACTMs or for NWP (data exchange with a limited time period); off-line or on-line access coupling, with or without the following iterations with corrected fields)
- Full feedbacks included on each time step (on-line coupling/integration).

For the realization of all aerosol forcing mechanisms in integrated systems it is necessary to improve not only ACTMs, but also NWP/MetMs. The boundary layer structure and processes, including radiation transfer, cloud development and precipitation must be improved. Convection and condensation schemes need to be adjusted to take the aerosol-microphysical interactions into account, and the radiation scheme needs to be modified to include the aerosol effects.

5. Concluding Remarks

The on-line integration of meso-scale meteorological models and atmospheric aerosol and chemical transport models enables the utilization of all meteorological 3D fields in ACTMs at each time step and the consideration of the feedbacks of air pollution (e.g. urban aerosols) on meteorological processes and climate forcing.

These on-line coupled model developments will lead to a new generation of integrated models for climate change modelling, weather forecasting (e.g., in urban areas, severe weather events, etc.), air quality, long-term assessment chemical composition and chemical weather forecasting.

Main advantages of the on-line modelling approach include:

- Only one grid; No interpolation in space;
- No time interpolation;
- Physical parametrizations are the same; No inconsistencies;
- All 3D meteorological variables are available at the right time (each time step);
- No restriction in variability of meteorological fields;
- Possibility to consider feedback mechanisms;
- Does not need meteo- pre/post-processors.

While the main advantages of the off-line approach include:

- Possibility of independent parametrizations;
- More suitable for ensemble activities;
- Easier to use for the inverse modelling and adjoint problem;
- Independence of atmospheric pollution model runs on meteorological model computations;
- More flexible grid construction and generation for ACTMs,
- Suitable for emission scenarios analysis and air quality management.

The COST728 overview shows a quite surprising number of on-line coupled MetM and ACTM model systems already being used in Europe. However, many of the on-line coupled models were not built for the meso-meteorological scale, and they (e.g. GME, ECMWF GEMS, MESSy) are global-scale modelling systems and first of all designed for climate change modelling. Besides, at the current stage most of the online coupled models do not consider feedback mechanisms or include only direct effects of aerosols on meteorological processes (like COSMO LM-ART and MCCM). Only two meso-scale on-line integrated modelling systems (WRF-Chem and ENVIRO-HIRLAM) consider feedbacks with indirect effects of aerosols.

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Chapter 1. On-Line Modelling and Feedbacks

1.1 On-line Coupled Meteorology and Chemistry Models in the U.S.: *Yang Zhang*

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Abstract
The climate-chemistry-aerosol-cloud-radiation feedbacks are important processes occurring in the atmosphere. Accurately simulating those feedbacks requires fully-coupled meteorology, climate, and chemistry models and presents significant challenges in terms of both scientific understanding and computational demand. This review focuses on history and current status of development and application of on-line models in the U.S. Several representative on-line coupled meteorology and chemistry models such as GATOR-GCMOM, WRF/Chem, CAM3, MIRAGE, and Caltech unified GCM are included. Major model features, physical/chemical treatments, as well as typical applications are evaluated with a focus on aerosol microphysics treatments, aerosol feedbacks to planetary boundary layer meteorology, and aerosol-cloud interactions. Recommendations for future development and improvement of online coupled models are provided.

1. Introduction

The climate-chemistry-aerosol-cloud-radiation feedbacks are important in the context of many areas including climate modelling, air quality (AQ) /atmospheric chemistry modelling, numerical weather prediction (NWP) and AQ forecasting, as well as integrated atmospheric-ocean-land surface modelling at all scales. Some potential impacts of aerosol feedbacks include a reduction of downward solar/thermal-IR radiation (direct effect); a decrease in surface temperature and wind speed but an increase in relative humidity and atmospheric stability (semi-direct effect), a decrease in cloud drop size but an increase in drop number via serving as cloud condensation nuclei (first indirect effect), as well as an increase in liquid water content, cloud cover, and lifetime of low level clouds but a suppression of precipitation (the second indirect effect). Aerosol feedbacks are traditionally neglected in meteorology and AQ modelling due largely to historical separation of meteorology, climate, and AQ communities as well as our limited understanding of underlying mechanisms. Those feedbacks, however, are important as models accounting (e.g., *Jacobson, 2002; Chung and Seinfeld, 2005*) or not accounting (e.g., *Penner et al., 2003*) for those feedbacks may give different results and future climate changes may be affected by improved air quality. Accurately simulating those feedbacks requires fully-coupled models for meteorological, chemical, physical processes and presents significant challenges in terms of both scientific understanding and computational demand. In this work, the history and current status of development and application of online models are reviewed. Several representative models developed in the U.S. are used to illustrate the current status of online coupled models. Major challenges and recommendations for future development and improvement of online coupled models are provided.

2. History of Coupled Chemistry/Air Quality and Climate/Meteorology Models

2.1 Concepts and History of Online Models

Atmospheric chemistry/air quality and climate/meteorology modelling was traditionally separated prior to mid. 1970's. The three-dimensional (3-D) atmospheric chemical transport models (ACTMs) until that time were primarily driven by either measured/analyzed meteorological fields or outputs at a time resolution of 1-6 hrs from a mesoscale meteorological model on urban/regional scale or outputs at a much coarser time resolution (e.g., 6-hr or longer) from a general circulation model (GCM) (referred to as offline coupling). In addition to a large amount of data exchange, this offline separation does not permit simulation of feedbacks between AQ and climate/meteorology and may result in an incompatible and inconsistent coupling between both meteorological and AQ models and a loss of important process information (e.g., cloud formation and precipitation) that occur at a time scale smaller than that of the outputs from the offline climate/meteorology models. Such feedbacks, on the other hand, are allowed in the fully-coupled online models, without space and time interpolation of meteorological fields but commonly with higher computational costs.

The earliest attempt in coupling global climate/meteorology and chemistry can be traced back to mid-late 1970's, when simple stratospheric chemistry (e.g., the Chapman reactions, the NO_x catalytic cycle, and reactions between hydrogen and atomic oxygen) was first incorporated into a GCM to simulate global ozone (O_3) production and transport (e.g., *Cunnold et al., 1975; Schlesimger and Mintz, 1979*). In such models, atmospheric transport and simple stratospheric O_3 chemistry are simulated in one model, accounting for the effect of predicted O_3 on radiation heating and the effect of radiation heating on atmospheric circulation, which in turn affects distribution of O_3. Since mid. 1980's, a large number of online global climate-chemistry models have been developed to address the Antarctic/stratospheric O_3 depletion (e.g., *Coriolle et al., 1986, 1990; Rose and Brasseur, 1989; Austin et al., 1992; Rasch et al., 1995; Jacobson, 1995*), tropospheric O_3 and sulfur cycle (e.g., *Roelofs and Lelieveld, 1995; Feichter et al., 1996; Barth et al., 2000*), tropospheric aerosol and its interactions with cloud (e.g., *Chuang et al., 1997; Lohmann et al., 2000; Jacobson, 2000, 2001a; Easter et al., 2004*). The coupling in most online models, however, has been enabled only for very limited prognostic gaseous species such as O_3 and/or bulk aerosol (e.g., *Schlesimger and Mintz, 1979*) or selected processes such as transport and gas-phase chemistry (i.e., incompletely- or partially-coupling). This is mainly because such a coupling largely restricts to gas-phase/heterogeneous chemistry and simple aerosol/cloud chemistry and microphysics and often neglects the feedbacks between prognostic chemical species (e.g., O_3 and aerosols) and radiation (e.g., *Roelofs and Lelieveld, 1995; Eckman et al., 1996; Barth et al., 2000*) and aerosol indirect effects (e.g., *Liao et al., 2003*), with a few exceptions after mid. 1990's when truly-coupled systems were developed to enable a full range of feedbacks between meteorology/climate variables and a myriad of gases and size-resolved aerosols (e.g., *Jacobson, 1995, 2000; Ghan et al., 2001abc*).

The earliest attempt in coupling meteorology and air pollution in mesoscale models can be traced back to early 1980s (*Baklanov et al., 2007* and references therein). Since then, a number of mesoscale online coupled meteorology-chemistry models have been developed in North America (e.g., *Jacobson, 1994, 1997a, b; Mathur et al., 1998; Côté et al., 1998; Grell et al., 2000*) and Australia (e.g., *Manins, 2007*) but mostly developed recently by European researchers largely through the COST Action 728 (http://www.cost728.org) (e.g., *Baklanov et al., 2004, 2007*, and references therein). The coupling was enabled between meteorology and tropospheric gas-phase

chemistry only in some regional models (e.g., *Grell et al., 2000*); and among more processes/components including meteorology, chemistry, aerosols, clouds, and radiation (e.g., *Jacobson, 1994, 1997a, b; Jacobson et al., 1996; Mathur et al., 1998; Grell et al., 2005; Fast et al., 2006; Zhang et al., 2005a, b; Hu and Zhang, 2006; and Korsholm et al., 2007*). Similar to global models, a full range of climate-chemistry-aerosol-cloud-radiation feedbacks is treated in very few mesoscale models (e.g., *Jacobson, 1994, 1997a, b; Grell et al., 2005*).

Two coupling frameworks are conventionally used in all mesoscale and global online coupled models: one couples a meteorology model with an AQ model in which the two systems operate separately but exchange information every time step through an interface (referred to as separate online coupling), the other integrates an AQ model into a meteorology model as a unified model system in which meteorology and AQ variables are simulated together in one time step without a model-to-model interface (referred to as unified online coupling). Transport of meteorological and chemical variables is typically simulated with separate schemes in separate online models but the same scheme in unified online models. Depending on the objectives of the applications, the degrees of coupling and complexities in coupled atmospheric processes in those models vary, ranging from a simple coupling of meteorology and gas-phase chemistry (e.g., *Rasch et al., 1995; Grell et al., 2000*) to the most sophistic coupling of meteorology, chemistry, aerosol, radiation, and cloud (e.g., *Jacobson, 1994, 2004b, 2006; Grell et al., 2005*). While online coupled models can in theory enable a full range of feedbacks among major components and processes, the coupling is typically enabled in two modes: partially-coupled where only selected species (e.g., O_3) and/or processes (e.g., transport and gas-phase chemistry) are coupled and other processes (e.g., solar absorption of O_3 and total radiation budget) remain decoupled; fully-coupled where all major processes are coupled and a full range of atmospheric feedbacks can be realistically simulated. At present, very few fully-coupled online models exist; and most online models are partially-coupled and still under development.

2.2 History of Representative Online Models in the U.S.

In this review, five models on both regional and global scales developed in the U.S. are selected to represent the current status of online coupled models. These include:

- one global-through-urban model, i.e., the Stanford University's Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model **(**GATOR-GCMOM) (*Jacobson, 2001c, 2002, 2004a; Jacobson et al., 2004*),
- one mesoscale model, i.e., the National Oceanic and Atmospheric Administration (NOAA)'s Weather Research Forecast model with Chemistry (WRF/Chem) (*Grell et al., 2005; Fast et al., 2006*), and
- three global models, i.e., the National Center for Atmospheric Chemistry (NCAR)'s Community Atmospheric Model v. 3 (CAM3), the Pacific Northwest National laboratory (PNNL)'s Model for Integrated Research on Atmospheric Global Exchanges **(**MIRAGE) (*Ghan et al., 2001abc; Easter et al., 2004),* and the Caltech unified GCM *(Liao et al., 2003; Liao and Seinfeld, 2005*).

All these models predict gases, aerosols, and clouds with varying degrees of complexities in chemical mechanisms and aerosol/cloud microphysics. The history and current status of these models along with other relevant models are reviewed below.

Jacobson (1994, 1997a, b) and *Jacobson et al. (1996)* developed the first unified fully-coupled online model that accounts for major feedbacks among meteorology, chemistry, aerosol, cloud, radiation on urban/regional scales: a gas, aerosol, transport, and radiation AQ model/a mesoscale meteorological and tracer dispersion model (GATOR/MMTD, also called GATORM). *Grell et*

al. (2000) developed a unified online coupled meteorology and gas-phase chemistry model: Multiscale Climate Chemistry Model (MCCM, also called MM5/Chem). Built upon MM5/Chem and NCAR's WRF, *Grell at al. (2002)* developed a unified fully-coupled online model, WRF/Chem, to simulate major atmospheric feedbacks among meteorology, chemistry, aerosol, and radiation. This is the first community online model in the U.S. Since its first public release in 2002, WRF/Chem has attracted a number of external developers and users from universities, research organizations, and private sectors to continuously and collaboratively develop, improve, apply, and evaluate the model. In WRF/Chem, transport of meteorological and chemical variables is treated using the same vertical and horizontal coordinates and the same physics parametrization with no interpolation in space and time. In addition to Regional Acid Deposition Model v.2 (RADM2) in MM5/Chem, WRF/Chem includes an additional gas-phase mechanism: the Regional Atmospheric Chemistry Mechanism (RACM) of *Stockwell et al. (1997)* and a new aerosol module: the Modal Aerosol Dynamics Model for Europe (MADE) (*Ackermann et al., 1998*) with the secondary organic aerosol model (SORGAM) of Schell et al. (2001) (referred to as MADE/SORGAM). Two additional gas-phase mechanisms and two new aerosol modules have been recently incorporated into WRF/Chem by external developers (*Fast et al., 2006; Zhang et al., 2005a, b, 2007; Hu and Zhang, 2006; Huang et al., 2006*). The two new gas-phase mechanisms are the Carbon-Bond Mechanism version Z (CBMZ) (*Zaveri and Peters, 1999)* and the 2005 version of Carbon Bond mechanism (CB05) of *Yarwood et al. (2005)*. The two new aerosol modules are the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (*Zaveri et al., 2005*) and the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (*Zhang et al., 2004*).

On a global scale, a number of climate or AQ models have been developed in the past three decades among which very few of them are online models. Since its initial development as a general circulation model without chemistry, CCM0 (*Washington, 1982*), the NCAR's Community Climate Model (CCM) has evolved to be one of the first unified online climate-chemistry models, initially with gas-phase chemistry only (e.g., CCM2 (*Rasch et al., 1995*) and CCM3 (*Kiehl et al., 1998; Rasch et al., 2000*)) and most recently with additional aerosol treatments (e.g., CAM3 (Collins et al., 2004, 2006ab; and CAM4 (http://www.ccsm.ucar.edu)). *Jacobson (1995, 2000, 2001a)* developed a unified fully-coupled Gas, Aerosol, TranspOrt, Radiation, and General circulation model (GATORG) built upon GATORM and a 1994 version of the University of Los Angeles GCM (UCLA-GCM). *Jacobson (2001bc)* linked the regional GATORM and global GATORG and developed the first unified, nested global-through-urban scale Gas, Aerosol, Transport, Radiation, General Circulation, and Mesoscale Meteorological model, GATOR-GCMM. GATOR-GCMM was designed to treat gases, size- and composition-resolved aerosols, radiation, and meteorology for applications from the global to urban (< 5 km) scales and accounts for radiative feedbacks from gases, size-resolved aerosols, liquid water and ice particles to meteorology on all scales. GATOR-GCMM was extended to Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR-GCMOM) in *Jacobson (2004, 2006)* and *Jacobson et al. (2004, 2006)*. Built up NCAR CCM2 and PNNL Global Chemistry Model (GChM), MIRAGE was developed and can be run offline or fully-coupled online (*Ghan et al., 2001abc and Easter et al., 2004*). Several online coupled global climate/aerosol models with full oxidant chemistry have also been developed since early 2000 but most of them do not include all feedbacks, in particular, aerosol indirect effects; and they are still being developed (e.g., *Liao et al., 2003*). Among all 3-D models that have been developed for climate and AQ studies at all scales, GATOR-GCMOM, MIRAGE, and WRF/Chem represent the state of science global and regional coupled models; and GATOR-GCMOM

appears to be the only model that represents gas, size- and composition-resolved aerosol, cloud, and meteorological processes from the global down to urban scales via nesting, allowing feedback from gases, aerosols, and clouds to meteorology and radiation on all scales in one model simulation.

3. Current Treatments in Online Coupled Models in the U.S.

In this section, model features and treatments for the five representative online coupled meteorology and chemistry models developed in the U.S. are reviewed in terms of model systems and typical applications, aerosol and cloud properties, aerosol and cloud microphysics and aerosol-cloud interactions. As shown in Table 1, four out of the five models are unified online models (i.e., GATOR-GCMOM; WRF/Chem, CAM3, and Caltech unified GCM) and one (i.e., MIRAGE) is a separate online model, all with different levels of details in gas-phase chemistry and aerosol and cloud treatments ranging from the simplest one in CAM3 to the most complex one in GATOR/GCMOM. Those models have been developed for different applications. As shown in Table 2, the treatments of aerosol properties in those models are different in terms of composition, size distribution, aerosol mass/number concentrations, mixing state, hygroscopicity, and radiative properties. For example, MIRAGE2 treats the least number of species, and GATOR/GCMOM treats the most. Size distribution of all aerosol components are prescribed in Caltech unified GCM and that of all aerosols except sea-salt and dust is prescribed in CAM3; they are predicted in the other three models. Prescribed aerosol size distribution may introduce significant biases in simulated aerosol direct and indirect radiative forcing that highly depends on aerosol size distributions. The mixing state of aerosols affects significantly the predictions of direct/indirect radiative forcing. The internally-mixed (i.e., well-mixed) hydrophilic treatment for BC is unphysical and reality lies between the externally-mixed, hydrophobic and core treatments. Among the five models, GATOR/GCMOM is the only model treating internal/external aerosol mixtures with a coated BC core. All the five models predict aerosol mass concentration, but only some of them can predict aerosol number concentration (e.g., GATOR/GCMOM, WRF/Chem, and MIRAGE2). For aerosol radiative properties, GATOR/GCMOM assumes a BC core surrounded by a shell where the refractive indices (RIs) of the dissolved aerosol components are determined from partial molar refraction theory and those of the remaining aerosol components are calculated to be volume-averaged based on core-shell MIE theory. MIRAGE2, WRF/Chem, and Caltech unified GCM predict RIs and optical properties using Mie parametrizations that are function of wet surface mode radius and wet RI of each mode. Volume mixing is assumed for all components, including insoluble components. The main difference between Caltech unified GCM and MIRAGE2 (and WRF/Chem) is that Caltech unified GCM prescribes size distribution, but MIRAGE2 predicts it. In CAM3, RIs and optical properties are prescribed for each aerosol type, size, and wavelength of the external mixtures.
Table 3 summarizes model treatments of cloud properties, reflecting the levels of details in cloud microphysics treatments from the simplest in Caltech unified GCM to the most sophistic in GATOR-GCMOM. GATOR/GCMOM uses prognostic, multiple size distributions (typically three, for liquid, ice, and graupel), each with 30 size sections. MIRAGE2 and WRF/Chem simulate bulk single condensate in single size distribution, with either a prescribed modal distribution (MIRAGE2) or a predicted sectional distribution (WRF/Chem-MOSAIC). CAM3 treats bulk liquid and ice with the same prognostic droplet size treatment as MIRAGE2. Caltech unified GCM treats bulk liquid and ice with their distributions diagnosed from predicted cloud water content. Among the five models, Caltech unified GCM is the only model that prescribes

cloud droplet number, which is predicted in the other four models. CAM3, MIRAGE2, and WRF/Chem use the same treatment for droplet number, with droplet nucleation parameterized by *Abdul-Razzak and Ghan (2000).* GATOR treats prognostic, size- and composition-dependent cloud droplet number from multiple aerosol size distributions. While an empirical relationship between sulfate aerosols and CCN is commonly used in most atmospheric models, CCN is calculated from Köhler theory using the aerosol size distribution and hygroscopicity in all models but Caltech unified GCM. Other than Caltech unified GCM that does not treat CCN and Ice Deposition Nuclei (IDN), all other four models treat the competition among different aerosol species for CCN but the hydrophobic species are not activated in CAM3 since it assumes an external-mixture. Among the five models, GATOR/GCMOM is the only model that simulates composition of IDN. MIARGE and CAM use a prognostic parametrization in terms of cloud water, ice mass, and number to predict cloud radiative properties. WRF/Chem also uses the same method but with sectional approach. Caltech unified GCM simulates cloud optical properties based on MIE theory and prescribed Gamma distribution for liquid clouds. GATOR/GCMOM simulates volume-average cloud RIs and optical properties based on MIE theory and an iterative dynamic effective medium approximation (DEMA) to account for multiple BC inclusions within clouds. The DEMA is superior to classic effective-medium approximation that is used by several mixing rules such as the volume-average RI mixing rule *(Jacobson, 2006*).

Table 4 shows model treatments of aerosol chemistry and microphysics that differ in many aspects. For example, Caltech unified GCM treats aerosol thermodynamics only, the rest of models treat both aerosol thermodynamics and dynamics such as coagulation and new particle formation via homogeneous nucleation. The degree of complexity varies in terms of number of species and reactions treated and assumptions made in the inorganic aerosol thermodynamic modules used in those models. The simplest module, MARS-A, is used in WRF/Chem-MADE/SORGAM, and the most comprehensive module, EQUISOLV II, is used in GATOR/GCMOM. For secondary organic aerosol (SOA) formation, both CAM3 and MIRAGE2 use prescribed aerosol yields for a few condensable semi-volatile organic compounds (SVOCs), which is the simplest, computationally most efficient approach but it does not provide a mechanistic understanding of SOA formation. GATOR/GCMOM simulates SOA formation from 10-40 classes SVOCs via condensation and dissolution based on Henry's law. Caltech unified GCM simulates SOA formation based on a reversible absorption of 5 classes of biogenic SVOCs and neglects that from anthropogenic SVOCs. In MADE/SORGAM and MOSAIC in WRF/Chem, SOA formation via reversible absorption of 8 classes SVOCs is simulated based on Caltech smog-chamber data. Two approaches are used to simulate SOA formation in WRF/Chem-MADRID (*Zhang et al., 2004*). MADRID 1 uses an absorptive approach for 14 parent VOCs and 38 SOA species. MADRID 2 combines absorption and dissolution approaches to simulate an external mixture of 42 hydrophilic and hydrophobic VOCs. Coagulation is currently not treated in CAM3 but simulated with a modal approach in MIRAGE2, sectional approach in GATOR/GCMOM, and both in WRF/Chem-MADE/SORGAM and MOSIAC. Different from other model treatments, GATOR accounts for van der Waals, viscous forces, and fractal geometry in simulating coagulation among particles from multiple size distributions (*Jacobson and Seinfeld, 2005*). For gas/particle mass transfer, CAM3, MIRAGE2, and Caltech unified GCM use the simplest full equilibrium approach. GATOR/GCMOM uses a computationally-efficient dynamic approach with a long time step (150-300 s) (PNG-EQUISOLV II) for all treated species (*Jacobson, 2005*). In WRF/Chem, full equilibrium approach is used in MADE/SORGAM, dynamic approach is used in MOSAIC. MADRID offers three approaches: full equilibrium, dynamic, and hybrid; their performance has been evaluated in

Zhang et al. (1999) and Hu et al. (2007). Hu et al. (2007) have shown that the bulk equilibrium approach is computationally-efficient but less accurate, whereas the kinetic approach predicts the most accurate solutions but typically with higher CPUs.

Table 5 summarizes the treatments of aerosol-cloud interactions and cloud processes. Aerosol activation by cloud droplets to form CCN is an important process affecting simulations of aerosol-cloud interactions, and aerosol direct and indirect forcing. CAM uses empirical, prescribed activated mass fraction for bulk CCN. MIRAGE and WRF/Chem use a mechanistic, parameterized activation module that is based on Köhler theory to simulate bulk CCN. Important parameters for activation such as the peak supersaturation, S_{max}, mass of activated aerosols, and the size of the smallest aerosol activated are calculated using a parametrization of *Abdul-Razzak et al. (1998)* and *Abdul-Razzak and Ghan (2000)* that relate the aerosol number activated directly to fundamental aerosol properties. GATOR-GCMOM also simulates a mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory. One difference between the treatments in GATOR/GCMOM and MIRAGE is that the MIRAGE activation parametrization neglects size-dependence of the water vapor diffusivity coefficient and mass transfer coefficient, which may lead to an underestimation of cloud droplet number concentration. In addition, the equilibrium Köhler theory may be inappropriate for larger particles due to the kinetic effect (i.e., mass transfer limitation). Such size-dependence and kinetic effect are accounted for in GATOR/GCMOM.

4. Major Challenges and Future Directions

Significant progress has been made in the past two decades in the development of online coupled climate- (or meteorology-) chemistry and their applications for modelling global/regional climate, meteorology, and air quality, as well as the entire earth system. Several major challenges exist. First, accurately representing climate-aerosol-chemistry-cloud-radiation feedbacks in 3-D climate- or meteorology-chemistry models at all scales will remain a major scientific challenge in developing a future generation of coupled models. There is a critical need for advancing the scientific understanding of key processes. Second, representing scientific complexity within the computational constraint will continue to be a technical challenge. Key issues include (1) the development of benchmark model and simulation and use of available measurements to characterize model biases, uncertainties, and sensitivity and to develop bias-correction techniques (e.g., chemical data assimilation); (2) the optimization/parametrization of model algorithms with acceptable accuracy. Third, integrated model evaluation and improvement and laboratory/field studies for an improved understanding of major properties/processes will also post significant challenges, as it involves researchers from multiple disciplinaries and requires a multidisciplinary and/or interdisciplinary approach. Key issues include (1) continuously operation of monitoring networks and remote sensing instrument to provide real-time data (e.g., AirNow and Satellite) for data assimilation/model evaluation and (2) the development of process-oriented models to isolate complex feedbacks. Finally, a unified modelling system that allows a single platform to operate over the full scale will represent a substantial advancement in both the science and the computational efficiency. Major challenges include globalization/downscaling with consistent model physics and two-way nesting with mass conservation and consistency. Such a unified global-to-urban scale modelling system will provide a new scientific capability for studying important problems that require a consideration of multi-scale feedbacks.

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Table 1 : Model Systems and Typical Applications of Online Models.

Model System/Scale	Met. Model	Chemical Transport Model (Main features)	Typical Applications	Example References
GATOR-GCMOM & Predecessors (Global-through-urban)	MMTD GCMM GCMOM	Gas-phase chemistry: CBM-EX: (247 reactions, 115 species); Bulk or size-resolved aqueous-phase sulfate, nitrate, organics, chlorine, oxidant, radical chemistry (64 kinetic reactions); size-resolved, prognostic aerosol/cloud with complex processes	Current/future met/chem/rad feedbacks; Direct/indirect effects; AQ/health effect	Jacobson, 1994, 1997ab, 2001, 2002, 2004; Jacobson et al., 2004, 2006, 2007
WRF/Chem (Mesoscale)	WRF	RADM2, RACM, CBMZ, CB05 (156-237 reactions, 52-77 species); bulk aqueous-phase RADM chemistry (MADE/SORGAM) or CMU mechanism (MOSAIC/MADRID; Three aerosol modules (MADE/SORGAM, MOSAIC, and MADRID with size/mode-resolved, prognostic aerosol/cloud treatments	Forecast/hindcast, Met/chem feedbacks; O_3, $PM_{2.5}$; Aerosol direct effect	Grell et al. (2005); Fast et al. (2006); Zhang et al. (2005ab, 2007)
CAM3 & Predecessors (Global)	CCM3/ CCM2/ CCM1	Prescribed CH_4, N_2O, CFCs/MOZART4 gas-phase chemistry (167 reactions, 63 species); Bulk aqueous-phase sulfate chemistry of S(IV) (4 equilibria and 2 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Climate; Direct/indirect effects; Hydrological cycle	Rasch et al., 1995; Kiehl et al., 1998; Collins et al., 2004, 2006ab
MIRAGE2 & 1 (Global)	CAM2/ CCM2	Gas-phase CO-CH_4-oxidant chem.; Bulk aqueous-phase sulfate chemistry (6 equilibria and 3 kinetic reactions); Mode-resolved simple aerosol treatment; Prognostic aerosol/cloud treatments	Trace gases and PM; Direct/indirect effects	Ghan et al., 2001abc Zhang et al., 2002; Easter et al., 2004
Caltech unified GCM (Global)	GISS GCM II'	Harvard tropospheric O_3-NO_x-hydrocarbon chemistry (305-346 reactions, 110-225 species); bulk aqueous-phase chemistry of S(IV) (5 equilibria and 3 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Global chemistry-aerosol interactions; aerosol direct radiative forcing; the role of heterogeneous chemistry; impact of future climate change on O_3 and aerosols	Liao et al., 2003; Liao and Seinfeld, 2005

Table 2 : Treatments of Aerosol Properties of Online Models.

Model System	Composition	Size Distribution	Aerosol Mixing State	Aerosol Mass/Number	Aerosol Hygroscopicity	Aerosol radiative properties
GATOR-GCMOM	47 species (sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, crustal)	Sectional (17-30): variable, multiple size distributions	A coated core, internal/external mixtures	Predicted/Predicted	Simulated hydrophobic-to-hydrophilic conversion for all aerosol components	Simulated volume-average refractive indices and optical properties based on core-shell MIE theory
WRF/ Chem	Sulfate, nitrate, ammonium, BC, OC, sea-salt	Modal (3): variable (MADE/SORGAM) Sectional (8): variable (MOSAIC/MADRID) single size distribution	Internal	Predicted/Diagnosed from mass or predicted	The same as MIRAGE2	The same as MIRAGE2
CAM3	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust	Modal (4): predicted dust and sea-salt, prescribed other aerosols; single size distribution	External	Prescribed or predicted/Diagnosed from mass	hydrophobic and hydrophilic BC/OC with a fixed conversion rate	Prescribed RI and optical properties for each aero. type, size, and wavelength, for external mixtures
MIRAGE2	Sulfate, BC, OC, sea-salt, dust	Modal (4): variable; single size distribution	Externally mixed modes with internal mixtures within each mode	Prescribed or predicted/Diagnosed or predicted	Simulated BC/OC with prescribed hygroscopicities	Parameterized RI and optical properties based on wet radius and RI of each mode
Caltech unified GCM (Global)	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust	Sectional (11) prescribed for sea-salt; Sectional (6) prescribed for mineral dust; Modal (1): prescribed size distribution for other aerosols; single size distribution for all aerosols	BC, OC, and mineral dust externally mixed with internally-mixed SO_4^{2-}, NH_4^+, NO_3^-, sea-salt, and H_2O; different aerosol mixing states for chemistry and radiative forcing calculation	Predicted aerosol mass; aerosol number not included	Simulated BC/OC with prescribed hygroscopicities	Simulated optical properties based on Mie theory with size- and wavelength-dependent refractive indices

Table 3 : Treatments of Cloud Properties of Online Models.

Model System	Hydrometeor types in clouds	Cloud droplet size distribution	Cloud droplet number	CCN/IDN composition	CCN/IDN spectrum	Cloud radiative properties
GATOR-GCMOM	Size-resolved liquid, ice, graupel, aerosol core components	Prognostic, sectional (30), multiple size distributions (3)	Prognostic, size- and composition-dependent from multiple aerosol size distributions	All types of aerosols treated for both CCN/IDN	Predicted with Köhler theory; sectional (13-17); multiple size distributions (1-16) for both CCN/IDN	Simulated volume-average refractive indices and optical properties based on MIE theory and a dynamic effective medium approximation
WRF/ Chem	Bulk single condensate	Prognostic, sectional, single size distribution (MOSAIC)	The same as MIRAGE2 (MOSAIC)	The same as MIRAGE2 but sectional; CCN only	The same as MIRAGE2 but sectional, CCN only	The same as MIRAGE2 but sectional (MOSAIC)
CAM3	Bulk liquid and ice	The same as MIRAGE2	The same as MIRAGE2	All treated species except hydrophobic species; CCN only	Prescribed; CCN only	The same as MIRAGE2
MIRAGE2	Bulk single condensate	Prescribed, modal, single size distribution	Prognostic, aerosol size- and composition-dependent, parameterized	All treated species; CCN only	Function of aerosol size and hygroscopicity based on Köhler theory; CCN only	Prognostic, parameterized in terms of cloud water, ice mass, and number
Caltech unified GCM (Global)	Bulk liquid and ice	Diagnosed from predicted cloud water content; single size distribution	constant cloud droplet number based on observations	None	None	Simulated based on MIE theory with different parametrizations for liquid and ice clouds

Table 4 : Treatments of Aerosol Chemistry and Microphysics of Online Models.

Model System	Inorganic aero. thermodynamic equilibrim	Secondary organic aerosol formation	New particle Formation	Condensation of gases on aerosols	Coagulation	Gas/particle mass transfer
GATOR-GCMOM	EQUISOLV II, major inorganic salts and crustal species	Condensation; Dissolution based on Henry's law (10-40 classes VOCs)	Binary homogeneous nucleation of H_2SO_4 and H_2O, T- and RH-dependent	Dynamic condensation of all condensible species based on growth law (e.g., H_2SO_4, VOCs) using the Analytical Predictor of Condensation (APC) with the moving center scheme	Sectional, multiple size distributions, accounts for van der Waals and viscous forces, and fractal geometry	Dynamic approach with a long time step (150-300 s) (PNG-EQUISOLV II) for all treated species
WRF/ Chem	MARS-A (SORGAM) MESA-MTEM (MOSAIC) ISORROPIA (MADRID)	Reversible absorption (8 classes VOCs) based on smog-chamber data (SORGAM and MOSAIC) Absorption (MADRID1) and combined absorption and dissolution (MADRID2)	Binary homogeneous nucleation of H_2SO_4 and H_2O; T- and RH-dependent; sectional; different eqs. in different aero modules	Dynamic condensation of H_2SO_4 and VOCs using the modal approach (SORGAM), of H_2SO_4, MSA, and NH_3 using the Adaptive Step Time-split Explicit Euler Method (ASTEEM) method (MOSAIC), and of volatile inorganic species using the APC with moving center scheme (MADRID)	Modal/Sectional (MADE/SORGAM, MOSAIC), single size distribution, fine-mode only	1. Full equili. In all aerosol modules 2. Dynamic in MOSAIC and MADRID 3. Hybrid in MADRID
CAM3	MOZART4 with regime equili. for sulfate, nitrate, and ammonium	Prescribed SOA yield for □-pinene, n-butane, and toluene	None	Instantaneous condensation of inorganic species	None	Full equilibrium involving $(NH_4)_2SO_4$ and NH_4NO_3
MIRAGE2	Simple equilibrium involving $(NH_4)_2SO_4$ and precursor gases	Prescribed SOA yield	Binary homogeneous nucleation of H_2SO_4 and H_2O; T- and RH-dependent	Dynamic condensation of H_2SO_4 and MSA based on Fuchs and Sutugin growth law	Modal, single size distribution, fine-mode only	Simple equilibrium involving $(NH_4)_2SO_4$ and precursor gases
Caltech unified GCM (Global)	ISORROPIA with regime equili. for sulfate, nitrate, ammonium, sea-salt, and water	Reversible Absorption for 5 biogenic SVOC classes	None	None	None	Full equilibrium involving $(NH_4)_2SO_4$ and NH_4NO_3

Table 5: Treatments of Aerosol-Cloud Interactions and Cloud Processes of Online Models.

Model System	Aerosol Water uptake	Aerosol activation aero-CCN/IDN	In-cloud Scavenging	Below-cloud Scavenging	Droplet Sedimentation
GATOR-GCMOM	Equilibrium with RH; ZSR equation; simulated MDRH; Hysteresis is treated	Mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory	Aerosol activation Nucl. scavenging (rainout) precip. rate dependent of aerosol size and composition	Aerosol-hydrometeor coag. (washout), calculated precip. rate dependent of aerosol size and composition	size-dependent sedimentation
WRF/ Chem	The same as MIRAGE2 but sectional (MOSAIC)	The same as MIRAGE2 but sectional (MOSAIC)	The same as MIRAGE2 but sectional	The same as MIRAGE2 but sectional	The same as MIRAGE2
CAM3	For external mixtures only, equilibrium with RH, no hysteresis	Empirical, prescribed activated mass fraction; bulk CCN only	Prescribed activation, autoconversion, precip. rate independent of aerosols	Prescribed scav. efficiency, no-size dependence	size-dependent sedimentation
MIRAGE2	Equilibrium with RH based on Köhler theory, Hysteresis is treated	Mechanistic, parameterized activation based on Köhler theory; bulk CCN only	Activation, Brownian diffusion (inters./activated), autoconversion, nucleation scavenging, precip. rate independent of aerosols	Prescribed scavenging efficiency with size dependence	no droplet sedimentation
Caltech unified GCM (Global)	ZSR equation, no hysteresis	None	Autoconversion nucl. scavenging with prescribed scavenging coefficient for sea-salt and dust and a first-order precipitation-dependent parametrization for other aerosols; precip. rate independent of aerosols	First-order precipitation-dependent parametrization; calculated scavenging efficiency with size dependence	implicitly accounted for in a parametrization of the limiting autoconversion rate

1.2 On-line Chemistry within WRF: Description and Evaluation of a State-of-the-Art Multiscale Air Quality and Weather Prediction Model: *Georg Grell, Jerome Fast, William I Gustafson, Steven E. Peckham, Stuart McKeen, Marc Salzmann, Saulo Freita*

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1. Introduction

Many of the current environmental challenges in weather, climate, and air quality involve strongly coupled systems. It is well accepted that weather is of decisive importance for air quality. It is also recognized that chemical species influence the weather by changing the atmospheric radiation budget as well as through cloud formation. However, a fundamental assumption in traditional air quality modelling procedures is that it is possible to make accurate air quality forecasts (and simulations) even while ignoring much of the coupling between meteorological and chemical processes. This commonly used approach is termed offline. Here we describe a modelling system – and some relevant applications – that include many of these coupled interactions, resulting in advanced research and forecast capabilities that lead to an improvement of the understanding of complex interactive processes that are of great importance to regional and urban air quality, global climate change, and weather prediction. The resulting improved predictive capabilities could lead to more accurate health alerts, to a larger confidence when using the modelling system for regulatory purposes, and to better capabilities in predicting the consequences of an accidental or intentional release of hazardous materials. In section 2 we first give a brief outline of the model capabilities. WRF/Chem is a community model and is being developed by many different groups. Whenever developers are willing to provide their new implementations back to the community, these implementations are subjected to rigorous evaluation before being officially released to the public. Section 3 describes some of the data sets that are being used for evaluation. Finally, in section 4 we describe ongoing and future development work.

2. The On-line Modelling Approach

In online modelling systems, the chemistry is integrated simultaneously with the meteorology, allowing feedback at each model time step both from meteorology to chemistry and from chemistry to meteorology. This technique more accurately reflects the strong coupling of meteorological and chemical processes in the atmosphere. The state-of-the-art tightly coupled modelling system described here is based on the Weather Research and Forecast (WRF) model. It is designed to be modular, and a single source code is maintained that can be configured for both research and operations. It offers numerous physics options, thus tapping into the experience of the broad modelling community. Advanced data assimilation systems are being developed and tested in tandem with the model. The model is designed to improve forecast accuracy across scales ranging from cloud to synoptic, which makes WRF particularly well suited for newly emerging Numerical Weather Prediction (NWP) applications in the non-hydrostatic regime. Meteorological details of this modelling system

can be found in *Skamarock et al. (2005)*; the details of the chemical aspects are covered in *Grell et al. (2005), Fast et al. (2006),* and *Gustafson et al. (2007).*

2.1. Grid-Scale Transport of Species

Although WRF has several choices for dynamic cores, the mass coordinate version of the model, called Advanced Research WRF (ARW) is described here. The prognostic equations integrated in the ARW model are cast in conservative (flux) form for conserved variables; non-conserved variables such as pressure and temperature are diagnosed from the prognostic conserved variables. In the conserved variable approach, the ARW model integrates a mass conservation equation and a scalar conservation equation of the form

$$\mu_t + \nabla \cdot (\mathbf{V}\mu) = 0,$$

$$(\mu\phi)_t + \nabla \cdot (\mathbf{V}\mu\phi) = 0.$$

In these equations μ is the column mass of dry air, V is the velocity (*u, v,* w), and ϕ is a scalar mixing ratio. These equations are discretized in a finite volume formulation, and as a result the model exactly (to machine roundoff) conserves mass and scalar mass. The discrete model transport is also consistent (the discrete scalar conservation equation collapses to the mass conservation equation when ϕ = 1) and preserves tracer correlations (c.f. *Lin and Rood (1996)*). The ARW model uses a spatially 5th order evaluation of the horizontal flux divergence (advection) in the scalar conservation equation and a 3rd order evaluation of the vertical flux divergence coupled with the 3rd order Runge-Kutta time integration scheme. The time integration scheme and the advection scheme is described in *Wicker and Skamarock (2002). Skamarock et al. (2005*) also modified the advection to allow for positive definite transport.

2.2. Sub-Grid Scale Transport

Typical options for turbulent transport in the boundary layer include a level 2.5 Mellor-Yamada closure parametrization (*Mellor and Yamada, 1982*), or a non-local approach implemented by scientists from the Yong-Sei University (YSU scheme, *Hong and Pan, 1996*). Transport in non-resolved convection is handled by an ensemble scheme developed by *Grell and Devenyi (2002).* This scheme takes time-averaged rainfall rates from any of the convective parametrizations from the meteorological model to derive the convective fluxes of tracers. This scheme also parameterizes the wet deposition of the chemical constituents.

2.3. Dry Deposition

The flux of trace gases and particles from the atmosphere to the surface is calculated by multiplying concentrations in the lowest model layer by the spatially and temporally varying deposition velocity, which is proportional to the sum of three characteristic resistances (aerodynamic resistance, sublayer resistance, and surface resistance). The surface resistance parametrization developed by *Wesely (1989)* is used. In this parametrization, the surface resistance is derived from the resistances of the surfaces of the soil and the plants. The properties of the plants are determined using land-use data and the season. The surface resistance also depends on the diffusion coefficient, the reactivity, and water solubility of the reactive trace gas.

The dry deposition of sulfate is described differently. In the case of simulations, in which aerosols are not calculated explicitly, sulfate is assumed to be present in the form of aerosol particles, and its deposition is described according to *Erisman et al. (1994).*

2.4. Photolysis Frequencies

Two options are available to calculate photolysis frequencies for the photochemical reactions of the gas-phase chemistry model. These are based on *Madronich (1987)* and *Wild et al. (2000)* and are also calculated online.
The profiles of the actinic flux are computed at each grid point of the model domain. For the determination of the absorption and scattering cross sections needed by the radiative transfer model, predicted values of temperature, ozone, and cloud liquid water content are used below the upper boundary of WRF. Above the upper boundary of WRF, fixed typical temperature and ozone profiles are used to determine the absorption and scattering cross sections. These ozone profiles are scaled with TOMS (Total Ozone Mapping Spectrometer) satellite observational data for the area and date under consideration.
The radiative transfer model in *Madronich (1987)* permits the proper treatment of several cloud layers, each with height-dependent liquid water contents. The extinction coefficient of cloud water $ß_c$ is parameterized as a function of the cloud water computed by the three-dimensional model based on a parametrization given by *Slingo (1989)*. For the Madronich scheme used in WRF/Chem, the effective radius of the cloud droplets follows *Jones et al. (1994)*. For aerosol particles, a constant extinction profile with an optical depth of 0.2 is applied.

2.5. Gas-Phase Chemistry: Hard-Coded Chemical Mechanisms

WRF/Chem can use two hard coded chemical gas-phase mechanisms. The first is an atmospheric chemical mechanism that was originally developed by *Stockwell et al. (1990)* for the Regional Acid Deposition Model, version 2 (RADM2) (*Chang et al., 1989*). The RADM2 mechanism is a compromise between chemical detail, accurate chemical predictions and available computer resources. It is widely used in atmospheric models to predict concentrations of oxidants and other air pollutants. For inorganic species, the RADM2 mechanism includes 14 stable species, 4 reactive intermediates, and 3 abundant stable species (oxygen, nitrogen and water). Atmospheric organic chemistry is represented by 26 stable species and 16 peroxy radicals. The RADM2 mechanism represents organic chemistry through a reactivity aggregated molecular approach (*Middleton et al., 1990*). Similar organic compounds are grouped together into a limited number of model groups through the use of reactivity weighting. The aggregation factors for the most emitted VOCs are given in *Middleton et al. (1990)*. A QSSA (Quasi Steady State Approximation) method with 22 diagnosed species, which includes 3 derived from a lumped group, 3 constant, and 38 predicted species is used for the numerical solution. The rate equations for 38 predicted species are solved using a Backward Euler scheme.
The second option is the CBM-Z mechanism. The implementation of the CBM-Z (*Zaveri and Peters, 1999*) is described in more detail in *Fast et al. (2006)*. The CBM-Z photochemical mechanism (*Zaveri and Peters, 1999*) contains 55 prognostic species and 134 reactions. CBM-Z uses the lumped-structure approach for condensing organic species and reactions, and is based on the widely used Carbon Bond Mechanism (CBM-IV). CBM-Z extends the original CBM-IV to include more long-lived species and their intermediates, revised inorganic chemistry, explicit treatment of lesser reactive paraffins such as methane and ethane, revised treatments of reactive paraffin, olefin, and aromatic reactions, inclusion of alkyl and acyl peroxy radical interactions and their reactions with NO3, inclusion of longer lived organic nitrates and hydroperoxides, revised isoprene chemistry, and chemistry associated with dimethyl sulfide (DMS) emissions from oceans.

2.6. Gas-Phase Chemistry: the WRF/Chem/KPP Coupler

Coupled state-of-the-art meteorology/chemistry models such as WRF/Chem (*Grell et al., 2005*) typically include hundreds of reactions and dozens of chemical species. Solving the corresponding huge systems of ordinary differential equations requires highly efficient numerical integrators. In the case of hard-coded manually "tuned" solvers, even minor changes to the chemical mechanism, such as updating the mechanism by additional equations, often require recasting the equation system and, consequently major revisions of the code. This procedure is both extremely time consuming and error prone. In recent years, automatic code generation has become an appreciated and widely used tool to overcome these problems. The Kinetic PreProcessor (KPP, *Damian et al. 2002; Sandu et al. 2003; Sandu and Sander 2006*) is a computer program which reads chemical equations and reaction rates from an ASCII input file provided by the user and writes the program code necessary to perform the numerical integration. Efficiency is obtained by automatically reordering the equations in order to exploit the sparsity of the Jacobian. Receently, some of the KPP capabilities have been adapted for WRF/Chem. For this purpose, the latest KPP version (V2.1) was slightly modified (i.e. an additional switch has been implemented) to produce Fortran 90 modules which can be used in WRF/Chem without further modifications. Furthermore, a preprocessor for WRF/Chem has been developed which automatically generates the interface routines between the KPP generated modules and WRF/Chem, based on entries form the WRF/Chem registry and on KPP input files. This WRF/Chem/KPP coupler can be executed automatically during build time and considerably reduces the effort necessary to add chemical compounds and/or reactions to existing mechanisms as well as the effort necessary to add new mechanisms using KPP in WRF/Chem. At present equation files are available for various version of the RACM and RADM2 mechanisms (also RACM-MIM, *Geiger et al., 2003*).

2.7. Aerosol Modules

WRF/Chem has several options for modelling aerosols. These are based on the Modal Aerosol Dynamics Model for Europe (MADE/SORGAM) (*Ackermann et al., 1998, Schell et al., 2001*), and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC). MADE/SORGAM is a modification of the regional particulate model (*Binkowski and Shankar, 1995*). Secondary organic aerosols (SOA) have been incorporated into MADE by *Schell et al. (2001)*, by means of the Secondary Organic Aerosol Model (SORGAM). Since the different components of the modules are well documented in the above cited references, only a brief summary of the most important features shall be given here.

In MADE/SORGAM, the size distribution of the submicrometer aerosol is represented by two overlapping intervals, called modes, assuming a log-normal distribution within each mode. The conservation equations used to predict the aerosol distributions are similar to those for the gas phase species, with additional terms characterizing the aerosol dynamics and are formulated in terms of the integral moments. The most important process for the formation of secondary aerosol particles is the homogeneous nucleation in the sulfuric acid-water system. It is calculated by the method given by *Kulmala et al. (1998)*. Aerosol growth by condensation occurs in two steps: the production of condensable material (vapor) by the reaction of chemical precursors, and the condensation and evaporation of ambient volatile species on aerosols. In MADE the Kelvin effect is neglected, allowing the calculation of the time rate of change of a moment for the continuum and free-molecular regime. The mathematical expressions of the rates and their derivation are given in *Binkowski and Shankar (1995)*. For coagulation, in MADE it is assumed that during the process of coagulation the distributions remain log-normal. Furthermore, only the effects caused by Brownian motion are considered for the treatment of coagulation. The mathematical formulation for the coagulation process can be found in *Whitby et al. (1991), Binkowski and Shankar (1995)*.

The inorganic chemistry system is based on MARS (*Saxena et al., 1986*) and its modifications by *Binkowski and Shankar (1995)*, which calculates the chemical composition of a sulphate/nitrate/ammonium/water aerosol on equilibrium thermodynamics. Two regimes are considered depending upon the molar ratio of ammonium and sulphate. For values less than 2 the code solves a cubic for hydrogen ion molality, and if enough ammonium and liquid water are present it calculates the dissolved nitrate. For modal ionic strengths greater than 50, nitrate is assumed not to be present. For molar ratios of 2 or greater, all sulphate is assumed to be ammonium sulphate and a calculation is made for the presence of water. The Bromley methods are used for the calculation of the activity coefficients.
The organic chemistry is based on SORGAM (*Schell et al., 2001*). SORGAM assumes that SOA compounds interact and form a quasi-ideal solution. The gas/particle portioning of SOA compounds are parameterized according to *Odum et al. (1996)*. Due to the lack of information all activity coefficients are assumed to be 1. SORGAM treats anthropogenic and biogenic precursors separately and is designed for the use of the RACM gas phase mechanism (*Whitby et al., 1991*).
The Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) was added as one of the aerosol chemistry options by *Fast et al. (2006)*. In contrast to the modal approach for the aerosol size distribution employed by MADE / SORGAM, MOSAIC employs a sectional approach, which is not restricted by physical and numerical assumptions inherent to the modal approach. The aerosol size distribution is divided into discrete size bins. Each bin is assumed to be internally-mixed so that all particles within a bin are assumed to have the same chemical composition. The number of size bins is flexible and eight bins were employed for out simulations with six bins for particle diameters less than 2.5 μm and two size bins for particle diameters greater than 2.5 μm. Both mass and number are simulated for each bin. Both mass and number are simulated for each bin. Particle growth or shrinkage resulting from uptake or loss of trace gases (H_2SO_4, HNO_3, HCl, NH_3, and eventually secondary organic species) is first calculated in a Lagrangian manner. Transfer of particles between bins is then calculated using either the two-moment approach (*Tzivion*, 1989), as in this study, or the moving-center approach (*Jacobson*, 1997) with these growth rates.
Aerosols in MOSAIC are composed of sulfate, nitrate, ammonium, chloride, sodium, other (unspecified) inorganics, organic carbon (OC), elemental carbon (EC), water, and optionally, calcium, carbonate, and methane sulfonic acid. Both the actual aerosol water content and aerosol water content at 60% RH (assuming complete deliquescence) for each size bin are treated as prognostic species and are used to diagnose whether the aerosol particles are on the upper or lower hysteresis curve at relative humidities between the deliquescence and crystallization points. Assuming eight size bins, 88-112 prognostic species are required for the aerosol composition species, water, and number.
MOSAIC contains three new components designed to be numerically efficient without sacrificing accuracy including 1) a new mixing rule, called the Multi-component Taylor Expansion Method (MTEM), to calculate the activity coefficients of various electrolytes in multi-component aqueous solutions (*Zaveri et al., 2005a*) 2) an efficient solid-liquid equilibrium solver, called the Multi-component Equilibrium Solver for Aerosols (MESA), to compute the solid, liquid, or mixed-phase state of aerosols (*Zaveri et al., 2005b*), and 3) a dynamic integration technique, called Adaptive Step Time-split Explicit Euler Method (ASTEEM), for solving the coupled gas-aerosol partitioning differential equations.

2.8. The Aerosol Direct Effect

A key component for online models is the available short-wave radiation parametrizations and how these are coupled to the various aerosol modules. In WRF/Chem, the radiation schemes

include a very simple scheme (*Dudhia, 1989*), which has aerosol effects included in a very rudimentary way (*Grell et al., 2005*), but also very sophisticated schemes, such as the short wave radiation package from NCAR's Community Atmospheric Model (CAM) Version 3 (http://www.ccsm.ucar.edu/models/atm-cam/index.html). *Fast et al. (2006)* were the first to incorporate a more sophisticated aerosol/radiation feedback into WRF/Chem. This approach is now fully implemented and tested within the WRF/Chem repository. In the current release it must be used together with the CBM-Z gasphase chemical mechanism. For release V3 (to be released in March of 2008) all other mechanisms will be available. Aerosol chemical properties and aerosol optical properties are related in the following way (*Fast et al., 2006*). The extinction, single-scattering albedo, and the asymmetry factor for scattering are computed as a function of wavelength and three-dimensional position. In MOSAIC, each chemical constituent of the aerosol is associated with a complex index of refraction. For each size bin, the refractive index is found by volume averaging, and Mie theory is used to find the extinction efficiency, the scattering efficiency, and the intermediate asymmetry factor as functions of the size parameter. Optical properties are then determined by summation over all size bins. Once the aerosol radiative properties are found, they are fed to a shortwave radiative transfer model to calculate the direct aerosol forcing (*Fast et al., 2006*).

2.9. The Aerosol Indirect Effect

Details of the implementation of the aerosol indirect effects can be found in *Gustafson et al. (2007)*. In short, the first and second aerosol indirect effects are implemented in WRF/Chem through a tight coupling of the MOSAIC aerosol module to the Cloud Condensation Nulei (CCN) and cloud droplets of at least on of the microphysics and radiation schemes (*Gustafson et al., 2007*). Included are also parametrizations for activation/resuspension and wet scavenging (*Easter et al., 2004; Ghan et al., 2001*), and aqueous chemistry (*Fahey and Pandis, 2001*). Work is in progress to add the capability to include the aerosol indirect effects into additional microphysics models that are available in WRF/Chem, as well as to the MADE/SORGAM aerosol module.

2.10. Fire Plumerise

A 1-dimesnional fire plumerise model has been implemented into WRF/Chem. This approach is described by *Freitas et al. (2006)*. The plumerise model is called at every grid point with a user specified time interval. Input data come from Satellite estimates, and an emissions injection height is determined by the model.

3. Model Evaluation and Scientific Applications

Each new implementation is subjected to rigorous evaluation, using two different test-bed data sets that include chemical constituents as well as meteorological parameters in three dimensions. These data sets are well documented and have been used for model evaluation with many different modelling systems, including WRF/Chem (*McKeen et al. 2006, 2007, Pagowski et al. 2005, 2006, Pagowski and Grell 2006, Kim et al. 2007, Wilczak et al. 2007*), with many more publications in preparation for the 2006 TEXAQS field experiment data set. As an example we show in Fig. 1 a comparison with observations of two particular set-ups of WRF/Chem as well as other modelling systems. The 2004 data set is based on the New England Air Quality field experiment (NEAQS2004) that took place in July and August of 2004. Displayed in Fig. 1 are the correlation coefficients comparing model forecasts (true forecasts, not hindcasts) of the 8-hr averaged peak ozone mixing ratios when compared to surface monitoring stations. Fig. 2 shows the horizontal distribution of WRF/Chem results

using the best performing set-up shown in red in Fig. 1). The horizontal plots are correlation coefficients, biases and root mean square errors averaged over a 30 day period (30 runs, each run is initialized at 00UTC).

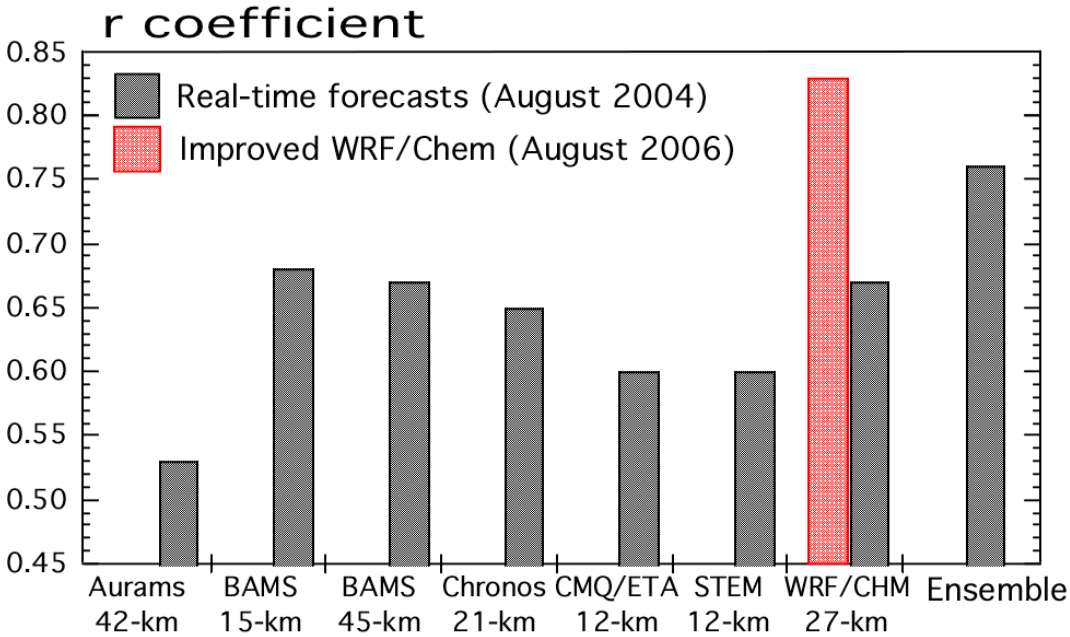

Figure 1 : Correlation coefficients for various model runs, comparing model forecasts of 8-hr averaged peak ozone mixing ratios with those observed by surface monitoring stations. The statistics include a month worth of forecast runs.

Fig.3 and Fig. 4 are shown here to indicate the spread of available observations for model evaluation. Fig. 3 shows a comparison of water vapor mixing ratio as predicted by WRF/Chem for the same time period, but compared to aircraft observations. Similar comparisons are available for other meteorological parameters as well as many chemical constituents, including Ozone, PM species and ozone precursors (Fig. 4). Detailed results are displayed on the WEB at http://www.al.noaa.gov/ICARTT/modeleval.

4. Ongoing Work with WRF/Chem

Ongoing and future work include the extension of the modelling system to global scales. The global WRF model will be available with the next release in March of 2008. An offline version exists and will soon be released for public use. Various groups are working on implementation of new aerosol modules as well as parametrizations (bin resolved and double moment bulk microphysics as well as radiation schemes) that include the aerosol direct and indirect effects. In addition, a version of the Model of Emissions of Gases and Aerosols from Nature (MEGAN) has been implemented into WRF/Chem (courtesy of Serena Chung, Jerome Fast, Christine Wiedinmyer) and will be released in March of 2008. The 2008 release will also include a new photolysis radiation scheme, known as the F-TUV scheme from Madronich. Finally, experiments using chemical data assimilation methods are underways to be able to deteremine a more optimal initial analysis state of the chemical and meteorological atmosphere.

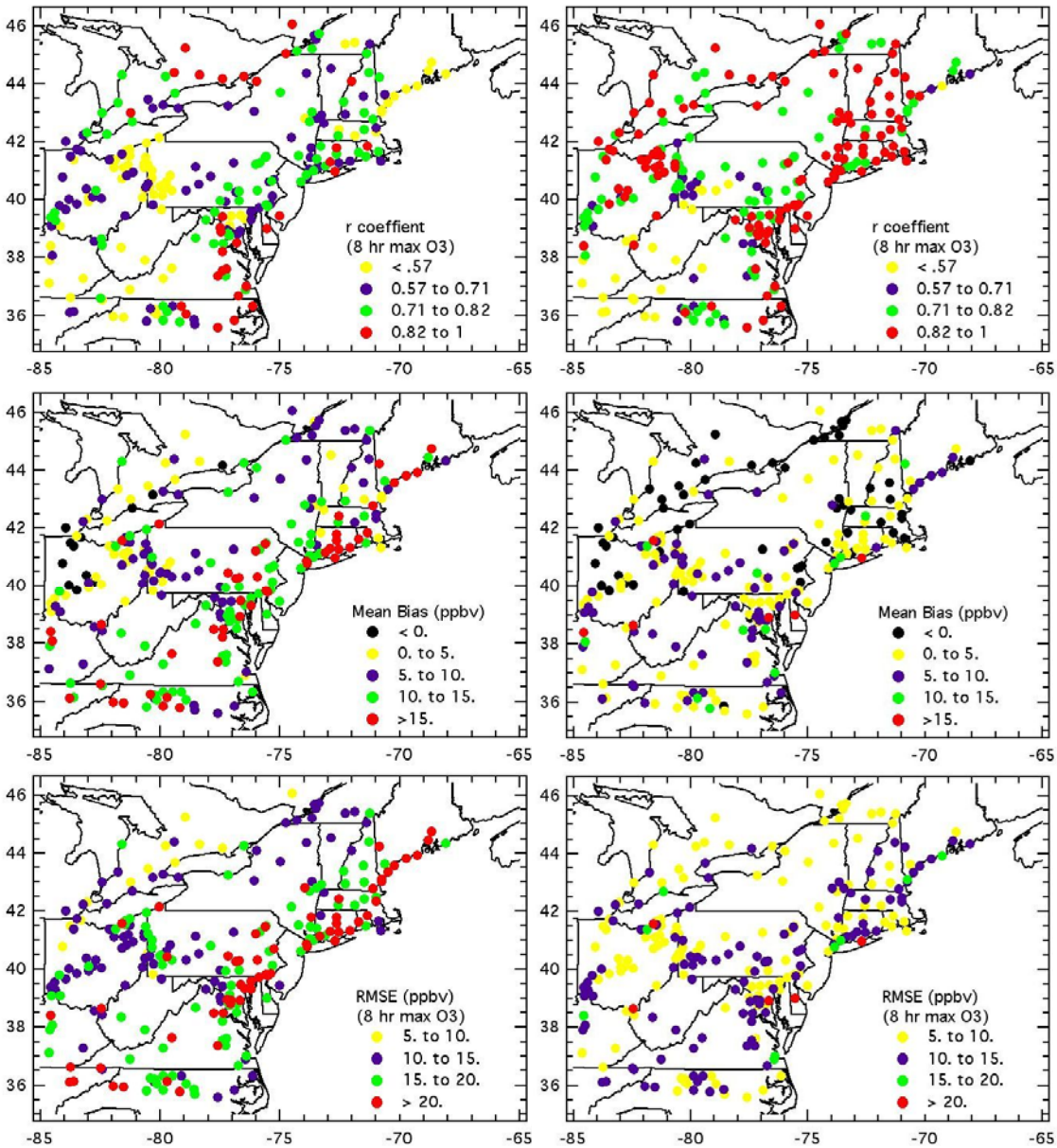

Figure 2 : Correlation coefficient, mean biases, and Root Mean Square Errors(RMSE) for WRF/Chem, comparing model forecasts of 8-hr averaged peak ozone mixing ratios with those observed by surface monitoring stations. The statistics span a time period of 30 days. The model was run once a day at 0000UTC.

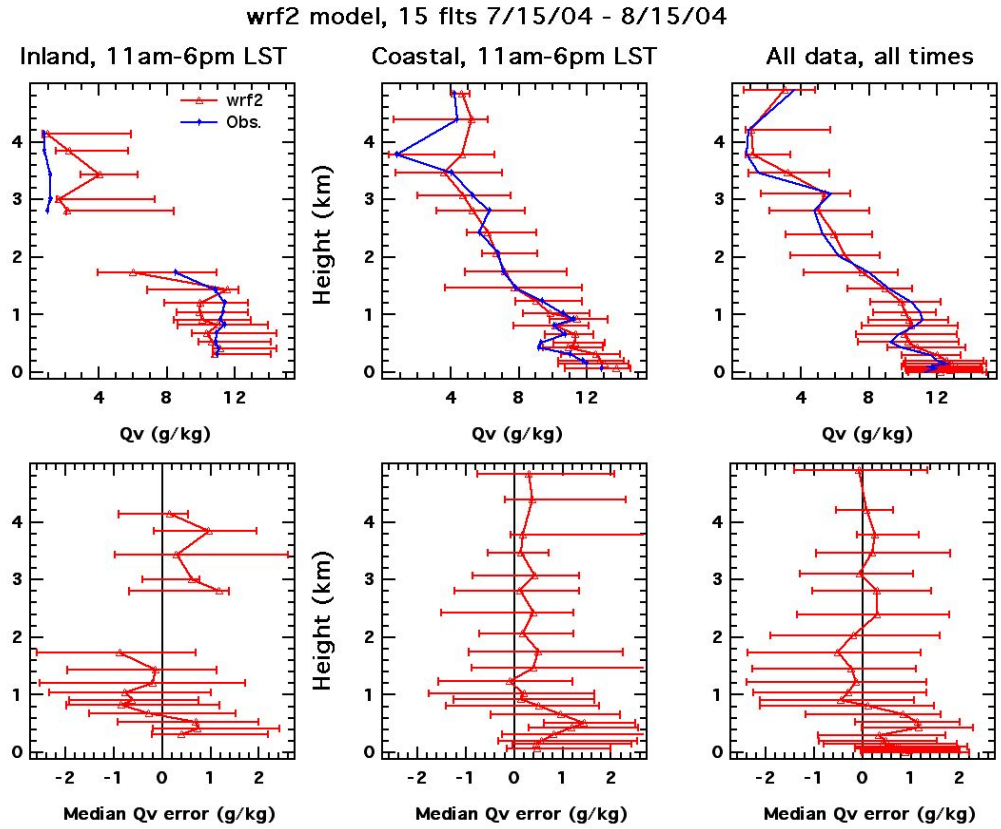

Figure 3 : Meteorological evaluation of WRF/Chem using the NEAQS2004 field experiment. Displayed are model predicted mean profiles of mixing ratio versus the observed profiles during 15 aircraft flights.

gas phase chemistry

	AURAMS	CHRONOS	STEM	WRF-2
O_3	√	√	√	√
CO			√	√
NO	√	√	√	√
NOx	√	√	√	√
NOy	√	√	√	√
PAN	√	√	√	√
Isoprene	√	√	√	√
SO_2	√	√	√	√
NO_3	√	√		√
N_2O_5	√	√		√
CH_3CHO	√			√
Toluene		√		√
Ethylene	√	√		√
NH_3				√

aerosols, radiation, meteorology

	AURAMS	CHRONOS	STEM	WRF-2
PM2.5	√	√	√	√
Asol SO_4	√	√	√	√
Asol NH_4	√			√
Asol OC	√	√	√	√
Asol EC	√		√	√
Asol NO_3	√			√
JNO_2				√
T	√	√	√	√
P	√		√	√
H_2O	√	√	√	√
winds	√	√	√	√
SST	√			√
Radiation				√

Figure 4 : Availability of chemical constituents as well as meteorological parameters for model evaluation (from http://www.al.noaa.gov/ICARTT/modeleval)

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1.3 Multiscale Atmospheric Chemistry Modelling with GEM-AQ: *Jacek Kaminski[1], Lori Neary[1], Joanna Struzewska[2] and John C. McConnell[1]*

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1. Introduction

The strategic objective of our project was to develop and evaluate a modelling system for tropospheric chemistry and air quality (AQ). In our design we have selected the Global Environmental Multiscale model (GEM) *Cote et al. (1998a)* as a host meteorological model for inclusion of AQ processes. The GEM model was developed at the Canadian Meteorological Centre and is used for operational numerical weather prediction (NWP) in Canada. The GEM model was augmented by implementing AQ chemistry, including the gas phase, aerosol and cloud particles, limited wet chemistry, emission, deposition, and transport processes.
The integrated model (so called GEM-AQ) serves as a platform for performing scientific studies on processes and applications. In order to develop an AQ modelling system which can accommodate various scales and processes, we have used the GEM model as a computational platform and environmental processes were implemented on-line. There is a growing recognition for on-line implementation of tightly coupled environmental processes. Similar implementation of environmental processes is done in WRF/Chem (Weather Research and Forecasting model with Chemistry) (*Grell et al., 2005*), MC2-AQ (Mesoscale Compressible Community model with Air Quality) *Kaminski et al. (2002)*, MESSy (Modular Earth Submodel System) *Joeckel et al. (2006)*, RAMS (Regional Atmospheric Modelling System) (*Marecal et al., 2006*) and Meso-nh (non-hydrostatic mesoscale atmospheric model) (*Tulet et al., 2003)*.
The on-line implementation of environmental processes in the GEM model allows running in global uniform, global variable, and limited area configurations, allowing for multiscale chemical weather forecasting (CWF) modelling. This approach provides access to all required dynamics and physics fields for chemistry at every time step. The on-line implementation of chemistry and aerosol processes will allow for introducing feedback on model dynamics and physics. The use of the GEM framework permits the incorporation of chemical data assimilation techniques into the model validation and application studies in a unified fashion.
The developed modelling system can be used to plan field campaigns, interpret measurements, and provide the capacity for forecasting oxidants, particulate matter and toxics. Also, it can be used to provide guidance to evaluate exposure studies for people, animals, crops and forests, and possibly for epidemiological studies.

2. Methodology

2.1. Host Meteorological Model

The host meteorological model used for air quality studies is the Global Environmental Multiscale (GEM) model. GEM can be configured to simulate atmospheric processes over a broad range of scales, from the global scale down to the meso-gamma scale.

2.2. Model Dynamics

The set of non-hydrostatic Eulerean equations (with a switch to revert to the hydrostatic primitive equations) maintain the model's dynamical validity right down to the meso-gamma scales. The time discretization of the model dynamics is fully implicit, 2 time-level (*Cote et al., 1998ab).* The spatial discretization for the adjustment step employs a staggered Arakawa C grid that is spatially offset by half a mesh length in the meridional direction with respect to that employed in previous model formulations. It is accurate to second order, whereas the interpolations for the semi-Lagrangian advection are of fourth-order accuracy, except for the trajectory estimation (*Yeh et al., 2002).* The vertical diffusion of momentum, heat and tracers is a fully implicit scheme based on turbulent kinetic energy (TKE). GEM version 3.1.2 was used in the current study.

2.3. Model Physics

The physics package consists of a comprehensive set of physical parametrization schemes (*Benoit et al., 1989).* Specifically, the atmospheric boundary layer (ABL) is based on a prognostic equation for TKE. The surface temperature over land surface is calculated using the force-restore method combined with a stratified surface layer. Deep convective processes are handled by a Kuo-type convective parametrization *(Kuo, 1974)* for the resolutions that we have adopted for this study. The infrared radiation scheme includes the effects of water vapor, carbon dioxide, ozone, and clouds. Gravity wave drag parametrization is based on a simplified linear theory for vertically propagating gravity waves generated in a statically stable flow over mesoscale orographic. GEM physics package version 4.2 was used in the current study.

2.4. Air Quality Modules

Air quality modules are implemented on-line in the host meteorological model. Currently, there are 37 advected and 14 non-advected gas phase species in the model. Transport of the chemically active tracers by the resolved circulation is calculated using the semi-Lagrangian advection scheme native to GEM. The vertical transfer of trace species due to subgrid-scale turbulence is parameterized using eddy diffusion calculated by the host meteorological model. Large scale deep convection in the host model depends on the resolution: in this version of GEM-AQ we use the mass flux scheme of *Zhang et al. (1995)* for tracer species.

2.5. Gas Phase Chemistry

The gas-phase chemistry mechanism currently used in the GEM-AQ model is based on a modification of version two of the Acid Deposition and Oxidants Model (ADOM) *Venkatram et al. (1988),* derived from the condensed mechanism of *Lurmann (1986).* The ADOM-II mechanism comprises 47 species, 98 chemical reactions and 16 photolysis reactions. In order to account for background tropospheric chemistry, 4 species (CH_3OOH, CH_3OH, CH_3O_2, and CH_3CO_3H) and 22 reactions were added. All species are solved using a mass-conserving implicit time stepping discretization, with the solution obtained using Newton's method. Heterogeneous hydrolysis of N_2O_5 is calculated using the on-line distribution of aerosol.

Although the model meteorology is calculated up to 10 hPa, the focus of the chemistry is in the troposphere where all species are transported throughout the domain. To avoid the overhead of stratospheric chemistry in this version (a combined stratospheric/tropospheric chemical scheme is currently being developed) we replaced both the ozone and NOy fields with climatology above 100 hPa after each transport time step. This ensures a reasonable upper boundary to the troposphere, while ensuring that the transport of ozone and NOy fields

to the troposphere is well characterized by the model dynamics. For ozone we used the HALOE (Halogen Occultation Experiment) climatology (e.g. *Hervig et al., 1993*), while NOy fields are taken from the CMAM (Canadian Middle Atmosphere Model).
Photolysis rates (J values) are calculated on-line every chemical time step using the method of *Landgraf and Crutzen (1998)*. In this method, radiative transfer calculations are done using a delta-two stream approximation for 8 spectral intervals in the UV and visible applying precalculated effective absorption cross sections. This method also allows for scattering by cloud droplets and for clouds to be presented over a fraction of a grid cell. Both cloud cover and water content are provided by the host meteorological model. The J value package used was developed for MESSy (*Joeckel et al., 2006*) and has been implemented in GEM-AQ.

2.6. Aerosol Package

The current version of GEM-AQ has 5 size-resolved aerosols types, viz. sea salt, sulphate, black carbon, organic carbon, and dust. The microphysical processes which describe formation and transformation of aerosols are calculated by a sectional aerosol module (*Gong et al., 2003)*. The particle mass is distributed into 12 logarithmically spaced bins from 0.005 to 10.24 microns radius. This size distribution leads to an additional 60 advected tracers. The following aerosol processes are accounted for in the aerosol module: nucleation, condensation, coagulation, sedimentation and dry deposition, in-cloud oxidation of SO_2, in-cloud scavenging, and below-cloud scavenging by rain and snow.

2.7. Gas-Phase Removal Processes

The effects of dry deposition are included as a flux boundary condition in the vertical diffusion equation. Dry deposition velocities are calculated from a `big leaf' multiple resistance model *(Wesely, 1989; Zhang et al., 2002)* with aerodynamic, quasi-laminar layer, and surface resistances acting in series. The process assumes 15 land-use types and takes snow cover into account.
GEM-AQ only has a simplified aqueous phase reaction module for oxidation of SO_2 to sulphate. Thus, for the gas phase species, wet deposition processes are treated in a simplified way. Only below-cloud scavenging of gas phase species is considered in the model. The efficiency of the rainout is assumed to be proportional to the precipitation rate and a species-specific scavenging coefficient. The coefficients applied are the same as those used in the MATCH model (Multiscale Atmospheric Transport and Chemistry Model) used by the Swedish Meteorological and Hydrological Institute (SMHI) *(Langner et al., 1998).*

2.8. Emissions

The emission dataset used for global simulations was compiled using EDGAR 2.0 (Emission Database for Global Atmospheric Research) (archived in 2000, valid for 1990) and GEIA (Global Emissions Inventory Activity) global inventories. The EDGAR 2.0 dataset was chosen for its detailed information on non-methane volatile organic compound speciation. Emission data compiled for GEM-AQ includes global fields of anthropogenic emission fluxes with 1° x 1° resolution and natural emissions with 5° x 5° resolution. Yearly averaged anthropogenic emissions contain different industrial sectors and non-industrial activity such as burning of agricultural wastes and fuel wood, for 14 gaseous pollutants. Monthly averaged biogenic, ocean and soil emission fluxes, as well as biomass burning (forest and savannah) emissions, have been derived for 9 species (7 VOC species, CO and NO_2)
In the upper troposphere/lower stratosphere (UTLS) region sources of NOx are small, from large scale convective updrafts, stratospheric sources, aircraft and lightning. We have used the monthly mean totals of lightning NOx from the GEIA inventory (scaled from 12.2 Tg/yr to 2

Tg/yr) and distributed them in the horizontal according to the convective cloud distribution of the model.

3. Model Applications

The GEM-AQ model has been run for a number of scenarios ranging from a global uniform domain (this study), global variable resolution for regional scenarios *O'Neill et al. (2006)*, to high resolution studies (*Struzewska and Kaminski, 2007*), global uniform long term simulations to derive a multi-year model climatology, to examine seasonal variation and regional distribution, evaluate global emissions, and provide chemical initial and boundary conditions for high resolution model simulations (*Kaminski et al., 2007)*. GEM-AQ has also been augmented to study persistent organic pollutants (POPs) globally (*Gong et al., 2007; Huang et al., 2007*).

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1.4 Status and Evaluation of Enviro-HIRLAM: Differences between On-line and Off-line Models : *Ulrik Korsholm, Alexander Baklanov and Jens Havskov Sørensen*

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Abstract
Online coupled model systems in which a meteorological model contains emissions, transport, dispersion, deposition, chemistry and aerosol physics of pollutants have recently received much attention, mainly due to the prospects of including feedbacks between pollutants and meteorological fields and quantifying indirect effects. There are, however, several other important differences between online and offline coupled models and in this context it is important to investigate when online models are useful. This study presents the current status of Enviro-HIRLAM, an online coupled model developed at the Danish Meteorological Institute, and investigates the importance of two of the main advantages of online models; the increased availability of meteorological information and the inclusion of feedbacks. Enviro-HIRLAM is shown to perform satisfactorily during tests of transport, dispersion and deposition, using data from the first European Tracer Experiment (ETEX-1) and measurements of deposited Cesium-137 made after the Chernobyl accident. Offline and online simulations of the ETEX-1 release are compared and it is shown that the offline coupling interval plays an important role in constraining the influence of meso-scale disturbances on (long range) plume development. Feedback in the form of the first indirect effect has been implemented in the model and comparison of simulations with and without feedback shows that a redistribution of pollutants occurs. The results are consistent with a change in atmospheric stability due to the aerosol cloud interactions.

1. Introduction

Chemical transport models have found usage within a wide range of disciplines serving as tools for basic research, emergency preparedness, air pollution forecasting and for decision support systems. Traditionally, these models have developed independently of meteorological short range weather forecast models and are generally either Gaussian plume, Eulerian, Lagrangian or hybrid (Eulerian-Lagrangian, Gaussian-Lagrangian) models. They are forced by pre-processed output from the meteorological models (typically every three hours) and their ability to predict the development of tracer clouds is strongly dependent on the quality of the meteorological output from the driver. This type of coupling between the meteorological driver and the chemical transport model is termed offline.

In recent years computer power has increased dramatically and short range meteorological models have reached cloud resolving scales (5 km and below). These have prompted the development of Eulerian models which integrate all the components of the chemical transport models in the meteorological driver (GATOR, WRF-CHEM, GEM-AQ, BOLCHEM, COSMO LM-ART). Hereby, the meteorological fields (wind, humidity, temperature, cloud water content, etc.) are available at each time step of the meteorological model. This type of model is termed online.

A formal definition of online and offline models may be given as: Offline models comprise separate chemical transport models forced by output from operational meteorological models, analyzed or forecasted meteorological data from archives or data sets, pre-processed meteorological data, measurements or output from diagnostic models. Online models comprise online access models, in which meteorological fields are available at each time step of the meteorological model and online coupled models in which feedbacks between meteorology and tracers are also accounted for.

At cloud resolving scales the meteorological models explicitly resolves more variability than corresponding statistical parametrizations may provide at coarser resolution. In order to utilize this variability for more precise transport, dispersion, deposition and transformation of pollutants online models are needed. Further advantages of online models include: meteorological and tracer fields are on the same grid, using the same physical and dynamical parametrizations thereby avoiding inconsistencies, no temporal or spatial interpolation or pre-processing is necessary, all two and three dimensional meteorological fields are available at each time step of the meteorological model, thereby avoiding loss of variability in the meteorological forcing fields, it is not necessary to handle large output files from meteorological models and there is a possibility of including feedback mechanisms. Offline models, on the other hand, are more suitable for ensembles, where the meteorological fields are reused for many perturbed runs (feedbacks are neglected), and are also easier to use for adjoint modelling. They allow for usage of many different parametrizations and may employ more flexible grid structures.

Pollution concentration fields are known to contain large temporal and meso-scale variability (*Anderson et al., 2003*). Such variability is typically generated by meso-scale influences in the mean flow including sea breezes, development of clouds and precipitation, frontal circulations (and associated rapid changes in wind direction), urban circulations and flow over and around orographical features. The horizontal scale of such disturbances extends from a few kilometres to several hundreds of kilometres, while the time-scale ranges from less than one hour to days. Online models have the ability to temporally and spatially resolve meso-scale disturbances and it is expected that this leads to greater accuracy in tracer distributions, especially at cloud resolving scales. The same holds true for feedbacks such as direct, indirect and semi-direct effects, due to more precise simulations of radiative fluxes and cloud development and precipitation.

The purpose of this study is to illustrate some important differences between online and offline model systems and to evaluate transport, dispersion and deposition of the online coupled meteorological and chemical transport and dispersion model Enviro-HIRLAM (High Resolution Limited Area Model), which is developed at the Danish Meteorological Institute (DMI).

2. Model Description

Enviro-HIRLAM is an online coupled meteorological, chemical transport and dispersion model (Fig. 1). It is based on a previous version HIRLAM-tracer and at its core lies DMI-HIRLAM, version 6.3.7 employed for limited area short range operational weather forecasting at DMI (*Chenevez et al., 2004*). For a detailed description of the features in HIRLAM the reader is referred to the HIRLAM reference guide (*Undén et al., 2002*).
Point sources are parameterized by assuming that the tracer distribution is uniform within the grid box containing the release site, an assumption which, depending on the spatial resolution, is fulfilled in a well mixed boundary layer. The emission is ascribed the grid point closest to the release site in the lowest model layer, corresponding to a height of approximately 30 meters above the surface. In a well mixed boundary layer this will not affect the results away from the emission grid box.

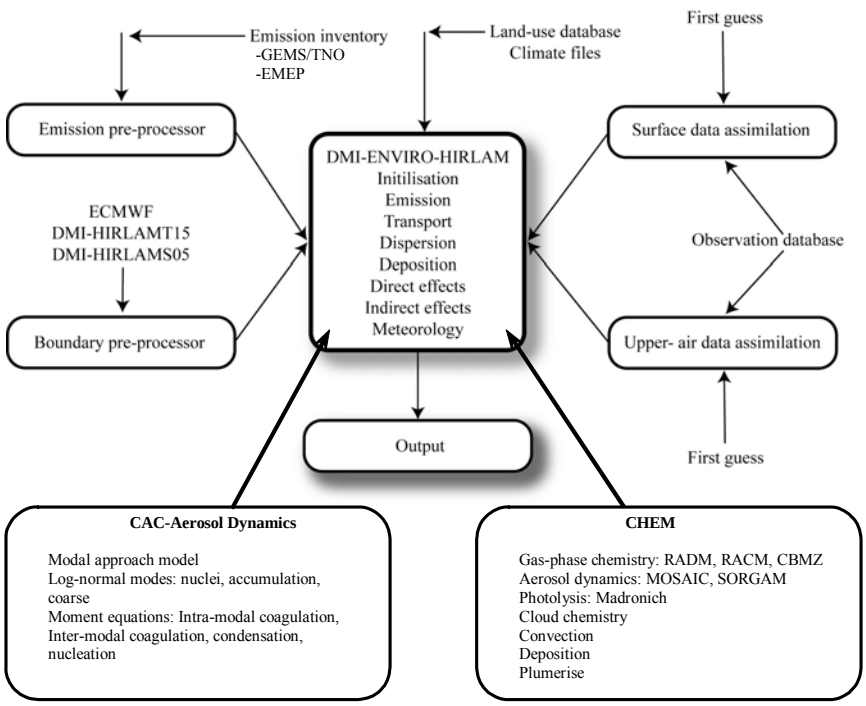


Figure 1 : Illustration of Enviro-HIRLAM structure, including meteorological, chemical and aerosol components.

For integrated atmospheric chemical transport models the requirement of consistency, monotonicity, positive definiteness and mass conservation of the numerical schemes for tracer transport is stronger than for numerical weather prediction models. To ensure the fulfilment of these requirements for chemical species and aerosols the schemes should be harmonised so that the same conservative schemes are used for meteorological and chemical quantities. Work is progressing along these lines with the implementation of the CISL (Cell Integrated Semi Lagrangian) advection scheme (*Nair and Machenhauer, 2002*) in Enviro-HIRLAM.

Several options of advection schemes for the tracers have previously been implemented (Central Difference, Semi-Lagrangian, Bott) (*Bott, 1989ab*); usage depends on the experiment at hand. In order to maintain large time-steps in the solution of the meteorological dynamical equations and at the same time ensure sufficient tracer mass conservation the BOTT scheme was used for the tracers while the semi-lagrangian scheme was used for meteorological quantities. This inconsistency did not affect tracer mass significantly during the runs presented here.

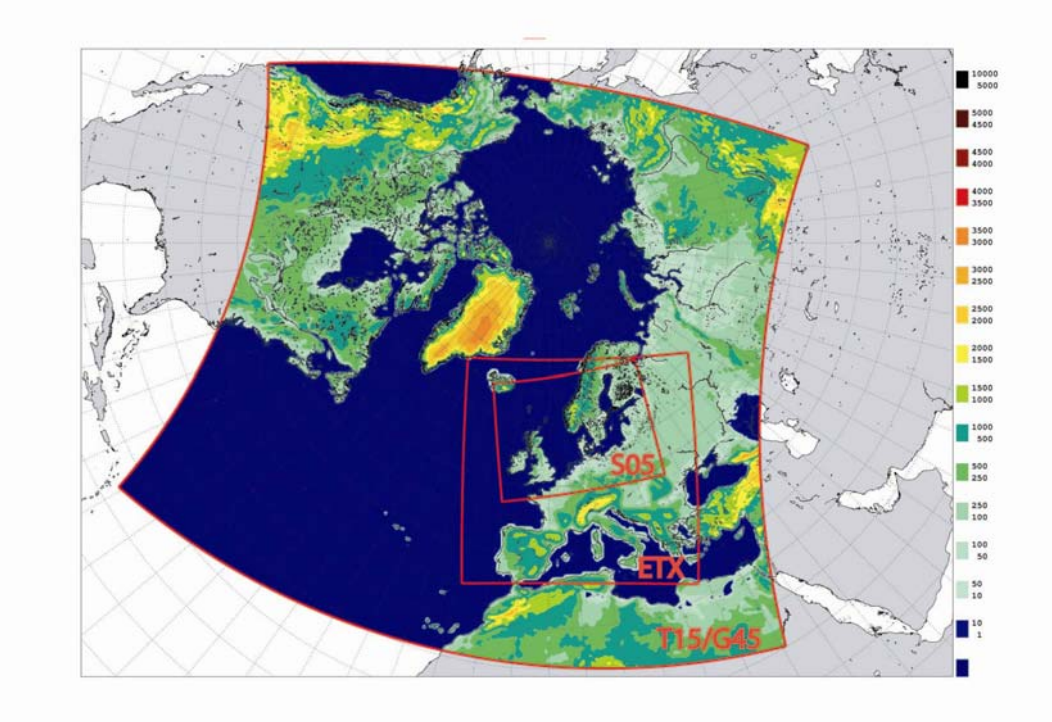


Figure 2 : Areas covered by the model during this study represented by surface geopotential height (meters). The domains T15 and G45 both cover the same area but the horizontal resolution is 0.15° x 0.15° and 0.45° x 0.45°, respectively. The ETX domain is in 0.40° x 0.40° resolution while S05 is in 0.05° x 0.05° resolution.

Dry deposition is parameterized via a resistance approach in which resistances depend on particle size and density, land-use classification and atmospheric stability (*Wesley, 1989; Zanetti, 1990*). Wet deposition is included via below cloud scavenging (washout), using a parametrization based on precipitation rates (*Baklanov and Sørensen, 2001*) and scavenging by snow is parameterized using the scheme by *Maryon et al. (1996)*. The terminal settling velocity is considered in both the laminar case, in which Stoke's law is used and the turbulent case in which a iterative procedure is employed (*Näslund and Thannng, 1991*). For very small particles a correction for non-continuum effects is used.

Photolysis is treated in the scheme by Madronich (*Madronich, 1987*) and the gas-phase chemistry schemes comprise: RADM2 (*Stockwell et al.., 1990*), RACM (*Stockwell et al., 1997*) and CBM-Z an extension of Carbon Bond-IV (*Zaveri and Peters, 1999*). A modal aerosol module containing three log-normal modes, developed at DMI, will be used to treat aerosol physics (*Gross and Baklanov, 2004*).

In the present study horizontal diffusion was switched off. Hence, the numerical diffusion arising from the Bott scheme was the only representation of sub-grid scale horizontal eddies. In the vertical a modified version of the Cuxart, Bougeault, Redelsperger (CBR)-scheme developed for HIRLAM is employed (*Cuxart et al., 2000*). It is based on turbulent kinetic energy, which is a prognostic variable in the model, and a stability dependent length scale

formulation. The model is hydrostatic and horizontal discretization is carried out on a rotated latitude-longitude Arakawa C grid, while in the vertical a hybrid between terrain following sigma and pressure coordinates is employed with 40 levels. A non-hydrostatic version of HIRLAM exists but is presently not used in Enviro-HIRLAM. Digital filter initialization is employed and the model may be run with surface and upper air (3DVAR/4DVAR) analysis. The three model areas used in this study consisted of a 0.40° x 0.40° domain termed ETX, a 0.15° x 0.15° domain termed T15 and a 0.05° x 0.05° domain termed S05 (Fig. 2).

3. Model Evaluation

3.1. Transport and Dispersion

During the first European Tracer Experiment (ETEX-1) a non-depositing tracer gas (Perflouro-Methyl-Cyclo-Hexane) was emitted from a site in Northern France (Brittany; 2°00'30''and 48°03'30''). The average emission rate was 7.95 g/s and it commenced on 23 October at 16:00 UTC lasting for 11 hours and 50 minutes. The spatial and temporal development of the tracer cloud was measured at 168 measurement stations in Europe and both real time and retrospective model inter-comparison projects were carried out (*Graziani et al., 1998; Mosca et al., 1998)*. The purpose of this experiment was to evaluate the models ability to transport and disperse a tracer.

Transport and dispersion was evaluated without any form of tuning by comparing a simulation of the ETEX-1 release to the official measurements of surface concentration. To facilitate comparisons with models evaluated during ATMES II (Atmospheric Transport Model Evaluation Study) an identical statistical methodology was employed (*Mosca et al., 1998*). Background values were subtracted so that only the pure tracer concentration was used. Measurements of zero concentration (concentrations below the background level) were included in time series to the extent that they lay between two non-zero measurements or within two before or two after a non-zero measurement. Hereby, spurious correlations between predicted and measured zero-values far away from the plume track are reduced.

The current version of Enviro-HIRLAM has not previously been evaluated against ETEX-1 measurements. The ETX domain (Fig. 2) was used with at time-step fixed at 10 minutes, and initial and boundary conditions were post-processed from the European Centre for Medium-Range Weather Forecasts operational model, IFS (Integrated Forecast System). No surface or upper air data assimilation was employed and the model was integrated 80 hours into the future. The start time was on 23 October 1994 at 12:00 UTC, four hours before the start of the release. Output was interpolated to measurement stations in order to compare to the observations and produce statistical measures.

Results and Discussion

The synoptic situation in the days following the ETEX-1 release has previously been described in detail by *Gryning et al. (1998)* and *Graziani et al. (1998)*. Correspondingly, the model plume was initially advected by a westerly flow, mainly influenced by synoptic-scale forcings, in a north-easterly direction (*Nastrom and Pace, 1998*). The spatial structure of the model plume resembled the observations (Fig. 3) and remained continuous throughout the forecast period. The plume is most sensitive to meso-scale perturbations during its initial development. Even though the bulk of the plume remains continuous the marginal structure may be affected by such disturbances and cause large errors in verification scores at specific stations. After 36 hours the model plume had attained a U-shaped deformation receding over Northern Germany. Although less distinct a similar structure is present in the observations

(Fig. 3). The model, thus, over-predicted the development of the deformation, which extended further to the North.

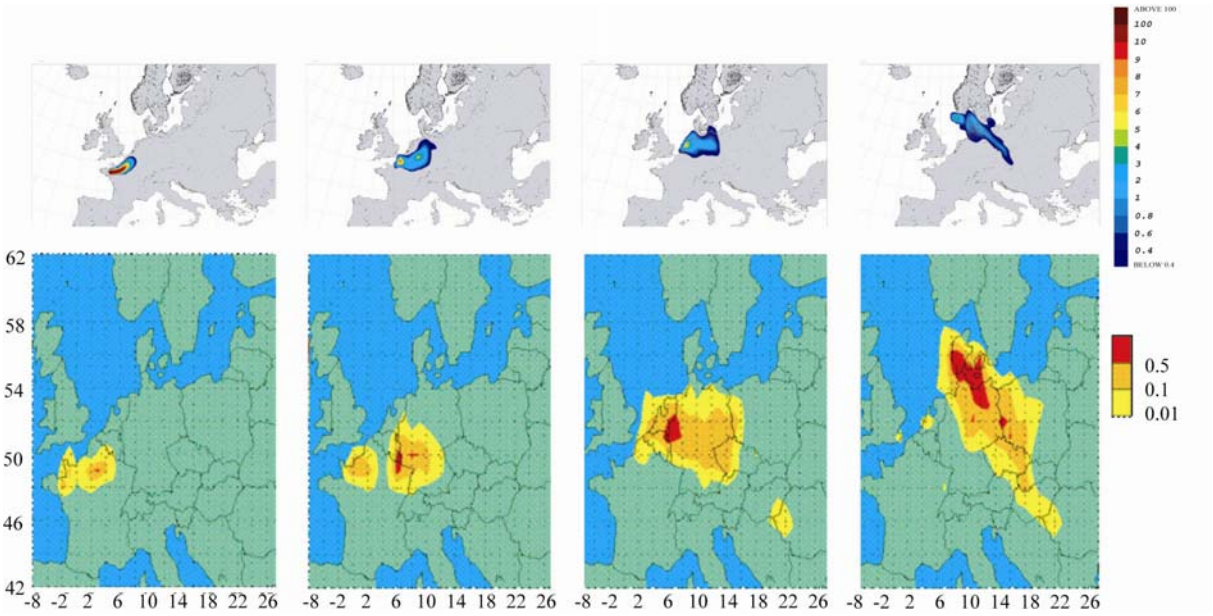


Figure 3 : Top panel: simulated development of the ETEX-1 tracer plume at 12, 24, 36 and 48 hours after start of release (ngm^{-3}). Bottom panel: Corresponding measurements (ngm^{-3}) (*Graziani et al., 1998*).

In line with the measurements the model plume stretched and its axis tilted, so it was oriented in a North-West to South-East direction, after 48 hours. The peak concentration, however, was located too far to the North. After 60 hours the largest concentration values were found in the North Sea, a feature which is also present in the observations. Following the methodology of ATMES II the time development of the model plume was evaluated at 11 selected stations (*Mosca et al., 1998*). These were chosen to constitute two arcs at different distances from the release site. The first arc (measurement stations: NL05, B05, NL01, D44) follows the Eastern border of Belgium. The arrival time at these stations ranged from 15 to 18 hours. The second arc (measurement stations: DK05, DK02, D42, D05, PL03, CR03, H02) extended from Denmark in the North to Hungary in the South and the arrival times at these stations ranged from 30 to 39 hours. The average correlation, normalised mean square error (NMSE), bias and figure of merit in time (FMT) (table 1) at the stations are 0.49, 5.25, 0.18 ng/m³ and 29.35 % respectively. These values are all acceptable when compared to the model scores during ATMES II. All the scores are degraded by the values at the stations in the first arc, suggesting worse performance close to the release site than further away from it, which was generally also found during ATMES II.

Table 1 : Statistical scores for the ETEX-1 simulation at 11 selected measurement stations.

Station	B05	CR03	D05	D44	DK02	DK05	H02	D42	NL01	NL05	PL03
Bias (ngm^{-3})	0.76	-0.08	0.02	0.45	-0.01	-0.11	-0.02	-0.14	0.48	0.65	-0.06
NMSE	12.9	7.95	2.00	4.54	0.93	4.77	1.05	2.25	4.46	14.8	1.95
Correlation	0.80	0.92	0.29	0.64	0.68	0.08	0.86	0.46	-0.05	0.29	0.43
FMT (%)	12.9	26.1	29.6	32.1	51.4	15.4	49.3	32.7	15.9	19.1	38.4

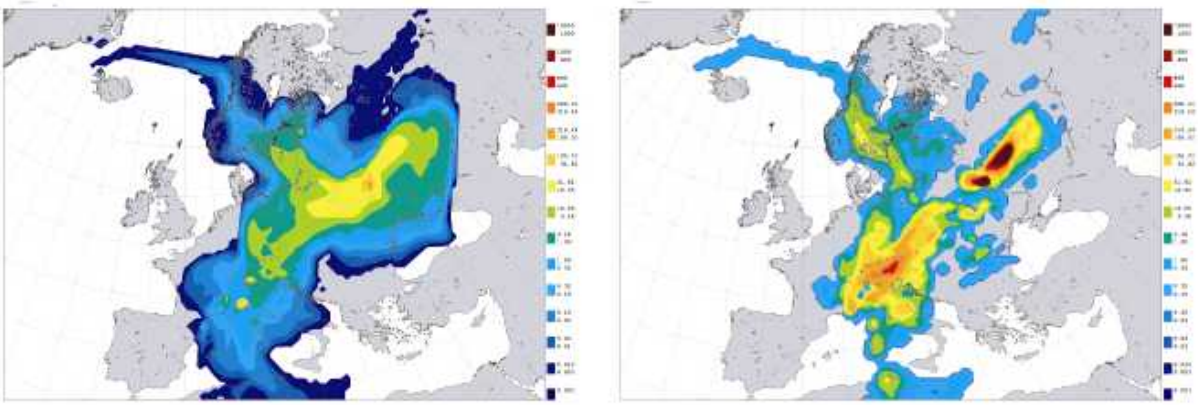
3.2 Deposition

To evaluate the deposition routines a simulation of the Chernobyl accident was carried out and compared to measurements of total deposited Cesium 137 (Cs-137). The measurements

were extracted from the Radioactivity Environmental Monitoring database at the Joint Research Centre, Ispra, Italy (http://rem.jrc.cec.eu.int/). The comparison date was chosen to be 1 May 1986 at 12:00 UTC, since at this time the greatest number of measurements was available. Statistical measures were calculated following the recommendations of the Atmospheric Transport Model Evaluation Study (ATMES) final report (*Klug et al., 1992*).
The total amount and corresponding temporal development of the Cs-137 emission has been estimated (*Devell et al., 1995; De Cort et al., 1998; Persson et al., 1986*) and is associated with at least 50 % uncertainty. The current simulation considered the transport, dispersion and deposition of Cs-137 and employed vertically stratified point sources in order to simulate the explosions and the following fire.
The size distribution of particles containing Cs-137 is not known and here only mono-disperse particles with a radius of 0.5 µm and a density of 1.88 g/cm³ were considered. The model area corresponded to the G45 domain (Fig. 2). The start time was at 25 April 1986 at 18 UTC and the model was run two days ahead and then reinitialized and restarted until 7 May at 18:00 UTC. Surface analysis and 3DVAR upper air analysis was used as initial conditions for the meteorology at the beginning of each cycle and six hourly boundaries were post-processed from the IFS model.

Results and discussion

Considering the large uncertainty of the emission data, the uncertainty in the measurements, the mono-disperse nature of the simulation and the coarse resolution the model reproduce (spatially) most features of the deposition field satisfactorily (Fig. 4). This includes the peaks close to the accident site, in southern Finland, Switzerland, Austria and Italia. The band of increased activity extending from Southern Finland across Sweden and Norway is not well captured and is known to be caused by wet deposition. In the model the precipitation falls in a band further south causing the shift in the wet deposition pattern (Fig. 4). The simulation is associated with large global bias (Table 2) which may be due to the misplacement of this band and the rest of the statistical scores are satisfactory considering that no attempts of tuning the model has been performed.



(a) (b)

Figure 4 : (a) Simulated accumulated dry deposition (kBqm^{-2}),(b) Simulated accumulated wet deposition (kBqm^{-2}).

Table 2 : Global (containing both temporal and spatial variability) statistical scores for the simulation of Cs-137 deposition after the Chernobyl accident.

Global statistical parameter	Calculated value
Observed mean (kBqm^{-2})	19.97

Predicted mean (kBqm^{-2})	56.74
NMSE	6.34
Bias (kBqm^{-2})	38.77
Pearson's correlation	0.59
FMT (%)	26.29

4. On-line / Off-line Comparison

4.1. Variability

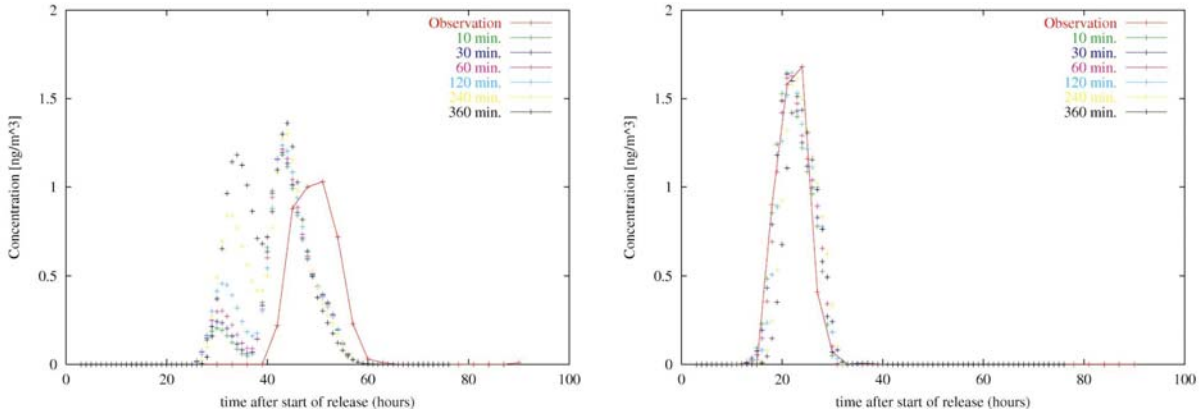
Offline models may not have the ability to temporally resolve the evolution of meso-scale disturbances which often have time scales well below the coupling interval (the time span between updates of meteorological fields for the offline model) even when sophisticated interpolation and diffusion procedures are employed to produce intermediate time steps. This may lead to errors in the simulation of tracer transport, dispersion, deposition, transformations and chemistry. The purpose of this experiment is to illustrate this difference between online and offline models.

For the current experiment a reference ETEX-1 simulation was run in online mode (using a 10 minute time step) while an identical set of experiments were run in offline mode using different coupling intervals of 30, 60, 120, 240 and 360 minutes. The results at a particular station, which is known to be influenced by meso-scale activity, were compared to measurements and conclusions regarding the effect of the coupling interval were drawn.

The set-up is identical to what is described in the evaluation of transport and dispersion section. The output has been interpolated to two ETEX-1 measurement stations, F15 and DK02, which were dominated by short and long range transport respectively.

Results and Discussion

At station F15 the measurements are dominated by a single peak which was captured well by the simulation (Fig. 5.1; run with a 10 minute coupling interval). The result was not sensitive to variations in the coupling interval of up to six hours. This suggests that the peak was generated by transport of the bulk of the plume over the site without any influence from short time scale disturbances (less than six hours), i.e the wind did not vary rapidly. At station DK02 the plume had traversed a region in which meso-scale disturbances are known to influence the dispersion (*Sørensen et al., 1998*). The predicted development of the concentration field had a phase error of a few hours on the arrival of the plume but was otherwise in good agreement with the observations (Fig. 5.1). A false (not in observations) peak preceding the plume existed and is indicative of meso-scale influences during plume development in the model. Notice that the first peak did not contribute to the statistical scores because the observations are zero. As the coupling interval was increased the main (second) peak remained unaffected while the amplitude of the first peak gradually increased. This suggests that the existence of the first peak is related to short time-scale disturbances in the forecasted meteorological fields, while the second peak is generated by transport of the bulk of the plume.



(a) (b)

Figure 5.1 : Measured and modelled time development of concentration (ngm^{-3}) at ETEX stations DK02 (a) and F15 (b) for coupling intervals 10 (online), 30, 60, 120, 240 and 360 minutes.

Meso-scale eddies are superposed on the mean flow generating cyclonic and anti-cyclonic perturbations in the plume. Eddies are visible as peaks in plots of relative vorticity (Fig. 5.2; eddies which influenced the plume development is marked with arrows). The eddies filled the boundary layer between the surface and at least 800 Hpa, persisted at least 15 hours, had maxima of +/- 6 s-1 respectively at the surface and tilted towards the NE and SW with height. 24 hours after the start of the release the plume maximum had split into two separate parts (Fig. 3). The head received cyclonic rotational momentum from a meso-scale disturbance and reached DK02 after 26 hours giving rise to the first peak. As the latter part of the cloud progressed it received anti-cyclonic momentum and after 36 hours the plume attained a U-shaped deformation which was advected towards DK02. The rotational time scale of the eddies was not large enough (compared to the advective time scale of the plume) to cause a full revolution in the plume

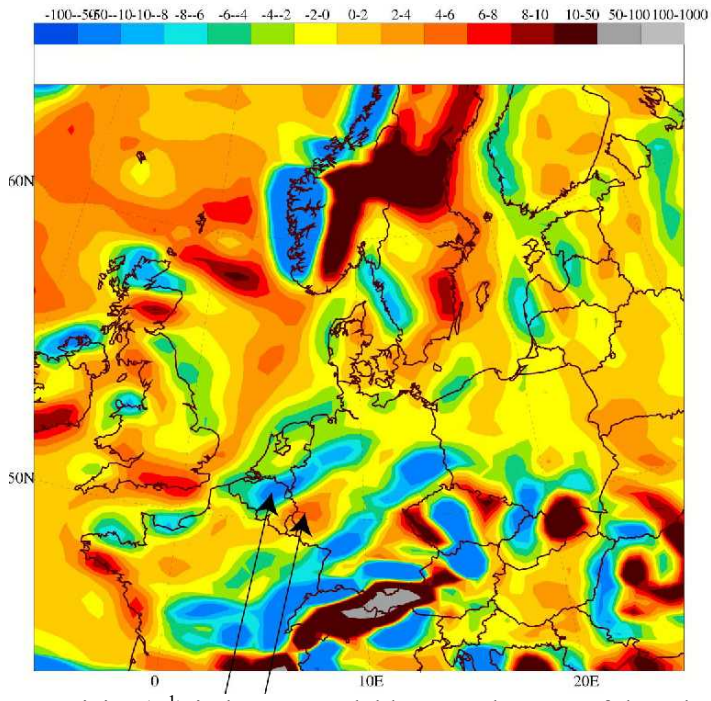


Figure 5.2 : Relative vorticity (s^{-1}) in lowest model layer at the start of the release. Arrows indicate eddies influencing the plume during transport.

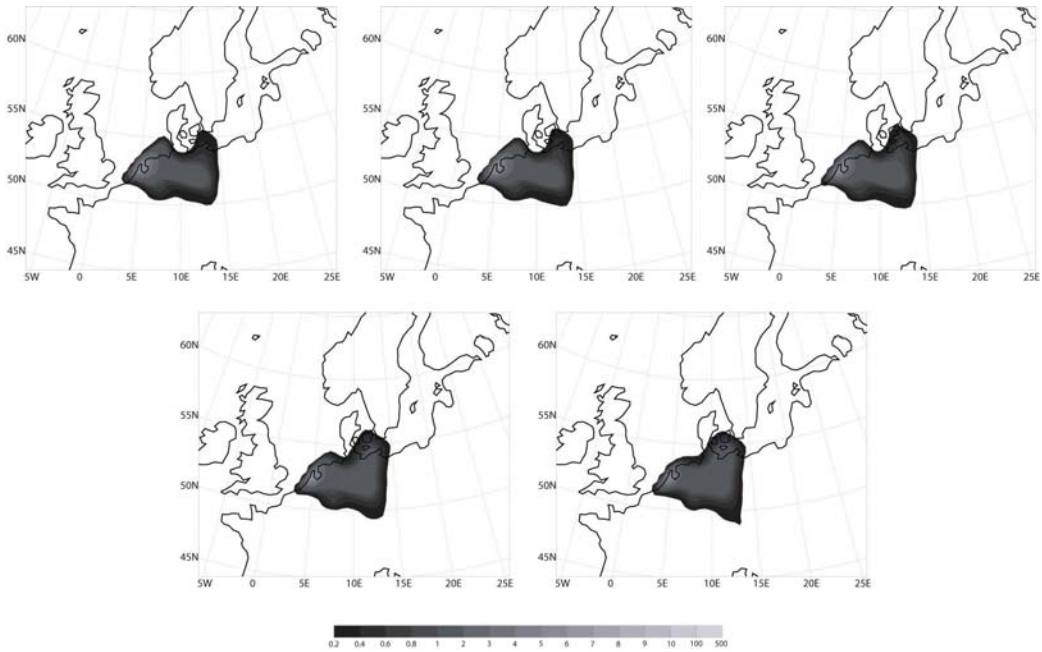


Figure 5.3 : Modelled concentration field (ngm^{-3}) 36 hours after start of release for a 30, 60, 120, 240 and 360 minute coupling interval respectively.

As the coupling interval was increased changes in the magnitude of eddies were not resolved and the U-shape extended further northwards leading to increased peak values in the concentration field at DK02 (Fig. 5.3). Increased temporal resolution constrained the evolution of the meso-scale disturbances, leading to better correspondence with measurements. Hence, even at coarse resolution it may be necessary to decrease the coupling interval in order to achieve correspondence with measurements at specific stations.
This experiment was conducted in a meteorological situation without strong dispersion and only horizontal effects were considered. However, previous studies have shown that very short coupling intervals are necessary to constrain vertical mixing processes (*Grell et al., 2004*). In general, the appropriate length of the coupling interval will depend on the application and the meso-scale activity. From these experiments it is not possible to give general recommendations along these lines; however, a coupling interval of three hours is not sufficient to constrain the development of the meso-scale disturbances.

4.2. Feedbacks

Both the direct effect, whereby aerosols redistributes, absorb and reemit incoming shortwave radiation and the first indirect effect, whereby the presence of aerosols in clouds cause a change in the droplet size distribution and thereby cloud albedo (making the cloud whiter) for fixed water content, have been implemented in the model. The purpose of this experiment is to demonstrate that feedbacks may induce important changes in the dispersion of tracers.
For the current experiment urban land-use fractions had been implemented in the model and the emission of sulphate particles from urban agglomerations was considered. The pollutant and meteorological fields were coupled via the first indirect effect and changes, between the reference (REF) simulation and the simulation including feedbacks (FED), were considered.
The simulation was conducted on the S05 domain (Fig. 2) covering northern Europe. Surface analysis and 3DVAR upper air analysis was conducted producing initial conditions for the simulations and hourly boundary files are post-processed using output from the operational model DMI-HIRLAM-T15 used for short range weather forecasting at DMI with a horizontal

resolution 0.15° x 0.15° (domain T15 in Fig. 2). The time-step was 2.5 minutes and runtime was 48 hours with analysis time 11 April 2007 at 06 UTC. This date was chosen due to the presence of a thick cloud over much of Northern Europe (Fig. 6.1). Urban fractions was extracted from the Global Land Cover Characteristics (GLCC) database (this information is normally ignored during climate file generation, i.e the model normally runs without any representation of urban areas) and concatenated in the first guess after surface analysis. The model had been modified to read the fractions from the first guess and keep this information in memory for use during emission. In these simulations the urban fractions were only used for emissions and no changes in albedo, orographical roughness or heat fluxes was considered. Time dependent emissions of industrial PM2.5 were extracted from the EMEP inventory and 20 % was assumed to be sulphate particles (*Pakkanen et al., 2001*). These were emitted from the urban areas.

In Enviro-HIRLAM cloud radiative properties are parameterized via an effective radius of cloud droplets: $R_{eff} = 3L/(4\pi\rho_l N)$ where L is the cloud condensate content, ρ_l is the density of liquid water and N is the number concentration of cloud droplets. Without any coupling between meteorology and pollutants N is fixed at the constant background values 10^8 m^{-3} over marine and $4\cdot10^8$ m^{-3} over continental regions. The sulphate particles in clouds give rise to perturbations in the background values: $\Delta N_{continental}=10^{8.06}\cdot C^{0.48}$ and $\Delta N_{marine}=10^{2.24}\cdot C^{0.26}$ where C is the concentration of the sulphate particles (here it is assumed that all particles act as cloud condensation nuclei) (*Boucher and Lohmann, 1995*), thereby decreasing the effective radius of the cloud droplet distribution.

Results and Discussion

The inclusion of the first indirect effect led to changes in dry deposition patterns, whereas the total amount remained constant, over the major polluted regions such as Ruhr (Northern Germany), greater London, mid England, greater Berlin and greater Dublin (Fig. 6.2). The domain-averaged dry deposition did not change between the two runs ($0.667\cdot10^5$ ngm^{-2}). Hence, the total amount of dry deposited material remained constant and the observed changes were due to a redistribution of the material. The largest changes, of approximately 7 %, occured over the Ruhr area, which is the most polluted region included in the experiment.

Wet deposition followed the precipitation patterns and changes in either precipitation or concentrations at any below-cloud level will lead to modifications in wet deposition. Hence, changes in stability, and therefore dispersion of the particles, due to the first indirect effect will be more apparent in this field (Fig. 6.2). Changes are found over the major polluted cities if precipitation is present. The largest change of approximately 7.5 % occurs over the Ruhr area in Northern Germany, where the concentration is largest. The domain-averaged amount of wet deposited material ($0.156\cdot10^6$ ngm^{-2}) was approximately constant (changes approximately 0.1 %) and the change in precipitation (of order 0.1 mm over 42 hours) is negligible. Therefore, the change in wet deposition is believed to be due to a redistribution of the particles.

Since dry deposition is directly related to the stability of the atmosphere, changes in dry deposition may arise if either the tracer distribution or the atmospheric stability varies. The daytime (12 to 18 UTC) average change in boundary layer height is of the order 50 meters above the polluted regions, while it is strongly reduced during night-time where particles are generally not mixed into the cloud environments (Fig. 6.3), indicating some influence on atmospheric stability.

Particles were transported to the Norwegian mountains and Finland in the top of the boundary layer at cloud level and therefore did not give rise to changes in dry deposition (but do contribute to wet deposition if the clouds precipitate (Fig. 6.2)). The thick cloud cover and

complex orography gave rise to large (100 meters) changes in boundary layer height. These changes were also strongly reduced during night, suggesting that they are not the result of nonlinear dynamics. In order to further exclude short-term nonlinear dynamics as the course of the changes in boundary layer height, averages were taken over the entire period (not including a six hour spin-up). This did not result in any changes in the conclusions.
The strength of the effect is related to the amount of cloud cover and the concentration of sulphate particles. The cloud cover over the Ruhr area is quite thin (Fig. 6.1), but the concentration of particles is extremely high (Fig. 6.2), leading to large changes in dry deposition patterns in this region. Hence, even with a thin cloud cover this effect may be of importance for dry deposition.

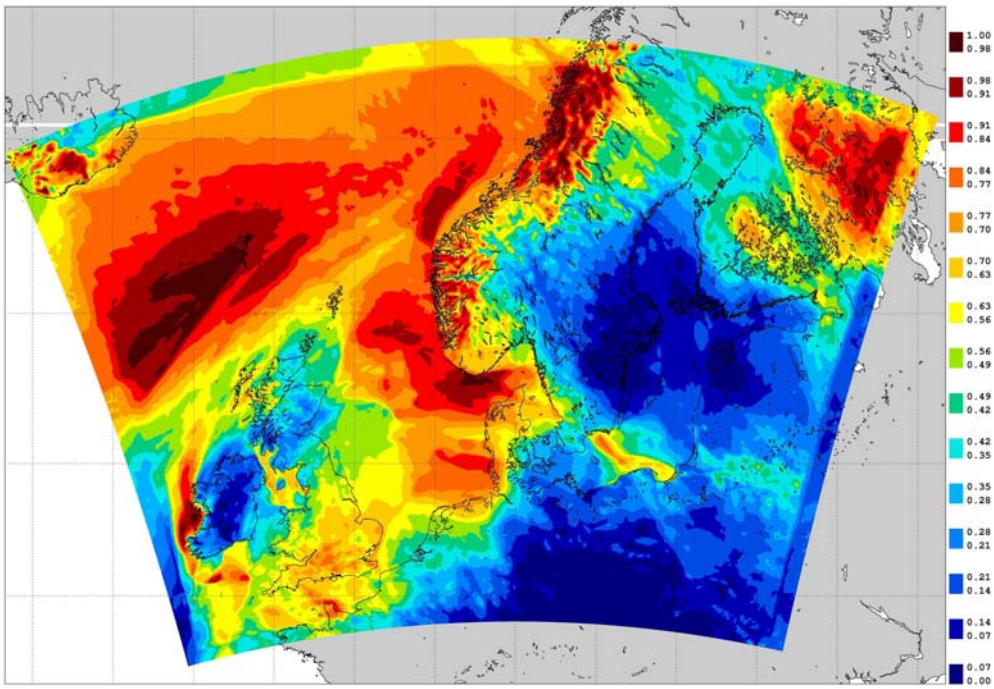


Figure 6.1 : Total cloud cover (%) time averaged over simulation period.

5. Conclusions

The online coupled model system, Enviro-HIRLAM, which integrates a meteorological model and an atmospheric chemical transport model and includes feedbacks between air pollution and meteorological processes, has been described and evaluated.

- Enviro-HIRLAM performs satisfactorily with regards to transport and dispersion when simulating the ETEX-1 controlled release.
- Enviro-HIRLAM performs satisfactorily with regards to deposition when simulating the Chernobyl accident.
- In situations with meso-scale activity the coupling interval, in offline models, is important in constraining meso-scale influences on plume development. Online coupling improves the results.
- Feedbacks through the first indirect effect lead to modifications of the order 7 % in dry and wet deposition patterns over major polluted areas in Europe. A consistent explanation is suggested; the first indirect effect affects the dispersion of pollutants through regulation of atmospheric stability, thereby leading to a redistribution of the pollutant.

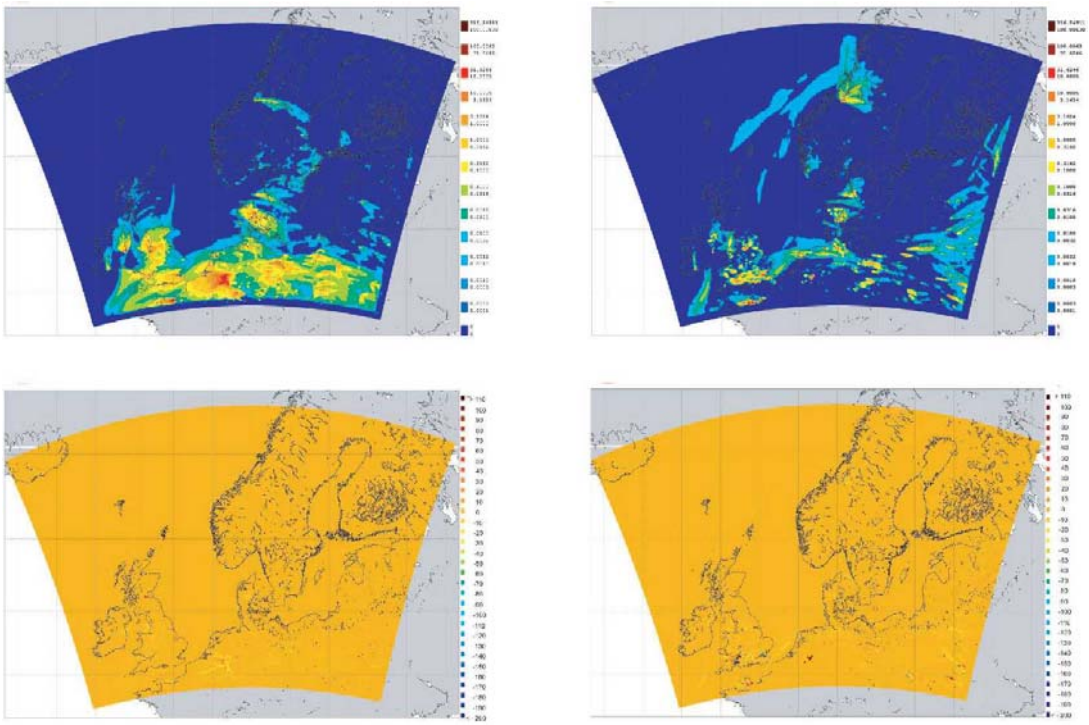


Figure 6.2 : Top panel: Dry (left) and wet (right) deposition (ngm^{-2}) at the end of simulation period. Bottom panel: changes (perturbation - reference) in dry (left) and wet (right) deposition (ngm^{-2}).

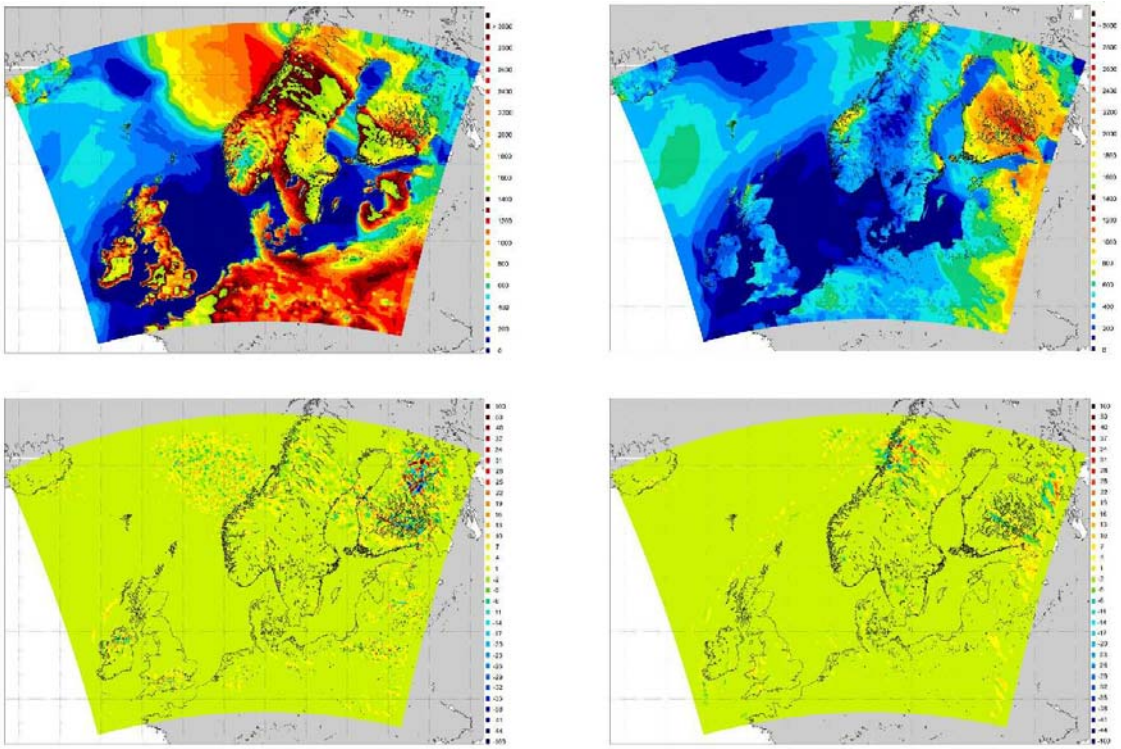


Figure 6.3 : Top panel: Reference day (12-18 UTC) (left) and night (right) averaged boundary layer height (meters). Bottom, panel: day (left) and night-time (right) averaged change (perturbation - reference) in boundary layer height.

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1.5 COSMO-ART – Aerosols and Reactive Trace Gases within the COSMO model: *Heike Vogel, D. Bäumer, M. Bangert, K. Lundgren, R. Rinke, T. Stanelle, B. Vogel*

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Abstract
We developed a fully on-line coupled model system composed of a numerical weather forecast model and a chemical transport model. With this model system we want to quantify feedback processes between aerosols and the state of the atmosphere and the interaction between trace gases and aerosols on the regional scale.

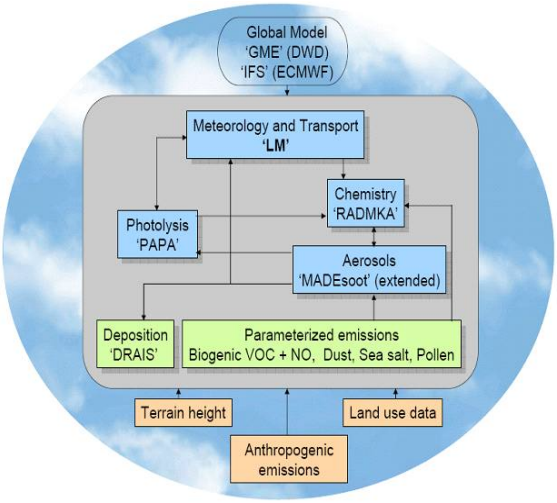
1. Introduction

Atmospheric aerosol particles modify the radiative transfer in the atmosphere and they have an impact on the cloud formation. Therefore, they alter the weather and they have an impact on climate. The anthropogenic part of this modification of the state of the atmosphere is currently not well understood and it raises the largest uncertainties with respect to climate change (see the IPCC report 2007).We developed a new on-line model system to investigate the aerosol-radiation-interaction on the regional scale.

2. Method

Based on the mesoscale model system KAMM/DRAIS/MADEsoot/dust (*Riemer et al., 2004, Vogel et al., 2006*) we developed an enhanced model system to simulate spatial and temporal distribution of reactive gaseous and particulate matter. The meteorological driver of the old model system (KAMM) was replaced by the operational weather forecast model COSMO model (= former **L**okal **M**odell, *Steppeler et al., 2003*) of the German Weather Service (DWD). The name of the new model system is COSMO-ART **(**ART stands for **A**erosols and **R**eactive **T**race gases; *Vogel et al., 2006*). The atmospheric chemistry transport model (ACTM) module was on-line coupled with the operational version of the COSMO model. That means that in addition to the transport of a non-reactive tracer the dispersion of chemical reactive species and aerosols can be calculated. Secondary aerosols which are formed from the gas phase, directly emitted components like soot, mineral dust, sea salt, and biological material are represented by log normal distributions. Processes such as coagulation, condensation, and sedimentation are also taken into account. The emissions of biogenic VOCs, dust particles, sea salt, and pollen are calculated also on-line, taking into account the dependencies on the meteorological variables. To calculate efficiently the photolysis frequencies a new method was developed using the GRAALS radiation scheme (*Geleyn and*

Ritter, 1992) which is already implemented in LM. With this model system we want to quantify feedback processes between aerosols and the state of the atmosphere and the interaction between trace gases and aerosols on the regional scale. To enable fully coupled model runs, the aerosol optical properties have been parameterized (*Bäumer et al., 2004*) since on-line Mie computations were too time-consuming.



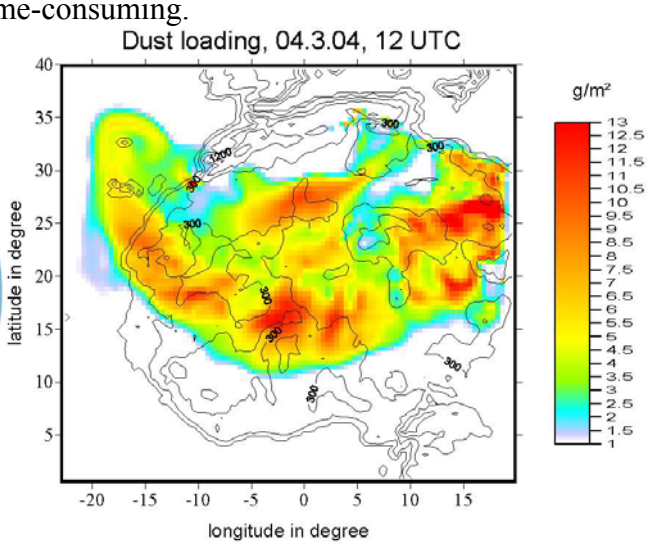


Figure 1 : The model system COSMO-ART. **Figure 2** : Horizontal distribution of the simulated dust loading over western Africa.

In the parametrization that is based on off-line Mie calculations, the aerosol optical properties for the eight spectral bands of the LM radiation scheme are calculated separately for the five modes of the aerosol model as a function of dry mass density, water content and soot content in each mode. For the simulations, the climatologically aerosol optical properties, which are used in the standard LM version, are replaced by these parameterized ones that take into account current modal aerosol mass densities. By comparing different simulation results obtained with parameterized and climatological aerosol optical properties, the impact of the aerosol can be quantified not only on the radiation, but also on other meteorological variables such as temperature. The model system can be embedded by one way nesting into individual global scale models as the GME model (Global model of the DWD) or the ECMWF model. Figure 1 gives an overview of new model system.

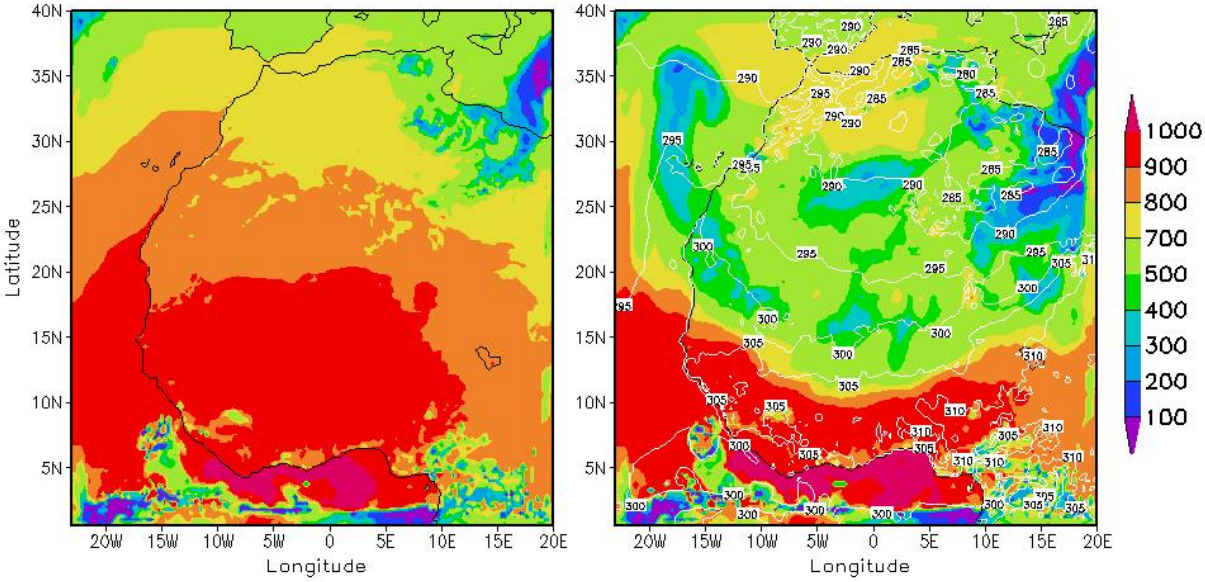
3. Results

In the following two case studies where the model was used to quantify the impact of natural and anthropogenic aerosol particles on the regional weather will be explained.

3.1. The Interaction of Mineral Dust with Radiation

The first application is the simulation of a mineral dust event over West Africa in March 2004. During this event there were high wind speeds and low temperature observed in the Sahara and heavy precipitations over Libya (*Knipperts and Fink, 2006*). Figure 2 shows the simulated dust loading for 4th March 2004 at 12 UTC. To investigate the impact of the dust aerosols on radiation two simulations were performed; one with no interaction between the actual aerosol concentration and radiation, and another that takes into account the interaction.
Figure 3 shows the results for the shortwave radiation balance at the surface for both cases. The high dust load of the atmosphere leads to a strong modification of the shortwave radiation balance. In contrast to the simulation without interaction between aerosols and radiation the

simulation using the actual values of the dust concentrations in the radiation scheme shows a reduced shortwave radiation balance up to a factor of 2 over western Africa, which has also an effect on the cloud formation and the dynamics (not shown here).



(a) (b)

Figure 3 : Horizontal distribution of the according shortwave radiation balance for the simulation taken into account the actual dust concentration (a) and the simulation using the climatological values (b), both for 04.03.2004, 12 UTC.

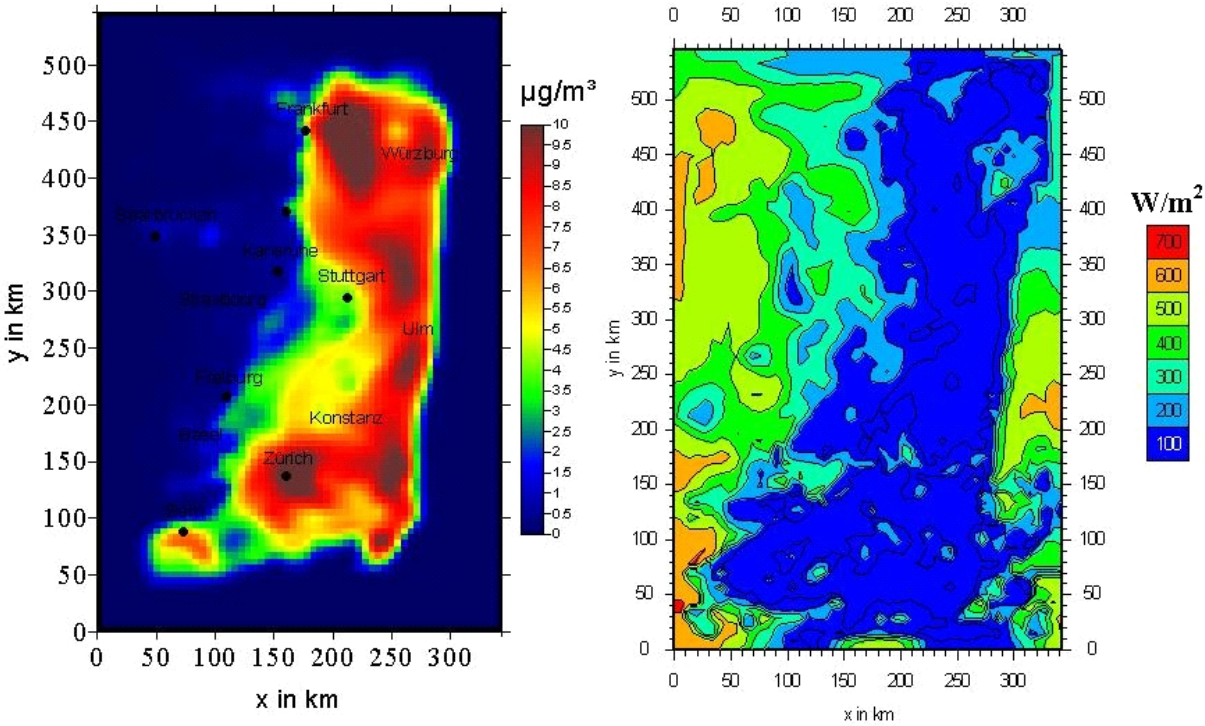
3.2. The Interaction of Anthropogenic Aerosols with Radiation

The model system was also applied to study the direct effect of anthropogenic aerosols on radiation. In addition the ageing process of the emitted soot particles was taken into account. The model domain for this study covered the south western part of Germany with adjacent areas. The simulation period was from 16.08.2005 to 22.08.2005. The emission data were available for this period with a temporal resolution of 1 h and a horizontal resolution of 7 km. As in the previous section, two model runs were carried out; one with the interaction of the actual aerosol concentration and the radiation, and one without. Figures 4-5 show results of these simulations. In contrast to the case study of the dust event the aerosol concentration is rather low. Nevertheless, the influence on the shortwave radiation balance is quite large due to the effect that the cloud formation is also influenced by the modifications in the radiation field caused by the aerosols. Although the aerosol concentration is much lower in the western part of the domain than in the eastern part (Fig. 4a), an effect on the shortwave radiation balance can be seen all over the model domain (Fig. 5a). In the less cloudy western part, there is a cooling effect of several tenth degrees dominating, whereas in the western part both areas with warming and cooling effects can be seen (Fig. 5b)

4. Conclusions

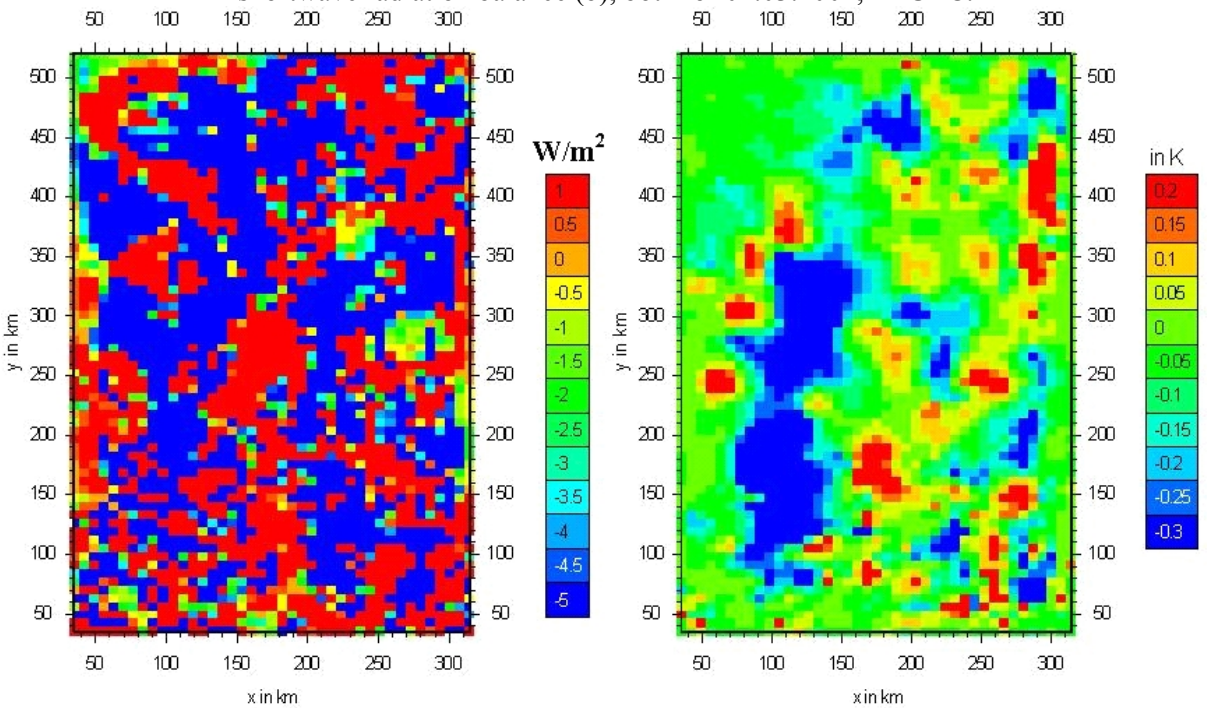
A new on-line coupled model system named COSMO LM-ART was developed. This model system contains, for example, a variety of natural and anthropogenic aerosols. The ageing process of soot is explicitly described. The treatment of their impact on the atmospheric radiation allows the quantification of feedback mechanisms. The simulation of a dust event occurring over West Africa gives rather high aerosol concentrations and consequently a strong effect on the shortwave radiation balance which leads also to differences in the cloud formation and the dynamics. The study concerning the anthropogenic aerosol-radiation-

interaction shows that despite of rather low aerosol concentrations the modification of the radiation balance causes a surprisingly strong effect on the cloud pattern that needs further investigations.



(a) (b)

Figure 4 : Horizontal distribution of the dry aerosol mass (a; at 20 m above ground) and the according shortwave radiation balance (b), both for 04.03.2004, 12 UTC.



(a) (b)

Figure 5 : Difference of the shortwave radiation balance (a) and the temperature (b) between the model run with aerosol-radiation interaction and the run without aerosol-radiation interaction.

Acknowledgements
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1.6 The On-Line Coupled Mesoscale Climate-Chemistry Model MCCM - a Modelling Tool for Short Episodes as well as for Climate Periods: *Peter Suppan, R. Forkel, E. Haas*

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Abstract
The on-line coupled regional meteorology-chemistry model MCCM (Mesoscale climate chemistry model, *Grell et al., 2000*), which is based on the non hydrostatic NCAR/Penn State University mesoscale model MM5, is presented. Recent applications of MCCM include short-term studies such as simulations of high pollution episodes for Mexico City, receptor analysis, as well as long term studies such as climate-chemistry simulations, simulations of yearly pollution conditions in the Alpine region, and daily real time forecasts of ozone and particulate matter for Germany

1. Introduction

Although on-line coupled models where meteorological and atmospheric chemistry processes are computed within one single model exist already since the 1990s and even earlear, off-line air quality models (where the chemical processes are treated independently of the meteorological model) are still widely used because of lower computational costs. However, due to this separation of meteorology and chemistry there can be a loss of possibly important information of atmospheric processes, as the meteorological information is transferred to the Atmospheric Chemistry Transport Model (ACTM) e.g. once or twice per hour. The simulation of atmospheric chemistry with an on-line coupled model can be regarded as more consistent than an offline treatment, as the chemistry part of the model receives all necessary meteorological information directly from the meteorological part of the model at each time step without any temporal interpolation. Especially on the regional scale with grid sizes down

to 1 km, the wind field and other meteorological parameters are highly variable and neglecting these variances may introduce certain errors. Although the advantages of on-line coupled meteorology-chemistry simulations against an off-line treatment are most effective for fine horizontal resolutions, effects already become significant at horizontal resolutions of around 30 km (*Grell et al., 2004*).

2. Description of MCCM

The on-line coupled regional meteorology-chemistry model MCCM (Mesoscale climate chemistry model, *Grell et al., 2000*) has been developed at the IMK-IFU on the basis of the non hydrostatic NCAR/Penn State University mesoscale model MM5 (*Grell et al., 1994*). The full coupling of meteorology and chemistry ensures that the air quality component of MCCM is fully consistent with the meteorological component. Both components use the same transport scheme, grid, and physics schemes for subgrid-scale transport. Similar to MM5 the MCCM model can be applied over a range of spatial scale from the regional (several thousand kilometers, resolution of 30-100 km) to the urban (100-200 km, resolution of 1-5 km) scales.
MCCM includes several tropospheric gas phase chemistry modules (RADM, RACM, RACM-MIM (*Stockwell et al., 1990, 1997; Geiger et al., 2003*)) and a photolysis module. Optional aerosol processes are described with the modal MADE/SORGAM aerosol module (*Schell et al., 2001*) which considers as single compounds sulphate, nitrate, ammonium, water, and four organic compounds. For the Aitken and the accumulation modes the gas/particle phase partitioning of the secondary sulphate/nitrate/ammonium/water aerosol compounds is based on equilibrium thermodynamics. The organic chemistry assumes that secondary organic aerosol compounds (SOA) interact with the gas phase and form a quasi-ideal solution.

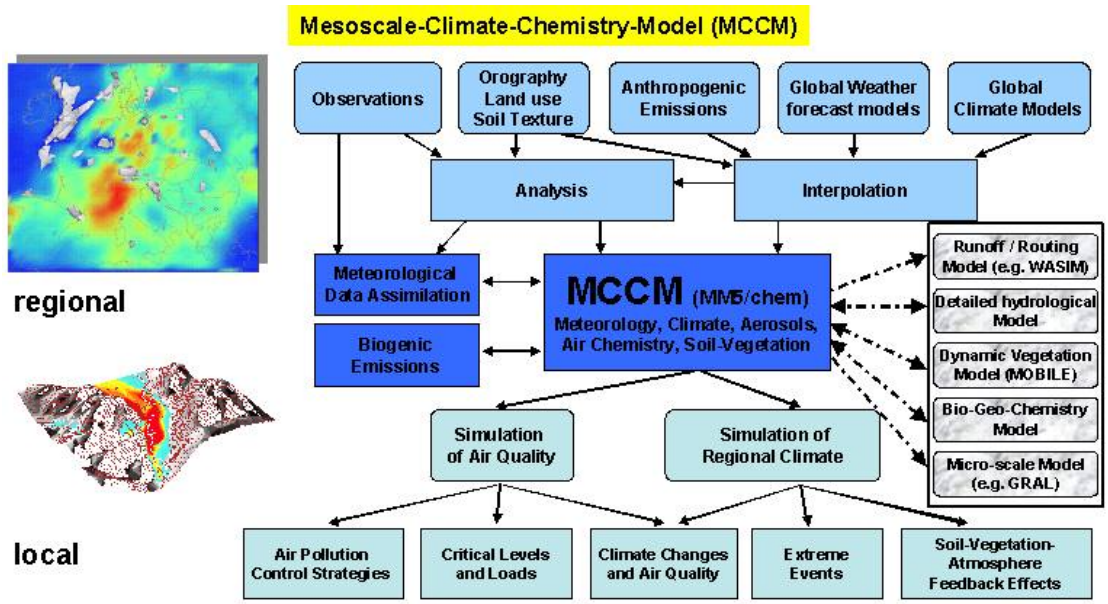


Figure 1: Schematic description of MCCM as used on the local and regional scales for air quality simulations, and for simulation of the regional climate including the introduced coupled models to hydrology and biosphere compartments.

Biogenic VOC and NO emissions are calculated on-line based on land use data, simulated surface temperature and radiation. Anthropogenic emissions of primary pollutants, like NO_x, SO_2, and hydrocarbons, as well as emissions of primary particulate matter have to be supplied

either at hourly intervals or as yearly data from gridded emission inventories. Validation studies with MCCM have shown its ability to reproduce observed meteorological quantities and pollutant concentrations for different conditions and regions of the Earth (*Forkel and Knoche, 2006; Forkel et al., 2004; Grell et al., 1998; Grell et al., 2000; Jazcilevich et al., 2003; Kim and Stockwell, 2007; Suppan and Skouloudis, 2003, Suppan and Schädler, 2004; Suppan, 2007*).

Furthermore, the model is linked to other models like hydrological or/and biosphere based models in order to describe the interactions and feedback mechanisms from each compartment to the air quality and vice versa. A schematic description of the model is given in Fig. 1.

The following examples show different applications of MCCM for short period simulations on air quality and emission reductions scenarios as well as simulations performed for the assessment of the impact from climate change to the regional air quality.

3. Applications

3.1. Evaluation Studies

In order to evaluate the performance of the three chemistry mechanisms included in MCCM, the results of simulations over a two month period in summer 2003 were compared with observations.

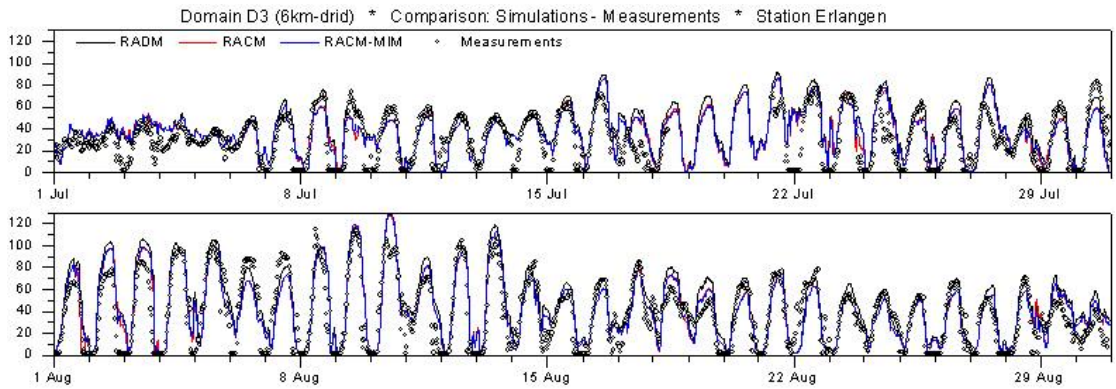


Figure 2: Calculated and measured time series of ozone at the Erlangen station for 3 different chemical schemes.

The mechanisms were constructed using a software engineering tool for chemistry kinetics (*Damian et al., 2002*). By using a pre-processor for the construction of the numerical integration code, the solver (numerical integration scheme) and the underlying mechanisms were decoupled and by using identical integrations schemes, the influence of the numerics to the species evolution is comparable. The simulations were performed with four nested domains (54, 18, 6 and 2 km grid resolution). They cover the time period from 1st July to 31st August 2003, and include episodes with high photosmog concentrations. The simulated species mixing ratios were compared with observations from the Bavarian measurement network (LFU). The evolution of the time series of the measured (LFU-Station Erlangen) and the simulated ozone (at 6 km resolution) are shown in Figure 2. As seen, this figure shows a good correlation between measurements and simulations. But on the other side, it is also seen that the RADM mechanism overpredicted ozone concentrations during the day, while the RACM and RACM-MIM simulation results fit better to the observations (*Haas et al., 2007*).

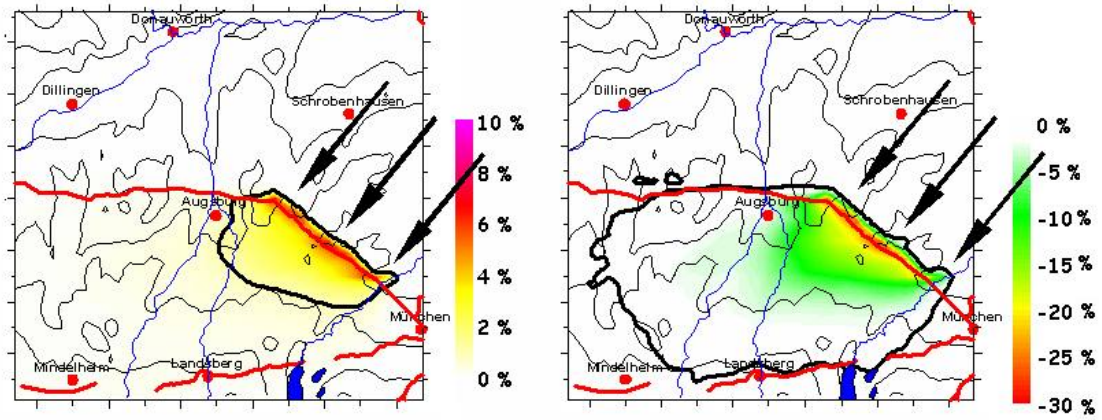
3.2. Air Quality Studies

Effect of highway emissions

A typical application for assessing the influence of specific emission sources to the air quality is demonstrated within the next example. To assess the influence of highway emissions to the ozone and nitrogen dioxide concentration fields, the line source (highway, ca. 35 km) between Munich and Augsburg was excluded and the emissions were set to the surrounding levels. This could be attributed to a reduction of 80% of the traffic emissions.

As seen in Figure 3, the increase of O_3 concentrations was up to 10%, whereas the NO_2 concentrations showed a decrease of 25%. The red line marks the region of influence of this specific emission reduction.

Compared to the NO_2-influenced region, O_3 has a smaller impact on the region (less than 800 km^2) but the impact is lasting longer (more than 12 h). The NO_2 influenced regions (close to 1000 km^2) are larger, but they existed for a shorter period (less than 6 h). The main time interval of influence for both NO and O_3 occurs during the night (*Suppan and Schädler, 2004*).



(a) (b)

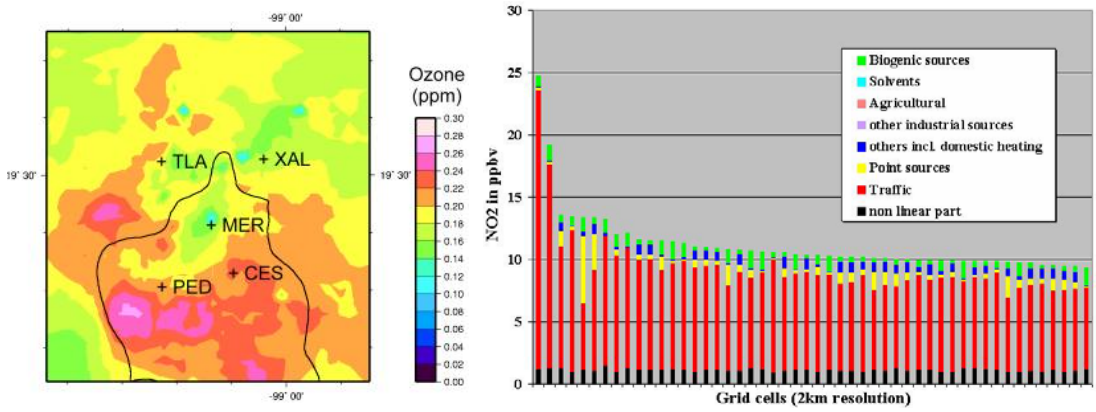
Figure 3: Traffic emission effect to O_3 (a) and NO_2 (b) concentrations at the nearby region of a highway during a 4 day period with strong north-easterly winds. The thick black line indicates the region of influence.

Scenario simulations for Mexico City

Mexico City suffers from severe air quality problems with maximum ozone values up to 250 ppb. In order to study the effect of different precursor emissions and possible mitigation measures on the ozone levels in Mexico City the MCCM was applied in the Greater Area of the city (*Jazcilevich et al., 2003; Forkel et al., 2004*). Figure 4a shows that, in agreement with observations, the simulated O_3 maxima occur in the southwest of the city (measurement station PED), which is down-wind of the city centre, as an uphill flow is prevailing during the afternoon. The minimum ozone concentrations are found in the centre of the city (station MER), where the NO emitted by traffic titrates the ozone, and in the northern part of the city at places where the NO emissions from industry and power plants locally reduce the ozone concentrations (near station XAL).

Compared to the ozone concentrations predicted for the 2010 baseline emissions, the emissions for a mitigation scenario including the replacement of old private cars, low sulphur diesel standards, the replacement of microbuses, and the relocation of two power plants (corresponding to a reduction of anthropogenic emissions of NO_x by about 20% and of VOC by 10%) result in a decrease of the daytime ozone concentrations between 5 and 25 ppb at most locations. However, in the centre of the city and for the locations where power plants are switched off, the noontime ozone concentrations are higher for the mitigation scenario than

for the baseline case since less ozone is titrated in case of the reduced NO emissions at these locations. The model results indicate that taken the projected emissions for 2010, extremely strong emission reduction measures for Mexico City would be necessary in order to significantly improve the air quality in the city



(a) (b)

Figure 4a: Average surface ozone concentration (in ppm) over 8 days at 16:00. Mexican summer time for the baseline 2010 emission scenario. **Figure 4b**: Source-receptor relationship for the 50 grid cells with highest NO_2-concentrations within the conurbation of Munich during a 4 day period.

Source receptor analysis

Source-receptor analysis was performed to allocate the air chemistry parameters to the individual emission sources. To estimate the impact of a source group on a certain pollutant, several simulations have to be accomplished. To minimize the associated uncertainties (non-linearity of chemical processes), the source group was suppressed. Due to the non-linear chemical processes, background concentrations and advection a "non-linear" fraction has to be introduced (*DG-ENV, 2001*). The source-receptor analysis is an important tool for abatement and emission reduction strategies.

In Figure 4b the source-receptor distribution for NO_2 is shown for a short time period in the greater area of Munich/Germany. The concentration at each grid cell includes several source categories. In accordance with the NO_x emission distribution the NO_2 concentrations caused by the traffic show also the highest values (*Suppan, 2007*).

Climate chemistry simulations

In order to investigate possible effects of global climate change on the near-surface concentrations of photochemical compounds in southern Germany, nested regional simulations with MCCM were carried out (*Forkel and Knoche, 2006*). The simulations with horizontal resolutions of 60 (for whole Europe) and 20 km (for central Europe) were driven by meteorological boundary conditions provided by a long-term simulation of the global climate model ECHAM4. Two time slices (representing 1990s and 2030s) of about 10 years each were compared.

For the region of southern Germany the simulations show an increase of the mean summer temperature by almost 2°C along with a decrease of cloud water and ice and a corresponding increase of the photolysis frequencies and the emissions of biogenic hydrocarbons. Under the model assumption of unchanged anthropogenic emissions this leads to an increase of the mean mixing ratios of most photooxidants. Because of the complex topography and the heterogeneous distribution of precursor emissions all parameters show pronounced regional patterns. The average daily maximum ozone concentrations in southern Germany increase for

the selected scenario by nearly 10% in summer months. Depending on the region the increase of the mean daily maximum ranges between 2 and 6 ppb. As a consequence, the number of days when the 8-hour mean of the ozone concentration exceeds the threshold value of 120 µg/m³ increases by 5 to 12 days per year (Fig. 5).

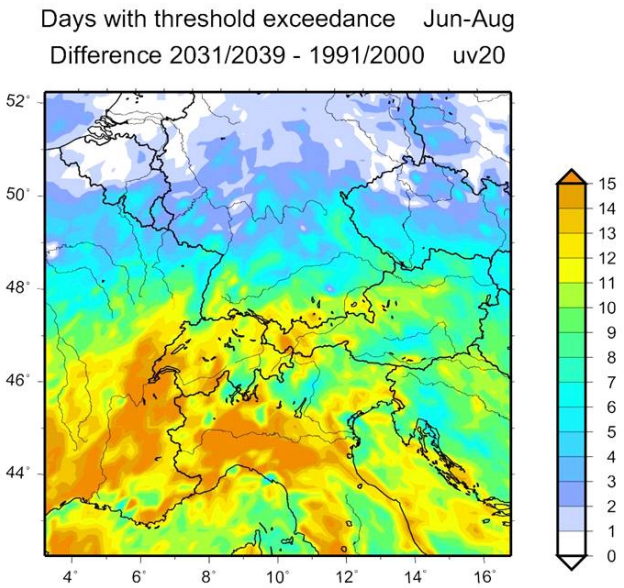


Figure 5: Effect of climate change on the exceedance of the ozone threshold value.

4. Conclusions

The on-line coupled model MCCM (which is based on well known and validated model MM5) has demonstrated its applicability to support and to address air quality issues, like emission reduction scenarios, abatement strategies, or the impact of climate change to the air quality in sensitive and urbanized areas both on the regional and local scales. Furthermore, this model is able to couple or to link also other models from different compartments like hydrology or biosphere.

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1.7 BOLCHEM, an Integrated System for Atmospheric Dynamics and Composition: *Alberto Maurizi, M. D'Isidoro and M. Mircea*

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Abstract
The on-line modelling system BOLCHEM consists of a meteorological limited area hydrostatic model (BOLAM) coupled with different gas chemistry models (SAPRC90 and CB-IV chemical mechanisms), an aerosol model (M7, work in progress) and a Lagrangian dispersion model for computation of forward and backward trajectories. The model is under development, but recent applications have shown its reliability for air quality studies. Future developments will focus on data assimilation of atmospheric composition and on feedback effects of composition on radiation and dynamics.

1. Introduction

Chemical composition at regional scale is a subject of increasing interest for both air quality and climatological issues. Several models exist, but no on-line Italian model was available some years ago. Different expertises are presented at ISAC-CNR on several aspects involved in air quality modelling: atmospheric chemistry, meteorology, microphysics and turbulence, and these were gathered to develop BOLCHEM. This model is a part of the European community project GEMS, sub-project Regional Air Quality models (RAQ), and it has been used to study different aspects of air quality related problems.

2. Model Description

The BOLCHEM model (BOLam + CHEMistry) is the result of an on-line coupling between the mesoscale meteorological model BOLAM (BOlogna Limited Area Model, http://www.isac.cnr.it/~dinamica/bolam/index.html) (*Buzzi et al., 1994; Buzzi et al., 2003*) and modules for transport and transformation of chemical species. BOLAM dynamics is based on hydrostatic primitive equations, with wind components, potential temperature, specific humidity, surface pressure, as dependent variables. The vertical coordinate system is hybrid-terrain-following, with variables distributed on a non-uniformly spaced staggered Lorenz grid. The horizontal discretisation uses geographical coordinates on the Arakawa C-grid. The time scheme is split-explicit, forward-backward for gravity modes. A 3D WAF (Weighted Average Flux) advection scheme coupled with semi-Lagrangian advection of hydrometeors is implemented. A 4th order horizontal diffusion of the prognostic variables (except for Ps), a 2nd divergence diffusion and damping of the external gravity mode are included. The lateral boundary conditions are imposed using a relaxation scheme that minimizes wave energy reflection. The initial and lateral boundary conditions are supplied from the ECMWF (European Centre for Medium-range Weather Forecasts) analyses available at 0.5° x 0.5° horizontal resolution. Hybrid model level data are directly interpolated on the BOLAM grid.
Transport (advection and diffusion) of tracers (both passive and reactive) is performed on-line at each meteorological time-step using WAF scheme for advection and a "true" (second order) diffusion, with diffusion coefficient carefully estimated from experiments (*Tampieri and Maurizi, In Review*). Vertical diffusion is performed using 1D diffusion equation with a diffusion coefficient estimated by means of an *k-l* turbulence closure scheme. Dry deposition is computed through the resistance-analogy scheme and is provided as a boundary condition to the vertical diffusion equation. Furthermore, vertical redistribution of tracers due to moist convection is parameterized consistently with the Kain-Frisch scheme used in the meteorological part for moist convection. Transport of chemical species is performed in mass units while gas chemistry is computed in ppm.
Physical/chemical processes are treated separately for the gas phase, aerosol classes, and generic tracers (e.g. radioactive species, Saharan dust, etc.). Gas phase is treated using the SAPRC90 or CB4 chemical mechanisms. Aerosols are modeled using M7 module from ECHAM5 (coupling is still in progress) and generic species are defined by the user case by case providing chemical/physical properties and equations. More technical details can be found in the COST-728/732 model inventory: http://www.mi.uni-hamburg.de/List_classification_and_detail_view_of_model_entr.567.0.html?&user_cost728_pi2 [showUid]=80.

3. Model Applications

The model has been used for a variety of situations in order to test the reliability of the choices made. It also currently runs at ECMWF in the frame of the GEMS Project for the ensemble near-real-time experiment. Some of the main results are briefly reported in the following sections.

3.1. Evaluation of Model Performances for Ozone

The performances of BOLCHEM on the ability to predict O_3 concentration over Italy were evaluated. The comparison between computed and measured concentrations for some periods

of 1999 showed that the model is capable to predict the diurnal cycle of O_3, in particular in summer. The agreement between modeled and measured quantities is good during daytime while at night there is some problem connected to O_3 destruction. However, US-EPA's criteria are met; so, that model results can be reliably used for air quality predictions. Some time series of O_3 computed with CBIV and SAPRC90 mechanisms compared with measurements are shown in Figure 1.

3.2. Ozone Sensitivity to Precursor Emission Reduction

An important aspect for emission reduction policies is the study of the regional sensitivity to precursor emission reduction. For this purpose indicator species are computed to asses reduction in NOx and VOC over the whole Italy and on subdomains centered on specific spots: the Milan and Rome areas and some of the major industrial areas. Different periods were selected, and runs with both chemical schemes were performed over Italy comparing indicator species computed for different reduction scenarios. For all the periods investigated, it was found that Italy, including the large islands Sicily and Sardinia, is mostly dominated by NOx chemical regimes, independently of the photochemical mechanism used. However, the effect of the NOx reduction predicted with the CB-IV mechanism is lower than that predicted with SAPRC90 mechanism. In addition, the urban areas of cities Milan, Rome, Naples or/and industrial areas around harbors of Genoa, Messina, and Venice are always in a marked VOC sensitive regime. Note that differences in the spatial distribution of the chemical regimes due to the photochemical mechanism used and due to the meteorological conditions are comparable. Examples of ozone sensitivity maps to reduction of VOC and NOx are presented in Figure 2.

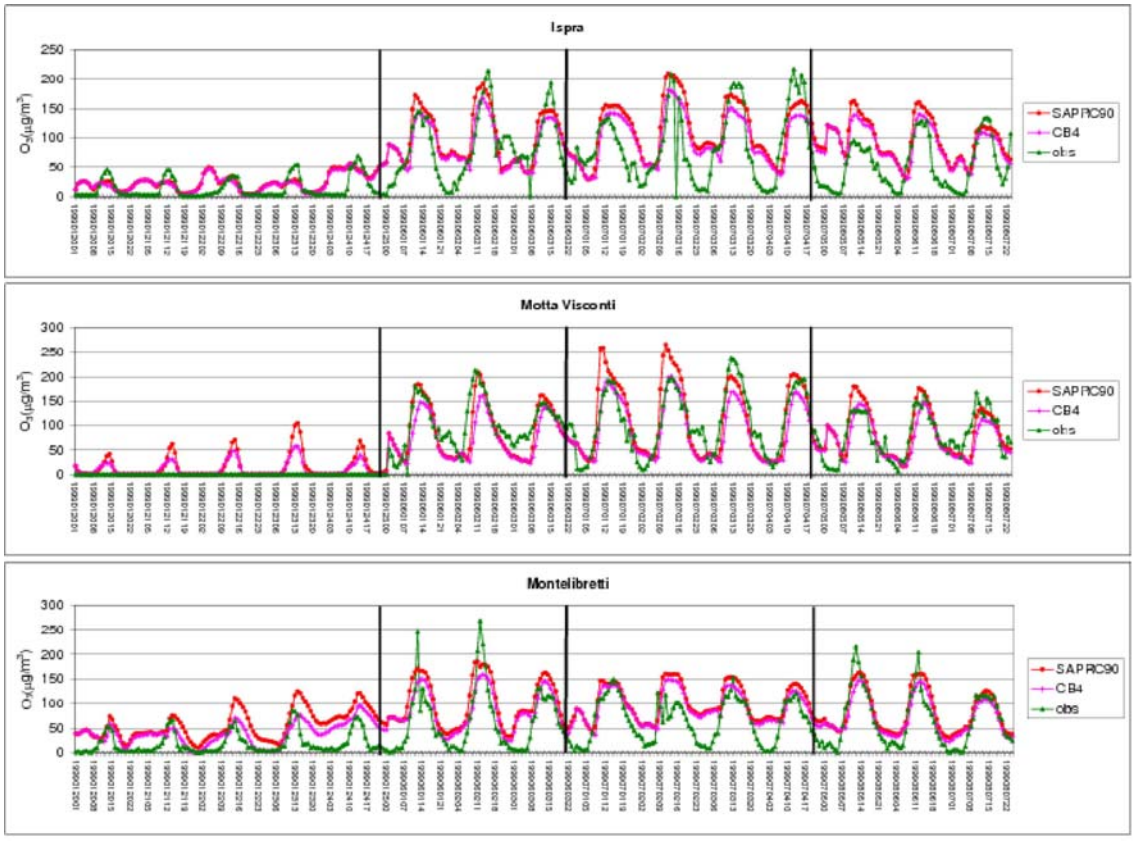
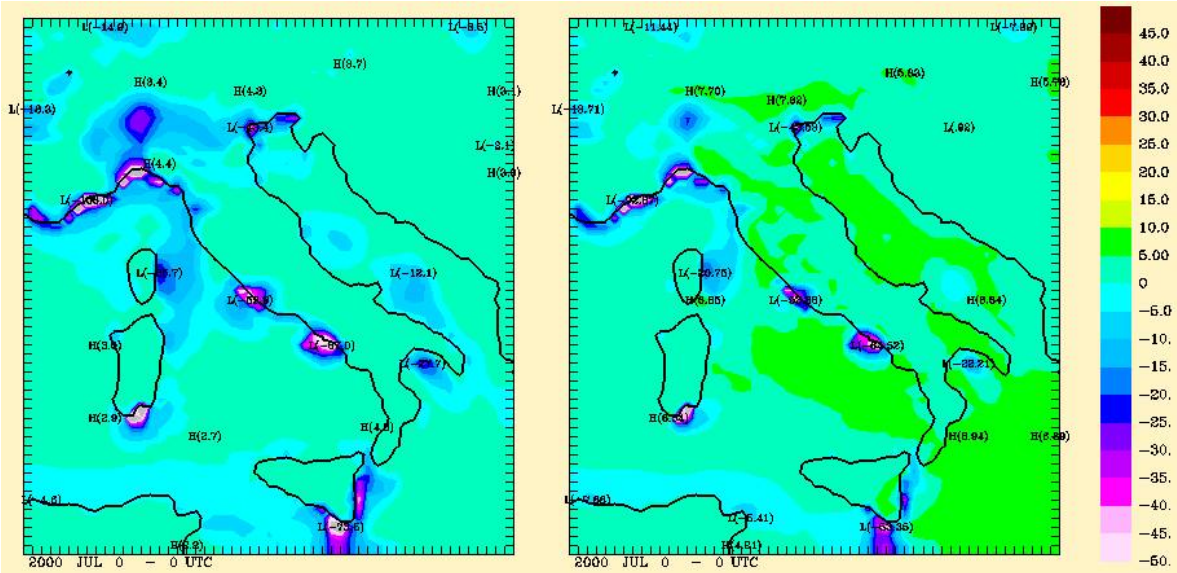


Figure 1 : Time series for ozone at three Italian locations: Ispra, Motta Visconti (MI) and Montelibretti (RM) for four months: January, June, July and August.

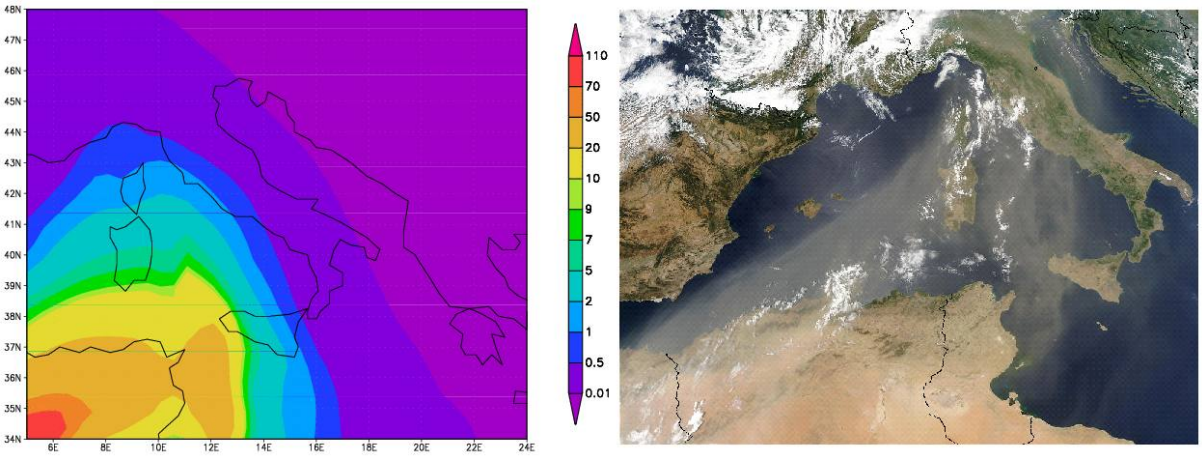


(a) (b)

Figure 2 : Maps of sensitivity of ozone to reduction of VOC (positive) and NOx (negative).

3.3. Saharan Dust Transport

Saharan dust is a major component of the aerosol load over Italy due to vicinity to the African continent. The direct forcing of dust aerosol may be comparable to or even exceed the forcing of anthropogenic aerosols. To correctly treat this aspect, a proper modelling of dust blowing sources is needed. A sensitivity experiment was carried out to test the sensitivity of the emission model (*Tegen et al., 2002*) to the friction velocity threshold during a strong Saharan dust outbreak that occurred from 15 to 19 July 2003, transporting the dust particles almost over the whole Italy (Figure 3). The comparison of model results with the observations (surface concentrations from EMEP stations and aerosol optical depth (AOD) from AERONET stations) allowed selecting the better threshold.



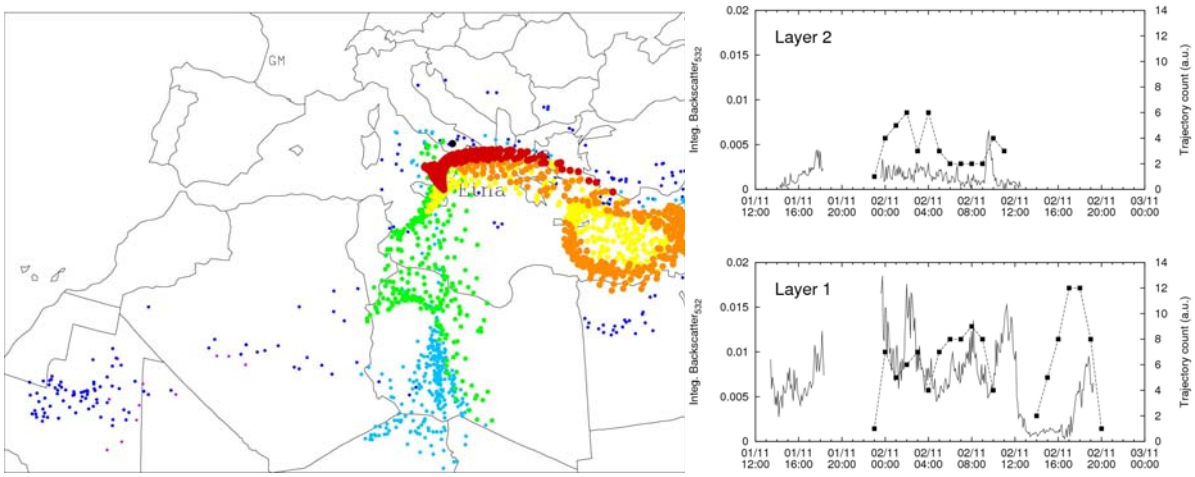
(a) (b)

Figure 3 : Vertically integrated aerosol load simulated by (a) BOLCHEM and (b) seen by MODIS AQUA.

3.4. Lagrangian Transport and Etna Eruption

A Lagrangian transport model implemented in BOLCHEM (BOLTRAJ variant) can be used in conjunction with the Eulerian part. This is useful as an analysis tool for a better interpretation of Eulerian simulations through the computation of the probability matrices of pollutant origin. It is also useful to study specific events of concentrated point sources when

the resolution of the Eulerian grid would be too small to represent dispersion at short time. The first application was the Mt. Etna eruption on autumn 2002 (*Villani et al., 2006*). The joint analysis of Lagrangian trajectories, satellite data, meteorology, and lidar measurements allowed to estimate the tropospheric dispersion coefficient and to clarify some features observed by lidar located near Potenza. Lidar measurements recorded a strong signal with a clear sulphate signature, along with weaker layers above. Satellite images gave no clear evidence, but trajectory analysis revealed the nature of the complex picture. Part of the trajectories traveled over Potenza taking a long passage through the Sahara region, possibly carrying some silicate and passing over Potenza at the same time (and at different heights) of those coming directly from Mt. Etna (Figure 4).



(a) (b)

Figure 4 : (a) Position of particles released at Mt. Etna from 26-10-2002 to 01-11-2002. Different colours denote different ages (in days); (b) Comparison of LIDAR signal measured over Potenza and simulated by BOLCHEM.

Acknowledgements

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Chapter 2. Off-Line Modelling and Interfaces

2.1 Off-line Model Integration: EU Practices, Interfaces and Possible Strategies for Harmonisation: *Sandro Finardi[1], Alessio D'Allura[1] and Barbara Fay[2]*

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1. Introduction

A survey of multiple and diverse modelling communities in European countries was performed in COST728 on a basis of partner contributions (*Baklanov et al., 2007*). Even if the model coverage remains incomplete and somewhat arbitrary the contributions represent a wide spectrum of modelling complexities and efforts in 16 European countries and about 40 institutions. The majority of the presented systems is based on mesoscale meteorological models (MetMs) available at the national weather services or weather forecasting consortia (i.e. HIRLAM, COSMO (Lokalmodell), ALADIN) and on international free community models developed by universities (i.e. MM5, WRF, MC2, RAMS). This approach allows the air quality (AQ) modelling community to take advantage and benefit from development, testing and model validations done for the purpose of numerical weather prediction (NWP). At the same time it provides users without large own development capacities or with the need for a standard system to apply model systems supported by a wider community.
The modelling components that deal with transport and transformation of atmospheric pollutants are more diverse than the MetMs, ranging from a simple passive tracer along a trajectory (i.e. CALPUFF) to a complex treatment of reactive gases in the Earth system (i.e. MESSy). The wide spectrum of model applications ranges from diagnostic or climatologic AQ assessments, episode analysis and source apportionment to AQ forecast at regional and urban scales and toxic and radioactive releases emergencies preparedness.
The communication between off-line coupled meteorological and AQ models is often a problem of underestimated importance. The variety of modelling systems previously introduced give a rise to different approaches and methods implemented within interface modules. Tasks covered by interfaces are minimized in coupled systems relying on surface fluxes, turbulence and dispersion parameters (i.e. eddy viscosity) provided by the meteorological driver. Other systems use interface modules implementing surface and boundary layer parametrizations to estimate dispersion parameters. Atmospheric physics parametrizations, and even default or limit values assumed for some key parameters, can have relevant effects on pollutant concentration fields in critical conditions (e.g. low wind and stable conditions). Interface modules can involve the evaluation of emissions of some relevant species that can be strongly influenced by meteorology, like biogenic VOC, pollen emission, windblown dust and sea salt spray. Moreover, mesoscale and urban scale AQ modelling systems are usually nested within larger scale application results, used to initialize and drive AQ fields at domain boundaries. This operation too can have relevant influence on AQ modeled fields, and especially, it is evident over complex topography.

2. Off-line Coupled Models and Interfaces

The major components of an integrated meteorological and AQ modelling system are sketched in Figure 1. Since the input data flow connects the meteorological modelling system and AQ model those two are generally defined as coupled models. Depending on the characteristics of this connection we can distinguish between off-line and on-line coupling. Off-line coupled MetMs and AQ models work separately, there is no feedback from Atmospheric Chemical Transport Models (ACTMs) to MetMs and meteorological input to the AQ model is usually limited to averages, either in time or space, of main variables defining the atmospheric status (fields are provided at any fixed times, e.g. 1 hour). This specific approach is a traditional way by which those complex systems have been developed until now.

The development of these modelling systems is usually focused on the scientific and technical features of emission, atmospheric flow and pollutant dispersion models, while comparatively little attention is devoted to the connection of different models. Meteorological and AQ models often employ different coordinate systems and computational meshes. In principle, interfaces should simply solve this grid system mismatch to connect MetMs output and AQ models input with minimum possible data handling.

Nonetheless, interface modules are often used to solve other system realization issues, e.g.:

- some AQ models rely on "standard" meteorological products which usually do not include turbulence, atmospheric stability, mixing height, and dispersion coefficients;
- MetMs cannot provide all the physical variables that are needed by AQ models (e.g. deposition velocities) or some meteorological fields may be estimated by parametrizations not compatible with modelling methods implemented in dispersion models;
- sometimes re-computation or "filtering" of dispersion parameters is considered more robust for practical applications;
- horizontal resolution of the meteorological forecast can be lower than that needed by AQ models, and insufficient to correctly estimate dispersion parameters.

To solve the above mentioned problems various tasks are often included within interface modules, as well as: data interpolation, meteorological fields downscaling, boundary layer parametrizations and estimation of dispersion coefficients, evaluation of meteorological driven emissions (e.g. biogenic, wind blown dust, sea salt), enhancement of physiographic data.

A multiplicity of off-line coupled modelling systems has been developed and applied all over the world. The most common systems features and interfacing strategies in Europe have been identified, within COST-728/WG2, using: COST-728/-732 model inventory (http://www.mi.uni-hamburg.de/index.php?id=539), a questionnaire on interfaces circulated among COST-728 participants and previous experiences and knowledge of COST-728/WG2 members (*Baklanov et al., 2007*).

Three main approaches have been identified:

a) joint development of coupled models, with interfaces built on specific models features and needs, this approach is mainly adopted by large institutions and Weather Services developing both MetM and AQ models;
b) use or customization of US Community modelling systems, e.g. MM5/WRF+CMAQ with MCIP interface module;

c) interfacing of self developed AQ models with EU Weather Services and US Community Meteorological Models through model specific or general purpose interfaces.

The first strategy implies the direct use of physical parameters estimated by MetMs, like e.g. Monin-Obukhov similarity theory (MOST) parameters, and limitation of the interface module tasks to the evaluation of missing variables. This approach is particularly attractive when meteorological and AQ models share the same computational grid system and data interpolation can be avoided. Note that changing grid system and topography makes necessary to re-compute the vertical wind component, to guarantee the mass conservation, which is essential for dispersion calculations.

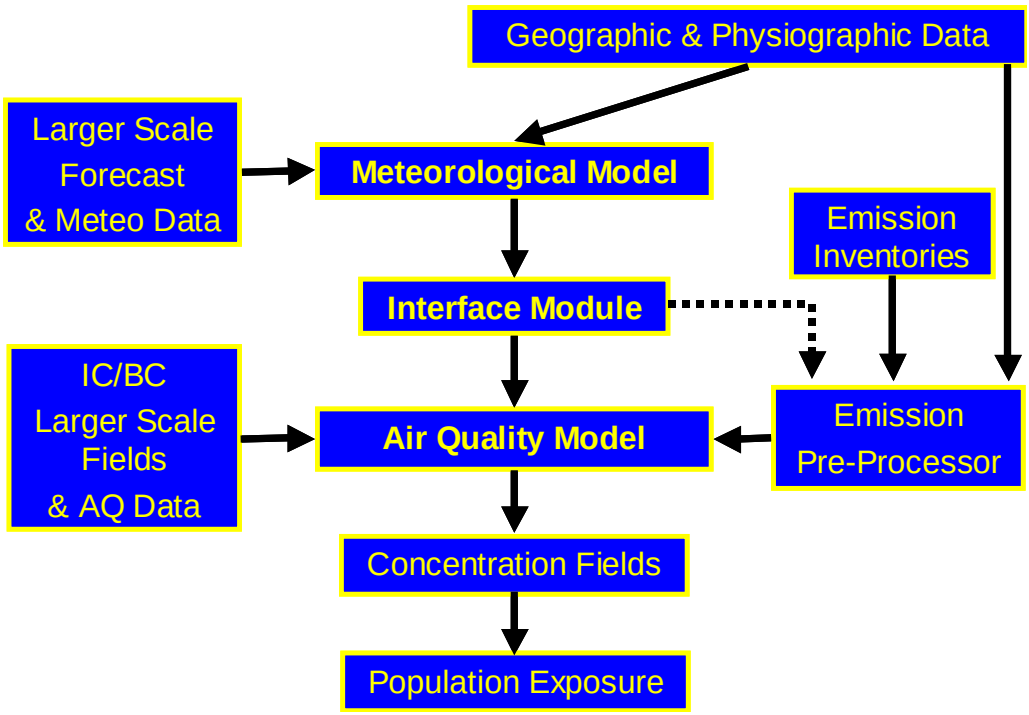


Figure 1: Integrated meteorological and air quality modelling system conceptual scheme.

The other approaches are mainly based on the development of meteorological pre-(post-) processors capable to evaluate surface and boundary layer scaling parameters, mixing height, atmospheric turbulence and dispersion parameters on the basis of the average meteorological variables provided by MetMs and possibly supplementary external data. This approach gives the possibility to interface meteorological and AQ models characterized by relevant differences that can make difficult a direct connection. Moreover, it can allow the introduction of additional high resolution information, like land-use, roughness length, or urbanized parametrization to be used by computations performed by the interface module. The use of boundary layer and dispersion parametrizations within interface modules or AQ models should take into account the effective resolution of MetMs to avoid parametrization of phenomena explicitly described by modeled meteorological fields, as it can happen when high resolution MetMs results are available.

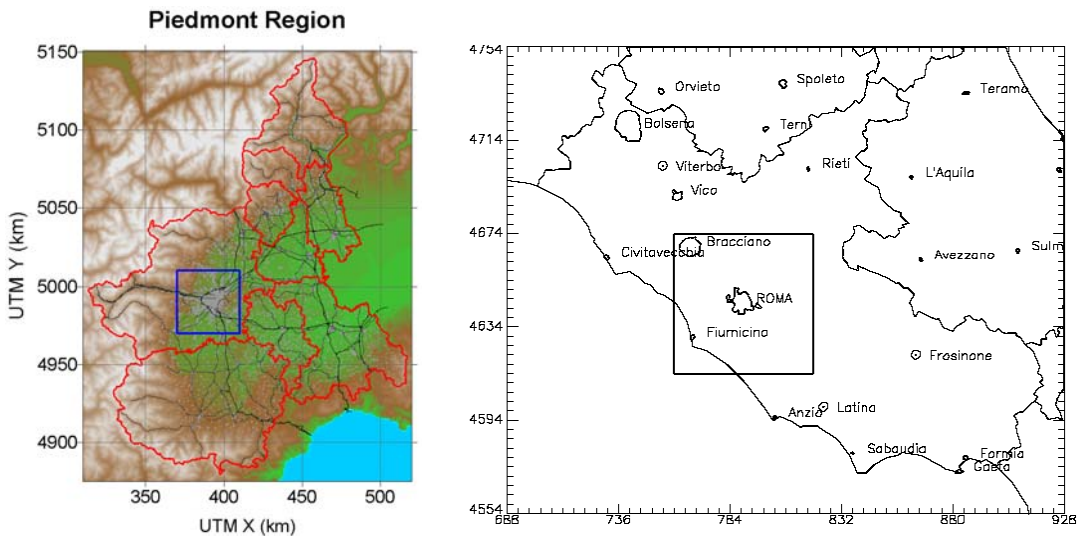
The AQ models have to be interfaced with pollutant emissions and initial and boundary conditions imposed from larger scale AQ forecast. Pollutant emissions can be influenced by meteorological conditions through different kind of processes of both anthropogenic and natural origin. Air temperature determines the amount of fuel consumption for house heating,

the meteorological conditions often influence people behavior (e.g. car or public transport usage) determining some features of pollutant emissions. Meteorological conditions influence natural emission processes like surface erosion, wind blown dust resuspension or biogenic emissions of Volatile Organic Compounds (VOC) from vegetation or pollen.
The ACTMs results depend on the initial conditions and inflow in the computational domain of background concentrations. The proper nesting of meso and local scale simulations within larger scale forecast results it is generally managed by an interface module that has to match grid and resolution differences and possibly different chemical schemes employed by considered models.
The following sections provide a few examples of the possible effects of the different interfacing issues on the AQ simulation results.

3. Air Quality Modelling System: Results

3.1. Interface Module and Model Nesting Effects on Air Quality Simulation

An integrated AQ modelling system similar to those previously sketched has been applied in two different urban environments with the intent to highlight its sensitivity to dispersion processes parametrizations and AQ model initialization implemented within the interface module. The modelling system, used for the following applications, is based on the MetM RAMS (*Pielke et al., 1992; Cotton et al., 2003*) and on the Eulerian chemical transport model FARM (*Calori and Silibello, 2004*). The cited meteorological and AQ models are connected by the interface module GAP/SurfPRO (*Calori et al., 2005; Finardi, 2005*). The Grid AdaPtor (GAP) is a grid interpolation tool with a capability to re-compute vertical velocities, it has been developed to interface FARM with any MetM. The SURFace-atmosphere interface PROcessor (SURFPRO) is a meteorological processor based on MOST designed to provide turbulence and dispersion scaling parameters, as well as eddy diffusivities and deposition velocities (*Beljaars and Holtslag 1991; Hanna and Chang 1992; Zilitinkevich et al. 2002b*).



(a) (b)

Figure 2 : Turin (a) and Rome (b) urban area air quality modelling systems computational domains.

The cities of Rome and Turin are respectively the first and fourth largest urbanized areas in Italy. They are exposed to severe air pollution episodes induced by complex air flow pattern and atmospheric boundary layer dynamics, due to both intrinsic complexity of the urban

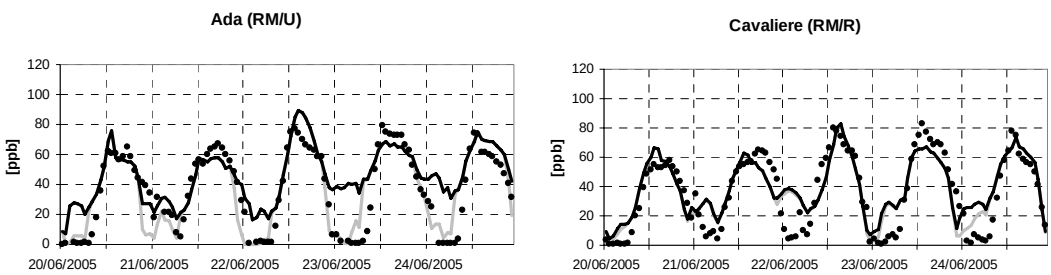
canopy and specific mesoscale flow features (e.g. sea breezes, catabatic flows, and air stagnation). The nested computational domains for both mentioned systems are depicted in Figure 2. Rome is the largest Italian city, characterized by a widely spread urbanized area, with a total population around 3.5 million. The city is often affected by high ozone and PM concentrations, likely to be detected in both summer and winter. During summer, the high insulation favors photochemical activities. In winter, persistent high pressure systems with very weak pressure gradients determine weak wind conditions and possibly temperature inversion causing pollutants accumulation in the lower layers of the troposphere.
Turin metropolitan area has a resident population of about 1.5 million. It represents the core of one of the major industrial areas in northern Italy. The city is located at the western edge of the Po Valley, and it is situated mainly on a flat topography between the Western Alps and a range of hills on its east side. Local circulation is strongly influenced by the shelter effect of the Alpine mountains chain, and it is dominated by the superposition of mesoscale (e.g. Po Valley stagnation, mountain/valley breezes and föhn) and urban flow features.

3.2. Dispersion Parametrization Effect

The modelling system introduced in the previous section has been applied to analyse a summer air pollution episode in the area of Rome (*Gariazzo et al., 2007*). The area is exposed to sea breeze circulation during daytime, while very week land breeze, turning to calm conditions within the city of Rome, characterises night time circulation. Surface turbulent fluxes and Eulerian dispersion coefficients (eddy diffusivities) used by the chemical transport model FARM are computed by SURFPRO. The similarity theory is based on the general assumptions of quasi-stationary and horizontally homogeneous flow, and constant (independent of height) turbulent fluxes within the surface layer (*Arya, 1988*). These assumptions are generally not fulfilled in urban areas and complex terrain. The MOST is nevertheless applied in many models even in these cases, mostly due to lack of other practical formulations (*Mahrt, 1999*). During low wind conditions like those observed in Rome at night time the values provided by parametrizations for the eddy diffusivity is very low and usually falls under minimum K_Z value. Different minimum values for K_Z are used by AQ models, normally ranging from 0.1 to 1 m^2s^{-1}. In SURFPRO different minimum values can be imposed as a function of land use:

$$K_z^{\min} = K_{rur}^{\min} \cdot (1 - f_{urban}) + K_{urb}^{\min} \cdot f_{urban}$$

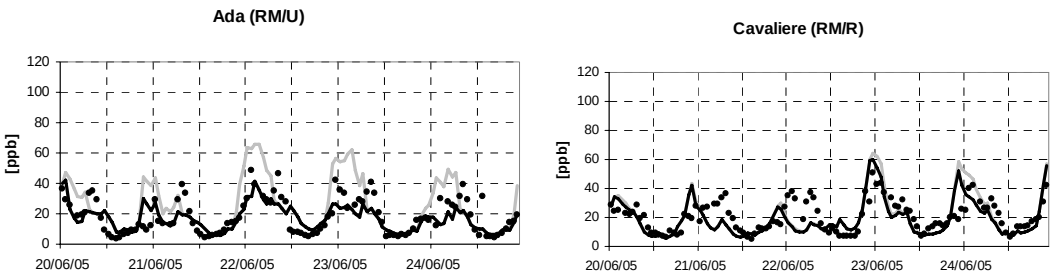
where f_{urban} indicates the urban land use fraction within each grid cell, $K_{rur}^{\min}$ and $K_{urb}^{\min}$ indicate minima of K_Z for rural and urban areas, respectively, for which values of 0.1 and 1 m^2s^{-1} are commonly used. This assumption can be justified due to the urban canopy effect that has the tendency to maintain neutral or slightly unstable conditions over the city during the night, consequently increasing pollutant dispersion with respect to rural conditions.



(a) (b)

Figure 3 : Comparison among O_3 observed (black dots) and computed concentration with K_Z minimum value set to 0.1 m^2s^{-1} (grey line) and 1 m^2s^{-1} (black line) at urban (a) and rural (b) stations.

Figures 3 and 4 show examples of the modelling system results for O_3 and NO_2 at an urban background (Villa Adda) and rural (Cavalieri) stations. The reference simulation (black line), with minimum K_Z defined by the previous formula, shows an overestimation of O_3 at the urban station during night time, while NO_2 is slightly underestimated. A second simulation has been performed imposing everywhere a minimum K_Z = 0.1 m^2s^{-1}. This run results (Figures 3-4, grey line) show an enhancement of O_3 at the urban station, and an excessive growth of NO_2 at night. As expected, no relevant change affects concentrations at rural location and during daytime. The stronger limitation imposed to vertical mixing by small K_Z makes rise NO_X concentrations and causes consumption of O_3 (ozone titration) in VOC-limited photochemical regimes.



(a) (b)

Figure 4 : Comparison among NO_2 observed (black dots) and computed concentration with K_Z minimum value set to 0.1 m^2s^{-1} (grey line) and 1 m^2s^{-1} (black line) at urban (a) and rural (b) stations.

Further tests and previous experiences in other geographic locations confirmed that minimum K_Z is a relevant (and often neglected) parameter to model properly the dispersion during weak wind and very stable conditions. Unfortunately no general value for minimum K_Z can be defined, while proper values depend on season and local climatology, as well as on numerical diffusion for advection scheme.

3.3. Surface Fluxes and Boundary Layer Parametrization Effect

In principle the direct interfacing methods consisting in the evaluation of dispersion parameters from meteorological models average fields and turbulent fluxes should be preferred to guarantee the modelling system consistency and to take advantage of the modelling capabilities of new generation MetMs, e.g. higher order turbulence closures, surface layer parametrizations and soil-surface-canopy models. On the other hand this approach can suffer the intrinsic weakness to be influenced by possible meteorological forecast errors or local scale flow features that can have relevant impact on surface fluxes, mixing height value and pollutant dispersion.

A test case to evaluate possible effects of different interfacing approaches has been run over the Torino area. The simulations covered a summer fair weather period, when thunderstorm activity occurred over the western Alps. The AQ model has been driven by two different set of turbulent surface fluxes and scaling parameter. The first set has been estimated using the surface fluxes produced by the MetM RAMS; the second - by SURFPRO interface module according to van Ulden and Holtslag formulation for surface fluxes and MOST.

Figure 5 shows the relevant differences obtained from the two test simulations for almost all the considered parameters (sensible heat flux, friction velocity, mixing height, and vertical diffusivity at the first vertical level) during the first day of simulation. The comparison of computed and observed concentrations (Figure 6) highlights the mismatch of NO_2 concentrations produced by the simulation using RAMS turbulent fluxes. The sensible heat

flux has been largely underestimated, producing a very limited boundary layer growth with relevant effects on local NO_X concentrations but limited influence on O_3. Further analysis pointed out as a localized convective precipitation event has been mispredicted by RAMS, affecting part of Torino city with a strong precipitation event that did not occur. The high uncertainty of storms location and intensity forecast is not surprising, it is due to the geographical complexity of the region and seasonal (July) thunderstorm frequency.

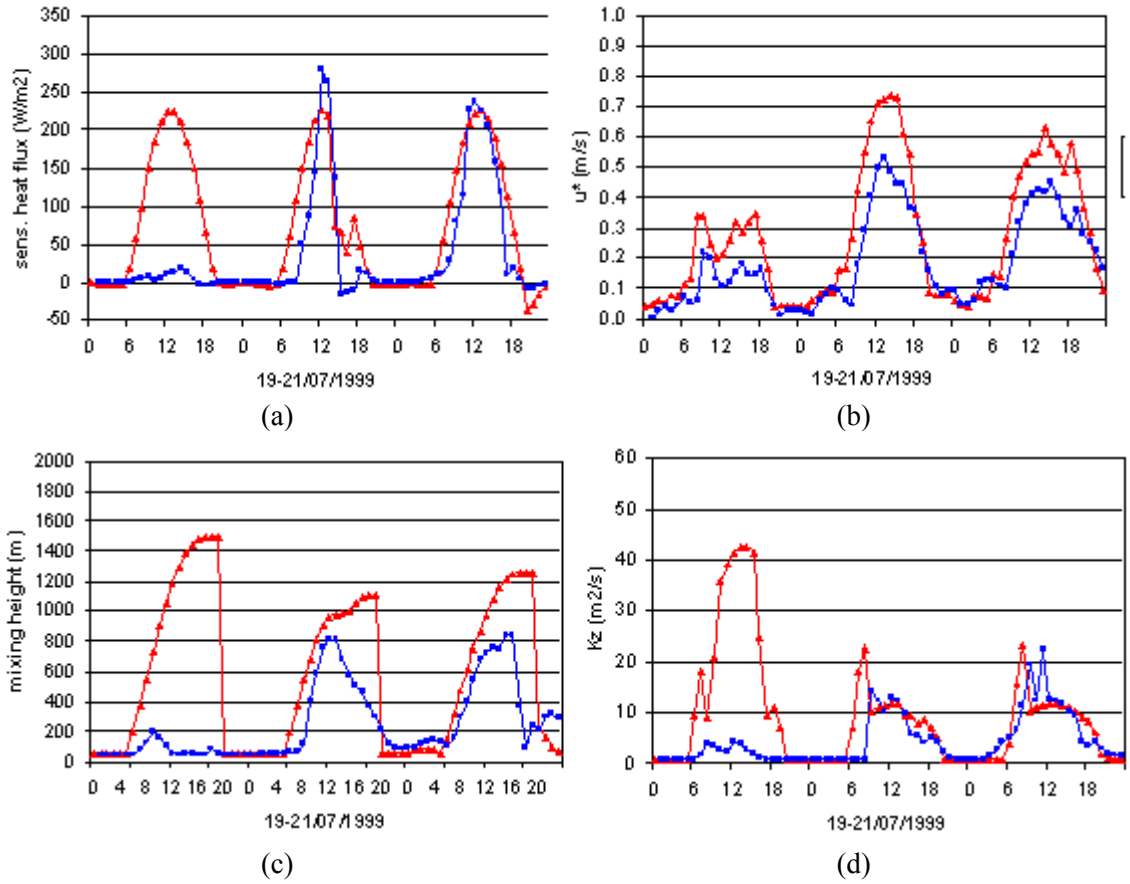


Figure 5 : Sensible heat flux (a), u_* (b), mixing height (c) and K_Z (d) computed by RAMS (blue line) and SURFPRO (red line) during summer thunderstorm episode in Torino.

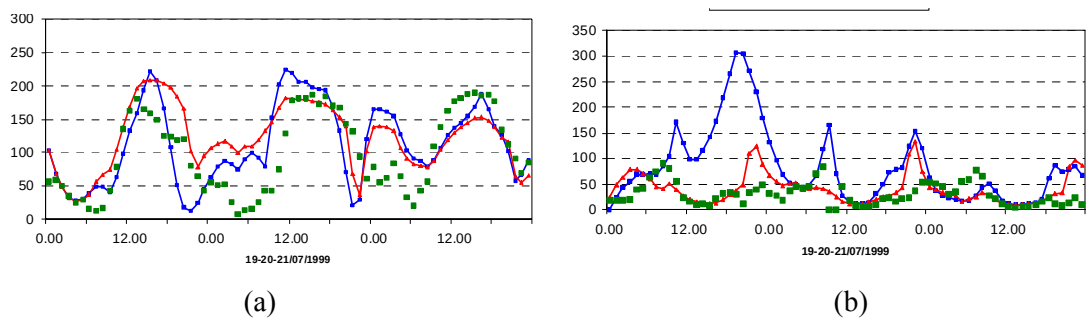


Figure 6 : Concentrations of O_3 (a) and NO_2 (b) computed using RAMS (blue line) and SURFPRO (red line) turbulent fluxes and scaling parameters vs. observations (green line).

During adverse meteorological events the use of an interface module to model dispersion parameters can have the advantage to reduce forecast error effects on predicted concentrations. Anyway, further analysis showed that the discussed results are strongly dependent on the radiation scheme used by RAMS model. Running the model with the

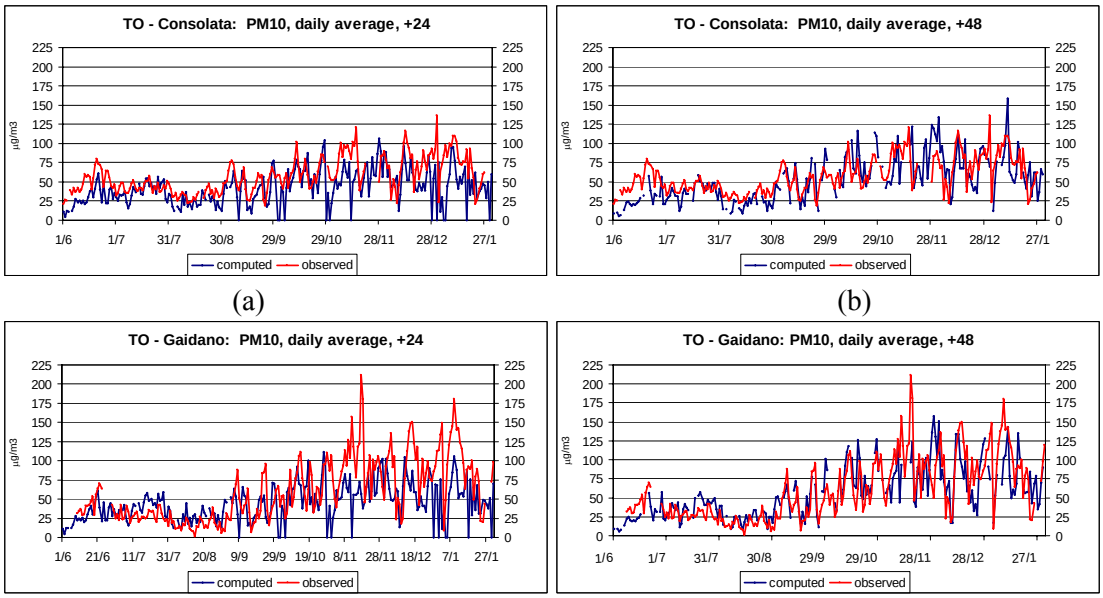
Harrington instead of Chen scheme (*Cotton et al., 2003*), the NO_X overestimation could be reduced due to larger values of surface radiation obtained.

3.4. Air Quality Initialisation at Regional and Urban Scale Effect

During the evaluation of the AQ forecasting system for the Torino metropolitan area *(Finardi et al., 2007*) the effect on model results of AQ initial and boundary conditions has clearly emerged. To define these data the modelling system relies on CHIMERE continental forecasts provided by *Prev'Air* European Scale Air Quality Service (http://www.prevair.org). This large scale AQ forecast was initially used to define both initial and boundary conditions. In principle local observations could be used to build more realistic initial concentration fields, but they are not yet available at the forecast simulation start time.

The AQ forecasting system results have been compared with observations over a period: from June 2006 to January 2007. The predicted concentration data have been divided in two time series obtained selecting the first (last) 24 h of each daily forecast cycle that covers 48 h period. Comparison of these two time series with observations (Figure 7) showed that the 48 h forecast obtains generally higher concentrations and better fits with observations with respect to the 24 h forecast. This behaviour was common to all pollutants except ozone (which was overestimated for 24 h simulation). The comparison of initial and 24 h concentration fields showed that differences were due to influence of initial conditions on the first day of simulation. The resolution difference between CHIMERE (50 km) and FARM (4 km) background domains didn't allow obtaining a proper initialisation. In CHIMERE topography, the city of Torino is located on the western Alps slope, at about 800 m asl. This feature clearly favours ozone overestimation and other pollutants underestimation, due to the city location outside of the Po valley.

This shortcoming identified induced to change a way to initialise AQ fields using previous day the forecasting system results (+24 h fields) and to prepare a simple AQ analysis tool, to be able to correct initial fields with local observations when they will be available. A resolution match problem is present, even if it is less evident, within the boundary conditions too. The final set up of the system is, therefore, programmed to move to 3 nested domains, introducing a larger background computational mesh enhancing background pollution simulation and reducing the effects of boundary conditions on the AQ forecast. I.e., as it was originally tested in the EC FP5 FUMAPEX project (*Finardi et al., 2007*).



(c) (d)

Figure 7 : Simulated (blue line) and observed (red line) PM10 daily average concentrations for the (a,c) first (0:+24) and (b,d) second (+24:+48) day of forecast at two Torino urban background stations (a,b) Consolata and (c,d) Gaidano. The comparison refers to June 2006 – January 2007 time period.

4. Summary and Discussion

The Working Group 2 of COST-728 started a survey of multiple and diverse coupled meteorological and air quality modelling systems developed and applied European countries. Our attention has been focused on off-line coupled modelling systems, which are by far the more numerous, even if the number of on-line coupled systems recorded is larger than expected. The important role of interface modules has been discussed and more common approaches followed in their development have been briefly described on the basis of COST-728 inquiry and known modelling systems analysis. In principle the direct interfacing methods consisting in the evaluation of dispersion parameters from meteorological models average fields and turbulent fluxes should be preferred to guarantee the modelling system consistency. On the other hand interface/processors allowing the user to reconstruct missing variables and dispersion parameters, to increase space resolution and possibly to enhance meteorological fields for lower atmospheric layers and for the urban atmosphere, are widely used by the air quality community. The influence of different interface modules on integrated air quality modelling system results has been shown through application examples taken from the author's experiences.

In the diverse landscape of European modelling, model harmonisation remains an important issue despite earlier efforts, e.g. COST-710 (1994-1998) which are continued in the regular Harmonisation conferences. Modular modelling, flexible input-output strategies and adaptable interfaces following agreed guidelines for off-line and on-line integrated modelling, which are applied by all including the large consortia and community models, would greatly facilitate model improvement and applicability for European users. The large variety of modelling systems can be considered a scientific richness but creates problems of model result inter-comparison and underlines difficulties in model development collaboration in Europe.

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2.2 Coupling Global Atmospheric Chemistry Transport Models to ECMWF Integrated Forecasts System for Forecast and Data Assimilation within GEMS: *Johannes Flemming[1], A. Dethof[1], P. Moinat[3], C. Ordóñez[2], V.-H. Peuch[3], A. Segers[7], M. Schultz[6], O. Stein[5], M. van Weele[4]*

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Abstract
The paper presents the implementation of a coupled forecast and assimilation system developed within the subproject on Global Reactive Gases (GRG) of the GEMS-project (Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data, FP6).
One of the main objectives of the GEMS project is to utilise ECMWFs 4D-VAR data assimilation system to assimilate satellite observations of atmospheric composition at the global scale. The GRG subproject focuses on the assimilation of the following gases: carbon monoxide (CO), ozone (O_3), nitrogen oxides ($NO_x = NO + NO_2$) formaldehyde (HCHO) and sulphur dioxide (SO_2). These gases play a key role in atmospheric chemistry and are observable from space.
ECMWF's integrated forecast system (IFS) is able to simulate the transport of these tracers but does not contain the modules for the simulation of chemical conversion, emission and deposition. Instead of directly integrating (on-line coupling) the relevant modules into the IFS, a coupled approach was taken on which links the IFS to already established Atmospheric Chemistry Transport Models (ACTMs). The coupled approach seemed to be a much smaller development effort, and it offers more flexibility in the choice of the modules for chemical conversion, emission and deposition by coupling different ACTMs to the IFS.

The two-way coupled system consists of the IFS and one of the chemistry transport models MOZART3, TM5, and MOCAGE. The coupling software OASIS4 has been implemented to facilitate the data exchange.
In the coupled system, IFS sends meteorological data at high temporal resolution to the ACTMs. The ACTMs provide concentration tendencies due to emissions and chemical conversion as well as initial tracer conditions to the IFS. The application of external tendencies is required in IFS because its 4DVAR data assimilation needs to account for tracer source and sink terms which are not simulated in the IFS model. Moreover, the tracer transport may benefit from the sophisticated vertical transport schemes of the IFS.
The coupled system has been applied in forecast mode for several months in 2003 in different configurations in terms of vertical transport and coupling synchronisation. Test assimilation runs of CO by MOPITT have been successfully carried out for several weeks. Experimental near-real time forecasts of the coupled system run since April 2007. However, this paper focuses on the design of the system.

1. Coupling of Earth-System Components Models

Numerical models simulating specific components and aspects of the earth-system have to exchange their results in order to study the many interactions within the earth-system.
A coupled model *A* provides more detailed information about processes which have been treated by simpler assumptions such as explicit or implicit climatologies in model *B*. If the system is two-way coupled, the response in model *B* is fed back to model *A*, leading to different result there, which may again influence model *B*.
Besides the scientific questions related to the coupling of models, the interaction of the numerical models is a big technical challenge. The transformation of data at different temporal and spatial resolutions as well as computational efficiency, memory consumption, data storage capacity, meta-data communication and code management are issues which have to be addressed.
The most common type of coupling is 2D in space, i.e. the coupled models cover separate 3D domains, such as atmosphere and ocean, which are connected to each other by a 2D interface. Less common is 3D coupling in which both models cover the same or an overlapping spatial domain, e.g. the atmosphere, but consider different aspects of it as in the case of weather forecasts models and chemistry transport models. The amount of data to be exchanged is bigger in 3D coupling and there are further consistency issues if both models simulate the same processes such as transport but in a different way (see section 0).
There are various options for the technical implementation of the coupling, which differ in the following aspects whether or not:

- a dedicated coupling software is used to facilitate the coupling;
- the coupled models stay independent as executables;
- the models or modules run concurrent or step-wise sequential.

The tightest way of coupling, often called “on-line” or “integrated” coupling, is the exchange by argument passing from subroutines simulating different components and aspects of the earth-system. These “integrated” models tends to have less consistency problems because the integrated modules have been aligned to the general model structure, in particular to model geometry and to decomposition for parallelisation. The integration may require a large coding effort, depending on the code structure of the modules, as well as substantial scientific testing to ensure the scientific integrity of the new modules in the existing model. Further, the integrated approach is less flexible in the choice of the coupled model and also requires

continuous code management in order to benefit from further development of the included models.
For these reasons many coupled systems try to keep some sort of independence of the component models, and the coupling is facilitated by a specific coupling-software. *Ford and Riley* (*2002*) give an overview of coupler software developed in North America and Europe.
The coupling-software mainly consists of two interconnected entities:

- A mechanism to let coupled models runs together and enable them to exchange data;
- An infrastructure to process mete-data needed for the communication of the models.

The model developer who wants to couple the models (i) has to include coupler-specific interfaces in the models, and (ii) has to provide the model and coupling meta-data according to standards of the coupling software. Both tasks can be time consuming, and the gain of interoperability has to be balanced against the costs of the implementation.
There are two basic design concepts for the coupling software: "Concurrent coupling" means that independent model executables or modules run at the same time on different computer resources. An additional coupler/driver executable controls the data exchange between the models. "Sequential coupling" relies on a "superstructure" which calls the components models sequentially for each coupling-time step using the same system resources. Sequential couplers can be considered as a partly automated procedure for the "manual" integration of component models as subroutines in one unified model code. Figure 1 shows the schematic of a concurrently coupled system (the GRG system) and integrated / sequential - coupled system. Sequential coupling tends to have less latency problems than concurrent coupling if the two components differ in their computation time. OASIS4 (*Valcke and Redler, 2006*) and OASIS3 (*Valcke, 2006*) are examples of concurrent couplers. The Earth System Modelling Framework (ESMF; www.esmf.ucar.edu) is an example for "sequential" coupling.

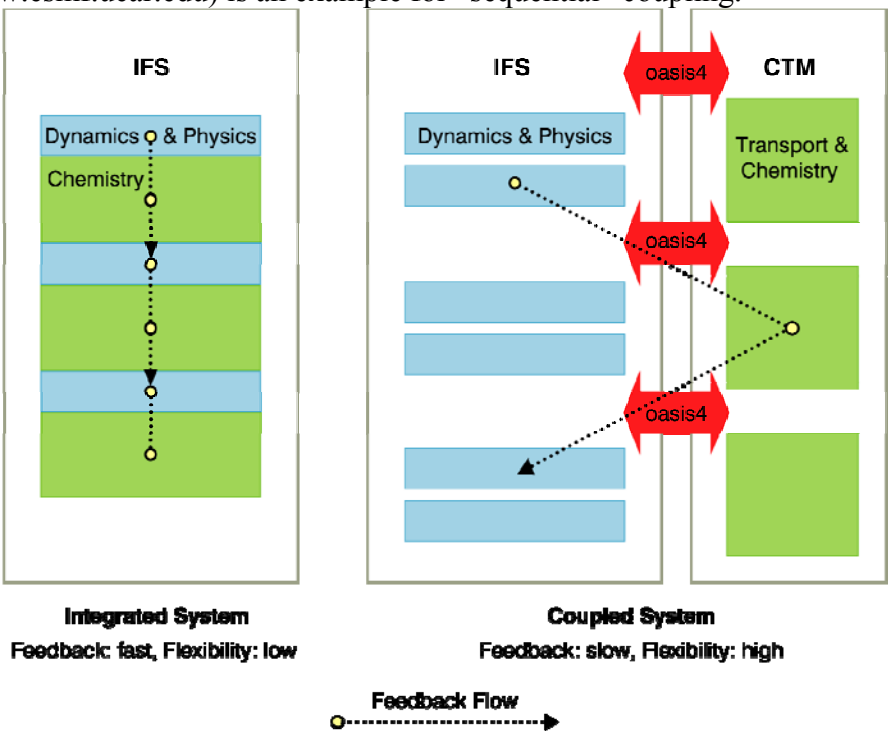


Figure 1 : Different designs of a coupled system: Concurrent coupling of two independent executables (left) and sequential coupling of model or integration of sub-routine calls (right).

2. The GEMS GRG Coupled System at ECMWF

As part of the GEMS subproject on global reactive gases (GRG), a system coupling ECMWFs integrated forecast system (IFS) with atmospheric chemical transport models (ACTMs) has been developed. The couplers software OASIS4, which is being developed as part of the prism project and the prism support initiative (http://www.prism.enes.org/), is used to couple the two components in concurrent fashion. The OASIS4 interfaces in the IFS and the run and configuration environment for the coupled experiments can easily be adapted to couple other earth system model to IFS.
The main motivation for the development of the GRG coupled system was the need to account for sink and source processes, namely chemical conversion, in the assimilation of satellite observation of chemical tracers within the 4D-VAR data assimilation of the IFS. Since it seemed to be costly to integrate complex chemical mechanisms in the IFS, tendencies describing sink and source processes should be provided by well established ACTMs, being coupled to the IFS. The three candidate ACTMs for the GRG coupled system are MOCAGE (*Josse et al., 2004*), MOZART (*Horowitz et al., 2003*), and TM5 (*Krol et al., 2005*).
A clear benefit of the coupled approach, in contrast to integration, is the flexibility in the choice of different coupled chemical schemes represented by the different ACTMs. However, the required three-dimensional coupling is less consistent than an integrated system because of feedback delay and dislocation due different transport representations (see section 2.4) in the IFS and the ACTM.

2.1. Configuration of the GRG Coupled System

The GRG coupled system is a three-dimensional two-way coupled system: IFS provides atmospheric fields at high temporal resolution to drive the ACTMs, and the IFS receives tracer concentration fields and tracer tendencies due to source and sink processes from the ACTM. A further coupling option is the feedback of concentration fields from IFS to the ACTM.
Depending on which tendency data are exchanged, the GRG coupled system can be run in three modes:

- ACTM forecast mode;
- IFS tracer forecast mode;
- IFS tracer data assimilation mode.

The ACTM forecast mode is a one-way coupling in which IFS provides the meteorological data on-line to the ACTM. The main difference to ACTM off-line runs is the high temporal resolution at which the ACTM gets the atmospheric data. Typical frequency for the coupling is one hour whereas the temporal frequency in off-line runs is six hours.
In IFS tracer forecast mode the ACTM provides initial condition for the chemical tracers (NO_x, NO_2, SO_2, CO, HCHO and O_3) and 3D fields of tracer tendencies due to emissions, deposition and chemical conversion to IFS. The IFS simulates the horizontal and vertical transport of these tracers and applies the ACTM tendency data in order to account for the source and sink processes not simulated in the IFS. The ACTM itself run as in ACTM forecast mode. The feedback option enables replacing the ACTM concentration fields, in particular the initial conditions, with the tracer fields of the IFS.
In IFS tracer data assimilation mode, the IFS tracer forecast mode is applied in the outer loops of ECMWF data assimilation system, i.e. the calculation of the trajectories runs of the "complete" model of the 4D VAR (*Mahfouf and Rabier, 2000*) The inner loops used in the minimisation step with the tangent linear and adjoint model are currently run uncoupled, i.e. without the application of the source and sink tendencies from the ACTM.
In the coupled system the IFS runs in a T159 spectral resolution and the grid point space is represented in the reduced Gaussian grid (*Hortal and Simmons, 1991*). The vertical coordinate

system is given by 60 hybrid sigma-pressure levels. In order to avoid difficulties in the vertical interpolation by the coupler, the ACTM use the same 60 vertical levels. The coupler only has to perform horizontal interpolations for which the bi-linear mode is applied. The resolution of the ACTM is lower (~T63) as the IFS resolution because of the high computational cost of the ACTMs (see Table 1).

Table 1 : Resources of the IFS and the ACTMs MOZART-3, MOCAGE and TM5.

Component	Resolution	Time step	Species	MPI / openMP	Run time 24h
IFS	T159, 60L	1800 s	5	8 / 2	(stand alone) 3 min
MOZART-3	T63, 60L	900 s	106	8 / 8	12 min
MOCAGE	2°x2°, 60L	900 s	126	1 / 12	188 min
TM5	2°x3°,60L	1200 s	54	12 / 1	31 min

The IFS is run on a higher horizontal resolution because of the quality of the meteorological forecasts and because a lower resolution would limit the acceptance of high resolution observations within data assimilation. The coupling frequency is 3600 s which is the largest acceptable time step for the IFS at a T159 resolution, and also the time step of some of the ACTMs. The exchange of data can be either provided via a master-process only or via a direct exchange via all processes involved in the simulations.
The following variables are covered by the OASIS4 interfaces in the IFS and the ACTMs:

- IFS to ACTM
  - T, Q, U, V (3D grid point);
  - O_3, NOx, SO_2, CO, HCHO - concentration (3D grid point);
  - ps, taux, tauy, shflx, qflx (2D grid point);
  - Vorticity, Divergence, ps (3D/2D spectral fields);
  - Wavenumber-info (3D/2D spectral fields).
- ACTM to IFS (3D grid point)
  - O_3, NOx, SO_2, CO and HCHO tendencies due to chemistry, wet deposition and atmospheric emissions;
  - O_3, NO_x, SO_2, CO and HCHO tendencies due to surface fluxes (emission, dry deposition);
  - O_3, NOx, SO_2, CO and HCHO – concentration.

2.2. Initial Condition Handling within Coupled Experiments and Feedback

The coupled long-term simulation and data assimilation runs are structured as a sequence of coupled runs (6 h in data assimilation, 24 h in forecast mode) because the IFS needs a re-start from a meteorological analysis as often as possible. The ACTM provides the tracer initial conditions of the IFS for the first forecasts. There are three modes of how the initial conditions for the GRG-tracers are obtained in the subsequent forecasts (see Figure 2).
In the “ACTM constrained” mode, the IFS gets the initial tracer conditions from the ACTM at the start of each forecast. The ACTM gets the whole set of initial conditions from the previous ACTM run.
In the “free running” mode, this exchange of initial conditions happens only at the first forecasts. Both the IFS and the ACTM use initial tracer conditions from their previous runs in all following coupled forecasts.
In “feedback” mode, the ACTM will use the tracer initial conditions provided by IFS after the first model run. The IFS tracer fields may now contain information from observations

(analysis mode) or may be different from the ACTM fields because of different vertical transport simulation in IFS.

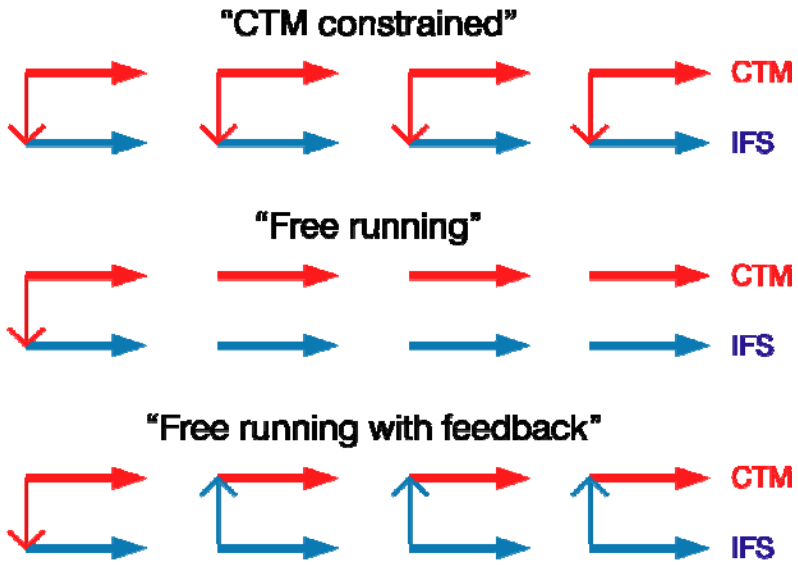


Figure 2 : Modes of initial condition handling in a sequence of short simulations.

2.3. Computational Performance of the GRG Coupled System

The main factor for the computational performance of the coupled GRG-system is the individual run time of the IFS and the ACTMs at ECMWF high performance computing facility (IBM power5). The computational cost of the ACTMs is clearly higher than the one of the IFS in forecast mode (see Table 1). The good scalability of the MOZART-3 model at ECMWFs computer led to acceptable run time within the coupled system. However, the MOZART-3 run time is still three times longer than the one of the IFS using only 25% of the CPUs. Further improvements in the run time of TM5 and MOCAGE are required to achieve acceptable run time within the coupled system.

The overhead because of the coupling can be attributed to the couplers set-up phase (only once per run) and the time of the data transfer and interpolation at every coupling time step. In the given setup the overhead is below about 3 % of the IFS stand-alone run time and about 1 % of the overall run time with coupled system IFS-MOZART-3.

A further constrained is the memory consumption of the component models and the OASIS4 coupler. The memory consumption of the coupler is occurs only temporarily during the exchange events but can reach up to 60 % of the IFS memory consumption, 15 % of the MOZART-3 consumption and 12 % of the total consumption. Figure 3 shows the memory consumption for each mpi-process of the coupled system IFS-MOZART-3.

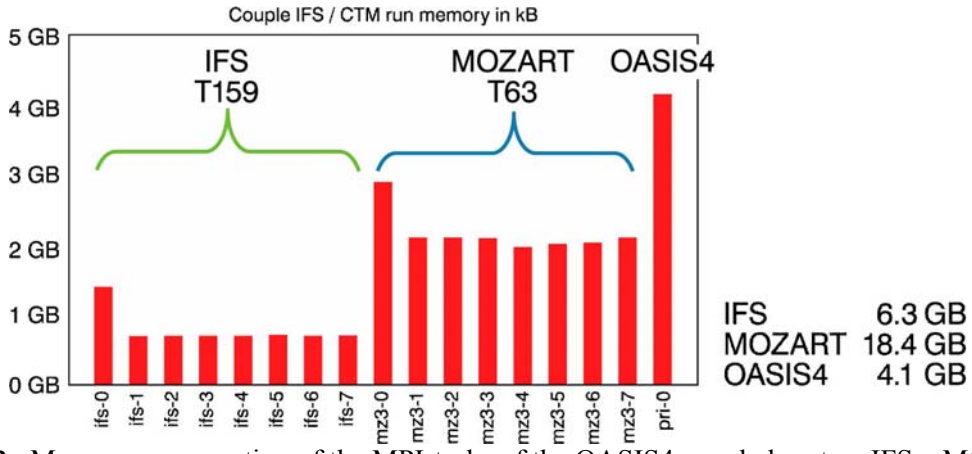


Figure 3 : Memory consumption of the MPI-tasks of the OASIS4 coupled system IFS – MOZART.

2.4. Dislocation and Feed-Back Delay

In the case of the GRG coupled system, both the IFS and the ACTM simulate both atmospheric transport processes. Different advections scheme or spatial and temporal resolutions may lead to different concentrations fields in the IFS and the ACTM. Thus, the applied ACTM tendencies can be inconsistent with the concentration fields in the IFS. The most annoying consequence would be negative concentration values in the IFS, due to un-balanced loss processes.

One example of the dislocation problem is depicted in Figure 4. O_3 tendency data due to chemical conversion (P&L, green circle) shall be given from a ACTM to IFS which does not simulate chemistry. If the O_3 fields in ACTM and IFS are dislocated, the tendencies data will be applied in the wrong part of the model domain.

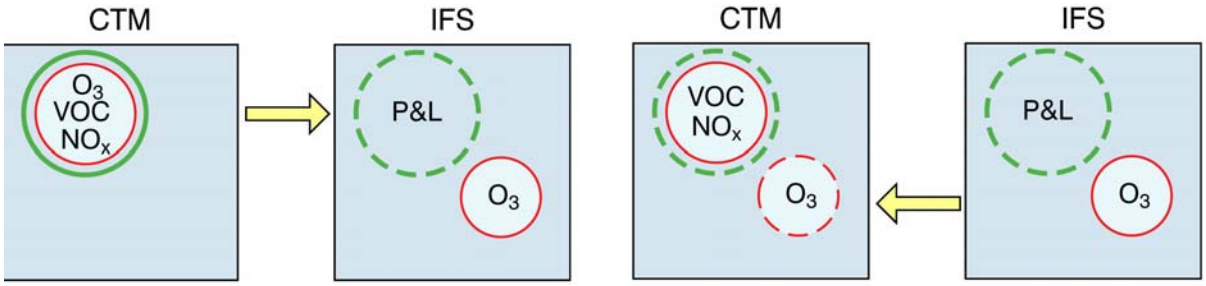


Figure 4 : Dislocation problem: A mismatch in the ozone fields (O_3, red) between ACTM and IFS causes a mismatch in the application of ozone tendency data (P&L, green) transferred from ACTM to IFS.

Two-way coupling is required if one wants to study the feedback of processes not included in the models. However, the time scales for the interaction is limited at least by two times the coupling interval. Two-way coupling requires a synchronous run of the two models. Lagged two-coupling, in which one of the component models runs ahead of the other model, is possible if the first model is not sensible to delayed input from the model running behind. Legged two-way coupling can be an option in atmosphere-ocean coupling but is was no option for the GRG-system.

In contrast to one-way coupling or lagged two-way coupling, the information for the next time step is not available in two-way coupling. This makes impossible to forward-interpolate the external data, e.g. meteorological fields, in time. Instead, they have to be assumed to be constant over the coupling interval.

3. Specific Issues of the GRG Coupled System

3.1. Formulation of Tendency Terms

The exchange of concentration tendencies, rather than concentrations, is a special and perhaps unique feature of the GRG coupled system. The formulation of the tendency terms has to reflect the operator splitting and time stepping in the both the ACTMs and the IFS as well as the relation between the tendency and the respective concentration value, and the cost (memory, time) of the exchange.

The ACTM use an operator-splitting approach in which chemistry, emission injection, diffusion and deposition are called in sequence and the update of the concentration follows directly within each subroutine.

The total tendencies T is given by the sum chemical loss L_C and production P_C, gain due to emissions P_E and loss L_E due to deposition.

$$T = P_C - L_C + P_E - L_D$$

Deposition L_D and chemical loss L_C and are proportional to the tracer concentration x and a relative formulation $L = l\ x$, i.e. a loss rate l, would better link tendency and concentration value and would help to avoid negative concentration. However, the output arguments of chemical routines provide total tendencies (P_C + L_C) for each time step and it would be difficult to distinguish production and loss. The relative formulation of the production is not advisable because it could cause high concentrations values to become even higher. One option would be to link the loss to OH concentrations only.

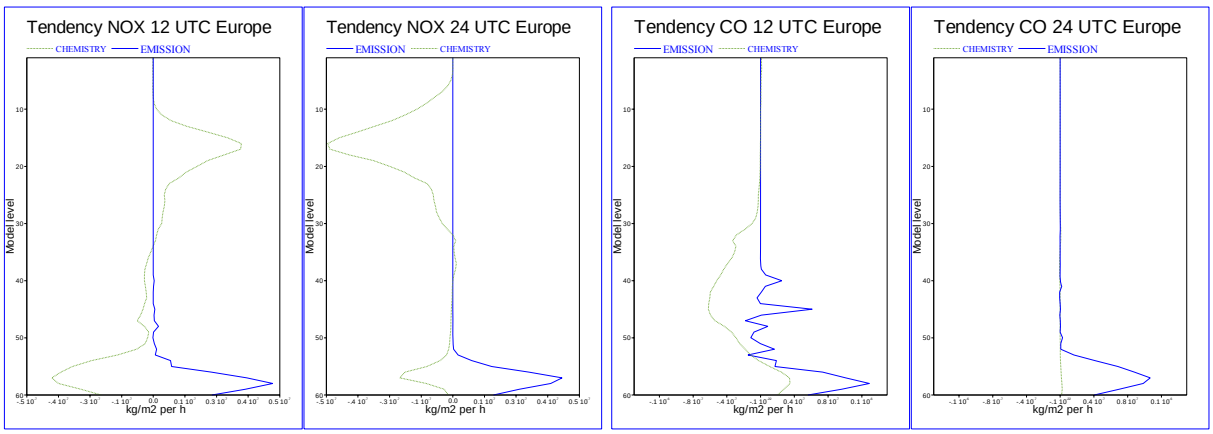
A disadvantage of separating production and loss, which tend to be much larger in absolute values than the resulting total, seems to be the separate interpolation of these fields. The sum of the interpolated production and loss terms may suffer from in-balances close to strong gradient, in particular if non-linear interpolation is applied.

Emissions are independent of the tracer concentration and can be considered as a surface flux. The injection of the emissions is integral part of the diffusion scheme on MOZART-3, i.e. as lower boundary for the fluxes, whereas TM5 and MOCAGE distribute the injected mass in a fixed ratio over selected layers in the boundary layer and apply their diffusion operator after the injection. The tendencies of the emissions P_E, therefore, have to be formulated either as 3D field including the diffusion or as 2D flux term. The diffusion in the IFS would have to be switched off if the 3D emissions-diffusion tendencies are applied. Air born emissions such as the ones from aircraft would have to be included in the 3D chemistry tendencies, if the surface emissions are expressed as a flux.

Dry deposition occurs at the lowest level and could be expressed both as tendency for the lowest layer or as a flux. Wet deposition would be a 3D tendency field.

The consideration of the arguments discussed above led to the following implementation of tendency extraction within the ACTM:

1. Process-specific 3D tendencies are determined by calculating the difference of the concentration fields before and after each of the chemistry, emission/diffusion and deposition subroutines.
2. The process-specific 3D tendencies are averaged over the coupling interval
3. The process-specific tendencies is either added up to **one** 3D total tendency field or added up to **two** 3D tendency fields containing (i) chemistry and wet deposition and (ii) emission and dry deposition.
4. The one or two 3D tendency fields are transferred to the IFS.



(a) (b) (c) (d)

Figure 5 : Profile of the area averaged (over selected domain) tendencies in kg/m^2 per model level due chemistry and emissions including vertical diffusion and convection for (a,b) NO_x and (c,d) CO at 12 and 24 UTCs.

Depending on a control switch, the three-dimensional emission and dry deposition tendencies can be converted into a surface flux by calculating the total column integral within the IFS.
Figure 5 shows profiles of the tendencies due to chemistry and wet deposition as well as emissions including vertical diffusion and convection for NO_x and CO at 12 and 24 UTC. The data is area-averaged over Central Europe (42N/-10W - 55N/10E) and shown in units of kg/m^{2}s to demonstrate the mass contribution of each model level. Model levels 60-50 cover the PBL, the tropopause is about at level 30. Clearly visible is the day-night difference of chemical loss and production. The emissions are a constant source term but the vertical tendency profiles are shaped by the vertical exchange in the boundary layer.

3.2. Implementation of GRG-Tracers Tendency Application in the IFS

The simplified sequence of the simulation of a passive tracer within an IFS time-step is as follows:

1. Calculate tendencies due to semi-Lagrangian advection scheme;
2. Calculate tendencies due to “physics”:
    a. Calculate tendencies due to surface flux injection and vertical diffusion within one routine;
    b. Calculate tendencies due to convection;
    c. Calculate tendencies due “other” processes (e.g. chemistry parametrization);
3. Update concentration fields from the start with accumulated tendencies of advection and “physics”.

Although the process-specific tendencies are stored for the update in a final step, the processes are not treated in an independent, i.e. parallel way. This is because the diffusion-routine uses the concentrations updated with dynamical tendencies and the convection routine uses the concentration updated with the diffusion (and surface flux) tendencies.
The position of the processes in the time loop is influenced by whether the process is fast or slow in respect to the time scale. More details on the implementation of the IFS physics can be found in *Beljaars, 2004*. Since the coupling interval (1-3 h) is larger than the model time step, the processes parameterised by ACTM input will appear as slow processes, even if the actual chemical conversion can be rather quick in the ACTMs.
In the IFS, emission injection and diffusion are part of one subroutine. Surface emissions, and likewise (dry) deposition, can be treated as surface fluxes. If the applied ACTM tendencies

already included the effect of diffusion and convection, the respective routine in the IFS physics would have to be switched off for this GRG-tracer.
The application ACTM source and sink information can be implemented in two modes:

1. IFS with complete ACTM "physics" for tracers: All "physics" tendencies (diffusion, convection, emission, chemical conversion, deposition) come from ACTM.
2. IFS with ACTM chemistry tendencies (3D) and ACTM surface fluxes (emission and dry deposition)

In the first mode, the IFS would only advect the GRG-tracers. The ACTM tendency field, consisting of the contributions of all source and sink processes would be consistent in itself. Dislocation could occur due to different advection in the ACTM and the IFS.
In the second mode, a consistent treatment of the emission injection and vertical transport would be achieved. In particular, the adjoint formulation of diffusion and convection in data assimilation would be consistent with the forward model. However, dislocation of the chemistry tendencies is more likely than in case 1 because the IFS concentration fields tend to differ more from the ACTM fields.

3.3. A Diagnostic NO_x Inter-Conversion Operator for Fast Reaction not Captured by the Coupled Approach

The fast and quickly moving diurnal NO_2 - NO inter-conversion caused by solar radiation in the upper stratosphere could not be handled by the coupled system with a coupling frequency of one hour. Instead of a steady movement of the day-night border, a "carved" stripe-shaped concentration fields were simulated. Therefore, it was decided to use NO_x as the model variable since the chemical development of the NO_x fields is not so strongly influenced by solar radiation and the development of the NO_x fields can be simulated by the coupled system.
Since the satellite observations to be assimilated are NO_2 data, a diagnostic NO_x to NO_2 inter-conversion operator *H* was developed. For the application in 4D VAR data assimilation it's tangent linear **H** and adjoint $\mathbf{H}^{\mathbf{T}}$ had to be coded.
The inter-conversion operator is based on a simple chemical equilibrium between the NO_2 photolysis rate j_{NO2} and the O_3 concentration:

$$\frac{[NO_2]}{[NO_x]} \approx \frac{k_2 [O_3]}{j_{NO_2} + k_2 [O_3]}$$

The diagnostic NO_2/NO_x ratio depends on the following variables:

- Solar zenith angle;
- O_3 concentration;
- Slant O_3 column above;
- Temperature.

A parameterised approach for the calculation of clear-sky NO_2 - photolysis j_{NO2} rates was used based on the band scheme by *Landgraf and Crutzen (1998)* in combination with actinic fluxes parameterised following *Krol and Van Weele (1997)*. The diagnostic operator does not reflect the influence of clouds on j_{NO2}, and the adjustments to the equilibrium because of hydro-carbons lower troposphere and abundant O-radical in the higher stratosphere and mesosphere.
The missing cloud influence might be tolerable since the NO_2 observations tend to be restricted to conditions with small cloud cover (*Boersma et al., 2004*).
The inter-conversion operator links the NO_2 to the O_3 concentration in data assimilation. It is the first step towards the consideration of more chemical relationships within the GRG data assimilation system.

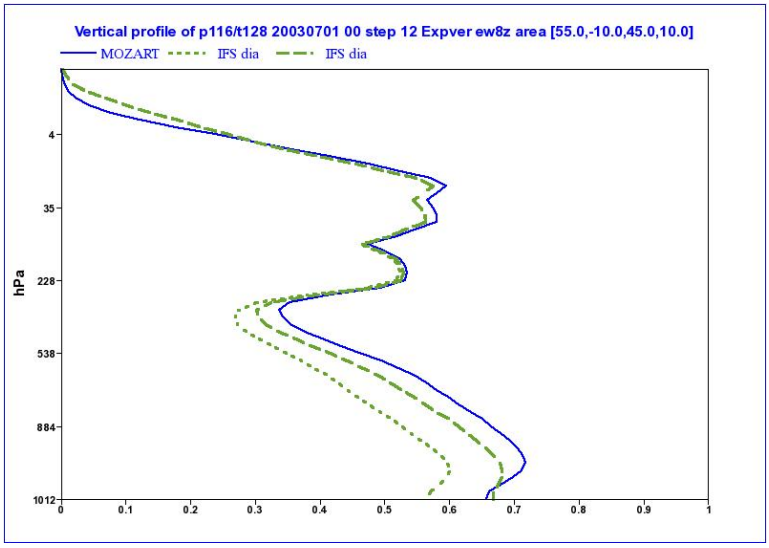


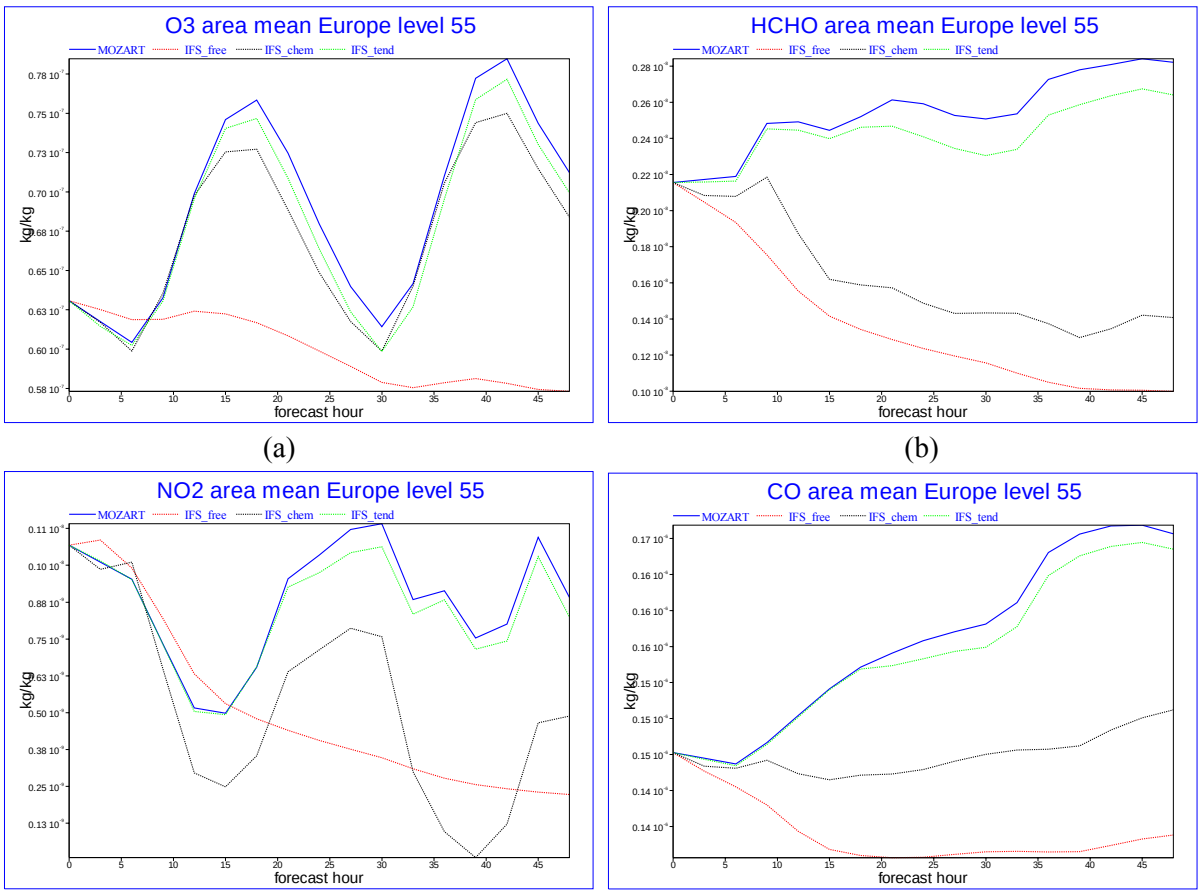
Figure 6 : NO_2-NO_x ratio profile averaged over Europe on 07 Jan 2002, 12 UTC, taken from MOZART directly and from the diagnostic inter-conversion operator without (IFS_dia dotted) and with and ad-hoc assumption of tropospheric peroxy-radical concentration (IFS_dia dashed).

Figure 6 shows a profile of the NO_2/NO_x ratio over Europe at 12 UTC calculated by the diagnostic operator and directly from the MOZART NO and NO_2 fields. An ad-hock approach of assuming a per-oxy-radical (HO_2 + RO_2) concentration 80 ppt (*Kleinman et al. 1995*) in the troposphere, multiplied by the cosine of the solar zenith angle to account of the diurnal cycle of the in the per-oxy-radical concentration, improved further the match of the NO_2/NO_x ratio between the diagnostic operator and MOZART.
Photolysis frequencies in MOZART-3 are based on tabulated values of the Tropospheric Ultraviolet and Visible radiation model ((TUV) version 3.0) (*Madronich and Flocke, 1998*) for clear sky conditions. The adjustment for cloudiness is described in *Brasseur et al. (1998)*.

4. Testing the Scientific Integrity of the GRG Coupled System in Forecast Mode

The integrity of the coupled system depends on whether the application of external tendency fields accounting for processes not included in IFS (chemistry, emission and deposition) give reasonable results of the forecast length. The objective is that the IFS is able to imitate the ACTM concentration developments and does not produce to many negative concentrations due to dislocated loss processes.
We studied area-averaged time series of tracer concentrations and spatial patterns of concentrations fields. The following IFS runs are compared with the MOZART concentrations:

- IFS_free: Initial conditions from MOZART, IFS transport;
- IFS_tend: Initial conditions from MOZART, IFS advection and ACTM sink& source tendencies including vertical transport;
- IFS_chem: Initial conditions from MOZART, IFS transport, ACTM sink & source tendencies excluding vertical transport.



(a) (b)

(c) (d)

Figure 7 : Time series of area mean of pollutant (a) O_3, (b) HCHO, (c) NO_2, and (d) CO concentration over selected European domain simulated with the MOZART, with IFS using no tendency information (IFS_free), with IFS using ACTM source and sink tendency information including diffusion and convection (IFS_tend) and with IFS using ACTM source and sink tendency and IFS vertical transport and emission injection (IFS_chem).

Figure 7 shows examples of time series of the area average over Europe of the GRG species for model level 55 (PBL niveau) of the three IFS runs and the MOZART simulation. If total ACTM tendencies (IFS_tend) are applied, the IFS can imitate the ACTM up to a forecast length of 48 h. Differences are obvious if the IFS vertical transport scheme is applied, because the vertical transport schemes differ between MOZART and the IFS. Spurious negative NO_x concentrations were detected during the night time, when the IFS vertical transport was applied (IFS_chem).

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2.3 The PRISM support initiative, COSMOS and OASIS4: *René Redler[1], Sophie Valcke[2], and Helmuth Haak[3]*

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Abstract
The increasing complexity of Earth System Models (ESMs) and computing facilities puts a heavy technical burden on the research teams active in climate modelling. The **PR**ogram for **I**ntegrated Earth **S**ystem **M**odelling (PRISM) provides the Earth System Modelling community with a forum to promote sharing of development, maintenance and support of standards and software tools used to assemble, run, and analyse ESMs based on state-of-the-art component models (ocean, atmosphere, land surface, etc..) developed in the different climate research centres in Europe and elsewhere. PRISM is organised as a distributed network of experts who contribute to five "PRISM Areas of Expertise" (PAE): 1) Code coupling and input/output, 2) Integration and modelling environments, 3) Data processing, visualisation and management, 4) Meta-data, and 5) Computing. Some of the tools and concepts developed within PRISM have been incorporated in the **CO**mmunity Earth **S**ystem **MO**del**S** (COSMOS) project, like the PRISM Compile and Runtime Environment and the OASIS3 coupler. Within PRISM further development is on-going, one example is the OASIS4 coupler, which targets next generation of the Earth system models.

1. The PRISM Concept, Goals, and Organization

PRISM (http://www.prism.enes.org/) was initially started as a project under the European Union's Framework Programme 5 (FP5, 2001-2004) and its long term support is now ensured by multi-institute funding of 7 partners (CERFACS, France; NEC-CCRLE-NEC, Germany; CGAM, UK; CNRS, France; MPI-M&D, Germany; Met Office, UK; and ECMWF) and 9 associate partners (CSC, Finland; IPSL, France; Météo-France, France; MPI-M, Germany; SMHI, Sweden; and computer manufacturers CRAY, NEC-HPCE, SGI, and SUN) contributing with IT and Earth science experts to the PRISM Support Initiative (PSI), currently for a total of about 8 person-years per year.

The PRISM concept, initially a Euroclivar recommendation, is to increase what Earth system modellers have in common today (compilers, message passing libraries, algebra libraries, etc.) and share also the development, maintenance and support of a wider set of ESM software tools and standards. This should reduce the technical development efforts of each individual research team, facilitate the assembling, running, monitoring, and post-processing of ESMs based on state-of-the-art component models developed in the different climate research centres in Europe and elsewhere, and therefore, promote the key scientific diversity of the climate modelling community. As demonstrated in other fields, sharing software tools is also a powerful initiative for increased scientific collaboration. It also stimulates computer manufacturers to contribute, thereby increasing the tool portability and the optimization of next generation of compute server for ESM needs, and also facilitating computer manufacturer procurement and benchmarking activities. The extensive use of the OASIS coupler illustrates the benefits of a successful shared software infrastructure. In 1991, CERFACS was commissioned to realise specific software for coupling different geophysical component models developed independently by different research groups. The OASIS development team strongly focussed on efficient user support and constant integration of the developments fed back by the users. This interaction snowballed and resulted in a constantly growing community. Today, the OASIS development and support (including OASIS3) capitalises about 25 person-years (py) of mutual developments and fulfils the coupling needs of about 15 climate research groups around the world. The effort invested therefore represents, at a first order, 25 py/15 groups = 1,7 py/group, which is certainly much less than the effort that would have been required by each group to develop its own coupler. In addition to that about 8 py have been invested so far into the development of OASIS4.

PRISM represents the first major collective effort at the European level to develop ESM supporting software in a shared and coherent way. This effort is recognised by the Joint Scientific Committee (JSC) and the Modelling Panel of the World Climate Research Programme (WCRP) that has endorsed it as a "key European infrastructure project". It is analogous to the ESMF project (http://www.esmf.ucar.edu) in the United States.

PRISM is lead by the PRISM Steering Board (one member per partner) that reviews each year a work plan proposed by the PRISM Core Group composed of PSI Coordinator(s), the leaders of the PRISM Areas of Expertise (see the next paragraph), and the chair of the PRISM User Group. The PRISM User Group is composed of all climate modelling groups using the PRISM software tools; given the dissemination of the OASIS coupler, the PRISM User Group is already a large international group.

1.1. PRISM Areas of Expertise

PRISM is organised around five PRISM Areas of Expertise (PAEs) having the following remits:

- Promote and, if needed, develop software tools for ESM. A tool must be portable, usable independently and interoperable with the other PRISM tools, and freely

available for research. There should be documented interest from the community to use the tool and the tool developers must be ready to provide user support.
- Encourage and organise a related network of experts, including technology watch.
- Promote and participate in the definition of community standards where needed.
- Coordinate with other PRISM areas of expertise and related international activities.

PAE "Code coupling and I/O"

The scope of the PAE "Code coupling and I/O" is to:

- develop, maintain, and support tools for coupling climate modelling component codes.
- ensure a constant technology watch on coupling tools developed outside PRISM.
- keep strong relations with the different projects involving code coupling in climate modelling internationally.

The current objectives are to maintain and support the OASIS3 coupler, continue the development of the OASIS4 coupler (see below), but also, through the organisation of workshops, help the community understand the different technical approaches used in code coupling, for example in the PALM coupler (*Buis et al., 2006*), in the UK Met Office FLUME project (*Ford and Riley, 2003*), in the US ESMF project (*Killeen et al., 2006*), and in the Bespoke Framework Generator (BFG) from U. of Manchester (*Ford et al., 2006*) used in the GENIE project. Currently OASIS3 is used for coupling between the COSMOS components (see section 2).

PAE "Integration and Modelling Environments"

This PAE targets the following environments:

- source version control for software development (including model development);
- code extraction and compilation;
- job configuration (how to set up and define a coupled integration);
- job running (how to control the execution of a coupled integration);
- integration with archive systems.

For source version control, PRISM promotes the use of subversion (*Pilato et al., 2004*) and recently moved from CVS to Subversion for its own software distribution server, now located at DKRZ in Hamburg.

Controlling the creation of executable and providing a suitable run time environment were seen as key integration activities within the EU FP5 PRISM project, resulting in the development of the PRISM Compile and Running Environments (SCE & SRE). These tools are built on the concept that software from multiple sources can use a single framework as long as those models can conform to a set of simple standards.

Different groups (ECMWF, IPSL, CERFACS) have also shown strong interest in the UK Met Office Flexible Configuration Management (FCM) tool for version control and/or compilation management. A further review will be conducted in those groups and this tool, together with Subversion, may be considered as a replacement to CVS and extend the simplicity of 'make' in the current SCE. The COSMOS components follow these standards and are integrated into the PRISM SCE and SRE (see section 2).

PrepIFS is a flexible User Interface framework provided by ECMWF that allows tailored graphical user interfaces to be built for the configuration of models and other software. It is integrated within the Supervisor Monitor Scheduler (SMS) for the management of networks of jobs across a number of platforms and both products are developed using Web Services technology. SMS and prepIFS have recently been packaged for use within the Chinese climate community. The power of these tools is recognised by PRISM, even if they are not

widely used in the European climate community because of the level of commitment and human resources required to run these sophisticated services.
ECMWF is currently developing "prepOASIS", a Graphical User Interface based on prepIFS but suitable to be run stand alone, to configure a coupled model using the OASIS4 coupler.

PAE "Data processing, visualisation and management"

The overall objective of this PAE is development of standards and infrastructure for data processing, archiving, and exchange in ESM and more general Earth System Research (ESR). The huge amounts of data in ESR do not allow for centralised data archiving. Networking between geographically distributed archives is required. Ideally the geographical distribution of federated data archives is hidden to the user by an integrative graphical WWW-based interface. Standards are required in order to establish a data federation and to work in network.
For data processing, this PAE currently analyses the Climate Data Analysis Tools (CDAT) and Climate Data Operators (CDO) respectively maintained and developed by PCMDI and MPI-M.
The M&D Group also develops the CERA-2 data model for the World Climate Data Centre, proposing a description of geo-referenced climate data (model output) and containing information for the detection, browse and use of data. An important collaboration is going on with the PAE Metadata and other international initiatives for the development and implementation of metadata standards for the description of model configuration and numerical grids.
Collaboration with PRISM related data archives in the development of data networking and federated archive architectures is also going on. ECMWF MARS software may be another candidate tool for meteorological data access and manipulation even if it is likely to be of interest only to major NWP sites due to its complexity.

PAE "Metadata"

In the last few years metadata has become a hot topic with new schemes and ideas to promote the interchangeability of ESMs or modelling components as well as data. The PRISM Metadata PAE provides a forum to discuss, develop, and coordinate metadata issues with other national and international projects. The fundamental objective is to develop, document, and disseminate ESM metadata schemes as well as tools for producing, checking and displaying metadata. Currently, this PAE offers an opportunity to ensure coherence between the following metadata definition efforts:

- Numerical Model Metadata (NMM; http://cgam.nerc.ac.uk/NMM), developed at University of Reading, is an evolving international metadata standard intended for the exchange of information about numerical code bases, and the models/simulations done using them.
- The CURATOR project (Earth System Curator; http://www.earthsystemcurator.org), a project similar to NMM in the US.
- Numerical grid metadata, developed by Balaji at GFDL, USA, for numerical grid description (http://mitgcm.org/eh3/people/balaji/balaji-gridmeta2005.pdf).
- The CF convention for climate and forecast metadata Interface (*Eaton et al., 2003*) designed to promote the processing and sharing of files created with the netCDF Application Programmer, developed by an international team and now managed by PCMDI and BADC.
- The metadata defined by the OASIS4 developers for description and configuration of the coupling and IO interface in a coupled model.

- The metadata currently defined in the UK Met Office FLUME project to manage and define the process of model configuration.

The goal of this PAE is therefore to integrate these emerging standards, ensure that they meet requirements and needs of the ESM community, and disseminate them as a "good practice".

PAE "Computing"

Experience has shown that a large variety of technical aspects related to computing are highly important for Earth system modelling. These techniques are in constant flow and evolve with new hardware becoming available. While computer vendors have to be kept informed about requirements emerging from the climate modelling community of Earth system modellers still have to be informed about computing issues to preview difficulties and evolutions. PRISM can play a role in that aspect through the new PAE "Computing" devoted to those technology trends. Possible technical topics are file IO and data storage, algorithmic development, portable software to fit the needs of parallel and vector systems.

It is first proposed to establish a group of people willing to contribute with their expertise via mailing list and a sharing of relevant information on the PRISM web site. In particular the activities will cover sharing of knowledge from the work on the Earth Simulator, establishment of links with the DEISA project (http://www.deisa.org), and providing information about important conferences and workshops. Depending on the number of volunteers and the input from this group, the list of tasks will be revised or extended in the next years.

2. COSMOS : COmmunity Earth System MOdelS

The idea behind the **CO**mmunity Earth **S**ystem **MO**del**S** (COSMOS) is that a single research institution alone cannot develop the most comprehensive models. COSMOS is a community network towards the development of a fully developed ESM. The complexity of ESMs requires the involvement of an interdisciplinary team of scientists to develop ESMs. This team has to be ready to work together intensively and share the knowledge and expertise of the team partners. So, COSMOS constitutes a team of experts to develop a flexible and portable model infrastructure, following and supporting the ideas of the PRISM initiative. The purpose is not only to develop models and their infrastructure, but also to use them to address challenging problems involving the interactions between different components of the Earth system. These models will be central tools to assess important feedback processes in the system, to assess environmental risks, and to develop mitigation and adaptation strategies.

2.1. Organisation

The most important body of the COSMOS network - community of Earth system scientists - is ready to work with scientific tools and endeavours to promote. The community meets, at least, once a year for the COSMOS General Assembly. At these meetings it takes strategic decisions, e.g. new scientific co-operation and projects. It also determines the configuration of the board (12 members). The members need to come from institutions having signed the COSMOS Memorandum of Understanding. Three of these members are one Chair and two Co-chairs; the board decides about these positions. The board calls activity heads for working groups and other activities. The activity heads assignment is to organise and run groups to think about and act upon projects, methods, aspects, etc. of ESM. The activity heads report about the activities of their enterprises to both the board and community. In their daily business they interact intensively with the COSMOS office.

The COSMOS office consists of the Science Director, an ex-official member of the board, and a Project Manager running day-to-day business. The office gets secretarial assistance for office issues and meeting organisation. Support positions for technical work on the model system as well as outreach activities are applied for. The office supports the chair and the co-chairs. It is assigned by the board, and it develops and proposes the strategies and plans for COSMOS.

2.2. The COSMOS Models

The COSMOS network crucially depends upon the availability and easy access to models. These models do not only have to be very well tested and mature, but also scientifically and technologically very advanced. The MPI-M has developed a suite of models in a framework, called "COSMOS v1" (http://www.mpimet.mpg.de/en/wissenschaft/modelle/model-distribution/available-models.html). The configurations possible with this modelling framework are indicated in Table 1.

Table 1 : Model Configurations with COSMOS v1.

	Model Components
Atmosphere GCM	ECHAM5
Ocean GCM	MPIOM
Ocean + Biogeochemistry	MPIOM-HAMOCC
Atmosphere / Ocean GCM	ECHAM5 / OASIS3 / MPIOM
Carbon Cycle	ECHAM5-JSBACH/OASIS3/MPIOM-HAMOCC
Aerosol system	ECHAM5-HAM / OASIS3 / MPIOM-HAMOCC

This is available to the COSMOS network. It is assembled following the PRISM philosophy:

- Coupling of atmosphere and ocean GCMs by OASIS3;
- Configuration of model types and building of executables for specific machines by the standard configuration environment (PRISM SCE);
- Running and data storage by using the standard runtime environment (PRISM SRE) where model configurations can be ported to, run, maintained and developed on supercomputers with relative ease.

This first version of the model of the COSMOS network, called COSMOS v1, has 28 users from 10 countries.

2.3. Atmosphere Chemistry

The chemistry of the atmosphere can be simulated with the new MESSy approach, the Modular Earth Submodel System (http://www.messy-interface.org). It has been successfully coupled to ECHAM5, and applied in multiyear integrations in different configurations and resolutions. ECHAM5/MESSy can be used to simulate ozone and related chemistry of the lower and middle atmosphere up to the mesopause at about 80 km, with no artificial boundaries applied, e.g., between the troposphere and stratosphere. The modular structure allows the selection of particular configurations to increase or decrease the level of details in describing processes such as tropospheric multiphase chemistry, aerosols, transport and deposition. The COSMOS v1 package integrates in a flexible and modular way models for the circulation of the atmosphere, ocean and sea ice, and optionally includes processes for aerosols, vegetation, and marine biogeochemistry. The integration of atmospheric chemistry into the COSMOS system will lead to the next generation: COSMOS v2. This will take profit from the ECHAM5 based atmospheric chemistry models:

- ECHAM5-MESSy for tropospheric and stratospheric chemistry (already distributed);

- ECHAM5-HAMMOZ for coupled aerosols and chemistry in the troposphere;
- HAMMONIA for the neutral and ionized chemistry, covering the entire atmosphere.

The PRISM standard environments of COSMOS v2 will allow for different model configurations, including atmospheric chemistry.

3. The OASIS4 Coupler

The OASIS coupler software allows synchronized exchanges of coupling information between numerical codes representing different components of the climate system.
OASIS3 (*Valcke, 2006*) is the direct evolution of previous versions of the OASIS coupler. In addition a new fully parallel coupler OASIS4 is developed within PRISM (*Valcke and Redler, 2006*). Other MPI-based parallel coupling software performing field transformation exists, such as the Mesh based parallel Code Coupling (MpCCI; http://www.scai.fraunhofer.de/mpcci.html) or the NCAR CCSM Coupler 6 (Cpl6; http://www.ccsm.ucar.edu/models/ccsm3.0/cpl6). The originality of OASIS in general lies in its great flexibility (as the coupling configuration is externally defined by the user) and for OASIS3 and OASIS4 in the additional add-on of the common treatment of coupling and I/O exchanges (again externally defined by the user).
As the climate modelling community is progressively targeting higher resolution climate simulations on massively parallel platforms with coupling exchanges involving a higher number of (possibly 3D) coupling fields at higher coupling frequencies, a completely new fully parallel coupler OASIS4 is developed within PRISM. OASIS4 is a portable set of Fortran 90 and C routines. At run-time OASIS4 acts as a separate parallel executable, the OASIS4 Driver-Transformer, and as a fully parallel model interface library, the OASIS4 PSMILe. The concepts of parallelism and efficiency like the parallel neighbourhood search drove OASIS4 developments, keeping at the same time in its design the concepts of portability and flexibility that made the success of OASIS3 and its predecessors.

3.1. Coupling Configuration

Each component model to be coupled via OASIS4 should be released with an eXtensible Markup Language (XML; http://www.w3.org/XML) file describing all its potential input and output fields, i.e. the fields that can be received or sent by the component through PSMILe ***get*** and ***put*** actions in the code. Based on those description files, the user produces, either manually or via a Graphical User Interface, the XML configuration files. As for OASIS3, the OASIS4 Driver extracts the configuration information at the beginning of the run and sends it to the different model PSMILes, which then perform the appropriate coupling or I/O actions during the run. OASIS4 is also highly flexible in the sense that any duration of run, any number of component models, any number of coupling and I/O fields, and particular coupling or I/O parameters for each field, can be specified.

3.2. Process management

In a coupled run using OASIS4, the component models remain separate executables. If only MPI1 is available (*Snir et al,. 1998*), the OASIS4 main processes (driver plus transformer routines) and the component models must all be started at once in the job script. If some or all of the components are programmed as subroutines of a main program (e.g. ocean and sea ice as subroutines of ocean general circulation model) these components can still be coupled via OASIS4 PSMILe routines provided that the subroutines are run concurrently. This can usually be achieved by running the subprograms on different processes which is configurable through the XML file and the use of appropriate PSMILe interface routines. If the MPI library

supports the MPI2 standard (*Gropp et al., 1998*) the user has the option to start only the OASIS4 driver process which then launches the different component models and transformer processes using the *MPI_Comm_spawn_multiple* functionality. The OASIS4 driver can spawn the different processes on different machines. In both cases, all processes are necessarily integrated from the beginning to the end of the run, and each coupling field is exchanged at a fixed frequency defined in the XML file for the whole run. In that sense, OASIS4 supports static coupling only.
Figure 1 illustrates the different ways of communication between the individual physical components with a coupled application. Ocean and sea ice are programmed as subroutines of a main program. In this example we assume that the two grids are identical and data can be exchanged directly through the PSMILe bypassing the transformer processes. Atmosphere and chemistry are programmed as separate executables. As they again work on identical grids data are exchanged directly (indicated by the lower red double arrow). As those grids are different from the ocean and sea ice data exchanged with those components go through the parallel transformer.

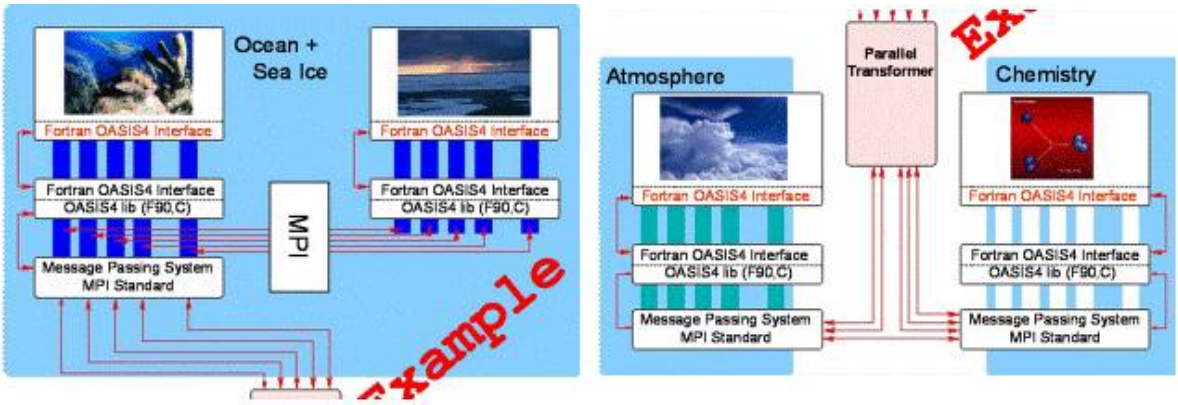

Figure 1 : Example for a possible coupled model configuration.

3.3. Coupling Field Transformation and Regridding

During the run the OASIS4 Driver (root) process takes over the functionality of the transformer and participates together with the other Transformer processes manages the transformation and regridding of 2D or 3D coupling fields. The (parallel) Transformer performs only the weight calculation and the regridding *per se*; the neighbourhood search, i.e. the search of the source points determination for each target point that contribute to the calculation of its regridded value, is performed in parallel in the source PSMILe.
During the simulation time stepping, the OASIS4 parallel Transformer can be considered as an automaton that reacts to what is demanded by the different component model PSMILes: receive data for transformation (source component process) or send transformed data (target component process). The OASIS4 Transformer, therefore, acts as a parallel buffer in which the transformations take place. Currently, only 2D and 3D nearest-neighbour, 2D and 3D linear, and bi-cubic regridding, and 2D conservative remapping techniques are implemented, but there are plans to implement also 3D cubic grid interpolation and 3D conservative remapping.

3.4. Communication: the OASIS4 PSMILe Software layer

To be coupled via OASIS4, the component models have to include specific calls to the OASIS4 PSMILe software layer. The OASIS4 PSMILe Application Programming Interface

(API) was kept as close as possible to OASIS3 PSMILe API; this ensures a smooth and progressive transition between OASIS3 and OASIS4.
The OASIS4 PSMILe supports fully parallel MPI-based communication, either directly between the models for those pairs of source and target grid points for which an exact match was found (including automatic repartitioning if needed) or via the parallel Transformer, and file I/O using the GFDL mpp_io library. Note, the mpp_io library has been extended to work optionally with the parallel NetCDF library (pNetCDF; http://cucis.ece.northwestern.edu/projects/PNETCDF) to allow for parallel file I/O via MPI-IO inside pNetCDF. The detailed communication pattern among the different component model processes is established by the PSMILe, using the results of the regridding or repartitioning neighbourhood search. This search is based on the source and target identified for each coupling exchange by the user in the XML configuration files and on the local domain covered by each component process. The search uses an efficient multigrid algorithm and is done in parallel in the source PSMILe, which ensures that only the useful part of the coupling field is extracted and transferred.
Besides these new parallel aspects, the OASIS4 PSMILe follows the same end-point communication and user-defined external configuration principles than the OASIS3 PSMILe.

3.5. The OASIS4 Users

OASIS4 portability and scalability was demonstrated with different "toy" models during the EU FP5 PRISM project. OASIS4 was also used to realize a coupling between the MOM4 ocean model and a pseudo atmosphere model at the Geophysical Fluid Dynamic Laboratory (GFDL) in Princeton (USA), and with pseudo models to interpolate data onto high resolution grids at IFM-GEOMAR in Kiel, Germany.
Currently, work is going on with OASIS4 at the:

- Swedish Meteorological and Hydrological Institute (SMHI) in Sweden for coupling regional ocean and atmosphere models (first physical case studies are already realized);
- European Centre for Medium-Range Weather Forecast (ECMWF), KNMI in the Netherlands, and Météo-France in the framework of the EU GEMS project, for 3D coupling between atmospheric and chemistry models;
- UK MetOffice for global ocean-atmosphere coupling;
- CERFACS and Météo-France;
- ACCESS.

After the current beta-testing phase, the first official OASIS4 version became available to the public in 2007.

4. Final Remarks

PRISM is already a success as it allows a community of developers facing similar technical problems in ESM to share their expertise and ideas. The difference in the level of buy-in for the different tools developed during the FP5 project helped identified for which tool standardisation is more or less achieved, for which tools convergence is wanted, and is currently not a target. The strength of the current decentralised PRISM organisation is to allow "best of breed" software tools to naturally emerge, although this means that PRISM relies on the developments done in the different partner groups to propose technical software solution to Earth system modellers. In the areas for which this philosophy does not apply and for which standards have to be pre-defined, for example for metadata definition, the big contribution of PRISM is to provide a visible entry point of the European ESM software

community for international coordination, for example with the American ESMF project within the WCRP framework. Given its institutional long term support, PRISM is now well placed to seek additional funding to support more networking and coordination activities or to help specific technical developments. And of course, more European or non-European collaborators are today most welcome to bring in additional expertise and to ensure a wider diffusion of the PAE tools and standards.

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2.4 Integrated Modelling Systems in Australia : *Peter Manins, M E Cope, P J Hurley, S H Lee, W Lilley, A K Luhar, J L McGregor, J A Noonan and W L Physick*

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The Commonwealth Scientific and Industrial Research Organization (CSIRO) is Australia's principal research organisation with over 6000 scientific, technical and support staff at 65 sites around Australia. A small group of scientists, in the Division of Marine and Atmospheric Research, has been working on air pollution models, developing, documenting, disseminating, and applying them to environmental issues around Australia and abroad. An extensive literature in both international journals and reports is available (see http://www.cmar.csiro.au/search/pubsearch.htm for author or keyword entry to these).

Motivated to explore air pollution in complex geographic settings, in 1986 Bill Physick coupled the Pielke mesoscale meteorological model (Colorado State University) with

McNider's Lagrangian particle model (University of Alabama) in off-line mode (REF???). After replacing the meteorological model with one developed locally (by John McGregor, REF???), speeding up the particle model near boundaries, and adding a new advanced display system, the result was Lagrangian Atmospheric Dispersion Model (LADM), used in numerous coastal industrial air pollution settings *(Physick et al., 1994*).
Seeing the strengths and weaknesses of the LADM approach, Peter Hurley built a test bed to explore various turbulence schemes, non-hydrostatic effects, and further ideas for speeding up Lagrangian particle models; by 1999 these had come together on a PC platform as the GUI-driven in-line integrated mixed-Lagrangian/Eulerian modelling system - The Air Pollution Model (TAPM) (*Hurley, 1999ab*).

TAPM uses the fundamental equations of atmospheric flow, thermodynamics, moisture conservation, turbulence and dispersion, wherever practical. For computational efficiency, it includes a nested approach for meteorology and air pollution, with the pollution grids optionally able to be configured for a sub-region and/or at finer grid spacing than the meteorological grid, which allows a user to zoom-in to a local region of interest quite rapidly. The meteorological component of the model is nested within synoptic-scale analyses/forecasts that drive the model at the boundaries of the outer grid. The coupled approach taken in the model, whereby mean meteorological and turbulence fields are passed to the air pollution module every five minutes, allows pollution modelling to be done accurately during rapidly changing conditions such as those that occur in sea-breeze or frontal situations. The model incorporates explicit cloud microphysical processes. The use of integrated plume rise, Lagrangian particle, building wake, and Eulerian grid modules, allows industrial plumes to be modelled accurately at fine resolution for long simulations. Similarly, the use of a condensed chemistry scheme also allows nitrogen dioxide, ozone, and particulate mass to be modelled for long periods.
TAPM has become an important integrated modelling system in Australia and abroad, evolving to the present Version 3 (*Hurley, 2005ab*): there are 154 active licences, involving 18 countries. It is successful because: 1) ease to use, and 2) self-imposed limitations that make it very practical:

- its application is limited to a few thousand kilometers; so, major inter-continental transports are not accommodated;
- because we are focussed on air pollution problems, high-impact weather is rarely of concern; so, its simplified approach to deep convection, absence of a stratosphere (maximum altitude is 8 km), and its explicit but simplified rain processes, are not significant;
- it includes wet and dry deposition processes and a simplified photochemical smog mechanism that is only applicable to regional and urban pollution.

The most striking thing about TAPM for most applications by its target user group is that it only requires information on pollutant emissions. I.e. datasets of the important inputs (e.g., terrain, land use and 3D synoptic meteorology) are needed for meteorological simulations and provided with the model, allowing model set up for any region, although user-defined databases can be connected to the model if desired. In fact TAPM has been shown to give very good results without any recourse to local meteorological data. Many demonstrations of the veracity of this claim have been made (see *Hurley et al. (2005b)* for an extensive set of case studies), and more recent references are given already at the CMAR Library Web address.
A major learning for Australian conditions is that recirculation of pollutants in the sea breeze is an important feature of coastal cities: the spatial resolution required of the meteorological

model needs to be adequate to resolve the sea breeze phenomenon. Resolution substantially finer than 10 km is required (i.e. 2–5 km seems as adequate).
Using a high quality emissions dataset, TAPM performance is very good for the prediction of extreme pollution statistics, important for environmental impact assessments, for both non-reactive (tracer) and reactive (nitrogen dioxide, ozone and particulate) pollutants for a variety of sources (e.g. industrial stacks and surface or urban emissions) — see *Luhar and Hurley (2003)*; *Hurley et al., (2005a)*. A study, with the attention to detail that makes a big difference, is by *Luhar et al., (2006a)*.
Other findings include:

- correctly specifying land-use and vegetation improves the meteorological predictions and has flow-through benefits for the air quality predictions;
- the vertical temperature profiles in standard synoptic analyses often have poor detail below 1000 m. For example, extra-stable layers between 200 and 400 m can be non-existent in the forecast model analyses that have only a few levels below 1000 m. This can greatly affect air quality predictions;
- it is important to use wind data only from well-sited anemometers for evaluating model performance, or for assimilating during a run.

Characteristics of urban areas that can affect flow properties include roughness length, building characteristics, thermal properties of the surface and anthropogenic heat flux. As *Luhar et al. (2006b)* showed by comparison with the Swiss BUBBLE data, although TAPM in particular accounts for these effects with a varying degree of complexity, the land-surface scheme in the model needs improvement to resolve the urban canopy layer and the roughness sublayer. The topic of urbanisation of meteorological models is an area of increasing interest, and has been a focus of COST-728. But there are also important new developments for rough boundary layers in which shedding shear layers slow the flow markedly (*Harman and Finnigan, 2007*). These may have implications for understanding flows in and above urban canopies.

As TAPM was being developed, the opportunity arose to integrate its chemical transport component with Australian Bureau of Meteorology's weather forecasting system to provide real-time weather and air pollution forecasts. TAPM already had been using historical six-hourly BoM global analyses to initialise that model; so, the extension was natural. In collaboration with the BoM and the major environment authorities of Australia, we developed the Australian Air Quality Forecasting System (AAQFS) in time to be run operationally for the 2000 Sydney Olympic Games (see *Cope et al. (2004), Hess et al. (2004), Tory et al. (2004))*. The major emphasis for Sydney was on predicting urban ozone *(Cope et al., 2005a*). It has been run twice daily ever since, producing hourly forecasts for the next 36 h for 21 chemical species on a 1 km grid for the major cities of Australia. We have learnt a lot and have made many improvements.

- A big learning was that it is best to have as much of the emissions inventory as possible on-line, described by algorithms that respond where appropriate to the forecast meteorology, and calibrated by relevant observational data. We now have emissions from motor vehicles, vegetation (*Azzi et al., 2005; Kirstine and Galbally 2004*), soils, wind-blown dust, bushfires, sea-salt spray, domestic wood heating, and some industry handled this way.
- As with the finer-scale TAPM experience, it is vitally important to accurately include biogenic emissions. In Australia, concentrations of up to 60 ppb ozone are measured in country areas with no evident industry or vehicle sources.

- Intrusion of bushfire smoke is a major cause of exceedences of air quality standards, in particular for ozone, in Australian cities. This is an extreme example of biogenic emissions, and is a major driver for the next point.
- Australia-wide forecasts are now done for wind-blown dust (e.g., *Wain et al., 2006*) and for bushfire smoke. Size-segregated dust is emitted and transported depending on historical land-use, soil-type and seasonal leaf area index (LAI). For bushfires, we use the Sentinel outputs of hotspots to locate emissions. Sentinel currently obtains MODIS data from the NASA EO Satellites Terra and Aqua (see http://sentinel2.ga.gov.au/acres/sentinel).

AAQFS provided the impetus to further explore complex chemistries for air pollution predictions, something we had been doing for special projects in and around Australia (e.g, *Cope et al., 2003; Malfroy et al., 2005*). We are currently gaining experience with the Carbon Bond 2005 mechanism and have incorporated this into a new Chemical Transport Model that runs optionally in both TAPM and AAQFS (it is called TAPM-CTM). This complexity is essential for addressing policy questions of urban planning and around the veracity of new transport fuels such as ethanol blends, and the effects of pollutants such as formaldehyde and benzene. Pollution by ozone and fine particles, particularly secondary particles, are the main questions ultimately being addressed, though air toxics and personal exposure and the interaction with indoor air pollution are also questions increasingly being asked.

Recognition that personal exposure is the really relevant air pollution question for human health has led to a lot of work on near-road air quality, both experimentally and by modelling. The Lagrangian Wall Model (LWM: *Lilley and Cope, 2005; Cope et al., 2005b*), a complex chemical transport model of a wall that is advected downwind of nominated anchor points can resolve pollution concentrations to 10 m. The model runs within a TAPM grid cell. Some thirty or so walls can be set up and tracked at once from a GUI, giving high resolution results for the effects of individual roads, intersections and terrain on the air quality. Other relevant work integrates the results of environmental monitoring data, high resolution modelling of pollution fields and hospital admissions to seek to improve understanding of pollutant exposure to increases in asthma (*Physick et al., 2006; Physick et al., 2007*).

On a wider front, for downscaling from large-scale climate models we have developed the Conformal-Cubic Atmospheric Model (CCAM), a global model that has spatially varying resolution (*McGregor, 1997; McGregor and Dix, 2005*). It is initialised from a single global analysis or climate change prediction to predict scenarios of weather down to a kilometre for hours to months. Embedding TAPM off-line and with the possibility of using TAPM-CTM on-line in CCAM gives us a powerful GUI-driven integrated weather and air pollution forecasting system that has wide application.

Our current direction is to merge our research with Australian Bureau of Meteorology. A particular development underway is ACCESS, the Australian Community Climate and Earth System Simulator, based in large part on HADGEM in collaboration with the UK Hadley Centre (*Martin, 2004; John, 2004*). CCAM is expected to become an alternative dynamical core (*McGregor, et al., 2007*), and AAQFS will be unified into the system. For air quality applications, ACCESS will play the major role, but TAPM and LWM will continue to develop in parallel, providing very convenient test beds for process and algorithm developments before they are considered for incorporation into ACCESS and possibly HADGEM.

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2.5 Coupling of Air Quality and Weather Forecasting - Progress and Plans at met.no : *Viel Ødegaard, Leonor Tarrasón and Jerzy Bartnicki*

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1. Introduction

Air quality modelling at met.no consists of three different systems, all coupled off-line to our numerical weather prediction (NWP) models. These are: 1) a nuclear emergency system, 2) an

urban air quality (AQ) forecasting system and 3) a long-term air quality chemical transport model routinely used in Europe to determine transboundary pollution fluxes.
The first system, the "Severe Nuclear Accident Program" (SNAP) model was developed at met.no to allow emergency risk assessment (*Saltbones et al., 1995, 1998*). This is a Lagrangian particle model transporting gases, noble gases, particles of different size and density. The modeled processes are advection and diffusion by random walk, dry deposition with gravitational settling velocity parametrization for particles and wet deposition as function of size and precipitation for particles. The model is operated by forecasters and the Norwegian Radiation Protection Authority (NRPA) in case of nuclear accident. It runs on meteorological input from operational HIgh Resolution Limited Area Model (HIRLAM) (10 and 20 km horizontal resolution) and from ECMWF (*Bartnicki et al., 2005*).
The second system, the urban air quality information system runs operationally at met.no and consists of the chemical dispersion model AirQUIS developed at Norwegian Institute for Air Research (NILU) and the non-hydrostatic NWP model MM5 in 1 km horizontal resolution nested in HIRLAM (*Berge et al., 2002*). AirQUIS is an Eulerian gridpoint model with point source emissions, line source emissions and area source emissions. The prognostic components of the model are PM_{10}, cities in 1x1 km horizontal resolution in both meteorological and AQ models. It is used $PM_{2.5}$ and nitrogen dioxide (NO_2). The system runs daily for 48 h forecasts for 6 Norwegian cities, with main focus in forecasting of urban air quality during winter season. The cities under study are located in low elevated areas surrounded by hills and mountains. Winter time inversions inhibit ventilation of pollution and thus, main exceedances of critical pollution levels occur in Norway during that time. The forecasts are distributed to end-users via newspapers, mobile network, and internet.
The third system is the ACTM supporting the modelling work under the Co-operative programme for monitoring and evaluation of the long-range transport of air pollutants in Europe (the EMEP programme). The model used, the EMEP Unified model, is developed at met.no for simulating atmospheric transport and deposition of acidifying and eutrophying compounds, aerosols as well as photo-oxidants over Europe. The model is a multi-layer Eulerian model and is now flexible with respect to the choice of horizontal grid projection, domain, and resolution. The model can thus, be run at local, regional, hemispheric, and global scales. Typically, the model simulates one year period of the transport and the current results of the regional model runs are available for the years 1980, 1985, 1990 and each year from 1995 to 2004. The EMEP Unified model in operational configuration is the regional version that uses presently HIRLAM PS (a dedicated version of HIRLAM, which has been frozen for last 10 years) which runs on a polar stereographic grid with a 50 km resolution and covers Europe and the Atlantic Ocean. In vertical, the model has 20 sigma layers reaching up to 100 hPa. Approximately 10 of these layers are below 2 km in order to obtain high resolution of the boundary layer. The polar stereographic projection is historically bound to the EMEP reporting grid for emissions over Europe. A detail description of the model can be found in *Simpson et al. (2003)* and validation results are available in *Tarrason (2003)*. The EMEP model is also run at regional scale in forecast mode with input from ECMWF Integrated Forecasting System (IFS). Initial results from these forecasts that have been running for 10 months are under evaluation and will be reported in due time. The hemispheric and global versions of the model are run based on ECMWF ERA data and ECMWF IFS archived data, respectively. A summary of the model performance in both hemispheric and global scales can be found in *Jonson et al. (2006; 2007)*.

2. Off-line Coupling of Meteorological and Chemical Transport Models

The Severe Nuclear Accident Program (SNAP) model runs with input from available operational HIRLAM. At present, the version runs on 20 km horizontal resolution and provides a sufficiently large domain for SNAP. The most important meteorological input is 3D wind and precipitation fields. The time resolution for meteorological input is 3-h in operational applications, and 1-h - historical simulations.

Parameter	Unit	Description	Main Purpose
3D fields - for 20 σ levels			
u,v	m/s	Wind velocity components	Advection
q	kg/kg	Specific humidity	Chemical reactions, dry deposition
$\dot{\sigma}$	s^{-1}	Vertical wind in σ coordinates	vertical advection
θ	K	Potential temperature	Chemical reactions, eddy diffusion
CL	%	Cloud cover	Wet removal, photolysis
PR	mm	Precipitation	Wet and dry deposition
2D fields - for Surface			
Ps	hPa	Surface pressure	Surface air density
T^2	K	Temperature at 2m height	Dry deposition, stability
H	Wm^{-2}	Surface flux of sensible heat	Dry deposition, stability
τ	Mm^{-2}	Surface stress	Dry deposition, stability
LE	Wm^{-2}	Surface flux of latent heat	Dry deposition

Table 1 : Variables transferred from HIRLAM to EMEP (from *Simpson et al., 2003*)

The AirQUIS model runs with input from MM5 in 1-h time resolution. The transferred 2D surface parameters are precipitation, total cloud cover, mixing height and surface temperature. 2D parameters from lowest model level are the air temperature, dew point, relative humidity, and vertical temperature gradient. The horizontal wind is the only 3D parameter used. A horizontal interpolation from polar stereographic grid to Universal Transverse Mercator (UTM) is taking place. At present no vertical interpolation is done. The vertical levels in AirQUIS are defined to be identical to the levels in MM5. A meteorological pre-processor calculates dispersion parameters based on the Monin-Obukhov similarity theory (MOST). The constant parameters topography and surface roughness are taken from the meteorological model.

The Unified EMEP model uses the 3D horizontal and vertical wind fields, specific humidity, potential temperature cloud cover, and precipitation. The transferred surface 2D fields for use in the chemical transport model are: surface pressure, 2 m temperature, surface flux of momentum, sensible and latent heat, and surface stress. All variables are given in 3-h interval. Table 1 lists the variables and their main purposes in the EMEP model. Inside the model different boundary layer parameters like the stability, eddy diffusion, and mixing height are calculated based on MOST.

3. Evaluation of Urban Air Quality Forecasts

Regular forecasts are produced with MM5/AirQUIS. The meteorological and air quality forecasts are evaluated against observations and reported on a yearly basis. Summary statistics and case studies are produced. In Figure 1 forecasts for the AQ station Alnabru are

compared to AQ observations at the same station and to meteorological observations at the two closest stations (Valle Hovin and Blindern). The missing peak in the NO_2 forecasts is not caused by errors in the meteorological forecasts. However, the AQ monitoring station is not located together with the meteorological station. Neither does the meteorological station measure all the parameters that used by the AQ model. Observations of inversion layer are limited to measurements of air temperature at 2 and 25 m in the presented case.
Experiments have been performed to address the error made by the pre-processor in AirQUIS. Figure 2 shows the resulting NO_2 forecasts from AirQUIS using dispersion parameters calculated by the pre-processor compared to AirQUIS using dispersion parameters calculated by the meteorological model MM5.
Both in the pre-processor and in MM5 the dispersion parameters are calculated using MOST. The difference is, therefore, due to a time step update of the parameters going into the parametrization scheme rather than an hourly update in the pre-processor. The figure shows that this difference has very small impact on the results.

4. Issues to Consider for NWP Models Providing Data for Air Pollution Models

All air pollution modelling systems at met.no are presently developing to include other types of meteorological drivers at finer resolution. For instance, a small scale SNAP version for simulating local effects in the range of 30-50 km is planned for 2007. Further a coupling to UK Met Office Unified model (UM) or small scale HIRLAM model will be made for even higher resolution SNAP. Also a full coupling of SNAP to regional HIRLAM model is in the line, i.e. SNAP as subroutine in HIRLAM code. During the winter season 2006-2007 the UM model was introduced to replace MM5, and a new interface to AirQUIS is built. Also the EMEP model is developing to use different main sets of meteorological data, ECMWF, HIRLAM and the non-hydrostatic models Weather Research and Forecasting Model (WRF), UM, and ALADIN.
For all these applications, special attention should be given to the interfaces between dynamical and chemical processes. While it is recognised that an on-line coupling of these processes will be ultimately necessary, there is still a series of processes that need special attention also under off-line applications, as named below.

4.1 Surface Classes

For consistent calculation of boundary layer parameters in off-line coupling the land-use classes in the meteorological model should ideally match the land-use classes presented in EMEP (Table 2). In the coupling with HIRLAM, where only five land-use classes are presented, and no parameters to distinguish needle leaf from broad leaf forest, is thus not fully consistent.

4.2 Physical Parametrizations

ACTM models can make use of atmospheric parameters that are output from some parametrization schemes. Boundary layer parameters have to be calculated inside ACTM models or in pre-processing if they are not available from the meteorological model. Entrainment and detrainment rates in cumulus clouds could be provided if sufficiently sophisticated cumulus parametrization is used in the meteorological model.

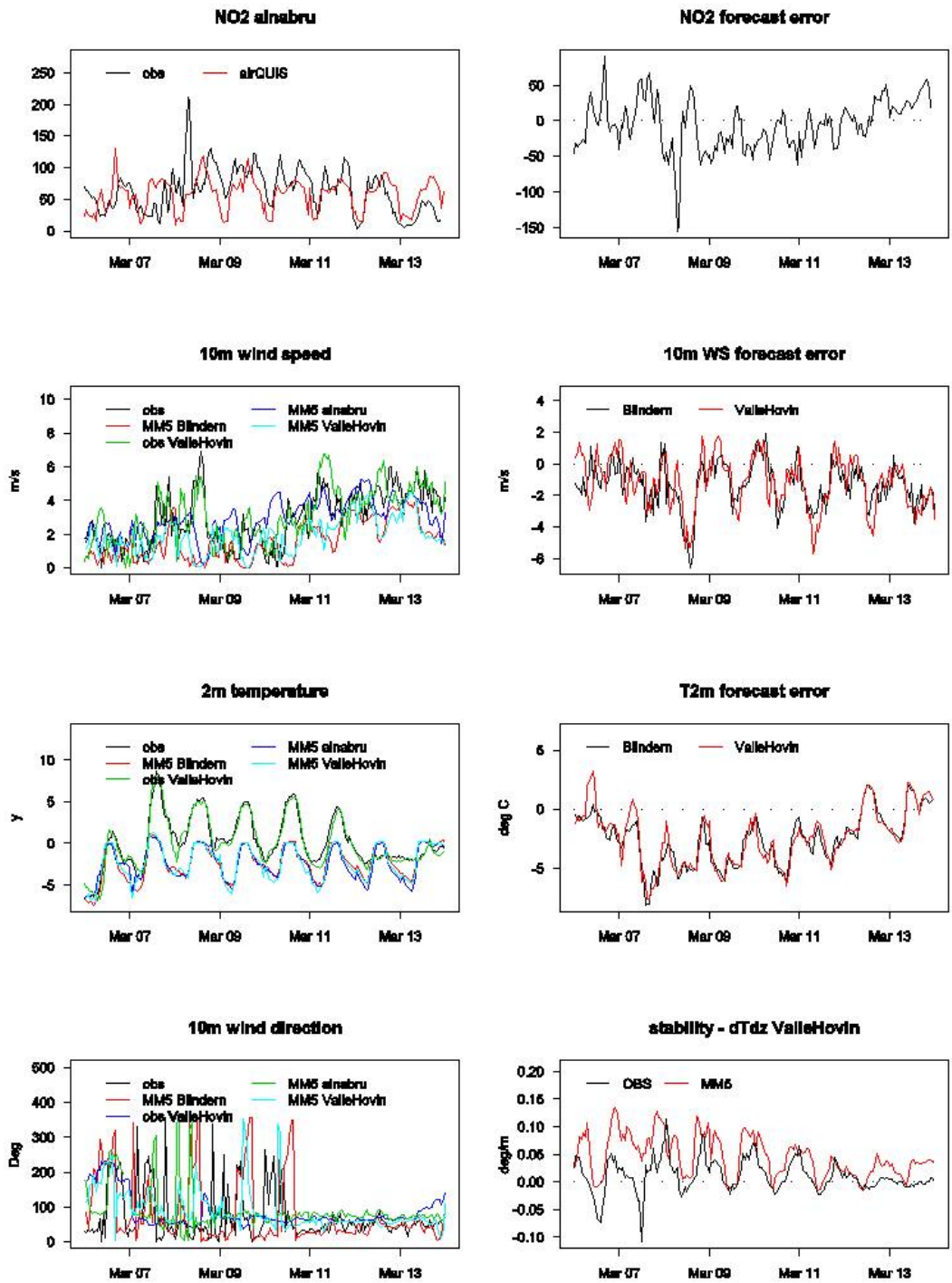

Figure 1 : Observations (black) and model forecasts of NO_2, wind speed, 2m temperature and wind direction (left), and forecast error for NO_2, wind speed and 2m temperature (right) at two different observations sites. Forecasts (red) and observations (black) of vertical temperature gradient 2-25m (bottom right) *(from Ødegaard et al., 2004).*

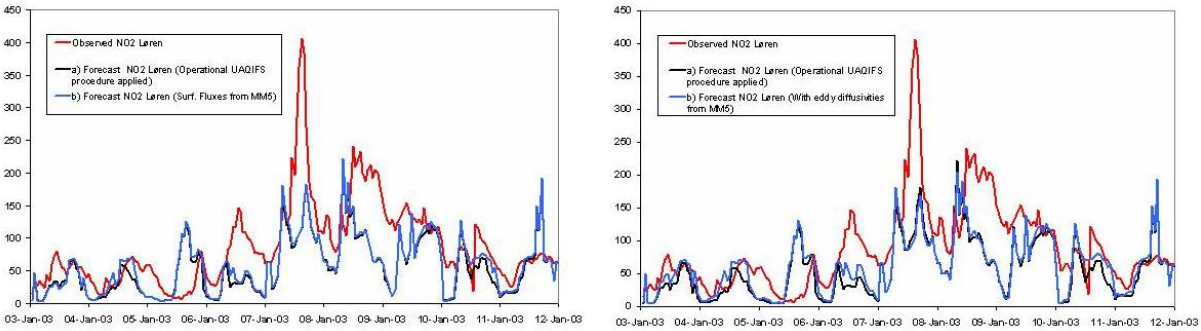

Figure 2 : NO2 forecasts from AirQUIS using surface fluxes (left) and eddy diffusivities (right) from pre-processor (black) and from MM5 (blue) compared to observations (red) *(from Slørdal and Ødegaard, 2005).*

Table 2 : Surface types used by the EMEP model.

Surface/vegetation type	H (m)	Albedo (%)
Temperate/boreal coniferous forests	>20	12
Temperate/boreal deciduous forests	>20	16
Mediterranean needle-leaf forests	15	12
Mediterranean broadleaf forests	15	16
Temperate crops	1	20
Root crops	1	20
Mediterranean crops	2	20
Semi-natural moorland	0.5	14
Grassland	0.5	20
Mediterranean scrubs	3	20
Wetlands	0.5	14
Tundra	0.5	15
Desert	0	25
Water	0	8
Ice	0	70
Urban	10	18

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2.6 A Note on Using the Non-Hydrostatic Model AROME as a Driver for the MATCH Model: *Lennart Robertson and Valentin Foltescu*

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Abstract
Non-hydrostatic weather data provide a challenge to off-line atmospheric chemistry and transport models concerning mass consistent transport that is essential especially for non-linear chemistry. The problem is well illustrated for the Multiple-scale Atmospheric Transport and CHemistry (MATCH) model where an initialization procedure of input meteorological data is proven necessary to ensure mass consistent transport. However, the initialization procedure used is based on hydrostatic assumption, and a question rises if the initialization deteriorates the non-hydrostatic information. We conclude that the initialization is necessary for non-hydrostatic, and corrections are of the same magnitude as for hydrostatic data and do not corrupt non-hydrostatic flow patterns.

1. Introduction

Mass conservation is one of the desired properties of transport schemes (*Willamson, 1992*). The design of the transport scheme as such is one of the parts to achieve this, but is necessarily not enough as inconsistent meteorological data may not fulfill mass conservation. The MATCH model is an off-line model with several options on output grids and flexible on input data on various resolutions. The vertical coordinates and resolution are however, adopted from the input meteorological data and restricted to hybrid sigma-pressure coordinates. The transport scheme is a modified flux oriented Bott scheme (*Robertson et al., 1998*). Another feature of the model is that the vertical winds are calculated internally of two major reasons: 1) the relative vertical wind is needed (normally not available in driving data), 2) internal interpolation of meteorological data (in time and space) demands recalculation of the relative vertical winds (*Robertson et al., 1998*). The vertical wind calculation is very sensitive to errors in mass divergence that do not correspond to pressure tendencies. A procedure for initialization is therefore, implemented based on the methodology proposed by *Heiman and Keeling (1989)*, where the horizontal winds are iteratively corrected until the vertically integrated mass divergence corresponds to the pressure tendency. The methodology has an inherent assumption of hydrostatic balance. The inconsistency in input data may arise from e.g. spectral numerical weather prediction (NWP) data interpolated to a grid, or induced by internal interpolations in time and space. There is always an interpolation associated with data from spectral NWP models provided on a grid-mesh, coming from the need of staggering horizontal wind components (to Arakawa C grid) demanded by the flux oriented transport

scheme. An additional source of inconsistency is truncation by compression of data in e.g. GRIdded Binary (GRIB) format. From this perspective non-hydrostatic NWP data will appear as “inconsistent” data.

2. Mass Conservation Test Run

It is rather straight forward to check the ability of mass conservation of the transport scheme, by initializing the model domain with a constant mixing ratio internally and on the boundaries, and during model run the mixing ratio should stay constant. The test setup works for schemes on flux-form but does not apply to semi-Lagrangian schemes (interpolation of equal numbers) or schemes on advective form (difference of equal numbers).
Figures 1 and 2 show test runs over 18 h non-hydrostatic driving data (AROME 2.5 km) and hydrostatic data (HIRLAM 5 km). The mass errors with no initialization ranges in both cases from -30 to +100 % (which means a rather fast deterioration of the mass conservation).
In Figure 3 the impact from the initialization on the horizontal winds is illustrated. The modifications are generally less than 0.1 m/s and over large areas less than 0.02 m/s, both for hydrostatic and non-hydrostatic data. The corrections are thus small and could not be expected to violate the general flow pattern. Note that the corrections seem to be very small in relation to the corresponding errors in Figure 1 and 2, which is a bit misleading as the latter results from integrated errors.

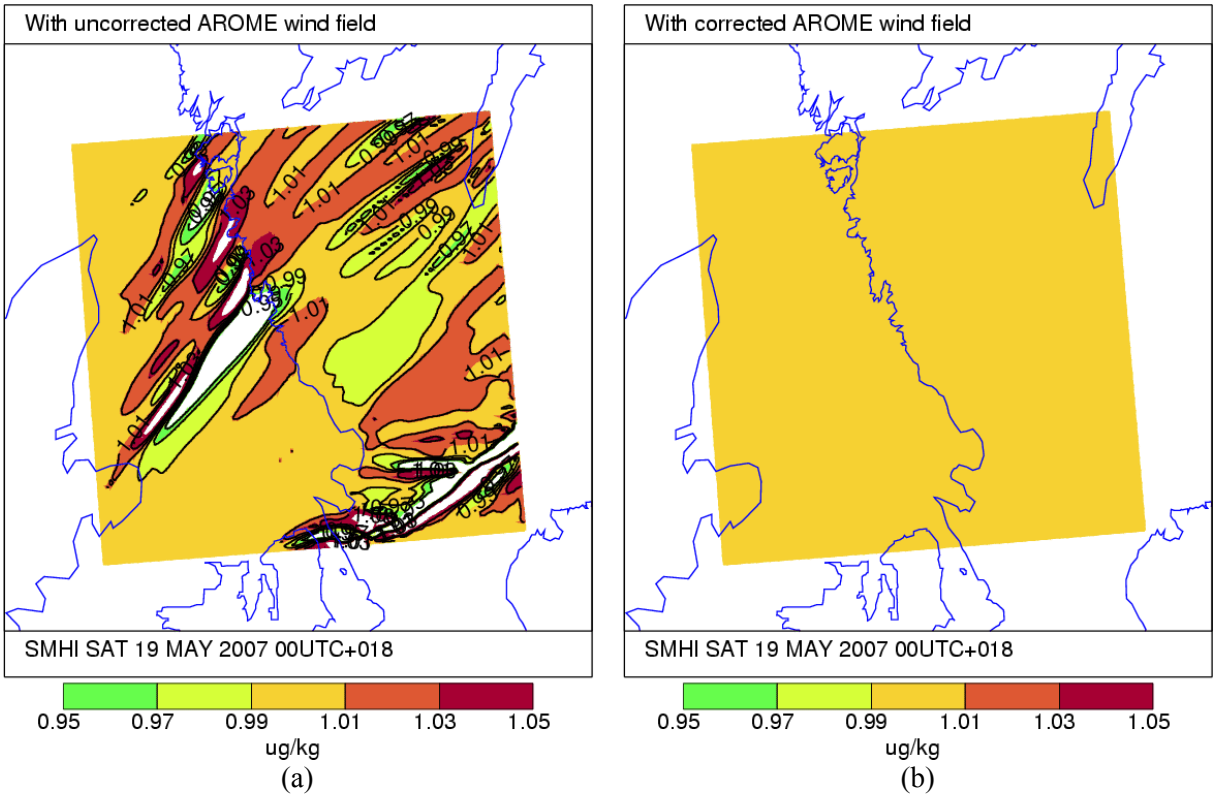
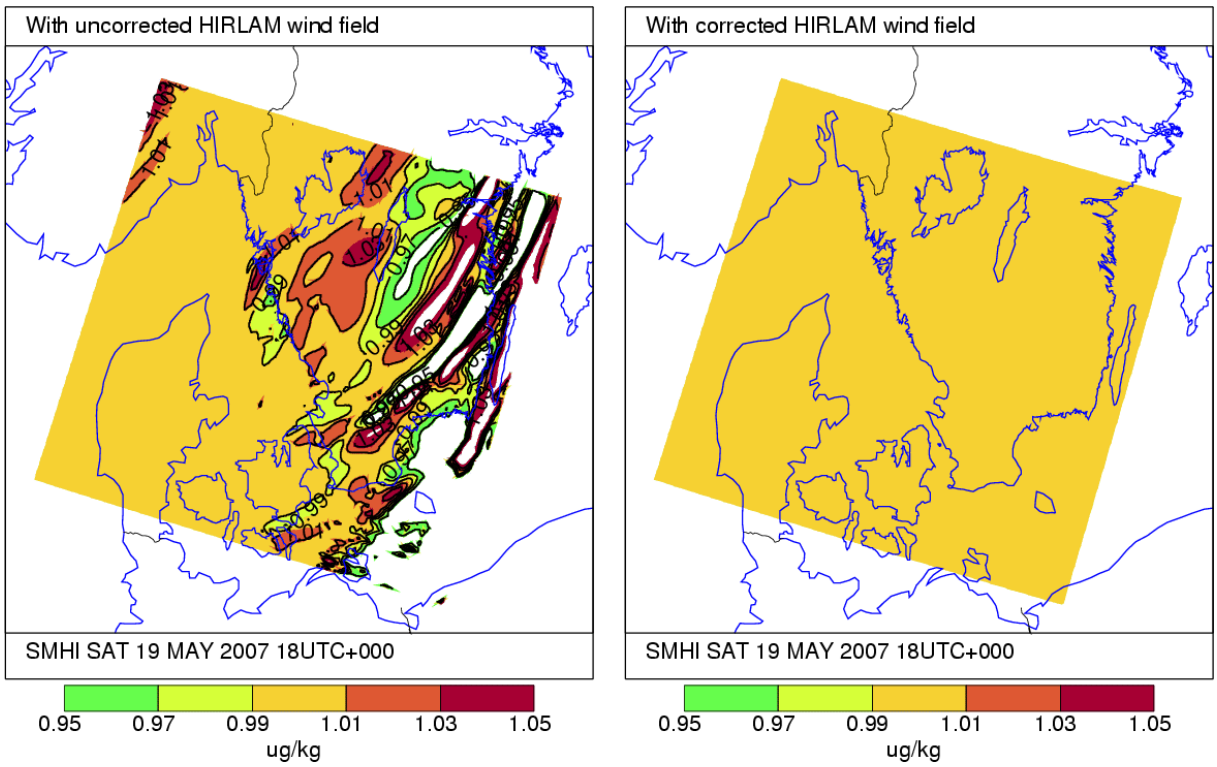
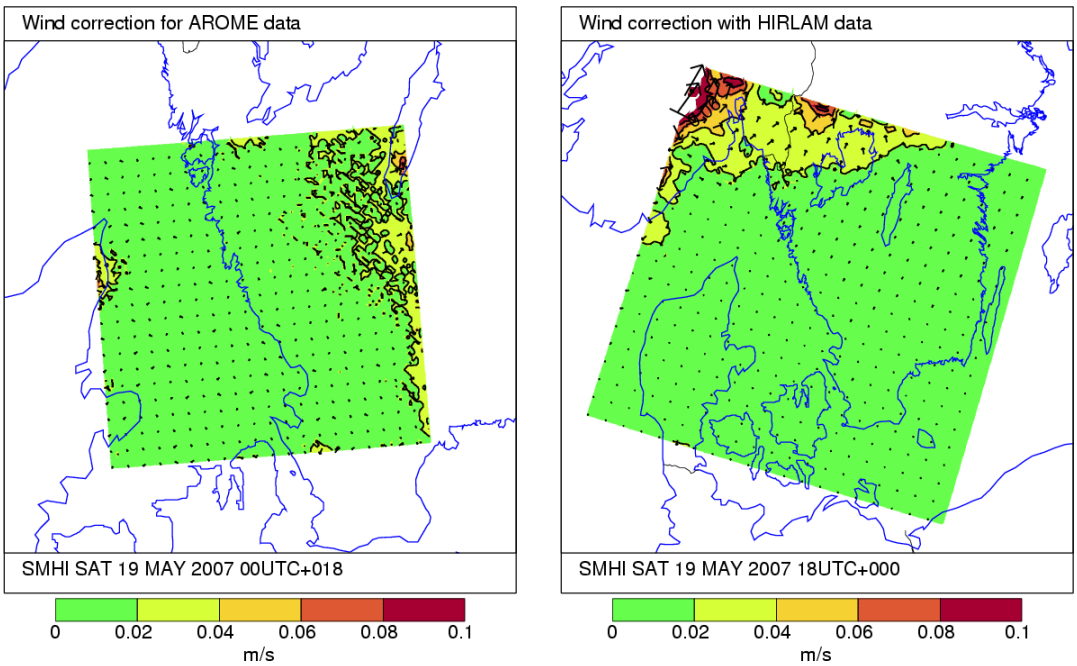


Figure 1 : Illustration of a mass conservation test then using non-hydrostatic AROME model data (2.5 km resolution) over a sub-area covering the Swedish west coast. The panels show simulations over 18 h without (a) and with (b) initialization of horizontal winds. The extreme values of the errors in the left panel ranges between -40 to +100 %.



(a) (b)

Figure 2 : Illustration of a mass conservation test then using the hydrostatic HIRLAM model data (5 km resolution) over a sub-area covering southern Scandinavia. The panels show simulations over 18 hours without (a) and with (b) initialization of horizontal winds. The extreme values of the errors in the left panel ranges between -30 to +90 % and probably associated with a frontal zone passage.



(a) (b)

Figure 3 : Illustration of the magnitudes of changes of the horizontal wind fields during initializations for AROME (a) and HIRLAM (b) data.

3. Conclusions

We have illustrated the need for initialization of non-hydrostatic as well as hydrostatic driving meteorological data for off-line atmospheric chemistry and transport models. The impact on the wind field from initialization is of the same magnitudes for both non-hydrostatic and hydrostatic data (i.e. less than a few dm/s), that indicates that no specific problem concerning initialization for mass conservation of non-hydrostatic data.

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2.7 Aerosol Species in the AQ Forecasting System of FMI: Possibilities for Coupling with NWP Models : *Mikhail Sofiev and SILAM Team*

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Correspondence to: Mikhail Sofiev (mikhail.sofiev@fmi.fi)

Abstract

This note outlines the current status of the regional air quality forecasting system (AQFS) of Finnish Meteorological Institute (FMI) and considers the means of bi-directional coupling of the current off-line system with meteorological models.

Current FMI regional AQFS is based on off-line coupled multi-model structure where the default meteorological driver is the reference HIgh Resolution Limited Area Model (HIRLAM), with a possibility to bypass it directly accepting the forcing of global data of European Centre of Medium-range Weather Forecast (ECMWF). The meso-to-regional scale chemistry transport model SILAM (Air Quality and Emergency Modelling System) is the atmospheric composition model, which is forced by the above meteorological fields, as well as by anthropogenic, biomass-burning, and natural emissions. The forecasting system evaluates several aerosol components: primary and secondary inorganic aerosols, sea salt, and biogenic pollen.

Options for closer coupling of the atmospheric chemical transport model (ACTM) and meteorological driver are based on bi-directional interfacing the SILAM and HIRLAM models, so that the chemical composition data are made available to meteorological model with a time lag or during the next forecasting cycle. However, the main obstacle on the way is the limitations of the HIRLAM physics, which do not allow any external aerosol fields – neither in radiative nor in cloud microphysics modules. Solution of these problems or accepting another model with higher flexibility, such as AROME, would pave the way for consideration of feedbacks.

1. Introduction

The regional air quality (AQ) forecasting system of FMI has been set up in 2005 and opened for public access via internet in 2006 (http//silam.fmi.fi). A primary goal of the system is to

evaluate and forecast the air pollution over the Finnish territory. Since Finland is a receptor of practically all main pollutants, the area of the simulations is necessarily covered the whole of Europe with a compromising resolution of 30 km. A nested domain was introduced in 2007 and it covers northern Europe with a resolution of 10 km.
The pollutants of primary concern are: particulate matter, first of all, fine particles, nitrogen oxides, and allergenic pollutants, such as birch pollen. Since in most cases the problems with NOx species are confined to Helsinki, the regional forecasting system originally did not include nitrogen chemistry limiting the chemical simulations with the sulphur oxides as an indicator of anthropogenic plumes. Currently, the new extension of the AQ forecasts is on the way to include operational simulations for all main anthropogenic pollutants including SOx, NOx, NHx, O_3, and VOCs. Corresponding setup has been created and is being tested in trial operational simulations.

2. Materials and Methods

2.1. The Forecasting System

Since February of 2006, the regional forecasting system of FMI covers three major types of sources (Figure 1): anthropogenic emission of sulphur oxides and primary particulate matter PM 2.5 and PM 10, biological sources of birch pollen and satellite-retrieved real-time information about the wild-land fires (based on hot-spots counts from MODIS instrument onboard NASA Aqua and Terra spacecrafts).

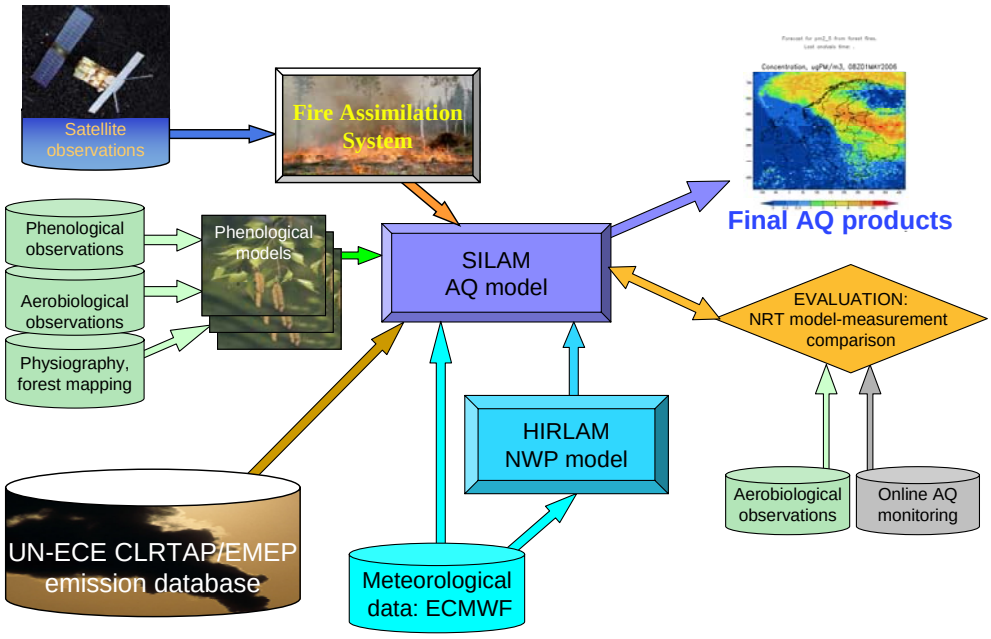

Figure 1 : A structure and main items of the regional air quality forecasting system of FMI.

2.2. The SILAM Model

SILAM is a dual-core modelling system with Lagrangian dispersion core based on an iterative advection algorithm of *Eerola (1990)* and a Monte Carlo random-walk diffusion representation *(Sofiev et al., 2006b)*, and an Eulerian dynamic core that applies the advection routine of *Galperin (2000)* with vertical diffusion scheme based on extended resistive analogy of *Sofiev (2002)* and parametrization of vertical diffusivity after *Genikhovich et al. (2004)*. The system can directly utilize the meteorological data from the HIRLAM and ECMWF

numerical weather prediction (NWP) models, as well as their archives. A typical time step accepted for the operational forecasts and most of hindcast studies is 3 h.
The operational forecasts included the following pollutants and source categories:

- sulphur oxides originated from anthropogenic and volcanic sources;
- primary particulate matter originated from anthropogenic sources (both fine particulate matter with diameter below 2.5 μm, $PM_{2.5}$, and the coarse fraction with diameter from 2.5 to 10 μm $PM_{2.5-10}$);
- primary $PM_{2.5}$ originated from biomass burning;
- birch pollen originated from birch forests during the flowering season.

Input emission data for the anthropogenic pollutants and Etna volcano are based on the database of the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int).
The near-real-time information on active biomass burning is extracted from the observations of the MODIS instrument onboard the NASA Aqua and Terra satellites (http://modis.gsfc.nasa.gov) with a spatial resolution of 1x1 km. The emission fluxes of $PM_{2.5}$ are defined from two algorithms. One is based on scaling the absolute temperatures of grid cells labelled as burning using their temperature anomalies (the temperature anomaly is defined as the difference of the observed and the long-term average temperatures) following the procedure described in *Saarikoski et al. (2007)*. The new method (currently under evaluation) is based on scaling the Fire Radiative Power following the methodologies of *Ichoku and Kaufman (2005)* and *Kaufman et al. (2003).*
Pollen model follows the approach suggested by (Sofiev *et al*., 2006a). The start day of the release is additionally adjusted to the conditions of the specific year using the near-real-time pollen observations of the European Aeroallergen Network (EAN, http://www.univie.ac.at/ean/). Specifics of the pollen grains as an atmospheric pollutant are taken into account in the parametrizations of the dry and wet deposition.

3. Evaluation of the Forecasting System

During pre-operational SILAM evaluation its results were compared with the data of the European Tracer Experiment (*Sofiev et al., 2006b*), Chernobyl and Algeciras accidental releases. For AQ related species, the system output was compared with the European AQ networks for the period of 2000-2002 and several campaigns and for some extreme episodes, such as the case of wild-land fires during spring and summer 2006.
An example of the long-term comparison for sulphates is shown in Figures 2 and 3, which are based on the year-long juxtaposition of the SILAM results and EMEP observational network. Comparing the results with e.g. EMEP simulations, one can conclude that SILAM accuracy is typical for such type of the simulations and the main features of distribution of anthropogenic sulphates are captured by the model. There are several problematic regions, however, such as Danish Straits and southern Baltic where the concentrations are substantially lower than the observed levels. Comparison of observed and modelled SO_2 concentrations and wet deposition (not shown) suggest a general deficit of SOx in this part of the model domain. One of possible reasons could be uncertain ship emission in the area.

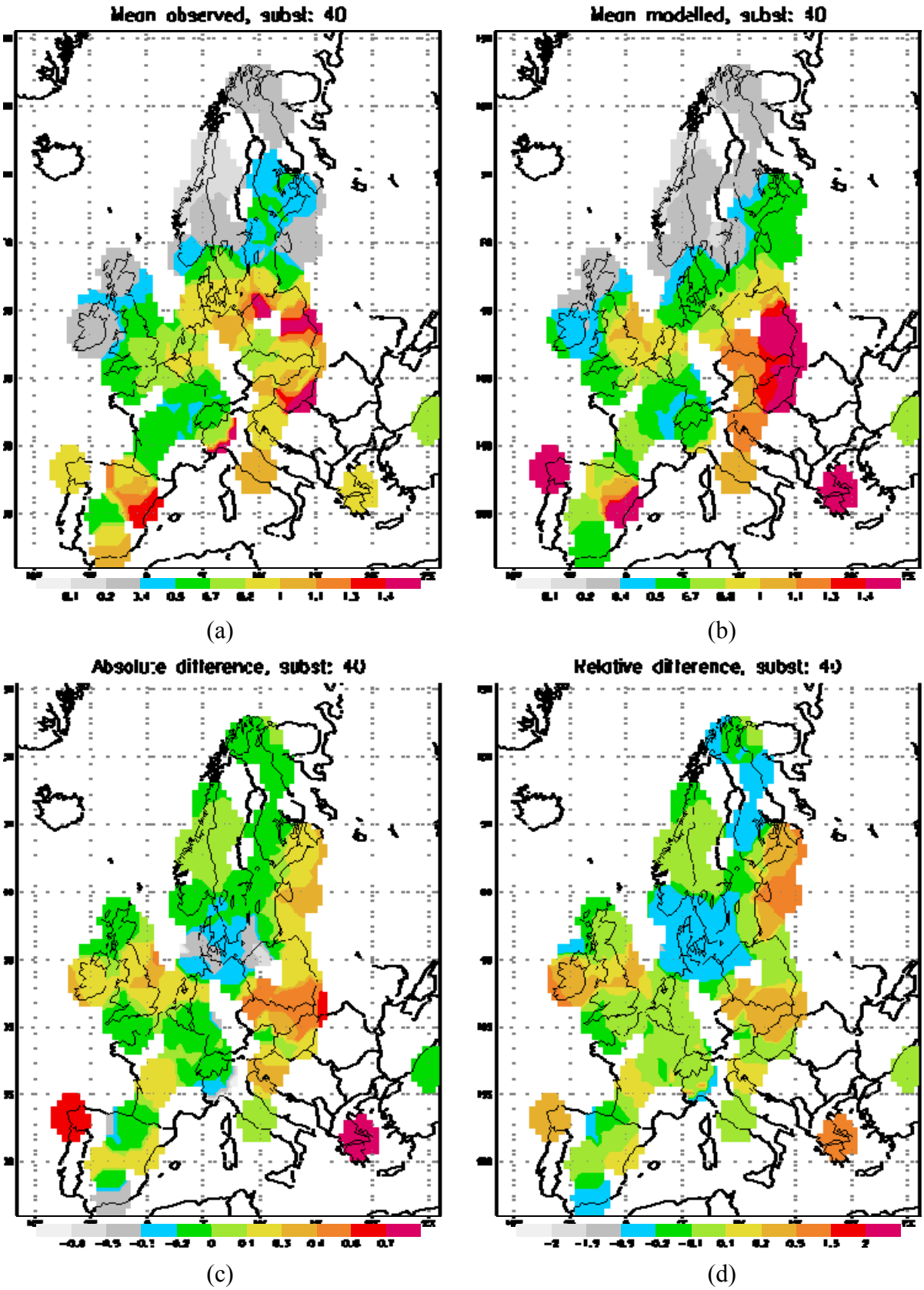

Figure 2 : Mean observed (a) and calculated (b) concentrations for sulphates in 2000, and absolute (c) and relative (d) differences. Unit: µg S m^{-3}.

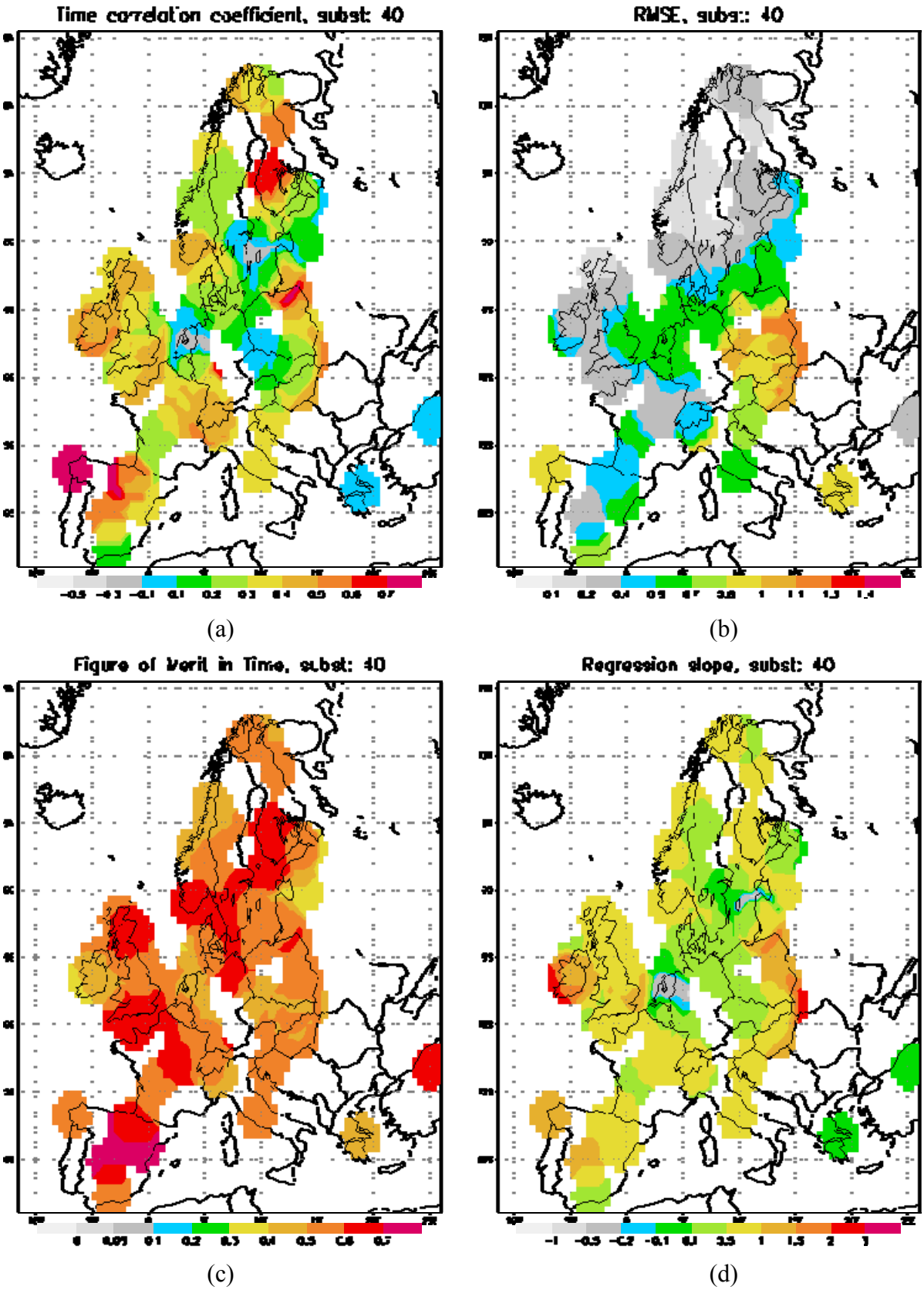

Figure 3 : Correlation coefficient over time (a), RMSE (b), Figure of Merit in Time (c) and regression slope (d) for sulphates in 2000. Unit: µg S m^{-3} for RMSE, relative for the other measures.

Comparison for total particulate matter is more problematic. Cases with dominant fire-induced pollution are reproduced comparatively well (Figure 4) while the contributions of dust and secondary organic aerosol are currently missing from the system. The resulting problems are illustrated by the Figure 5, which represents a full-year time series for a continental EMEP station from Germany. It is seen that for wet and cold seasons the model results for PM_{10} are very good; so, as the sulphate concentration throughout the year. However, during the period of strong agricultural activity the system misses more than 50% of PM_{10}.

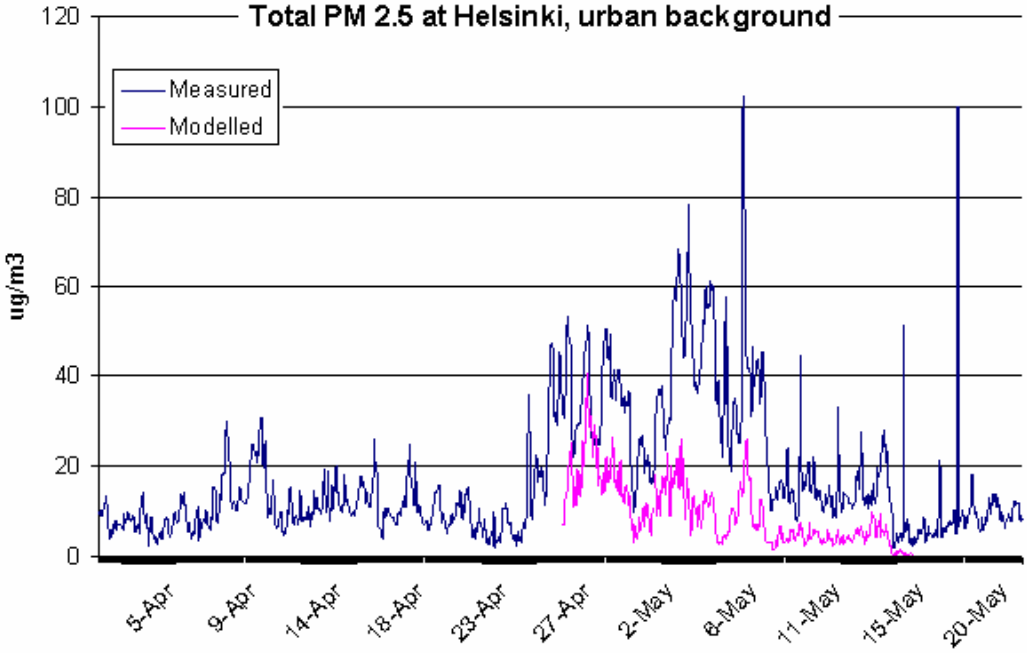

Figure 4 : Comparison of predicted $PM_{2.5}$ from fires and anthropogenic sources with total-$PM_{2.5}$ observed at Kumpula station in April-May 2006 (urban background in Helsinki). Peaks are attributed to fire smoke).

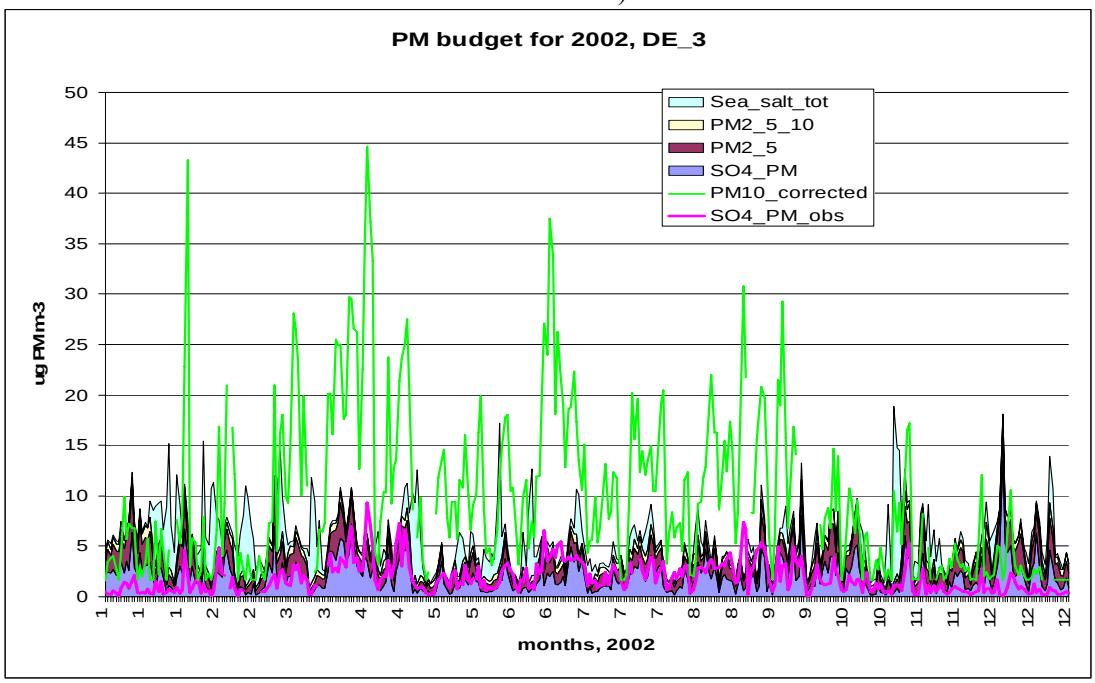

Figure 5 : Comparison of observed PM_{10} and total modelled PM_{10} composition for 2002. Unit : µg PM m^{-3}

4. Feedbacks with the Off-line Coupled Modelling Systems

The above forecasting system, upon completion of tests of the new chemistry scheme, will be able to simulate up to 80-90% of fine-size atmospheric aerosols. Such coverage would make it feasible to consider the feedbacks to meteorological simulations. Consideration of such feedbacks using the existing modelling systems, which are usually independent and can only be interfaced with each other, is feasible but challenging task.

Technically, the transfer of the information from the ACTM to meteorological model is straightforward. For the operational forecasts, the next cycle meteorological calculations can use the previous cycle chemical composition results. For the case of nested simulations, both re-analyses and forecasts, the next-step nested meteorological computations can utilise the previous-step composition data.

The main challenge, however, comes from the other side. It is not enough to transfer the data from ACTM to NWP model, it is necessary to create the corresponding structures and algorithms inside the NWP model itself, so that it is capable of making use of this information. Most of classical NWP models, including HIRLAM, have far-reaching parametrizations of most of radiation transfer and cloud microphysics processes. These parametrizations do not account for any actual information on aerosol content or gaseous composition of the atmosphere. To the opposite, they are based on “average” numbers and tuned to ensure the best score of the meteorological forecasts. In such a situation, replacement of the well-tuned parametrizations deeply embedded into the model physics with something new will be quite laborious and inevitably worsen the quality scores, at least, at the beginning. An alternative approach would be to utilise the models where some elements of external forcing is foreseen. Development of such systems can be easier.

A limitation of off-line feedback mechanisms is an implicit assumption that a single iteration NWP-ACTM-NWP is sufficient to reflect the bulk of the impact of chemical composition onto meteorology. This assumption is fulfilled in almost all cases but larger number of iterations might be needed in case of very strong deviation of the atmospheric composition from the default values assumed in the NWP model. Importance of this limitation and reasonable number of iterations needed for e.g. a dust storm simulations need investigation.

5. Conclusion

The current setup of the FMI chemical weather forecasting system follows the standard approach of the off-line coupling of meteorological and chemical transport models. It creates the possibility of utilising different meteorological drivers and to perform ensemble-type simulations.

The forecasting system covers most types of the atmospheric aerosols, except for dust and secondary organic particles and shows good results in comparison against observations, both long-term and dedicated campaigns. Full-chemistry model version with nitrates and ammonium is at the final evaluation stage.

Possibilities for the feedback from chemical composition to meteorology using the off-line models exists and comparatively straightforward from technical point of view. The obstacle, however, exits in the formulations of most of existing NWP models where the influence of the atmospheric composition is parameterised and hard-coded into the corresponding schemes, thus making it difficult to assimilate the chemical and aerosol data coming from the air quality models.

Acknowledgements
The study was performed within the scope of EU-GEMS project (Global Earth-System Monitoring using satellite and in-situ data) and the ESA PROtocol MOniToring for the GMES Service Element: Atmosphere (PROMOTE) project (http://www.gse-promote.org). Pollen simulations are based on outcome of the Finnish Academy POLLEN project.

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2.8 CAC: An Air Pollution Model used from Regional to Urban Scale modelling: *Allan Gross, Bjarne Amstrup, Alexander Baklanov, Thomas Lorenzen and Jens Havskov Sørensen*

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Abstract
The multi-module-structured tropospheric off-line Chemistry Aerosol Cloud (CAC) model developed at the Danish Meteorological Institute (DMI) is presented. The physics, chemistry, and aerosol modules which are currently included in CAC are described, and recent and present applications of the model are presented. Main results from the CAC simulations showed promising applicability.

1. Introduction

The model development strategy of the Environmental Meteorology group at the Danish Meteorological Institute (DMI) includes both on-line and off-line integrated models. Several off-line Atmospheric Chemistry-Aerosol-Transport (ACAT) models have been developed at DMI: the Multi trajectOry Ordinary differential Numerical (MOON), the Chemistry Aerosol Cloud (CAC), and the Danish EmeRgency MAnagement (DERMA), and the on-line Enviro-HIgh Resolution Limited Area (Enviro-HIRLAM) models. All the off-line models can be integrated with the DMI-HIRLAM model or other meteorological models, or utilizing

Numerical Weather Prediction (NWP) output data as meteo-drivers. In Figure 1 these deterministic models are presented:

- The MOON model is a chemistry aerosol model used to model the air pollution both in the boundary layer and free troposphere, and for stratospheric modelling (*Gross et al., 2005; Madsen, 2006*) (Red arrows in Figure 1), the advection is Lagrangian.
- The CAC model (*Gross and Baklanov, 2004*) (Red arrows in Figure 1) is a further development of the MOON model. The main difference between CAC and MOON models is that the advection in the CAC model is Eulerian.
- The Enviro-HIRLAM is an on-line ACAT model (*Korsholm et al., 2008*). Enviro-HIRLAM is also used for pollen forecasting (*Mahura et al., 2006; Rasmussen et al., 2006*) (Green arrows in Figure 1),
- The Micro-scale Model for Urban Environment (M2UE) Computational Fluid Dynamics (CFD) 3D obstacle-resolved air flow (Nuterman and Baklanov 2007).
- The Danish Emergency Response Model of the Atmosphere (DERMA) (*Sørensen, 1998; Sørensen et al., 2007*) (Blue arrows in Figure 1).

These modelling systems are developed in order to fulfil several duties at DMI, e.g. (i) smog and ozone preparedness, (ii) nuclear, veterinary, and chemical emergency preparedness and risk assessment, (iii) birch pollen forecasting, and (iv) research and development activities.

2. Model Description of the Tropospheric Chemistry Aerosol Cloud (CAC) Model

The CAC model (*Gross and Baklanov, 2004*) is a highly flexible multi-module based system. This model is a further development of the chemistry aerosol model MOON (*Gross et el., 2005*). The module concept makes it easy to perform chemical transformations, and apply the model with other emission inventories and/or meteorological datasets. The major difference between the two models is that in MOON the advection is Lagrangian whereas in the CAC model it is Eulerian. Furthermore, the aerosol physics in CAC is more advanced.

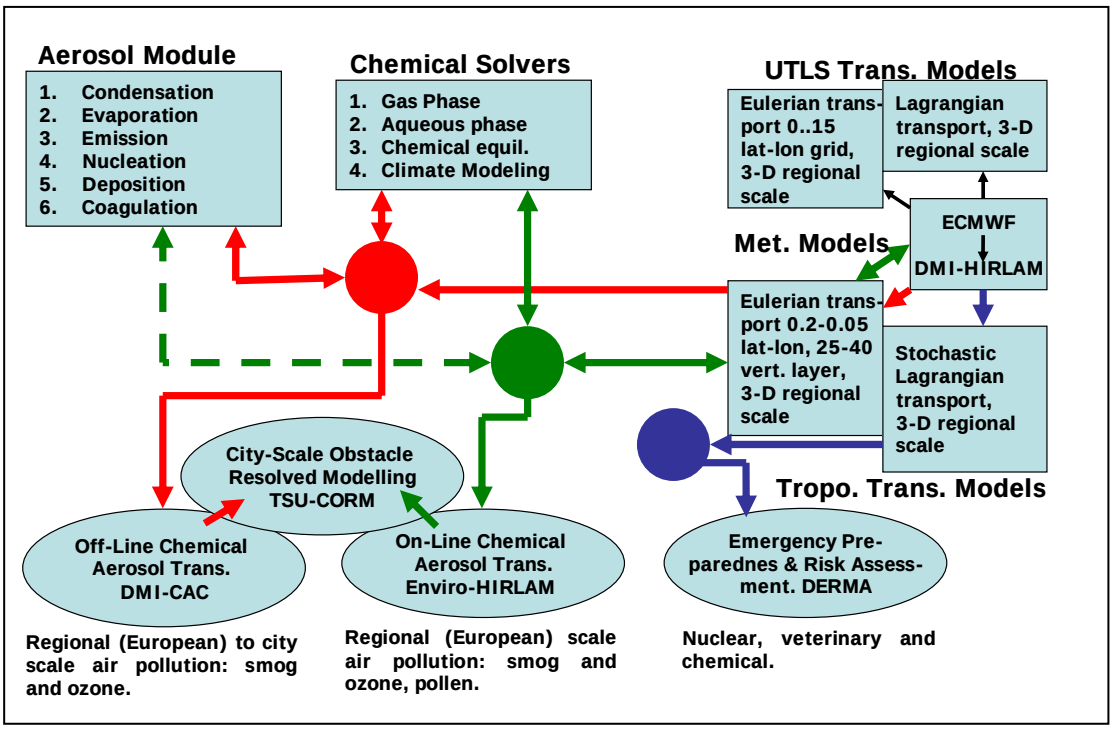


Figure 1 : Deterministic model systems developed at and used by the environmental meteorology group at DMI.

In the present version of CAC two chemical schemes can be used: the Regional Acid Chemistry Mechanism (RACM) (*Stockwell et al., 1997*) and an updated version of Carbon Bond IV (CB-IV) Mechanism (*Gery et al., 1989*) with improved isoprene chemistry (see Figure 2). Both mechanisms are used together with the Tropospheric Ultraviolet and Visible radiation model (TUV) (*Madronich, 2002*) to calculate photolysis rate coefficients, and emissions from EMEP.

The aerosol module treats condensation, evaporation, nucleation, deposition, and coagulation of aerosols, and a simple aqueous phase mechanism where the ISORROPIA module is used to insure chemical equilibrium between the aqueous phase and particle phase is used (see Figure 2). The numerical evolution of aerosols in the CAC model can be solved either by treating the aerosol distributions as bins or normal distributions.

The horizontal and vertical resolutions of the model depend on a resolution of the meteorological and emission data. At present the model is run over a 0.2º×0.2º longitude vs. latitude grid (Figure 3b). The current 3D version of the model has a vertical resolution of 25 levels. They are selected to cover the lowest 3 km of the troposphere. The amount of chemical compounds, which is transported from the free troposphere into the atmospheric boundary layer, is determined by the meteorological information and the concentration of the chemical compounds in the free tropospheric. These concentrations depend on the longitude, latitude, land/sea and month (*Gross et al., 2005*). The advection is solved using the Bott scheme.

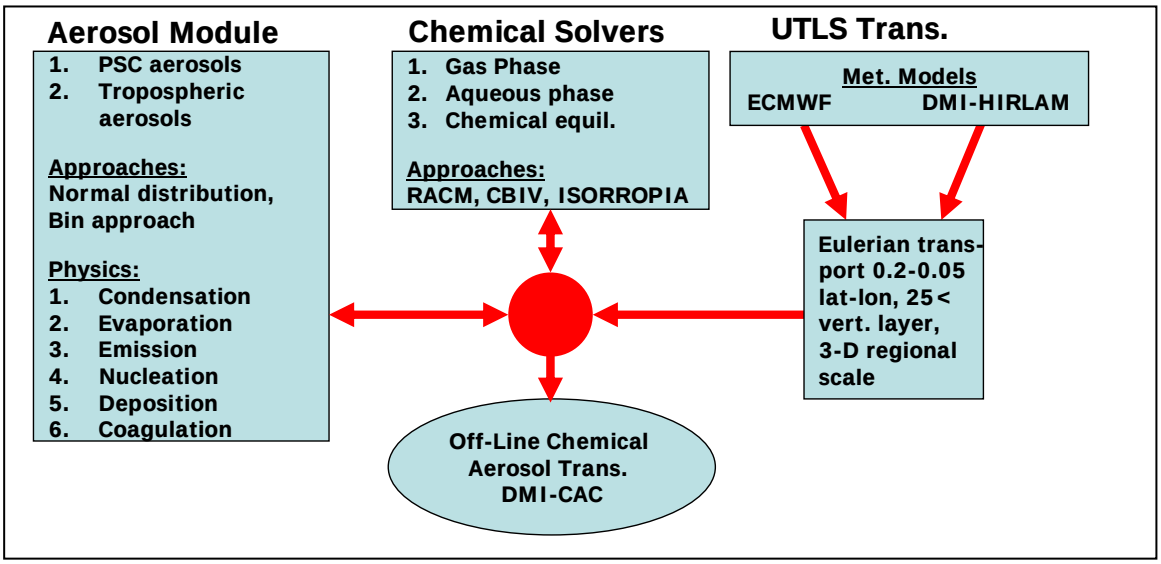


Figure 2 : Schematic description of the CAC model systems developed at DMI.

The model system is developed to simulate aerosols and gas-phase compounds from regional to urban scale of ground-level gas-phase air pollutants. It has been used for air quality forecasts of ground-level gas-phase air pollutants and modelling of historical data.

3. Meteorological Driver (DMI-HIRLAM) and Interface

Currently the CAC model is an off-line model. Therefore, depending on a study objective, different meteorological re-analysed archive datasets, numerical weather prediction, NWP (such as DMI-HIRLAM (*Sass et al., 2002*) or ECMWF) models or climate models can be used as meteorological driver for the CAC model.

At present, the following nested versions of DMI-HIRLAM are used as meteorological driver for the CAC model (Figure 3):

- T15 – 15x15 km, 40 vertical layers;
- S05 – 5x5 km, 40 vertical layers;
- Q05 – 5x5 km, 40 vertical layers;
- U01 – 1.4x1.4 km, experimental version for Denmark.

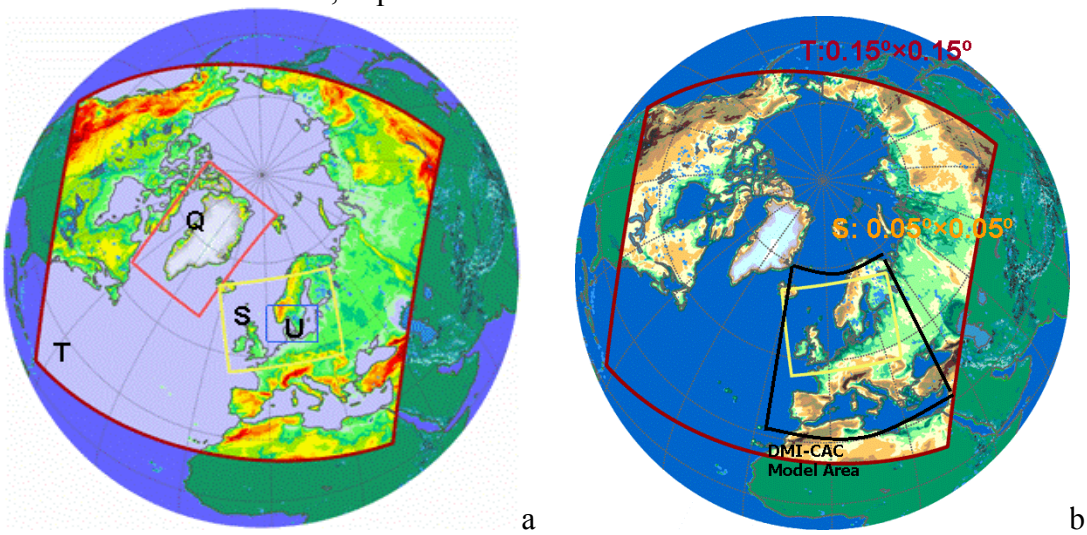


Figure 3 : (a) Operational DMI-HIRLAM modelling areas and (b): operational CAC model area.

The DMI-HIRLAM modelling system consists of the pre-processing, climate file generation, data-assimilation and analysis, initialization, forecast, post-processing, and verification. A forecast integration starts by assimilation of meteorological observations whereby a 3D state of the atmosphere is produced, which as well as possible is in accordance with the observations.

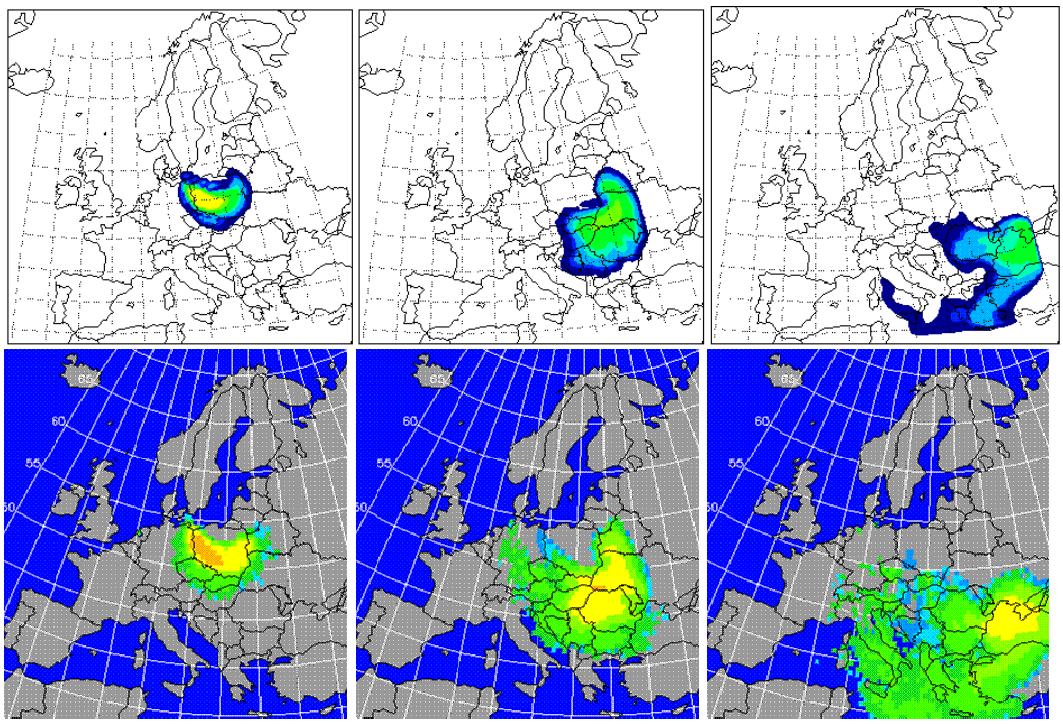
An interface between the DMI-HIRLAM and CAC models is build. The interface extracts necessary information from HIRLAM which then is used by CAC. A comprehensive script system is build to couple the CAC modules and the meteorological information from DMI-HIRLAM together followed by an air quality forecasts.

4. Case Studies

The CAC model is developed to simulate particulate and gas-phase air pollution from regional to urban scale. The model has been used to model artificial releases of passive tracers and air pollution episodes. At present the model is run in a pre-operational mode four times a day based on meteorological forecasts from the DMI-HIRLAM meteorological model.

4.1. Passive Tracer Release

Simulations for several passive tracer releases have been performed employing the CAC model. In Figure 4, the tracer release simulations from ENSEMBLE JRC project (experiment N13) after 18, 36 and 54 hours are presented. This release is a homogeneous release from the ground up to 500 meter above sea level (asl) over the city of Malmo (13°01’E, 55°36’N; Sweden).



18 hours **36 hours** **54 hours**

Figure 4 : Simulation of ENSEMBLE JRC project (experiment N13). The simulation is started at November 13, 2004, and ends November 25, 2004. Surface concentrations are shown after 18, 36, and 54 h. Upper panel results from the CAC model. Lower panel the average concentration from four individual models (DK3, DE1, and FR2), see text.

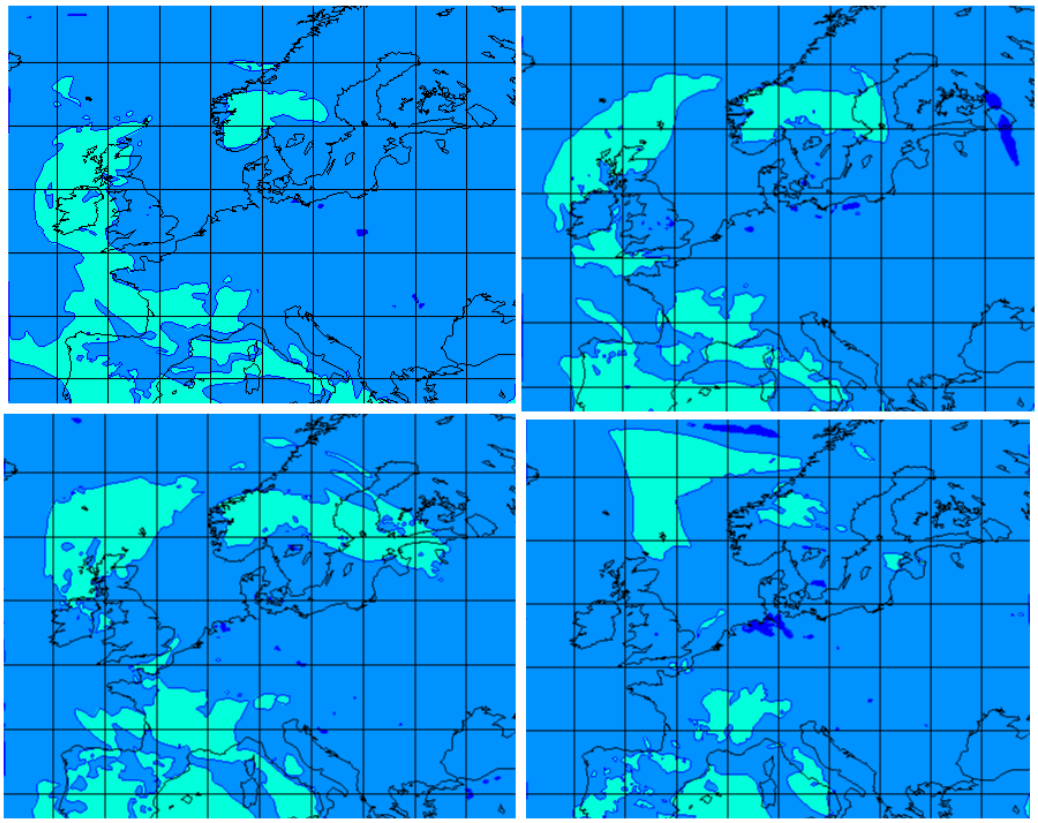


Figure 5 : 00 UTC O_3 forecast from December 20, 2007 using the CAC model. The plots are the forecasts from 24, 36, 48, 60 UTC.

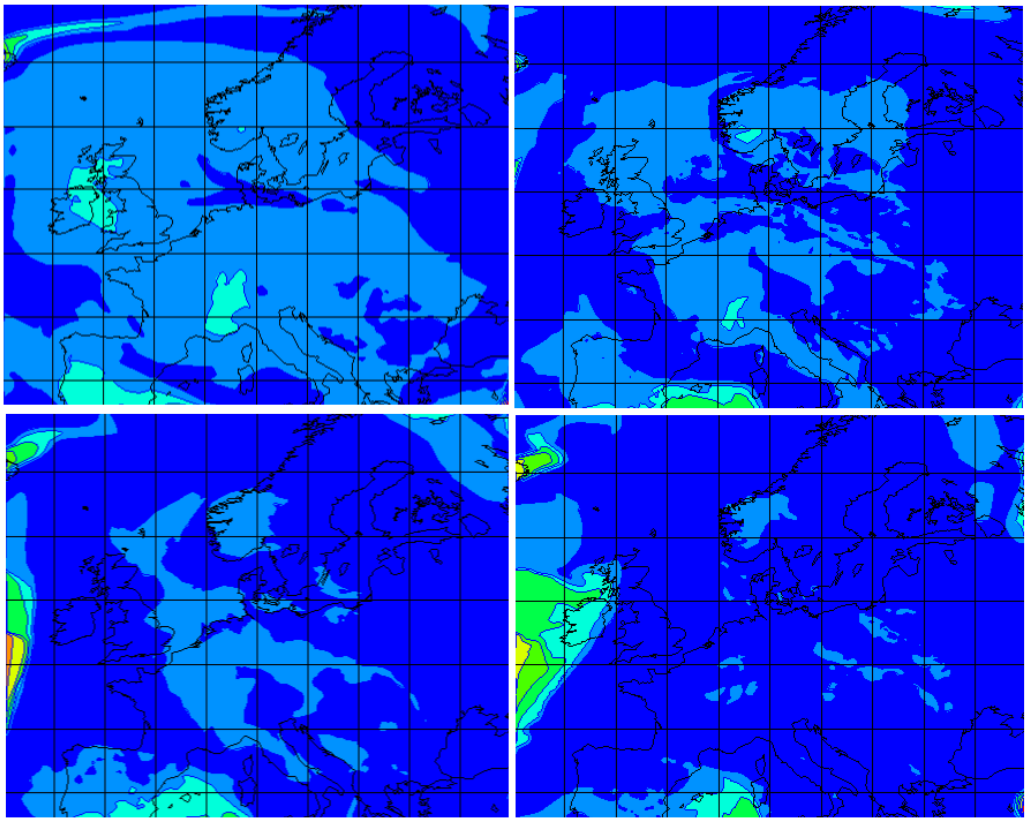


Figure 6 : 00 UTC NO_2 forecast from December 20, 2007 using the CAC model. The plots are from the forecasts from 24, 36, 48, 60 UTC.

The upper panel in Figure 4 is the surface concentrations from the CAC simulation while the lower panel is the average surface concentrations obtained by DERMA (DK3, Danish), DE1 (German) and FR2 (French) models. The results from the DERMA model are based on the same meteorological data as the CAC simulation. From Figure 4 we observe that the shape and concentration profile during the development of the plume from Malmo (Sweden) to over the south-west corner of Europe looks very similar for the CAC model compared with the average concentration profile from the DK3, DE1, and FR2 models.

4.2. Chemical Weather Forecasts

Currently the CAC model is setup to make chemical weather forecasts four times a day for O_3, NO, NO_2, CO, SO_2, Rn, Pb, $PM_{2.5}$ and PM_{10}. In Figures 5-6 examples of a 00 UTC forecast for O_3 and NO_2 are presented. The plots are the forecasts from 24, 36, 48, 60 UTC produced 20 Dec 2007. The simulation shows transport of pollutants over the North Sea from southern to northern parts of Europe. Furthermore, a natural correlation between O_3 and NO_2 is observed. NO_2 is higher over land than sea. The opposite is for O_3, since it is a winter simulation with low photochemical activity the emission of NO reduces O_3 over land.

5. Conclusion

We have presented the first results from a newly developed atmospheric chemical aerosol transport model CAC. These results show the expected behavior of the model. However, the air pollution forecasts are from winter 2007/2008, i.e. a low pollution period. In a future the CAC model will be also validated in details at higher polluted events.

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Chapter 3. Validation and Case Studies

3.1 Chemical Modelling with CHASER and WRF/Chem in Japan: *Masayuki Takigawa[1], M. Niwano[1], H. Akimoto[1], and M. Takahashi[1,2]*

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Abstract
In this paper results of studies with WRF/Chem and CHASER models are presented. The CHemical Atmospheric general circulation model for Study of Atmospheric Environment and Radiative forcing (CHASER) is a global chemical transport model (cf. *Sudo et al., 2002ab; Takigawa, 2005*). The gaseous and aerosol chemistry module is implimented in the CHASER in an on–line mode. CHASER is based on CCSR/FRCGC/NIES AGCM 5.7b, and the meteorology and radiation can be simulated in CHASER itself. The radiative feedback through the distribution of chemical species is taken into account. Daily forecasts have been available on internet since 1 January 2002. This forecasting system was developped for the use of daily flight planning for the PEACE–A (January 2002) and PEACE–B (April–May 2002) compaigns. A regional–scale chemical weather forecasting system based on WRF/Chem has been also developed. The lateral boundary for chemical species is taken from the 3–hourly output of CHASER. The modelled surface ozone was compared with the ground–based observations.

1. Introduction

Recently, atmospheric chemical transport models (ACTMs) have enhanced flight planning by providing direct information on the expected state of important 3D atmospheric chemical structures on timescales from hours to days, *i.e.*, the "chemical weather". The first chemical weather forecasts (CWFs) was performed during the Airborne Southern Hemisphere Ozone Experiment (ASHORE) and the Second European Stratospheric Arctic and Middle-Latitude Experiment (SESAME) in 1994 and 1995, respectively (*Lee et al.*, *1997*). The use of CWFs for field campaigns is expanding rapidly. Global CWFs from the Model of Atmospheric Transport and Chemistry (MATCH; of the Max–Planck–Institute for Chemistry, MPIC) were used during the Indian Ocean Experiment (INDOEX) in 1999, and the Mediterranean Intensive Oxidants Study (MINOS) and the Convective Transport of Trace Gases into the Upper Troposphere over Europe (CONTRACE) in 2001 (*Lawrence et al.*, *2003*). In contrast to regional CWFs, global CWFs can predict intercontinental transport. For example, *Lawrence et al. (2003)* estimated the frequency of intercontinental pollution plumes from North America and Asia to Europe. Chemical weather forecast systems using regional scale models have an advantage - a higher horizontal resolution – compared with global models. The Chemical Weather Forecasting System (CFORS) was used during the TRACE–P and the ACE–Asia campaign in 2001 *(Uno et al., 2003). Uno et al. (2003*) showed that changes in synoptic–scale weather patterns greatly influence continental–scale pollution transport in spring over East Asia.
This paper describes a newly developed global CWF system based on CHASER model that can be used to support atmospheric chemistry field campaigns. The model includes radiative

and dynamical processes, and comprehensive chemical schemes for the troposphere and lower stratosphere. This paper assesses the quality and estimates the value of CWFs from the model that was used in flight measurement campaigns.

2. Global Chemical Weather Forecasting System

The chemical weather forecast (CWF) system includes the coupled tropospheric chemistry climate model CHASER, which is described and evaluated in *Sudo et al. (2002ab)*. Physical and dynamical processes are simulated following the Center for Climate System Research/National Institute for Environmental Study/Frontier Research Center for Global Change (CCSR/NIES/FRCGC) atmospheric GCM (*Nakajima et al., 1995; Numaguti, 1993; Numaguti et al., 1995*). In this study the CHASER model is based on CCSR/NIES/FRCGC AGCM version 5.7b. Advective transport is simulated with a 4-*th* order flux–form advection scheme using a monotonic Piecewise Parabolic Method (PPM) (*Colella and Woodward*, *1984*) and a flux–form semi–Lagrangian scheme (*Lin and Rood*, *1996*). Subgrid–scale vertical fluxes of heat, moisture, and tracers are approximated using a non–local turbulence closure scheme based on *Holslag and Boville (1993)* used in conjunction with the level 2 scheme of *Mellor and Yamada (1974)*. The cumulus parametrization scheme is based on *Arakawa and Schubert (1974)* with several simplifications described in *Numaguti et al. (1997).* The closure assumption is changed from the diagnostic closure used in *Numaguti et al. (1997)* to a prognostic closure based on *Pan and Randall (1998),* in which cloud base mass flux is treated as a prognostic variable. An empirical cumulus suppression condition introduced in *Emori et al. (2001)* is adopted. Note that both the updraft and downdraft of chemical species by cumulus convection are included in the model. The large–scale condensation scheme is based on *Treut and Li (1991)*, in which subgrid probability distribution of total water mixing ratio in each grid box is assumed to have a uniform distribution. Spectral coefficients are triangularly truncated at wavenumber 42 (T42), equivalent to a horizontal grid spacing of about 2.8°. The model has 32 vertical layers that are spaced at about 1 km intervals in the free troposphere and lower stratosphere. The chemical side of the model is based on *Sudo et al. (2002a, 2003)*, and includes a detailed on-line simulation of tropospheric chemistry involving the O_3 –HO*x* –NO*x* –CH_4 –CO system and oxidation of NMHCs. The chemical model time step is 10 minutes. The model includes detailed dry and wet deposition schemes and heterogeneous reactions on the surface of sulfate and nitrate aerosols.

In addition to the extensive chemical reactions, forecast runs include tagged CO tracers. Such tracers are emitted normally over selected regions (north and south China, Japan, south Asia, northern America, Europe, and Siberia) and evolved subject to model transport schemes and normal chemical loss processes for CO. Anthropogenic surface emissions of CO are taken from the *Streets et al. (2003)* inventory over Asia (except China), and from the Emission Database for Global Atmospheric Research (EDGAR) (*Olivier et al.*, *1996*) over other regions. Surface CO emissions over China are taken from *Streets et al.* (*personal comm.*, *2005*). The estimated annual amount of emissions over China is 146 TgCO, a figure about 40 TgCO larger than that in *Streets et al. (2003)*. In this study, the timing of CO emission from biomass burning was estimated by using the average of the hot spot data from 1995 to 2001 from Along Track Scanning Radiometer (ATSR) (*Arino et al.*, *1999*) for the daily forecasts. ATSR hot spot data for 2002 were used in the post–analysis study. Tagged CO tracer is also considered for CO which is chemically produced from the oxidation of CH_4, isoprenes, and other NMHCs.

Each daily run is fully automated and consists of two parts: a “quasi–real–time” run and forecast run. The quasi–real–time run is derived from the NCEP final analysis (FNL) data,

and steps forward one day at a time as soon as the previous day's data are available. Forecast runs use the NCEP Global Forecast System (GFS) data instead of the NCEP FNL data. The NCEP data are re–gridded from 1° ×1° to 2.8° ×2.8° in horizontal, and from 24 to 32 layers in vertical. The relaxation time for nudging the CHASER model meteorological field to the NCEP meteorological data is 1 day in the free troposphere and lower stratosphere. The relaxation time approaches 0 at the surface. Sea surface temperature (SST) is based on the WMO Distributed Data Bases managed by the Japan Meteorological Agency. Winds from 10 to 3 hPa are calculated by using dynamical and physical procedures in the CHASER model. They are not nudged by NCEP data because the maximum height of NCEP data is lower than the top of the CHASER model. Humidity is calculated using the hydrological cycle (surface sources and sinks, transport, convection, diffusion, condensation, and precipitation). Consequently, the temperature and humidity fields do not produce destabilization or discontinuity because of inconsistencies between the CHASER temperature and NCEP humidity. The forecast run is initialized from a restart file written at the end of the previous day's quasi–real-time run. Automated runs normally start at 05:00 of the local (Japanese) standard time, JST (i.e. 20:00 UTC of the previous day). Therefore, the 1 day forecast was available for pre–flight briefing during the PEACE campaign. Pre–formatted figures are automatically made from output from both the quasi-real-time and forecast runs. Archived output data can be used to make custom figures via the web interface (http://www.jamstec.go.jp/frcgc/gcwm).

This forecast system evolved from a prototype run with lower resolution (T21) that started in November 2000. Then it switched to higher resolution (T42) with a spin–up time of 3 months for the global distribution of chemical species. Daily forecasts and archived output data have been available on a web page at the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) since 1 January 2002.

3. Results

The climatological and seasonal distributions of chemical species (i.e., the chemical climate) calculated by CHASER have already been evaluated in *Sudo et al. (2002b, 2003*). The focus here is on the validation and interpretation of the chemical structures on timescales from hours to days. The targeted features of the flights based on CWFs can be characterized by: a) pollution plumes that were affected by intercontinental transport, and b) the outflow of a polluted air mass from nearby populated/industrial regions. The global CWF can predict anywhere where enhanced mixing ratios may occur on a day–to–day bias. However, such predictions would benefit from global or regional model forecasts with higher resolution, especially, if they are used to plan flight paths.

3.1. Meteorological Fields

An accurate characterization of transport processes is of critical importance to flight planning and analysis of observations. Figure 1 compares PEACE G–II observations of meteorological parameters (wind direction and speed, air temperature, and relative humidity) with CHASER simulated meteorological output. Model output was extracted along the G–II aircraft flight path and compared to aircraft observations averaged over 60 seconds. The color of each dot denotes the hight. Wind speed, and air temperature well agreed with the G–II measurements. The modeled relative humidity shows a bias in the low–humidity regions. In addition, the model did not reproduce the supersatulated air seen in flight 12 of PEACE–A on 21 January and flight 11 of PEACE–B on 15 May. The flight paths on these dates passed through a low–pressure system, and because of a coarse resolution the model failed to resolve fine structures.

By the same reason, the model could not reproduce the wind direction in the lower troposphere (red dots in Figure 1), especially in PEACE–B. The correlation coefficients between modelled and observational data for all G–II flights were 0.81, 0.91, 0.99, and 0.66 for the wind direction, wind speed, temperature, and relative humidity, respectively. The CHASER global CWF accurately captured many of important observed meteorological features during PEACE. The results presented are driven by the NCEP FNL data. The forecast and hindcast meteorological fields were similar during both the PEACE–A and PEACE–B compaingns.

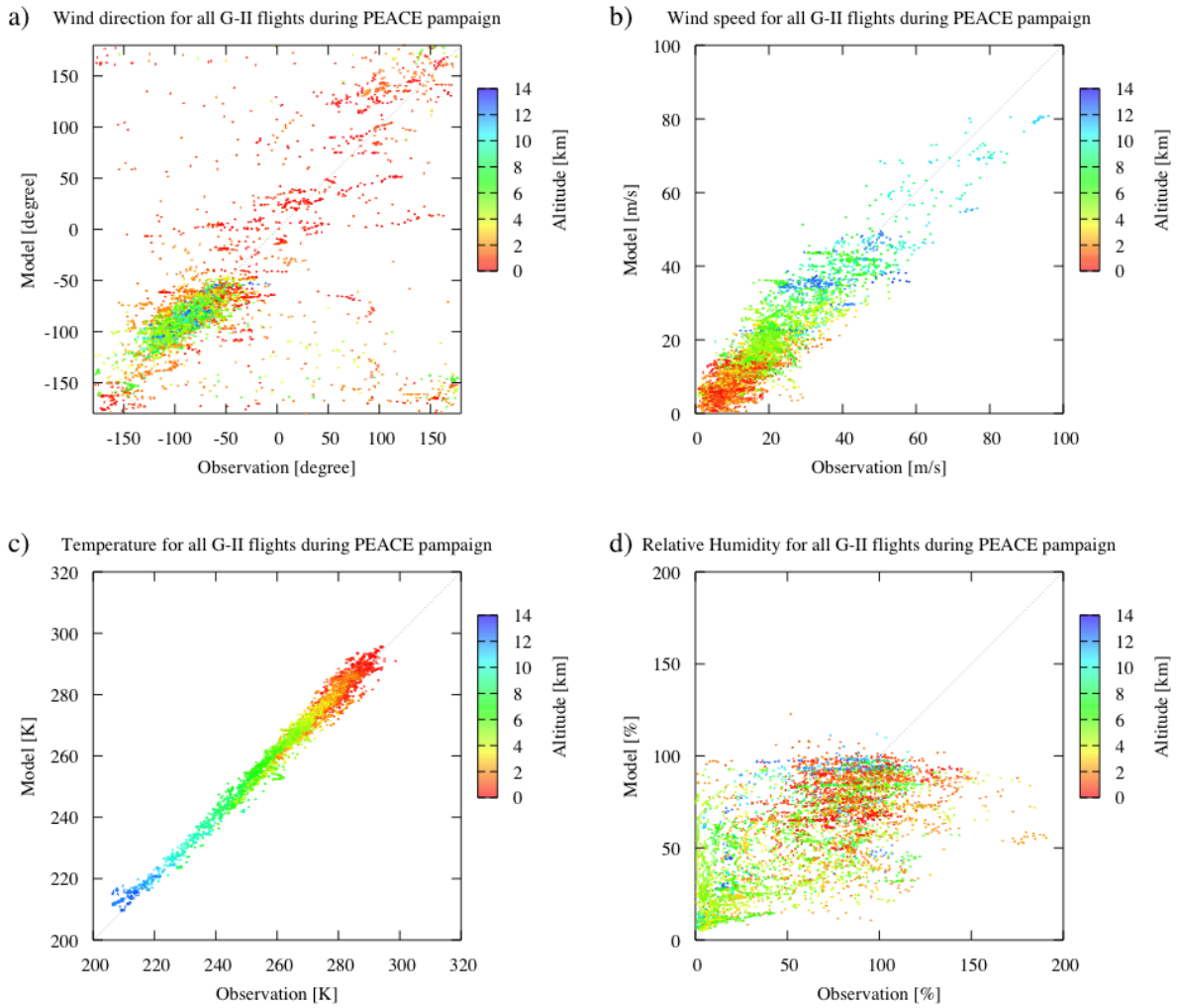


Figure 1 : Comparison between PEACE G-II airborne meteorological parameters and global chemical weather forecasting system output fields along the flight paths. Colors denote altitudes. Dots and crosses denote PEACE-A and PEACE-B data, respectively.

3.2. Comparison with Ground-based Observations

The mixing ratios of chemical species calculated by the CWF system during PEACE were compared with values observed at three ground–based observational sites in the major sampling region. Figure 2 shows CO at Minamitorishima (24.2°N, 153.6°E), Yonagunijima (24.3°N, 123.1°E), and Ryori (39.2°N, 141.5°E). The model reproduced the observed temporal variations of each site except small peaks at Yonagunijima, especially in winter. Although biomass–burning emissions based on the 2001-2002 ATSR hot spot data were incorporated into the model, the model still underestimated CO mixing ratio by about 20 ppbv at these sites in spring. The hot spot data are well correlated with emission anomalies,

especially for Siberia (*Yurganov et al.*, *2005*). Figure 2 shows the regional CO tracers calculated by CHASER. Siberian CO made small contribution to the CO concentrations in late spring at these sites.

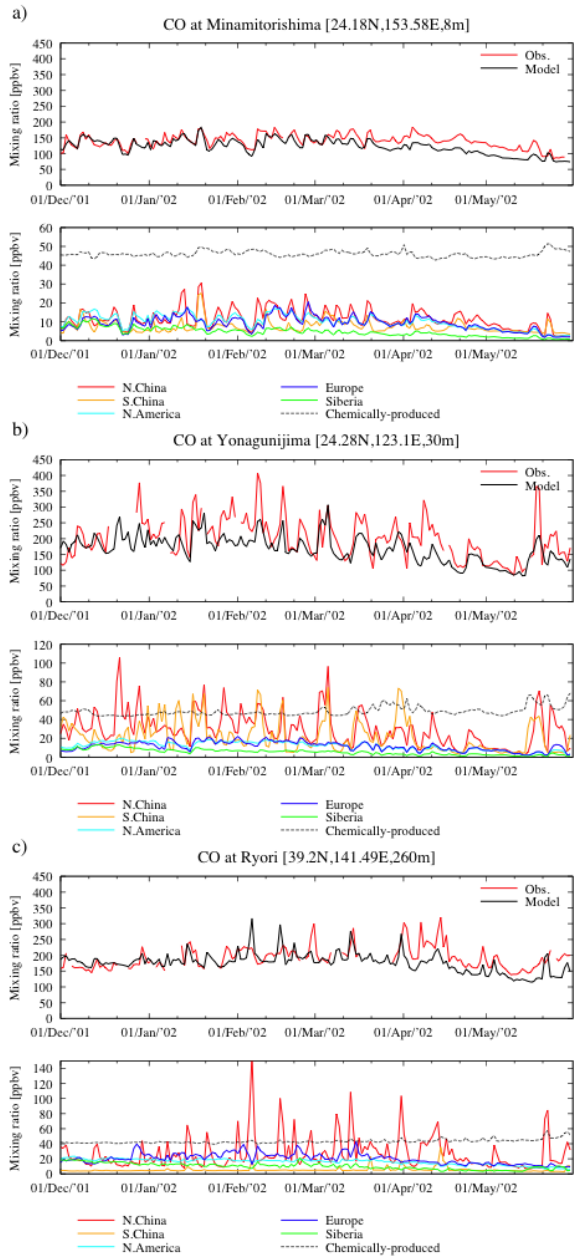


Figure 2 : Observed (red lines) and modeled (black lines) surface CO mixing ratio at measurement stations: (a) Minamitorishima (24.2°N, 153.6°E), (b) Yonagunijima (24.3°N, 123.1°E), and (c) Ryori (39.2°N, 141.5°E). Regional tagged CO tracers for northern China and Korea (red), southern China (orange), south Asia (gray), Europe (blue), North America (cyan), and chemically production (dotted lines) are also shown.

The concentration of Siberian CO does not exceed 5 ppbv in late spring. Minamitorishima is in the southeast of Japan and is affected by maritime air and outflow from Asia. Tagged CO tracers suggest that the enhanced CO level observed at Minamitorishima on 18 January is linked to emissions from northern and southern China. The CO level at Yonagunijima is strongly related to the Asian CO tracers. Some events with increased CO levels (greater than

300 ppbv) observed at Yonagunijima. Corresponding increases occurred in the China CO tracers in such events. Tracers of CO that is chemically produced from the oxidation of CH_4, isoprenes, and other NMHCs, increased by about 5–10 ppbv in the CO elevated event in the spring. Increases in CO tracers, linked to chemical production from hydrocarbons in polluted air masses, reflect the enhanced chemical activity in the spring.

3.3. Convective Outflow during PEACE–B Campaign

Convective activity can play an important role in late spring, when PEACE–B was conducted. *Oshima et al. (2004)* evaluated the origin of air parcels sampled by the aircraft during PEACE–B from altitudes between 4 and 13 km using backward trajectories and estimated that 69% of those that originated at/or below 800 hPa experienced convective uplifting. CCSR/NIES/FRCGC AGCM and CHASER consider tracer updraft and downdraft corresponding to deep cumulus convections. Figure 3 shows the modeled mass change of CO tracers in the free troposphere (above 2 km) caused by convective transport over northern China (poleward of 30°N) and Korea, southern China (equatorward of 30°N), and Japan. The model results show greater vertical transport resulting from deep convection over China in late spring. A clear increase also occured in January over Japan. It is related to passages of mid–latitude cyclones over Japan. The monthly budget of convective transport for May is estimated to be about 3 and 3.3 TgCO over northern and southern China, respectively. These values are about half of all the surface emissions over China in this month. The monthly budgets of surface emissions in the model over these regions are about 6.5 and 6.4 TgCO, respectively.

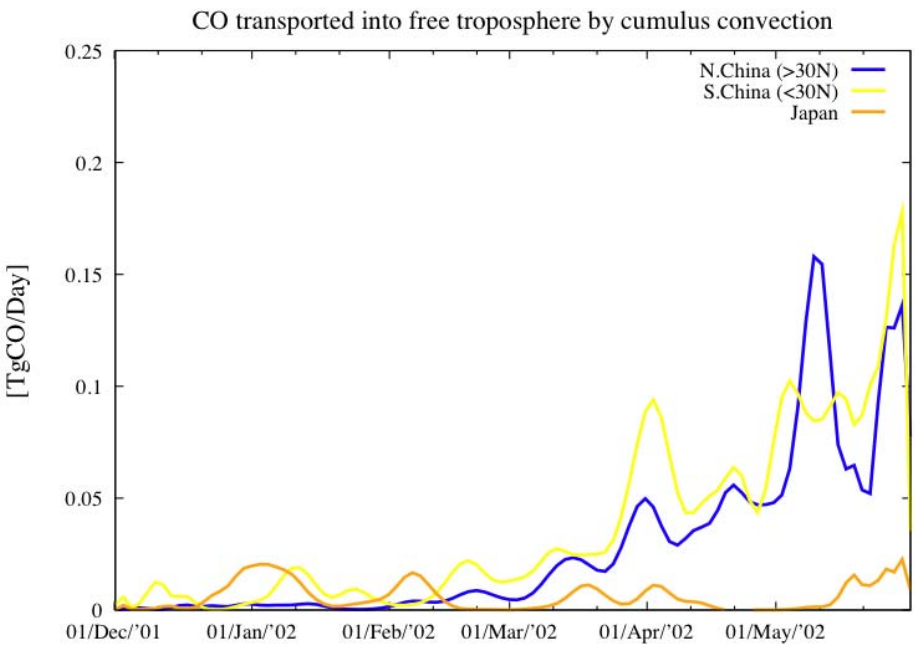


Figure 3 : Ten-day running mean of CO transport by convection over northern China (blue line), southern China (yellow line), and Japan (orange line) during PEACE as calculated by CHASER.

Several PEACE–B flights were carried out in May 2002 when convective CO transport was active. Figure 4 shows the vertical profiles of the observed and modeled CO mixing ratios on 14 May 2002 (flight 10, PEACE–B). CO increased at levels between 300–400 hPa. The model typically underestimated background CO level by about 20–40 ppbv compared to the PEACE–B observations, but for this flight, the model showed similar UT CO enhancement.

The correlation coefficient between the observed and calculated ΔCO for this flight is 0.56 in the FT. The observed CO concentration has increased from about 120 ppbv (outside the plume) to around 300 ppbv (inside the plume). There is a similar increase in the modeled CO profile, although the modeled CO increase is smaller (i.e., from 80 to 145 ppbv), and it is restricted to 300–350 hPa. Tagged CO tracers suggest that emissions from southern China are responsible for this enhanced UT plume. Emissions from southern and northern China contribute 18–21 and 8-10%, respectively, of the total CO mixing ratio in the plume. Figure 5 shows the sea level pressure, and the CO mixing ratio tendency forced by convective transport, CO fluxes, and modeled CO mixing ratio (all given at 300 hPa) calculated by CHASER.

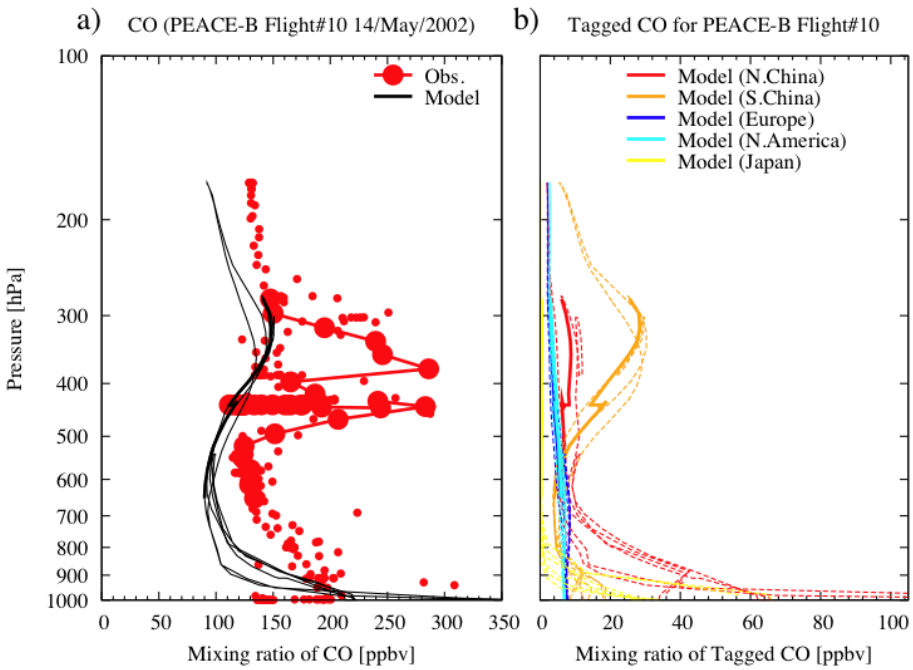


Figure 4 : a) Vertical profiles of the observed CO mixing ratios (red circles), along with the model output corresponding to the flight tracks for PEACE-B flight 10 on 14 May 2002. Black line denotes the total CO calculated by CHASER. B) Regional tagged CO tracers for northern China and Korea (red), southern China (orange), Europe (blue), North America (cyan), Siberia (green), Japan (yellow), and biomass-burning outside from Asia (gray). The bold lines indicate CO values from 06:48 UTC to 07:22 UTC, respectively.

Figure 5 also shows the track of flight 10 during PEACE–B on 14 May 2002. A mid–latitude cyclone was located over central China on 13 May. Warm and moist southerly winds in the lower troposphere intensified ahead of this surface cyclone over southern China, converging into a front extending between 20°N, 100°E and 30°N, 115°E. Deep convective clouds, with tops above 10 km around 30°N, 105–120°E, are found in an infrared (IR) image obtained by the Geostationary Meteorological Satellite (GMS)–5 (see Figure 6d of *Oshima et al. (2004)*). CO over southern Asia in the UT was transported by southerly LT winds over southern China, and trapped by the cumulus convection in that area. The model reproduces similar convective activity around the cyclone and models the CO changes influenced by convective transport, as shown in Figure 5a. Convective transport changes the CO concentration at an estimated rate of 0.5–3 ppbv/hour at 300 hPa over central China. The mass change of CO in the free troposphere caused by the uplift of cumulus convection was calculated by using cloud base mass flux and detraining mass flux in the model. The mass change of CO by the downdraft is

also considered, but the effect is small compared to the uplift over central China. The convective processes associated with this cyclone began on 12 May 2002. Consequently, convection transported about 0.3 TgCO on 12–13 May 2002, and the maximum height of the CO concentration increase caused by convective transport is between 300–400 hPa. A high CO region with the value of 150 ppbv or higher can be seen over northern China and Korea in Figure 5b. The modeled CO increase by convective transport has not been seen at 450 hPa over central China. The model tends to underestimate the detrainment of middle convection, and it is the probable reason why the model was not able to reproduce the CO increase at 450 hPa during the flight. Five–day backward trajectories for air parcels in which the highest CO mixing ratios were observed during this flight indicate that these high–CO air parcels were located over central China (around 30°N, 115°E) 24 hour prior to the measurement (see Figures 5ab of *Oshima et al. (2004)*). The CO over central China between levels of 300-400 hPa increased from 0.98 Tg on 11 May to 1.28 Tg on 13 May. The plume over central China was transported from the surface by the cumulus convection during 12–13 May. It was subsequently advected by westerly winds and moved over central Japan on 13–14 May.

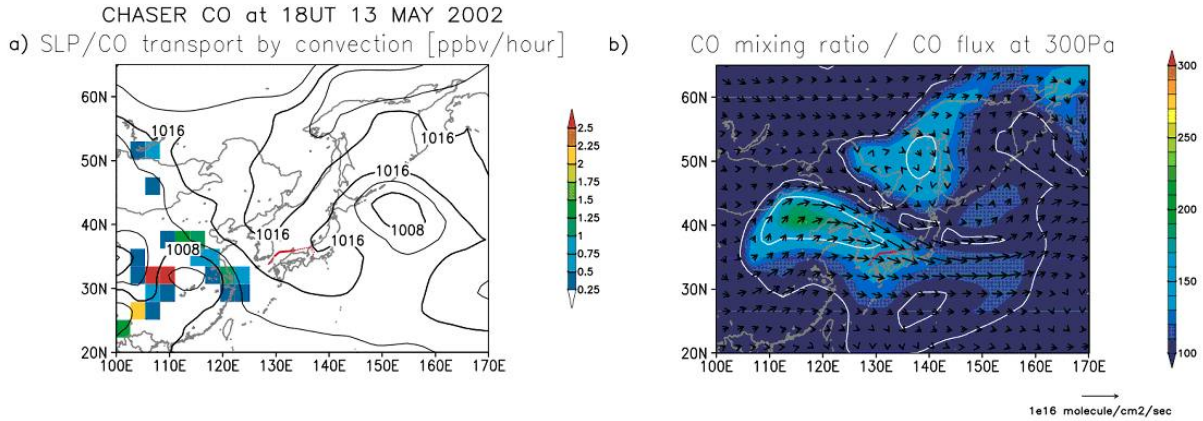


Figure 5 : a) Sea level pressure (contours, units of hPa), and CO tendency by convective transport at 300 hPa at 18:00 UTC on 13 May 2002 (color tones, units of ppbv/hour), b) CO mixing ratio and horizontal CO flux at 300 hPa at 18:00 UTC on 13 May 2002. The contour interval is 50 ppbv. The unit length is shown below the figure. Red lines denote the track of flight 10 of PEACE-B on 14 May 2002, and the leg from 06:48 UTC to 07:22 UTC is shown with the bold line.

4. Summary and Conclusions

Global chemical weather forecasts made with CHASER can be useful in planning measurement flights targeting different types of synoptic scale phenomena, such as near–surface outflow from nearby polluted regions, or intercontinental plumes of pollution in the middle and upper troposphere. The chemical forecast modelled was able to reproduce the CO values observed during the PEACE–A and PEACE–B campaigns. The values were within 10–20% of the observed mixing ratios at three ground–based observational sites in the major sampling region of PEACE, and within 20–30% of the airborne observed mean mixing ratios. The ability to reproduce spatial and temporal variability is critical for planning of measurement flights. Although the model underestimates the background CO level by 20–40 ppbv compared with observations in late spring, it is still capable to reproduce the transport of polluted air masses in the free troposphere. The model estimated the convective transport of CO. The results suggest that about a half of the emissions over China are affected by the cumulus convection in late spring.

Appendix A: One-Way Nested Global Regional Model based on CHASER and WRF/Chem

We are now also developing a one-way nested global-regional air quality forecasting (AQF) model system with full chemistry based on the CHASER (*Sudo et al., 2002a*) and WRF/Chem (*Grell et al., 2005*). Here, description and evaluation of model system are biefly given.

The global atmospheric chemistry transport model (ACTM) part is based on the CHASER model, which is based on CCSR/NIES/FRCGC atmospheric general circulation model (AGCM) version 5.7b. The basic features of the model have been already described in Section 2. The regional ACTM part is based on WRF/Chem (*Grell et al., 2005*). The databases used are the following:

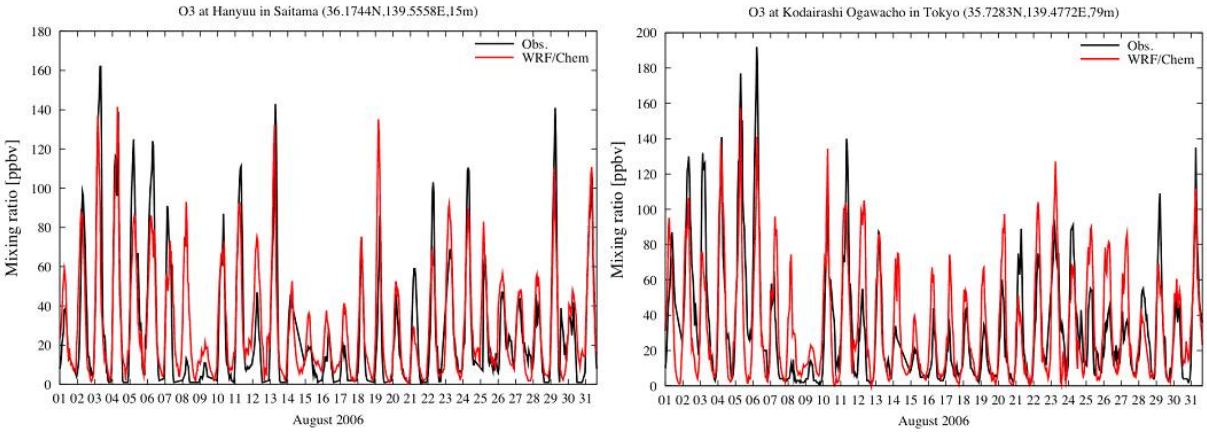
- Anthropogenic emission data over Japan, except those from automobiles, are from the JCAP (Japan Clean Air Program) with 1×1 km resolution (*Kannari et al., 2007*);
- Anthropogenic emissions from automobiles over Japan are from EAgrid2000 (East Asian Air Pollutant Emissions Grid Inventory) with 1×1 km resolution (*K. Murano, personal communication*);
- Surface emissions over China and North and South Korea are from REAS (Regional Emission Inventory in Asia) with 0.5×0.5° resolution (*Ohara et al., 2007*);
- Surface emissions over Russia are from EDGAR (Emission Database for Global Atmospheric Research) with 1×1° resolution (*Olivier et al., 1996*).

Diurnal and seasonal variations in surface emissions are taken into account in the JCAP and EAgrid2000 data, and diurnal variations are also parameterized in emissions from REAS and EDGAR following averaged variations of JCAP. Weekly variation between workdays and holidays is also taken into account in the EAgrid2000 automobile emission data. Note here that emissions based on the statistics in 2000 are applied in the present study. Biogenic emissions are based on *Guenther et al. (1993)*. The outer domain covers Japan with 15 km horizontal resolution (152×52 grids for chemical species), and the inner domain covers the Kanto region with 5 km resolution (111×111 grids for chemical species). The inner and outer domains in the regional ACTM have 31 vertical layers (up to 100 hPa). The two–way nesting calculation is applied in the regional ACTM part.

The lateral boundary of chemical species in the regional ACTM is taken from the global ACTM. The output of the global ACTM is linearly interpolated from the Gaussian latitude and longitude grid to a Lambert conformal conic projection for use in the regional ACTM. The lateral boundary is updated every 3 hour and linearly interpolated for each time step. We did not include feedback from the regional ACTM to the global ACTM; that is, the one–way nesting calculation was done between the global and regional ACTMs. The system is driven by meteorological data from NCEP for the global ACTM part and from the mesoscale model (MSM) of the Japan Meteorological Agency (JMA) for the regional ACTM part. A 15–hour forecast has been produced four times daily at 00, 06, 12, and 18 UTC with a lead time of 8–10 hours since July 2006, following a spin–up of 1 month for the global distribution of chemical species. The initial condition of the meteorological field for the regional ACTM was taken from the MSM for each forecast, and the initial condition of chemical species was taken from the model output driven by the analysis meteorology.

To evaluate the model–calculated ozone, the surface ozone mixing ratio was compared to that observed at air quality monitoring stations. There are 251 stations observing surface ozone within the inner domain of the regional ACTM as of August 2006. For the comparison of temporal variation, hourly averaged values of observed and modeled surface O_3 mixing ratios in August 2006 are shown in Figure A-1. Observed ozone exceeded 100 ppbv from 3 to 6 August at Hanyuu in Saitama Prefecture (36°10’28”N, 139°33’21”E, Figure A-1a), which is

downwind of the Tokyo metropolitan area. The maximum value in the observation was 162 ppbv at 16 UTC on 3 August. The model successfully reproduced the ozone maximum on 3 August. The maximum simulated value was 137 ppbv in the model. The model also successfully captured the decrease from 3 to 7 August, but failed to show the rapid decrease on 8 August. Three typhoons (Maria, Somai, and Bopha) occurred during this period, and the difficulty of predicting the meteorological field may have led to the overestimation of ozone on 8 August. Both the model and observations indicate low levels of ozone from 14 to 17 August as typhoon 200610 (Wukong) approached Japan. The observed and modeled ozone exceeded 100 ppbv on 11 and 13 August, and the model overestimated the ozone mixing ratio on 19 August. The modeled ozone mixing ratio was 135 ppbv, whereas the observed ozone mixing ratio was 86 ppbv. Daily variation in the ozone mixing ratio at nighttime was well reproduced by the model. The daily minimum of observed and modeled ozone exceeded 10 ppbv on 12, 15, 27, and 28 August; except for these days, the ozone level was almost zero during nighttime. The comparison between the modeled and observed daily variation in surface ozone at Kodaira in Tokyo (35°43’42”N, 139°28’38”) is shown in Figure A-1b.



(a) (b)

Figure A-1: Hourly observed (black) and modelled (red) surface ozone mixing ratio in August 2006 at Hanyuu in Saitama prefecture (a) and Kodaira in Tokyo (b).

Maxima of observed and modeled surface ozone at Hanyuu appeared on 3 August, and the observed and modeled ozone mixing ratios at Kodaira were 140 ppbv or higher on 5 and 6 August. The model tended to overestimate the daytime ozone maximum especially for cloudy days, and the discrepancy of daily maximum is larger in urban area compared to that in rural area. To evaluate the model performance, a set of statistical measures provided by the U.S. Environmental Protection Agency (*US EPA, 1991*) was evaluated for stations in the inner domain of the model. The mean normalized bias error (MNBE), the mean normalized gross error (MNGE), and the unpaired peak prediction accuracy (UPA) were 7.1, 9.5, and 9.4%, respectively. These values are within the criteria range suggested by the U.S. EPA (MNBE< ±10–15%, MNGE< ±30–35%, and UPA< ±15–20%).

Acknowledgements

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3.2 Operational Ozone Forecasts for Austria : *Marcus Hirtl[1], K. Baumann-Stanzer[1] and B. C. Krüger[2]*

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1. Introduction

Daily ozone forecasts for Austria have been run in an operational mode during summer 2005 and 2006. The model system has been set up for Austria in cooperation with the Central Institute for Meteorology and Geodynamic (ZAMG) and the University of Natural Resources and Applied Life Sciences (BOKU). The meteorological fields are supplied by the limited area model ALADIN-Austria (run twice a day at ZAMG). The 48-h forecasts are computed on domains with a horizontal resolution of 9.6 km (covering Austria) and 29 km (covering Central Europe). The dispersion modelling is done with the Comprehensive Air quality Model CAMx (version 4.20) with the SAPRC99-mechanism for gas phase chemistry.

2. Description of the Modelling System

The new air quality model system consists of three parts that are linked off-line together (Figure 1). The combination of the two major parts, the meteorological input provided by ALADIN and the chemical model CAMx, was implemented for the first time in this study.

CAMx (Comprehensive Air qualtity Model with extensions, http://www.camx.com) simulates the emission, dispersion, chemical reaction, and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on a system of nested 3D grids. A two grid nesting is used with a coarse grid over Europe and a finer grid for the core area covering Austria with the best possible spatial resolution of 9.6 km (according to the present grid of ALADIN-Austria).

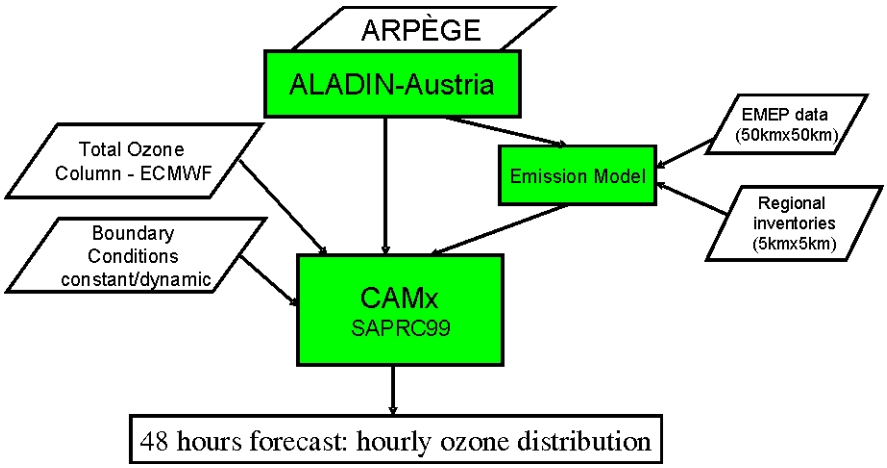

Figure 1 : Structure of the new air quality model system.

The meteorological fields are supplied by the limited area model ALADIN-Austria (http://www.cnrm.meteo.fr/aladin/). It is run twice a day at the ZAMG and renders forecasts for 48 h. The meteorological fields have a temporal resolution of 1 hour. The data is provided on 45 levels, and model has a horizontal resolution of 9.6 km. Fields of wind, temperature, pressure, convective and large scale precipitation, snow cover, solar radiation and specific humidity are extracted directly from the ALADIN dataset. The other fields, cloud optical depth, cloud water- and precipitation water content have to be parameterised (*Seinfeld, 1998*) from the ALADIN output.
The model system generally uses EMEP emissions. For the countries - Austria, Czech Republic, Slovakia and Hungary - the original 50x50 km data are downscaled to 5x5 km based on an inventory from 1995 (*Winiwarter and Zueger, 1996*). The EMEP data for 1999 (used for 2005 forecast) have been substituted by data for 2003 (*Vestreng et al., 2005*). In addition, a new highly resolved emission inventory for the city of Vienna, Austria (*Orthofer et al., 2005*) is used for this area.
The boundary conditions of the coarse grid were estimated from the forecast of the previous day. This method is compared with constant boundary conditions using average summer values. Total ozone column data was obtained from ECMWF data.

3. Operational Forecasts 2006

High ozone values are most frequently observed in the eastern parts of Austria, where warnings for values above the information or the alarm threshold are launched for ozone region 1 (covering lower Austria, Vienna and Burgenland).
Figure 2 shows the predicted maximum concentrations for ozone region 1. The course of ozone concentration from 1 day (Tag 1) and 2-days (Tag 2) model forecasts as well as the results from a backup run which considers constant boundary conditions only are compared to measurements (at 43 air quality stations).

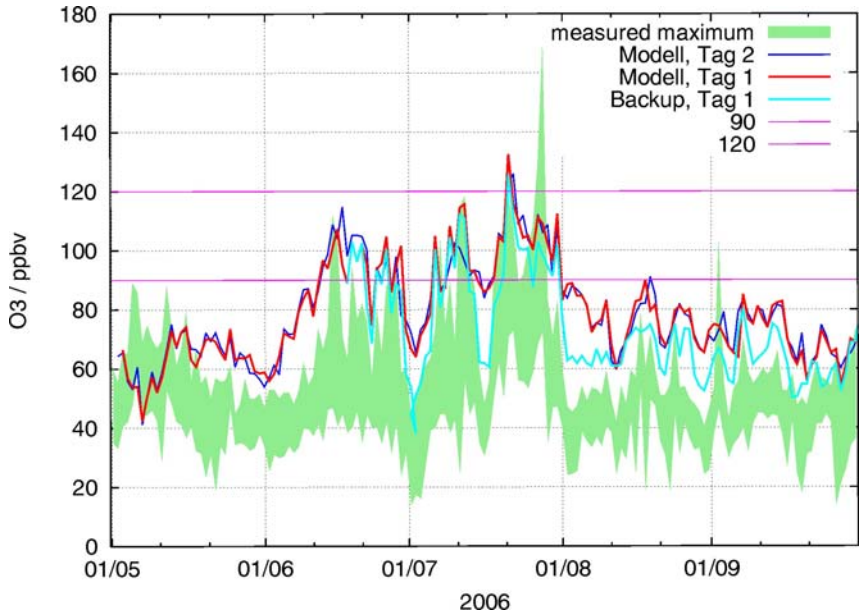

Figure 2 : (Green area: range between highest and lowest maximum observations (hourly average) at stations within the region; Blue line: maximum predicted on same day; Red line: maximum predicted on previous day; Yellow line: backup run (constant boundary conditions)).

In 2006 the exceedances of threshold (i.e. 180 µg/m^3 (≈90 ppbv)) have occured between middle of June and end of July. After that period the predicted concentrations were higher than the measurements. The yellow line shows that the model run considering constant boundary conditions performs slightly better than the dynamic approach during that period.
According to the station measurements, the information threshold has been exceeded on 17 days in the period depicted in Figure 2. Note that 16 of these exceedances were forecasted correctly. The alert exceedance predicted for July 21 was not observed but the measured values were just below the threshold value. The daily maxima above the alert threshold which were observed on 27-28 July 2006, were not predicted by the model. On these days, local peak emissions obviously caused a sudden and local increase of ozone concentrations around noon for 1-2 hours.
Figure 3 shows a scatter diagram of observed vs. modelled ozone in region 1. Depicted are daily maxima for the two forecast days and the backup run. Information and alarm threshold are marked by magenta lines. Most of the values lie above the 1:1 line which means that the model tends to predict higher concentrations than observed.

Table 1 summarizes the metrics of some selected stations in ozone region 1. The correlation and the standard deviation between observations and model prediction of the two forecast days as well as the backup run are depicted. The last line shows the values for all 43 stations in ozone region 1.

The correlation between the two forecast days with the measurements is practically the same. It seems that the backup run correlates better with the observations for the selected stations as well as for the whole region studied. The standard deviation is also smaller for backup run.
The statistical comparisons between ozone forecasts and measurements for all stations in eastern Austria show correlation coefficients between 0.4 and 0.7 and standard errors around 12 ppbv for 2006.

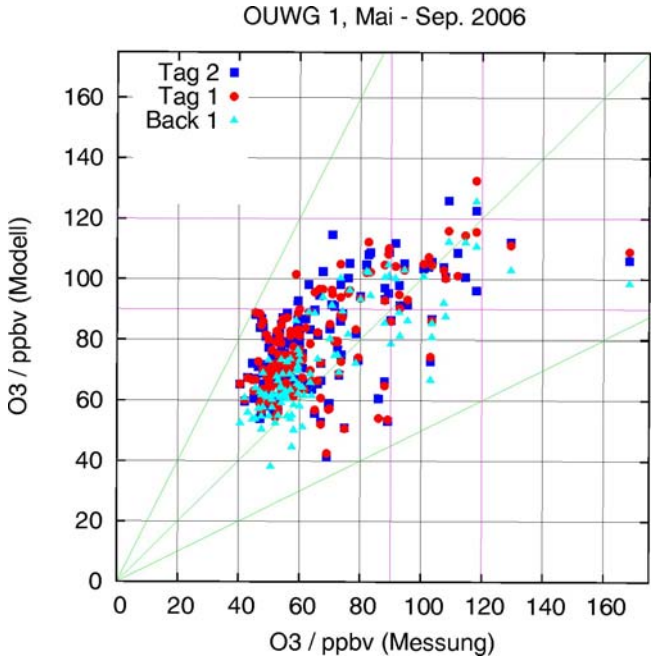

Figure 3 : Scatter-diagram of daily ozone maxima predicted versus observed in ozone region 1 for 2006 (Blue: maximum predicted on previous day; Red : maximum predicted on same day Light blue: maximum predicted by back up run; Magenta lines: information and alert threshold (*Directive 2002/3/EC*)).

	forecast day 1		forecast day 2		Backup day 1	
	r	s	r	s	r	s
Eisenstadt	0.62	12.16	0.61	12.54	0.73	10.26
Kittsee	0.63	13.47	0.64	13.15	0.72	11.75
Himberg	0.69	13.10	0.69	13.29	0.76	11.51
Schwechat	0.71	13.85	0.71	13.99	0.78	12.10
Tulln	0.65	14.27	0.65	13.84	0.70	12.49
Wiener Neustadt	0.54	12.23	0.55	12.27	0.64	10.48
Hohe Warte	0.68	13.33	0.68	13.53	0.75	11.56
Lobau	0.71	13.71	0.71	13.99	0.78	12.03
Average	**0.59**	**13.03**	**0.59**	**13.00**	**0.70**	**10.93**

Table 1 : Correlation (r) and standard deviation (s) of selected stations in ozone region 1.

It was investigated how accurate exceedances of the information threshold could be predicted by the model. The following combinations were considered, i.e. days when:

- the highest value of the observations as well as the model forecasts are bellow the information threshold (90 ppbv) in ozone region 1;
- observation and model lie above the threshold;
- the observations lie below the threshold, the model above it;
- the observations lie above the threshold, the model below it.

Table 2 shows the values for the respective months. Operational forecasts for 2006 and 2005 are compared with results of the backup run.

Although the predicted ozone values tend to be higher than the observations in 2006 the hit rate to predict the exceedance of the information threshold is 88%. This is slightly better than

the backup run in 2006 (84.8%) and it is worse than in 2005 (90.9%). These values include also months with low ozone values in Austria. During May, August, and September, no exceedances of the information threshold occured due to meteorological conditions. Considering only June and July 2006 (in total 61 day) the hit rate would be 70.5 and 63.6% for the operational and backup runs, respectively.

Obs.	*Mod.*	*May*	*June*	*July*	*Aug.*	*Sep.*	*Total*	*%*
Forecast 2006, day 1								
<90	<90	28	16	11	31	30	116	88.0
>90	>90	0	3	13	0	0	16	
<90	>90	0	11	6	0	0	17	12.0
>90	<90	0	0	1	0	0	1	
Total		**28**	**30**	**31**	**31**	**30**	**150**	
Backup-run 2006, day 1								
<90	<90	-	7	10	31	30	78	84.8
>90	>90	-	1	10	0	0	11	
<90	>90	-	4	7	0	0	11	15.2
>90	<90	-	1	4	0	0	5	
total		-	**13**	**31**	**31**	**30**	**105**	
Forecast 2005, day 1								
<90	<90	-	3	25	30	27	84	90.9
>90	>90	-	2	4	0	0	6	
<90	>90	-	2	0	1	2	6	9.1
>90	<90	-	0	2	0	1	3	
Total		-	**7**	**31**	**31**	**30**	**99**	

Table 2 : Hit-rate for the exceedance of the information threshold.

4. Conclusion

Daily ozone forecasts for Austria have been run during summer 2005 and 2006. The results of the forecasts have been evaluated with measurements of the Austrian air quality network for eastern parts of Austria. Generally the observed exceedances of the information threshold are reproduced by the model. Days with exceedances of the information threshold were predicted by the model with a probability of 88% during the summer period in 2006. The daily maxima above the alert threshold were not predicted by the model probably due to rather low resolution. The ozone forecasts are continued with an improved model system in the ozone season 2007. The emission inventories as well as the CAMx model will be updated. Additionally a new approach to obtain the boundary conditions from climatological average values will be applied and test runs with increased spatial resolution will be conducted.

Acknowledgements
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Landesregierung, Burgenländischen Landesregierung, Steiermärkischen Landeregierung, Kärntner Landesregierung, Oberösterreichischen Landesregierung, and Salzburger Landesregierung.

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3.3 Impact of Nesting Methods on Model Performance : *Ursula Bungert and Heinke Schlunzen*

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1. Introduction

The air quality directive of the European commission demands maps on concentrations and exceedances in different detail. For this purpose numerical models can be used. Some model systems are already adjusted to deliver the corresponding maps. For instance, the model system M-SYS consists of three mesoscale and one microscale model areas and applies one-way-nesting for meteorology and chemistry (*Trukenmüller et al., 2004*).
To calculate concentration data, reliable meteorological fields are needed, which should be calculated with the same resolution as the concentration maps. *Lenz et al. (2000)* have shown that the concentration fields are more sensitive to the description of the meteorological fields at the lateral boundaries than to the concentration fluxes over these boundaries. Nesting the meteorological fields, corresponding to the use of meteorological model results received on a coarser grid as time-dependent boundary values, needs to prescribe the boundary values as realistic as possible. In this work, we consider only the nesting of the meteorological fields and the influence of different update intervals of the forcing data on the model performance of nested simulations.

2. Method

The multiscale meteorology and chemistry model system M-SYS consists of the mesoscale models: MEsoscale TRAnsport and Stream (METRAS; *Schlünzen, 1990; Schlünzen and Katzfey, 2003*) and MEsoscale Chemistry Transport Model (MECTM; *Müller et al., 2000; Schlünzen and Meyer, 2007*), which are used in different resolutions, and the obstacle-resolving microscale models MITRAS and MICTM (*Schlünzen et al., 2003; Lopez et al., 2006*). Meteorology and chemistry transport are coupled off-line. Because the same grid, the same model physics and parametrizations are used, no interfaces are needed for the

meteorology-chemistry coupling. Model results are, beside others, maps of concentration fields on different scales with different resolutions.
Although, the nesting is applied in M-SYS at every time step, the forcing fields are only available at time intervals that are much longer than the time step used in the model. During these intervals, the forcing fields are linearly interpolated. To investigate, if the linear interpolation represents the development of the atmospheric fields in a realistic way, the mesoscale atmospheric model METRAS is used in different resolutions. The results of coarser grid simulations are used as forcing fields for the nested simulations in higher resolution. The time interval for writing the coarser model results determines the duration in which the forcing fields in the higher resolution simulations can be updated. By using the same model for calculating forcing data and performing the nested simulations on a higher resolving grid it is possible to perform well-controlled sensitivity studies on the effect of update interval on high resolution model results.
For nesting of meteorological models and for coupling of meteorology and chemistry transport it is important to know, how often the atmospheric forcing fields should be updated to sufficiently represent the non-linear processes triggered from the boundaries. If the update interval for the forcing fields is very small, the non-linear processes in the atmosphere should be well represented during these short intervals where a linear interpolation is applied. By continuous updating we eventually receive a model with multiple grids, which is a valid (but expensive) approach. However, despite costs this approach is not always possible, since limited area models need at some point lateral boundary values. At the outermost domain the forcing data are only available at specific time intervals. It needs to be known in which frequency these forcing data should be available. In addition, reading of forcing data needs additional time on the computer and thus, should be reduced to the necessary amount. Data update should be performed as much as necessary and as little as possible.
Changes in the atmospheric fields happen on different time scales and with different speed. Therefore, we adapt the time intervals for writing the model results (that are the forcing data for nested simulations) to the time scales, in which the atmospheric fields change, instead of using short but constant time intervals. If significant changes happen on short time scales, the results should be written more often than for more or less steady conditions.
The horizontal wind components, potential temperature and specific humidity are used in METRAS as forcing fields. Changes in the scalar quantities temperature and humidity are mainly induced by advection and diffusion, i.e. processes that depend on the wind. Therefore, we define the conditions for writing the model results only in dependence of changes in the horizontal wind components. The model results were alternatively written at regular intervals (3 h, 6 h), if the accelerations (changes in velocity) in 80 and 20% of the grid points are less than $5 \cdot 10^{-5}$ and $5 \cdot 10^{-6}$ m s^{-2}, respectively

3. Simulation Set-up

The outlined method of a situation-controlled writing of the model results is applied to a period in August 2003. During the simulated period (29-31 August 2003), a trough was extended from Spain to the Arctic northeast of Finland, lying between a high-pressure systems over the Atlantic Ocean and south-eastern, and then later Eastern Europe. In between this trough, a small scale low has developed originally lying over Belgium on 28th August, and then passed in west-east direction over northern Germany. Caused by this low, a significant precipitation occurred in this area.
The model areas used for simulating this case are shown in Figure 1. The large area (Figure 1a) covering significant parts of Europe has a horizontal resolution of 18 km, while a grid size

in the small area (Figure 1b) is 6 km. Model simulations are started for 20 CET, 28 August 2003. Comparisons are performed starting 1 hour after simulation begins (from 21 CET, 28 August 2003).

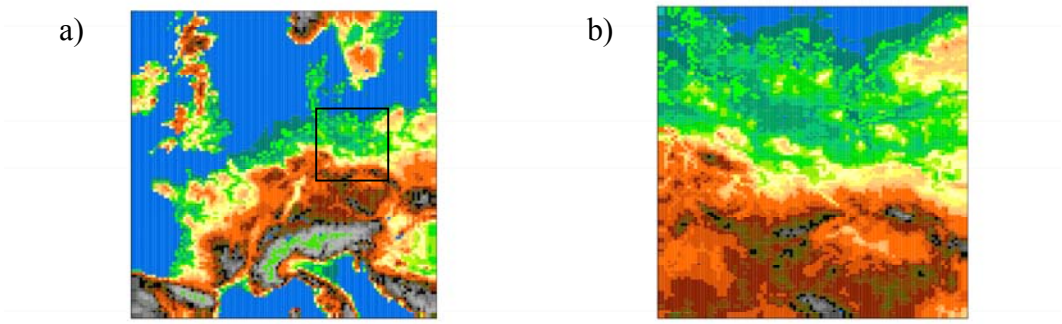

Figure 1 : The model areas used for the coarse grid run yielding the forcing data (a) and for the nested simulations for the case „low pressure system over Europe“. The positioning of the nested area is shown by the frame in (a).

Table 1 : Simulations performed with the high-resolution model area shown in Figure 1b.

	Simulation name	Forcing data from	Update interval
1	eu_6km_3h	coarse grid METRAS run	3 h
2	eu_6km_6h	coarse grid METRAS run	6 h
3	eu_6km_lcout80	coarse grid METRAS run	depending on results (80%)
4	eu_6km_lcout20	coarse grid METRAS run	depending on results (20%)
5	eu_6km_ana	analysis	12 h
6	eu_6km_nonesting	----	----

Several simulations on the 18 km grid that use different conditions for writing the model output results produce the forcing data for the nested simulations that use a 6 km resolution. The 6 km simulations are summarized in Table 1. As seen, besides the four simulations that are nested in METRAS 18 km results, two additional simulations were also performed. In the simulation 5 (eu_6km_ana) the forcing fields are derived from analyses that are available every 12 h; thus, a very large update interval is used. This update interval cannot be changed. Simulation 6 (eu_6km_nonesting) uses no nesting and no heterogeneous initialisation, but only integrates the initial profile forward in time. This is a sensitivity study to allow evaluating, if the large-scale situation has any impact on the high-resolution results.
In the following we compare results of the all simulations given in Table 1 to each other and to a reference case. This reference case is a simulation with high resolution in the whole domain (Figure 1a).

4. Simulation Results

The hit rates (in %) for wind speed and direction, and temperature were calculated for every hour of the simulation using allowed deviations (see Table 2). All hit rates are based on comparisons with the reference case and include all grid points (about $3 \cdot 10^6$) of the model domain in Figure 1b.

Values for the hit rates are shown in Figure 2 for the six different simulations. The simulation 6 has by far the largest deviations from the reference case. This shows that the result is indeed

sensitive to changes in the large-scale situation. In addition, also simulation 5 yields quite low hit rates. There are two probably reasons for this: a) the update interval of the forcing data is too small compared to the changes in the large-scale situation; b) the forcing data derived from the analysis are differ from the coarse grid simulation results and therefore lead to the low hit rates which are based on the results of the reference simulation.

Table 2 : Desired accuracy DA used for calculating the hit rate (from *Cox et al., 1998*).

Variable	Temperature (°C)	Dew point depression (°C)	Wind speed (m s^{-1})	Wind direction	Pressure (hPa)
Desired accuracy DA	± 2	± 2	± 1 for ff< 10m s^{-1} ± 2.5 for ff> 10m s^{-1}	± 30°	± 1.7

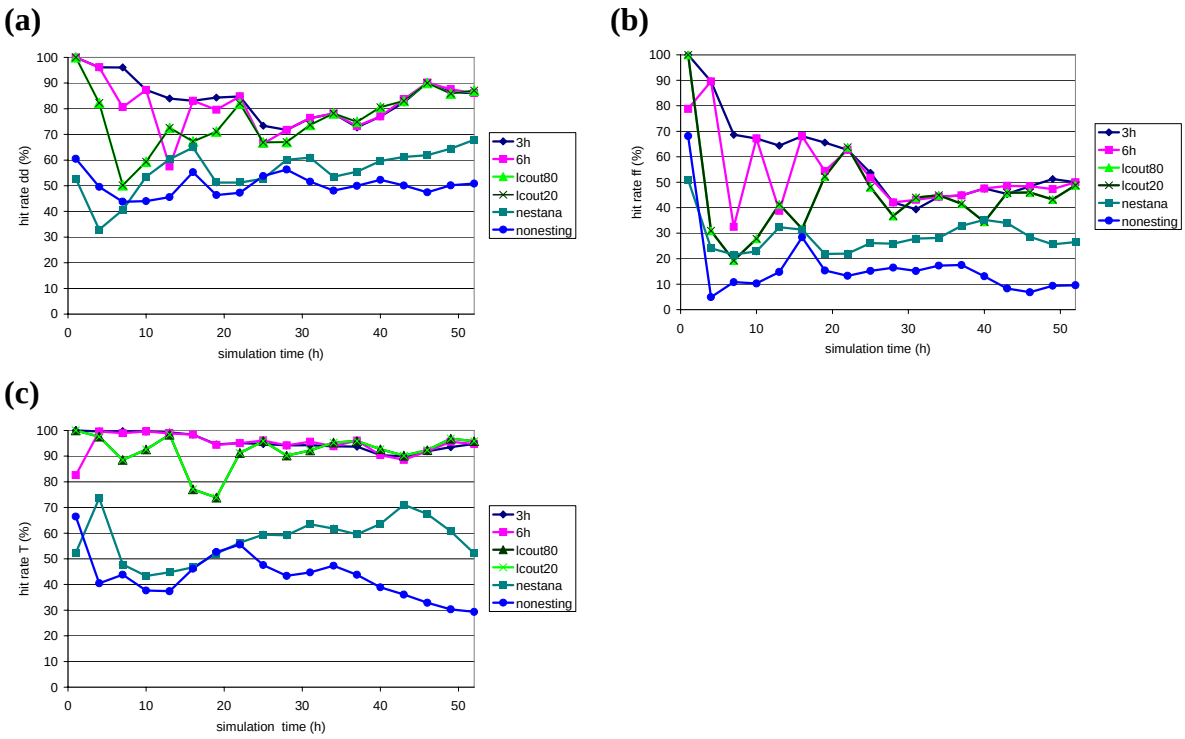

Figure 2 : Hit rates for wind speed (a), wind direction (b) and temperature (c). The hit rates are calculated by comparing results of the simulations with the high-resolution reference case.

The simulations nested in the coarse grid METRAS results agree in a similar way with the reference simulation in the second half of the simulation time (after about 21 h; i.e. 18 CET of 29 August 2003). At this time the performance of simulation 5 is somewhat closer to the other nested simulations than before. This might be a hint that the nesting becomes less relevant and the situation is more locally driven. In the first 21 hours of the simulation the two simulations with constant update intervals (3 h, 6 h) are closest to the reference case, while the adaptive update simulations (3, 4) show a high variability in performance. This is a hint that the acceleration is probably not a reliable measure to determine update intervals. The best performance is received in the present case study for a nesting every 3 h.

5. Conclusions and Outlook

METRAS is able to yield simulation results on different scales and details. Nesting is a helpful to improve the model performance if the forcing data represent the meteorological situation in a realistic way. The current studies on the impact of update interval lengths on model performance lead to the following conclusions:

a) In general, shorter update intervals lead to higher model performance;
b) Consecutive, but very different intervals seem to reduce the positive effect;
c) With increasing forecast time, the uncertainty in the forecast (that is resulting from the update of the boundary values) seems to decrease.

Especially the last result is surprising, since it is generally assumed that for longer forecast with limited area models the boundary values become more relevant. Therefore, it needs to be evaluated, if this is a result only true for the situation studied or if it is more general.

A next step to further investigate the influence of different update intervals will be to define a characteristic time for the occurring changes and using this characteristic time to control the writing of the data that are used as forcing fields in the high resolution simulations. First results seem to be promising in enhancing model performance compared to the constant update interval of 3 h.

Acknowledgments:
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3.4 Running the SILAM Model Comparatively with ECMWF and HIRLAM Meteorological Fields - a Case Study in Lapland: *Marko Kaasik[1], M. Prank[1] and M. Sofiev[2]*

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Abstract
The reproduction of an excessive aerosol event recorded during a measurement campaign carried out in April-May 2003 at the Värriö monitoring station (Finland, Eastern Lapland) was studied with the SILAM model. Both the Lagrangian and Eulerian kernels and ECMWF and HIRLAM (of FMI) meteorological fields were used. Applying adjoint modelling technique, the source of aerosol peak was identified as the Cu-Ni smelters of the Pechenganickel Enterprize (city of Nickel, Murmansk region, Russia) Nikel Metallurgy factory, Russia, about 200 km north of measurement site. In forward runs, the plume from Nikel matched the measurement site, except the Lagrangian run with ECMWF fields. Differences in concentrations and duration of modeled peak were significant between the Lagrangian and Eulerian runs and different meteorological data.

1. Introduction

This case study is intended to clarify the range of uncertainties in meso-scale atmospheric dispersion modelling due to different dispersion schemes (i.e. Lagrangian and Eulerian) and meteorological input. Propagation of a plume from a single point source was recorded in a monitoring station with a high timporal resolution. Thus, this case gives a deeper insight into variability of reproduction of instantaneous advection and dispersion conditions without diluting impact of numerous sources and time averaging.

2. Methods

This modelling study is based on the aerosol measurement campaign carried out during April-May 2003 at the Värriö monitoring station (67°46'N, 29°35'E; Finland, Eastern Lapland) located close to the Russian border (*Ruuskanen et al., 2004*). On the generally low aerosol background of Arctic spring only a few pollution episodes were observed (Fig. 1). In this paper we will focus on the highest episodes (i.e. up to 30 μg/m^3 of PM10 on May 2-3).

First, the SILAM model (*Sofiev et al., 2006*) was applied in adjoint mode to identify the potential sources of pollution. It was found that the aerosol peak of May 2-3 most probably originated from the Nikel metallurgy factory (Kola Peninsula, Russia) located about 200 km north from Värriö (*Kaasik et al, 2007*). Then the SILAM model was applied in a forward mode comparatively with the ECMWF and HIRLAM (FMI) meteorological datasets: EMEP emission data on sulphate and PM, sea salt emissions calculated by SILAM, emission model based on (*Mårtensson et al., 2003*). The well-tested Lagrangian kernel (Lagrangian particle model, version 3.7) and new Eulerian kernel (test version 4.1 released in 2007, presently under evaluation) of SILAM were applied with both meteorological datasets; thus, producing ensemble of four runs. Parameters of a highly buoyant plume from high stack were assigned: source height distributed from 200 to 1000 m.

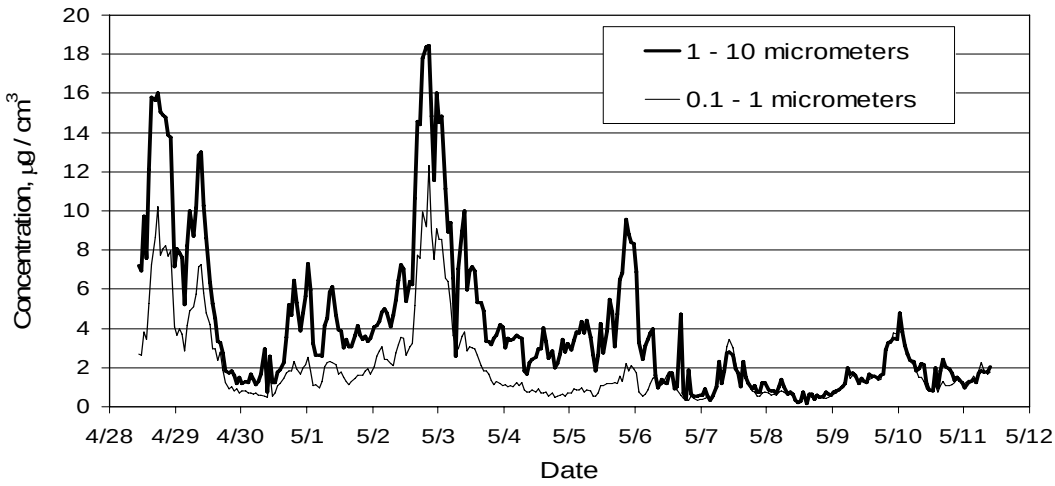


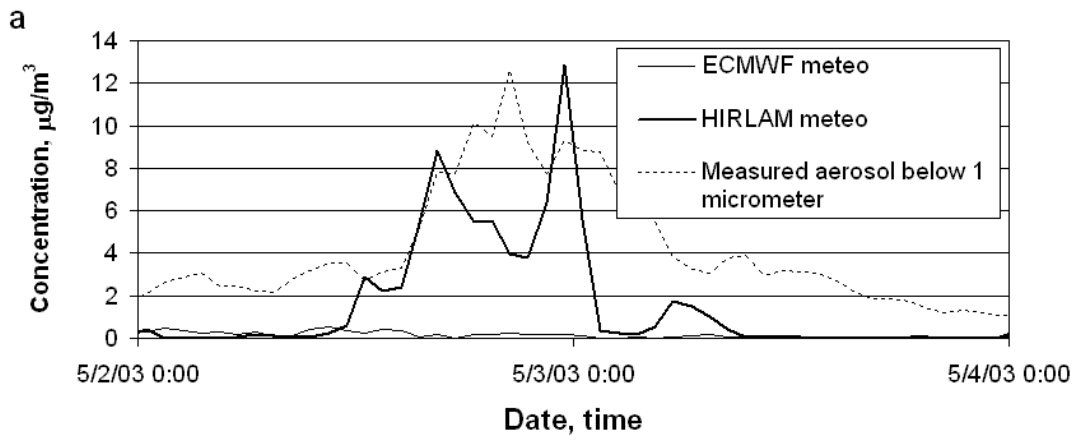
Figure 1: Measured surface-level concentrations of PM10 and PM1 in Värriö during April-May 2003.

3. Results

In the large scale the concentration patterns produced in all four runs were rather similar, but some differences concerning the May 2-3 peak appeared critical for local measurement-modelling comparison (see Fig. 2):

- the Lagrangian plume with ECMWF data narrowly missed the monitoring station passing eastwards; but with HIRLAM data plume matched the monitoring station, predicted concentrations and timing rather similar to measured ones;

the Eulerian plume with ECMWF data matched the monitoring station with slight delay, predicted concentrations were overestimated; but with HIRLAM data plume matched the monitoring station with a slight delay, predicted concentrations as highly overestimated.



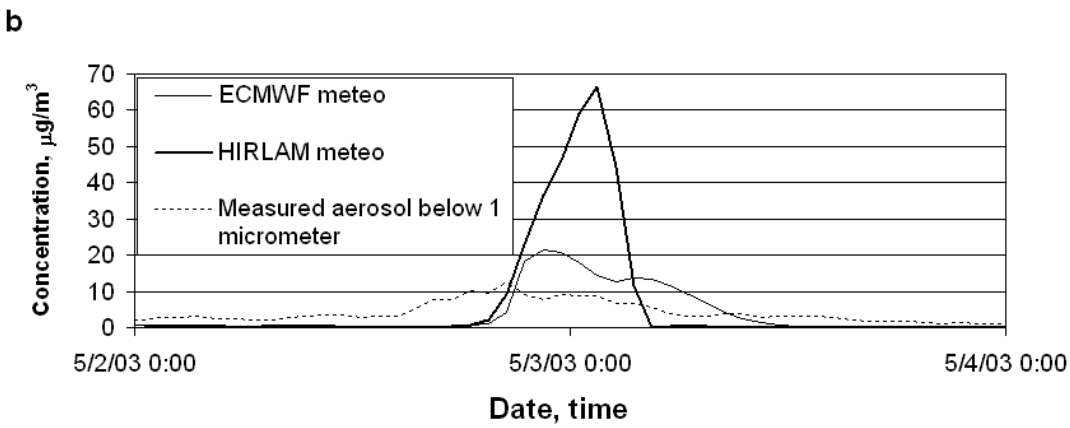


Figure 2: Surface-level concentrations of sulphate in Värriö during the peak event, modeled with SILAM: (a) Lagrangian kernel, (b) Eulerian kernel with ECMWF and HIRLAM meteorological input, compared to the measured concentrations of aerosol particles with diameter below 1 m.

It appeared that contribution of other emissions besides sulphates to this particular event was negligible. Thus, only the measured aerosol below 1 μm size limit (typical for sulphate aerosol) was taken for comparison.

Surface-level concentration maps of sulphate computed with (Lagrangian) SILAM 3.7 are presented in Fig. 3. The ECMWF data run gives a narrow plume directly to south from the plant, which misses the Värriö site just by one grid cell (20 km). The HIRLAM data run forces the plume to travel, at first, to south, and then to south-west passing over Värriö.

In general, the Eulerian scheme (version 4) produced much wider horizontal spread than the Lagrangian (Fig. 4), despite the higher concentration peaks. Runs with both meteorological drivers showed the sulphate polluted plume over Värriö, but with about 5-h delay compared to both measurements and Lagrangian (HIRLAM data) run.

Differences were found in vertical spread as well. In the Lagrangian run (Fig. 5) the concentration fields in the lower free troposphere followed the surface-level concentrations in general. This reflected the simplified vertical structure of this kernel, in particular, well-mixing assumption for the boundary layer and fixed diffusion term in the free troposphere. In the Eulerian run (Fig. 6), the higher-level modeled concentrations were patchy and less correlated with the near-surface fields in comparison with the Lagrangian run.

4. Discussion

The weather situation during the peak was complicated due to a high-pressure system with a centre located close to the monitoring site. Thus, wind was weak, changing rapidly in space and time. In such conditions small discrepancies between the meteorological fields and ways to treat these in the dispersion model become essential.

In this case study the Lagrangian run with HIRLAM data appears to match the observations better than the others. However, its plume was too narrow and its dispersion velocity seems to be somewhat over-predicted (Fig. 2a). Also, it is obvious that the Lagrangian run showed higher variability of the concentrations than the Eulerian one. The Eulerian plume appears much wider, traveling slower due to larger mass fraction near the surface and, thus, matching the measurement site more certainly. It also resulted in a rather smoother shape of concentration time series (Fig. 2b).

The Eulerian run with ECMWF meteodata appeares closer to the shape of observed time series while the stronger vertical motions predicted by the HIRLAM system caused a strong

peak in the concentrations.

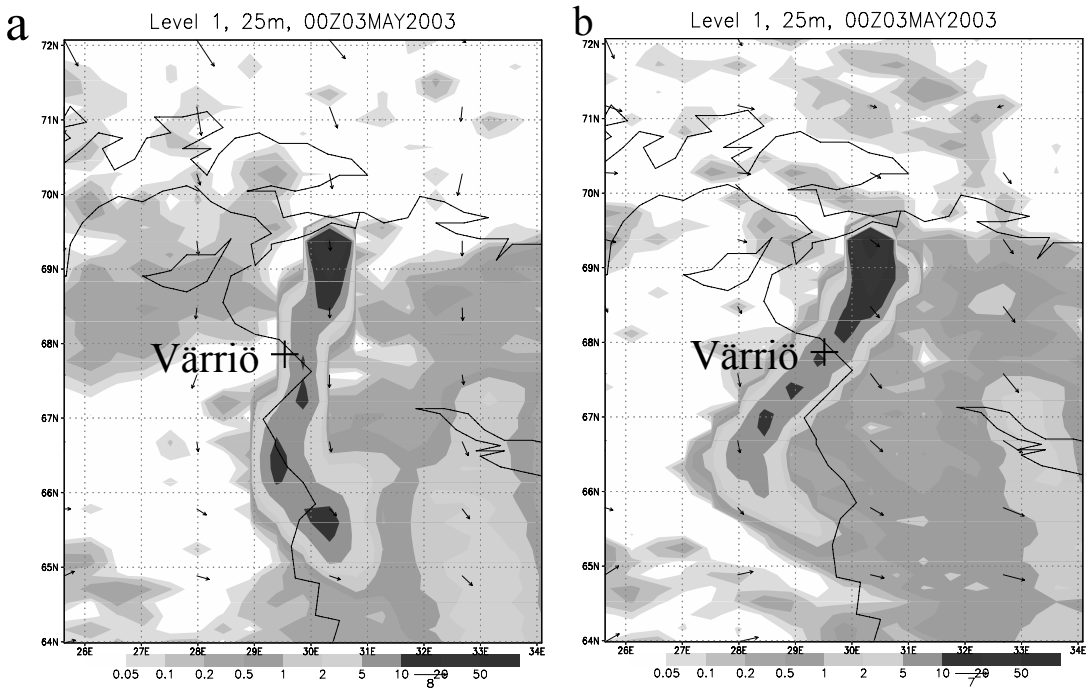


Figure 3: Surface-level concentration fields of sulphate, 00:00 at May 3, 2003, calculated with Lagrangian SILAM 3.7: a) with ECMWF meteorological fields, b) with HIRLAM (FMI) meteorological fields. Arrows – wind at 25 m.

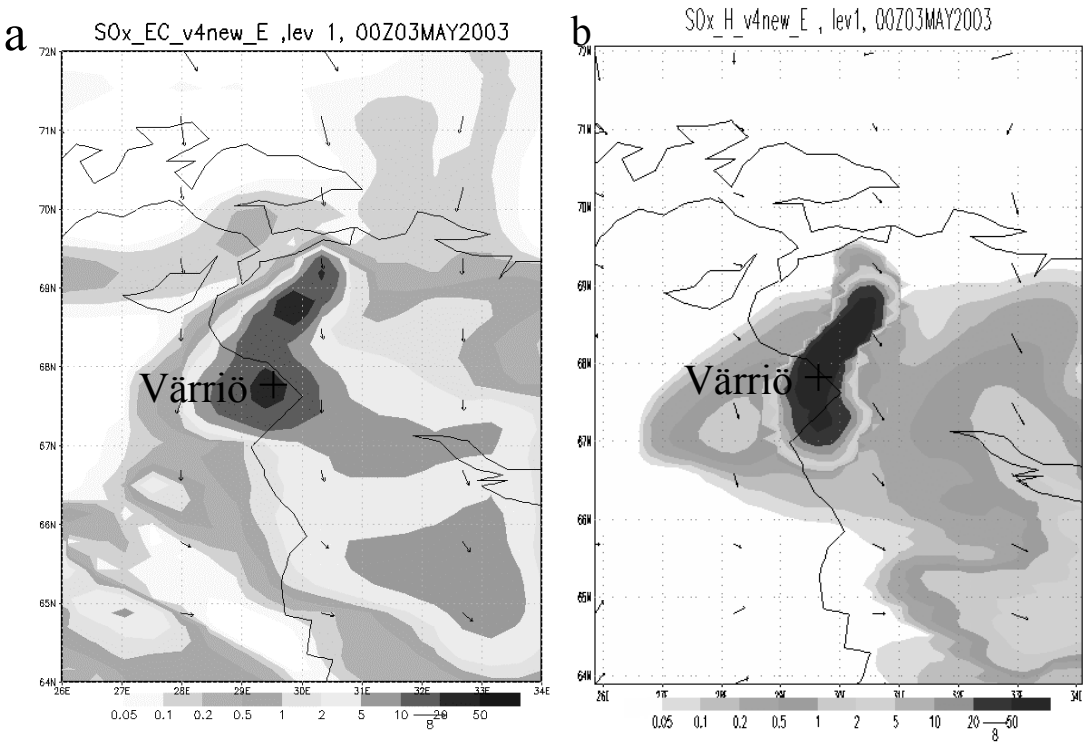


Figure 4: Surface-level concentration fields of sulphate, 00:00 at May 3, 2003, calculated with Eulerian SILAM 4: a) with ECMWF meteorological fields, b) with HIRLAM (FMI) meteorological fields. Arrows – wind at 25 m.

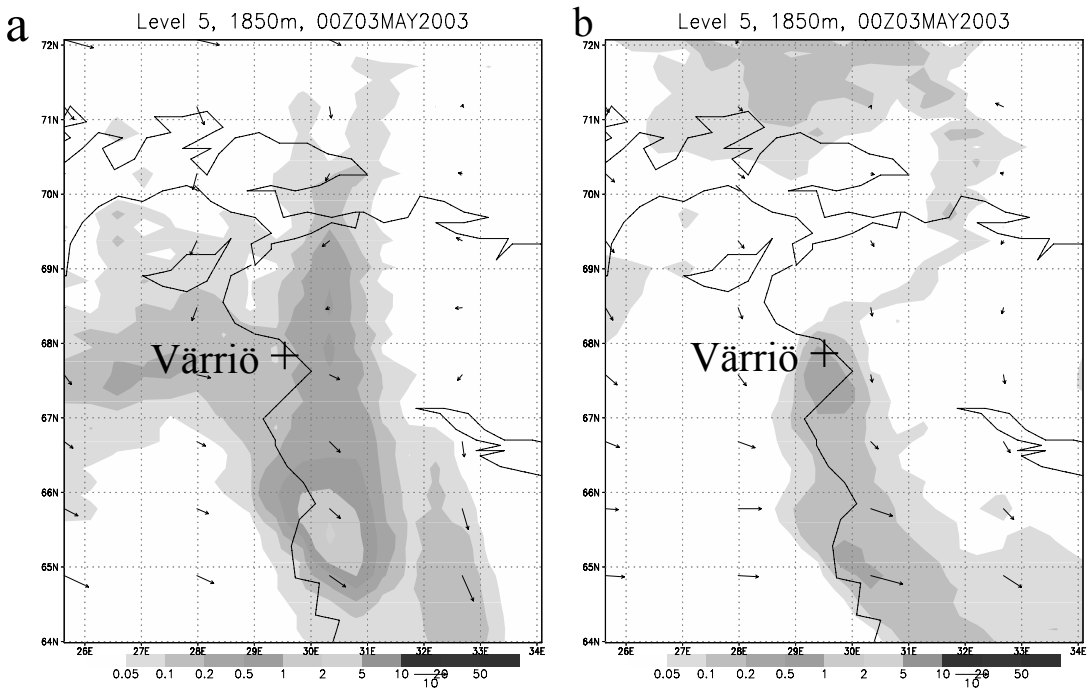


Figure 5: 1850 m fields of sulphate, 00:00 at May 3, 2003, calculated with Lagrangian SILAM 3.7: a) with ECMWF meteorological fields, b) with HIRLAM (FMI) meteorological fields. Arrows – wind at 1850 m.

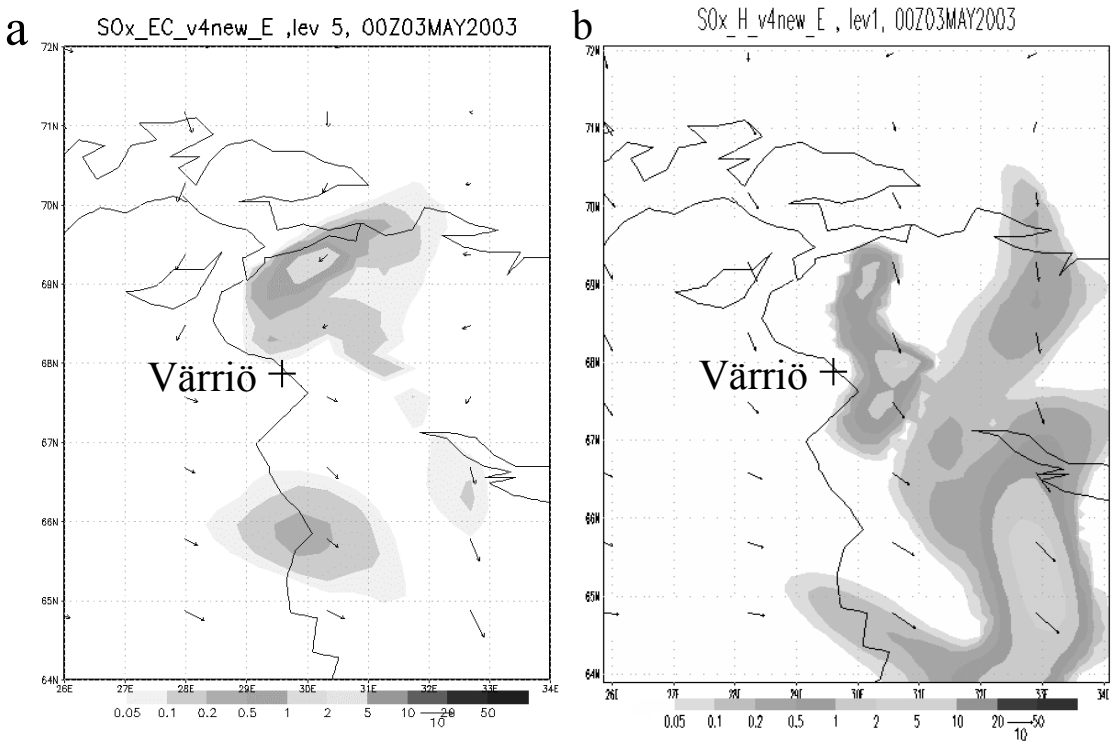


Figure 6: 1850 m fields of sulphate, 00:00 at May 3, 2003, calculated with Eulerian SILAM 4: a) with ECMWF meteorological fields, b) with HIRLAM (FMI) meteorological fields. Arrows – wind at 1850 m.

5 Conclusions

The current exercise highlights the objective difficulties in predicting the short-term episodes originated from a single nearly-point source and observed at a single site. It is, however, evident that an ensemble of four partly independent chemical-weather predictions appeared able to show high probability of the episode, its stochastic features, and a potential range of uncertainties in the results of simulations.

Acknowledgements

This study was supported by the Estonian Science Foundation, Grant 7005 and performed within the scope of the SILAM system evaluation for EU-GEMS and ESA-PROMOTE projects.

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Chapter 4. HIRLAM/HARMONIE - Atmospheric Chemical Transport Models Integration Session

The 'HIRLAM/HARMONIE-ACTMs integration' discussion session was arranged on Tuesday 22 May 2007 during the COST728-NetFAM 'Model Integration' workshop. Additionally to two overall introductory presentations by Alexander Baklanov, DMI (see on: http://netfam.fmi.fi/Integ07/baklanov_prese2.pdf) and Sander Tijm, KNMI (see on: http://netfam.fmi.fi/Integ07/tijm_prese.pdf) and round table discussions the following six specific presentations of the HIRLAM partners models were done (see the corresponding papers published in this volume):

- Status and Evaluation of Enviro-HIRLAM: Differences between On-line and Off-line Models : Ulrik Korsholm, Alexander Baklanov and Jens Sørensen (page 44)
- Coupling of Air Quality and Weather Forecasting - Progress and Plans at met.no : Viel Ødegaard, Leonor Tarrasón and Jerzy Bartnicki (page 106)
- A Note on Using the Non-Hydrostatic Model AROME as a Driver for the MATCH Model: Lennart Robertson and V. Foltescu (page 112)
- Modelling of Air Pollution from Urban to Regional Scale: DMI-CAC : Allan Gross et al. (page 115)
- Aerosol Species in the AQ Forecasting System of FMI: Possibilities for Coupling with NWP Models : Mikhail Sofiev (page 115)
- Running the SILAM Model Comparatively with ECMWF and HIRLAM Meteorological Fields - a Case Study in Lapland: Marko Kaasik, M. Prank and M. Sofiev (page 143).

4.1. Notes on 'HIRLAM/HARMONIE-ACTMs integration' discussion session: *Laura Rontu[1] and Alexander Baklanov[2]*

[1] { Finish Meteorological Institute (FMI), Helsinki, Finland}
[2] {Danish Meteorological Institute (DMI), Lyngbyvej 100, 2100 Copenhagen, Denmark}
Correspondence to:Laura Rontu (laura.rontu@fmi.fi)

Section aim and reasons of the meeting

The main idea of the section is to get together for NWP and ACTM modellers, discuss and build our joint strategy for developing integrated system(s) based on HIRLAM.
The HIRLAM consortium and ACTM modellers in HIRLAM-organisations have some interest and initiatives in such integration, but they work separately and have very low level of coordination and cooperation.
There are also several attempts in this direction, including the following.
- DMI is actively working with development of the on-line integrated system Enviro-HIRLAM, considering aerosol forcing mechanisms, etc.
- Most of HIRLAM-members institutes are using national HIRLAM NWP outputs as meteo-drivers for their ACTP modelling and air quality forecasting activities. They already have attempts to build off-line integrations of HIRLAM with own ACTMs (CAC, Chimere, DERMA, EMEP, MATCH, SILAM).

- Such work was also included in the HIRLAM-A development plan (S4.10/4.5 Task: Coupling with atmospheric chemistry).
- The 'Integration' WG2 in COST 728 involves 5 HIRLAM-member institute representatives (DMI, FMI, Met.no, SMHI, Estonian Tartu Univ.), and they are willing to consolidate and coordinate joint efforts in this work for coupling HIRLAM with ACTMs.

Notes from the discussion

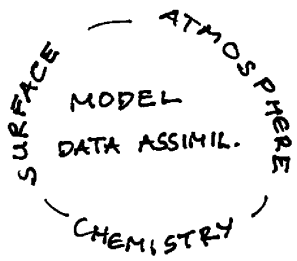
In the discussion session, short presentations by Alexander Baklanov, Sander Tijm, Lennart Robertson and Allan Gross were given (available at the workshop site at http://netfam.fmi.fi/Integ07). The discussion roughly followed a list of items suggested in Sander's presentation: interfaces, NWP output parameters needed by ACTMs, species influencing in weather prediction, suggestions for a chemical module of HIRLAM. In the following notes, individual discussion topics are shown in free order, adding also some elements that were touched outside the HIRLAM-HARMONIE session.

Should the NWP model (HARMONIE) produce air quality forecast in addition to the weather forecast?

- No exact answer was agreed for the current stage, however, some steps forward in this direction should be done and an interface allowing both on- and off-line coupling of HIRLAM/HARMONIE and air quality models should be built.
- In perspective the integrated NWP-ACTM modelling is very promising way for future atmospheric simulation systems leading to a new generation of models for improved meteorological, environmental and “chemical weather” forecasting.
The online integration of MetM and ACTM models gives a possibility to consider feedbacks of air pollution (e.g. aerosols) on meteorological processes and climate forcing, therefore it could also improve the weather forecasting itself.

Interfaces and coupling methods

- Use universal, well defined and documented software like OASIS4 ?
- Coding standards, interface rules, definition of the coupling parameters.
- Coupling of atmospheric model, chemistry and surface scheme both in data assimilation and prognostic regimes.
- Within HARMONIE cooperation, working group on (surface-related) interfaces is working: include chemistry aspects?



What is the importance for ACTM and NWP models of the good description?

- Initial state (data assimilation),
- Surface description (orography, land use, vegetation...),
- Sources and sinks (surface fluxes and emissions).

Use the same physiography data base for surface, atmosphere and chemistry, depending on application?

Importance of data assimilation in the ACTMs

- In the discussion, the importance of data assimilation was stressed.
- Very different amount and character of available meteorological and chemical observations.
- Using 4DVAR to include and combine all information.

Consistency of numerical algorithms (advection schemes) between ACTM and NWP models

- possibly different mass conservation requirements in ACTM and NWP models,
- improvement harmonisation of advection schemes is important.

What do the ACTM expect from NWP (HIRLAM/HARMONIE) models? - experience of ENVIRO-HIRLAM:

- microphysics, which would allow building of advanced cloud-aerosol interactions (indirect aerosols effects);
- radiation parametrizations, which would allow inclusion of direct and indirect aerosol interactions;
- improved 3D output of cloud information for off-line coupling with atmospheric CTM;
- improved PBL properties, especially PBL height;
- modular structure allowing easy modification of needed parametrizations.

How would the ACTM utilize improved information if NWP model would produce it?

Boundary layer parameters of NWP and ACTM models

Should the ACTMs use the (boundary layer) parameters of NWP models as such or use own meteorological preprocessors to calculate them? Which parameters are suitable for common use? Is the consistency more important than differences resulting from different methods of calculation?

Boundary layer height:
- A parameter crucial for ACTMs (mixing height), also central in new turbulence parametrizations suggested for NWP (Zilitinkevich et al).
- Not a well defined parameter, different and sensitive to input data methods of calculation.
- Not observed directly (except new possibilities of satellite-based information, ceilometers, etc.).

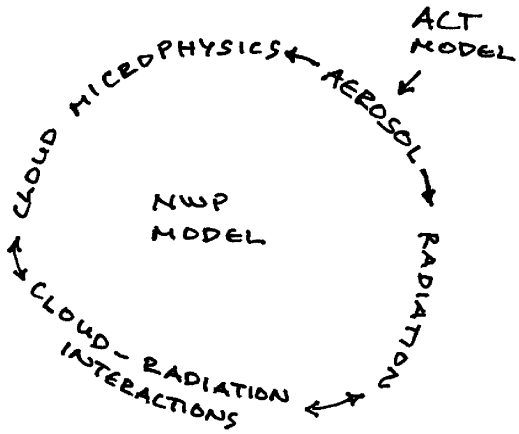
What species are expected to influence the weather?

- All greenhouse gases warm near-surface air.

- Aerosols: sea salt, dust, primary and secondary particles of anthropogenic and natural origin.
- Some aerosol particle components warm and others cool the air. Warm the air (by absorbing solar radiation and thermal-IR radiation): black carbon, iron, aluminium, polycyclic and nitrated aromatic compounds. Cool near-surface air (by backscattering incident solar radiation to space): water, sulphate, nitrate, most of organic compounds.
- Different mechanisms of aerosols and other chemical species effects on meteorological parameters (direct, indirect effects, etc.).

Sensitivity studies are needed to understand the relative importance of feedbacks. How to prepare a portfolio of feedbacks? First experience of ENVIRO-HIRLAM indicates some sensitivity to effective droplet size modification in radiation.

ECMWF experience: about five basic aerosol parameters, each with an integrated effective size have been included into the NWP model - sea salt, dust, primary and secondary aerosol species. Also data assimilation poses limit to the number of variables included.



Urbanization

In ENVIRO-HIRLAM:
- Surface improved description for urban areas: roughness, albedo, urban heat sources.
- Properties of urban aerosol used to modify the albedo characteristics and the effective radius of cloud droplets for the SW radiation (in the HIRLAM radiation scheme).

In FUMAPEX two other more sophisticated urban schemes: BEP (Martilli) and SM2-U modules were tested. They are more expensive computationally.

Town energy balance (TEB) module is a part of SURFEX, available in HARMONIE framework.

Handling of the finest-scale details of momentum fluxes in town (forest) canopy could be developed.

Inline chemistry in integrated models

Advantages
- Atmospheric model takes care of the transport (advection, vertical movement);
- Consistency of the numerical algorithms and parameter definitions follows automatically;

- Feedbacks may be included with maximum time resolution;
- Input-output and data storage can be kept in a minimum level.

Disadvantages
- Loss of flexibility in combining different atmosphere-surface-chemistry modelling systems and components;
- Possibly heavy and complicated all-purpose-systems, whose quick application to simple tasks is not straight-forward.

Within HARMONIE, SURFEX has recently *externalized* from the surface parametrizations in inline model environment, to allow greater flexibility.

Candidates for chemical module in HARMONIE

- No need to invent everything from the beginning - use experience of WRF and others.
- A WRF initiative, open to all developers: working group on physical and chemical parametrizations in integrated models (e-mail georg.a.grell@noaa.gov).
- ENVIRO-HIRLAM with the CAC chemical model could be a good base for this item.

Conclusions, next steps

- Preparation of discussion paper about integration of NWP and ACTM models for HIRLAM
- HARMONIE management, to form a starting point for further planning.
- Reporting ENVIRO-HIRLAM development progress, strategy and comparative analysis of on-line vs. off-line coupling and model runs with vs. without aerosol feedbacks.
- Report and further work within WG2: 'Integration' in COST 728 involving 5 HIRLAM-member institute representatives (DMI: A. Baklanov, FMI: M. Sofiev, Met.no: V. Ødegaard, SMHI: V. Foltescu, Tartu Univ.: A. Mannik).
- Organisation of an initial working group in HIRLAM-ACTM for further work and a sub-program (not completely inside the HIRLAM-A plan) for the ENVIRO-HIRLAM development cooperation.
- Meeting/training course about fine-scale integration aspects within NetFAM 2009.

4.2. Follow-up document: Air Chemistry Transport Modelling and Numerical Weather Prediction: *Sander Tijm[1] and Alexander Baklanov[2]*

[1] {KNMI, The Netherlands}
[2] {Danish Meteorological Institute (DMI), Lyngbyvej 100, 2100 Copenhagen, Denmark}
Correspondence to Sander Tijm (tijm@knmi.nl)

1. Introduction

The new strategy for development of a new generation integrated Meteorology (MetM) and Atmospheric Chemical Transport Model (ACTM) systems for predicting atmospheric composition, meteorology and climate change became more and more perspective in the future. This combination is reasonable due to the facts that: (i) meteorology is an important source of uncertainty in air pollution and emergency preparedness modelling, (ii) there are complex and combined effects of meteorological and pollution components on human health (e.g., hot spots in July of 2003 in Paris, France), (iii) pollutants have effects, especially

aerosols, on climate forcing and meteorological phenomena (precipitation, thunderstorms, etc.). So, this way of integrating modelling can be beneficial for model improvements in both communities: NWP and atmospheric environment forecasting.
To discuss this strategy on the European level the COST-728/NetFAM workshop on "Integrated systems of meso-meteorological and chemical transport models" was organized at the Danish Meteorological Institute (DMI) in Copenhagen on. 21-23 May 2007 (the workshop web-site is http://netfam.fmi.fi/Integ07). Within the workshop a special section 'HIRLAM/HARMONIE-ACTM integration' was arranged on Tuesday 22 May.
The main idea of the section is to get together for HIRLAM NWP and ACTM modelers, discuss and build our joint strategy for developing integrated system(s) based on HIRLAM.
The HIRLAM consortium and ACTM modelers in HIRLAM-organisations have some interest and initiatives in such integration, but they work separately and have very low level of coordination and cooperation.
There are also several attempts in this direction, including the following.
DMI is actively working with development of the on-line integrated system Enviro-HIRLAM, considering aerosol forcing mechanisms, etc.
Most of HIRLAM-member institutes are using national HIRLAM NWP outputs as meteo-drivers for their ACTP modelling and air quality forecasting activities. They already have attempts to build off-line integrations of HIRLAM with their own ACTMs (CAC, Chimere, DERMA, EMEP, MATCH, SILAM).
Such work was also included in the HIRLAM-A development plan (S4.10/4.5 Task: Coupling with atmospheric chemistry).
The 'Integration' WG2 in COST 728 involves 5 HIRLAM-member institute representatives (DMI, FMI, Met.no, SMHI, Estonian Tartu Univ.), and they are willing to consolidate and coordinate joint efforts in this work for coupling HIRLAM with ACTMs.
This results in possibilities for improving the use of HIRLAM in ACTM and ACTM results utilization in HIRLAM for NWP improvement.

2. On-line and Off-line Coupling

The integration/coupling of the HIRLAM NWP and ACTM models could be realized by different ways using the online and offline modelling approaches (in more details the definition and specifics of the approaches are discussed in AB presentation/abstract on the web-site). It could be realized for HIRLAM using the following variants.
One-way integration (off-line):

1. HIRLAM meteo-fields as a driver for atmospheric CTM (this way is used already by many air pollution modelers);
2. ACTM chemical composition fields as a driver for R/GCM (e.g. for aerosol forcing on meteo-processes, it could also be realized for NWP, e.g. HIRLAM).

Two-way integration:

1. Driver + partly feedbacks, for ACTP or for NWP (data exchange with a limited time period coupling: off-line or on-line access coupling, with or without second iteration with corrected fields);
2. Full feedbacks included on each time step (on-line coupling).

There is a clear difference in needs for the online coupling of chemistry transport models and the offline coupling. For the offline coupled models it would already be good to improve the meteorological quality, especially in fair weather conditions (claim: too little attention to these conditions, focus on extremes) and to include parameters that can be used directly in offline coupled models and that are important for processes like rainout. (It is important to

remember that the couplings can be done in 2 directions: not only from NWP to atmospheric CTM, but also from atmospheric CTM to NWP, see above).
For the online coupling it would be good to have a more modular setup of Hirlam to make it easier to plug in the chemistry modules. Also the convection and condensation schemes need to be adjusted to take the aerosol-microphysics interaction into account. Finally, also the radiation scheme needs to be adjusted to include the aerosol effect.
In more details the advantages and disadvantages of the online and offline modelling approaches are described in Alexander's presentation/abstract on the web-site.

3. Improvements in Meteorology and Output

Meteorological:

- PBL height (h), especially in very stable conditions (when h can be < than the lowest model level) and over inhomogeneous surfaces (like urban or forest areas), where internal boundary layers play important role and request using prognostic equations for PBL height.
- Inclusion of urban characteristics (only for high resolution, important for stable conditions and weak winds).
- Improve surface drag over forest and city. Recent studies show that it may be twice as strong as currently in the models. Diagnostic estimation of U10m and T2m over urban areas should be improved.
- Inclusion of aerosol and microphysics interaction.

Chemical species influencing the weather:

- All greenhouse gases warm near-surface air.
- Aerosols: sea salt, dust, primary and secondary particles of anthropogenic and natural origin.
- Some aerosol particle components warm and others cool the air. Warm the air (by absorbing solar radiation and thermal-IR radiation): black carbon, iron, and aluminum, polycyclic and nitrated aromatic compounds. Cool near-surface air (by backscattering incident solar radiation to space): water, sulphate, nitrate, most of organic compounds.
- Different mechanisms of aerosols and other chemical species effects on meteorological parameters (direct, indirect effects, etc.).
- Sensitivity studies are needed to understand the relative importance of feedbacks. First experience of ENVIRO-HIRLAM indicates some sensitivity to effective droplet size modification in radiation.

Output from HIRLAM to ACPM:

- Time averaged parameters (wind, temperature, cloud, precipitation, cloud top and bottom);
- Cloud top and bottom, Convective cloud top and bottom, Convective mass flux (if available);
- Stability parameters (MOL, W_*);
- PBL height as an output 2D field;
- Availability of the same physiography data base for surface, atmosphere and chemistry, depending on applications.

4. Importance of Data Assimilation and Problems with DA for Chemistry

Data assimilation (DA) is important for daily chemistry forecasts, but it also is a challenge. At the moment there are much fewer stations with chemistry observations than meteorological observations, and the observations of these stations may not always be available in real time. Also, the gradients in chemical species often are very sharp, which the current observation network and data assimilation schemes are not capable of representing. And last but not least, it is difficult to determine the assimilation increments with so many possible species and so few detailed observations. This is why ECMWF has limited the chemistry in their model and data assimilation to 5 species (in addition to the cost in computer power).
The adjoint modelling technique is a good instrument to improve on the climatological emissions that are currently used for many species.

5. Short-term Perspectives and Long-term Plans

1) In a very short term it may be possible to include the parameters that are requested and that may improve the offline coupling for ACTMs purposes. Here we are thinking about the average fields (accumulated) of wind components, temperature, specific humidity, cloud water and turbulent kinetic energy, and the parameters like 2D fields of cloud base and cloud top (to accurately determine where the scavenging is taking place). In addition some measures of stability (like the Monin-Obukhov Length, PBL height, etc.) could also be included. The things in this part may already be included in Hirlam version 7.2, which should be released in the autumn of 2007.
2) The vertical structure of PBL and SL is very important for ACTMs, so increasing the vertical resolution and improved parametrizations of BL are necessary. The meteorological improvements are already worked on for some aspects (stable PBL) and the urban characteristics should be included in the new surface scheme. When this scheme is ready, the probably not too large changes necessary for this will be included in the scheme. In the meso-scale model the urban parametrization already is available (Town Energy Budget in SURFEX).
3) At the moment the microphysics aerosol interaction is included in a very simple way in the convection and condensation schemes, where the cloud condensation nuclei have a lower concentration over the sea than over land. Here it would be very interesting and probably relatively easy to include the aerosols from a ACTM. The use of aerosol may be prepared by making a 3D field of aerosol that has the characteristics of the currently prescribed values, then the extension to a real 3D distribution of aerosols that can interact with the microphysics is relatively straightforward.
4) Off-line way of integration with reading ACTM output files by HIRLAM (see above the 'One-way off-line integration' #2) can also improve the NWP if aerosol feedback mechanisms are incorporated into HIRLAM. However, this way could be too expensive if high-resolution ACTM forecast is reading by HIRLAM on each time step. For example, some tests can be done using the CAC, DERMA, EMEP or MATCH models inputs for NWP runs with ENVIRO-HIRLAM feedback mechamisms. SMHI will test the OASIS4 coupler software for this work.
5) ENVIRO-HIRLAM gives a good perspective for the future and it is a good candidate for inclusion in the reference system, however the online coupling to the Hirlam model is more long-term work (not for Hirlam 7.2). It may be quite straightforward if e.g. the ENVIRO-HIRLAM parts can be included in the reference as an option. It can only be included as an option because it may be too expensive for operational use. It would be good to start with a separate HIRLAM-CHEM branch as soon as possible.

6) Organisation of an initial working group (from HIRLAM and ACTM representatives from different Hirlam-countries) for further HIRLAM-ACTM integration work and a sub-program (not completely inside the HIRLAM-A plan) for the ENVIRO-HIRLAM/HARMONIE (on-line and off-line versions) development cooperation.

Action 1 can be included in the work before the release of HIRLAM 7.2, action 2 is dependent on the progress of the surface scheme work, Action 3 may be planned for HIRLAM 7.3 and Action 5 is dependent on the scale of the work and the way ENVIRO-HIRLAM is set up.

6. Other Next Practical Steps

- Preparation of discussion paper about integration of NWP and ACTM models for HIRLAM
- HARMONIE management, to form a starting point for further planning.
- Reporting ENVIRO-HIRLAM development progress, strategy and comparative analysis of on-line vs. off-line coupling and model runs with vs. without aerosol feedbacks.
- Report and further work within WG2: 'Integration' in COST 728 involving 6 HIRLAM-member institute representatives (DMI: A. Baklanov, FMI: M. Sofiev, T. Vihma, KNMI: G. Geertsema, Met.no: V. Ødegaard, SMHI: V. Foltescu, Tartu University: A. Männik, etc.).
- Meeting/training course about fine-scale integration aspects within NetFAM 2009.

Chapter 5 Summary and Round Table Discussion

This chapter summaries the main discussion points arising from the topics of the workshop, namely:
1. Online and offline coupling of meteorological and air quality models
2. Implementation of feedback mechanisms, direct and indirect effects of aerosols
3. Advanced interfaces between NWP and ACTM models
4. Model validation studies, including air quality-related episode cases

It also draws together some conclusions that have strategic implications in this research area. In order to review progress in offline and online coupled models It is important to have exchange of experience from a wide range of groups involved in the development and application of these systems. The sections summarises inputs from a number of groups from USA, Canada, Japan, Australia and Europe. It is hoped that these discussions will lay te foundation to formulate a framework for an integrated mesoscale modelling strategy for Europe.

*The round table discussion inevitably examined the main r*equirements of such a strategy. The points discussed included:
- The large number of models and applications within Europe;
- The need for modelling communities to to have closer interaction (such as air quality, NWP, climate and model developers);
- The importanthat are the science questions to be addressed;
- The implications for users and for operational applications;
- Strategy should allow for these diversity and include off line and on line systems;
- The importance to learn from international group eg WRF community;
- The limitations of such a strategy.

5.1. On-line and Off-line Coupling of Meteorological and Air Quality Models: *Valentin Foltescu[1] and Jacek Kaminski[2]*

[1] {SMHI, Sweden}
[2] {SMHI, York University, Canada}

The aim of the workshop session on "coupling" was to discuss and, if possible, make recommendations on the best practice and strategy for further developments and applications of integrated modelling systems concerning Numerical Weather Prediction and/or Meso-meteorology (NWP/MM) and Atmospheric Chemical Transport Model (ACTM).

The following types of coupling can be considered:
- off-line
- on-line access (with availability of meteorological data at each time step)
- on-line integrated (with feedbacks possible to consider from ACTM to NWP/MM)

Feedback mechanisms – especially from aerosols – are considered to be important for a coupling strategy. Aerosol forcing mechanisms influence radiative and optical properties as well as cloud processes, leading for instance to changes of precipitation and circulation.

The strategic recommendations proposed during the "On line coupling and feedbacks" session of the workshop can be supplemented by:

- Need to identify key examples of significant feedbacks that were identified so far in sensitivity studies performed with on-line systems;
- Communicate and encourage exchange information within the NWP/MM modelling community on key feedback processes and their magnitudes.

Model interfaces include many aspects of the interoperability of NWP/MM and ACTM models. Modular coding is advocated in order to ease implementation of different algorithms/routines serving the same purpose. Standards for data exchange are also important.

Further recommendations to complement those proposed during the "model interface" session are listed below:

- It is important to make the modules available on request;
- Further study is required of the dislocation problem in coupling interfaces dealing with chemical-data assimilation;
- The computational efficiency when using different couplers needs to be investigated;
- Prepare a list of variables and parameters used in interfacing;
- Recommend when a choice should be made between time averaged values vs. instantaneous values of meteorological variables driving the ACTM routines;
- Continue to highlight opportunities to develop further coupling interfaces between NWP/MM and ACTM.

Other points which need more attention in respect to "coupling" NWP/MM and ACTM models involve:

- Inclusion of the sea breeze process which is crucial for chemical weather modelling in coastal regions. The meteorological driver needs to describe properly the process of sea breeze.
- Improved parametrization of urban effects on the atmospheric boundary layer (BL) is needed. NWP/MM models despite their increased resolution, still have shortcomings. For instance, the description of sub-surface, surface and urban BL for urban areas is similar to that of rural areas. Thus, the urban dynamics and energetics are not properly described. NWP/MM models are not primarily developed for air pollution modelling, and their outputs have to be made suitable input for urban-scale ACTMs.

5.2. Implementation of Feedback Mechanisms, Direct and Indirect Effects of Aerosols

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One of the important tasks is to develop a modelling instrument of coupled 'Atmospheric chemistry/Aerosol' and 'Atmospheric Dynamics/Climate' models for integrated studies, which

is able to consider the feedback mechanisms, e.g. aerosol forcing (direct and indirect) on the meteorological processes and climate change.

Chemical species influencing weather and atmospheric processes include greenhouse gases which warm near-surface air and aerosols such as sea salt, dust, primary and secondary particles of anthropogenic and natural origin. Some aerosol particle components (black carbon, iron, aluminium, polycyclic and nitrated aromatic compounds) warm the air by absorbing solar and thermal-IR radiation, whereas others (water, sulphate, nitrate, most of organic compounds) cool the air by backscattering incident short-wave radiation to space.

It is necessary to highlight, the effects of aerosols and other chemical species on meteorological parameters which have many different pathways (direct, indirect, semi-direct effects, etc.) and they have to be prioritised and considered in online coupled modelling systems. Sensitivity studies are needed to understand the relative importance of different feedback mechanisms.

A particular area of development for online coupled models is in the area of parametrizations that allow for interactions of physics with chemistry. With the realization of the increasing importance of science questions related to global and regional climate change, two-way interactions between meteorology and chemistry are becoming a necessity in complex 3-D models. Complex 3D models are increasingly being used not only for meteorological predictions, but to better understand and simulate the wide range of processes and atmospheric feedbacks that influence climate. In contrast to global climate models, the flexible grid structure of high resolution nonhydrostatic models enables the simulation of climate processes at spatial and temporal scales compatible with measurements, providing a framework in which to test new parametrizations of climate processes that are either treated in a simple way in current global climate models or neglected entirely.

The coordination (summary, overview, availability, development of new….) of parametrizations that allow for interactions between aerosols, radiation, chemistry, and clouds, as well as the coordination of new treatments for meteorological process modules (e.g. boundary layer, clouds) that are needed to improve predictions of atmospheric chemistry is a very useful subject. Model development in atmospheric chemistry and weather prediction has so far developed separately from each other, leading - for example - to meteorological parametrizations that have no treatment of any chemical species. A well known example linkage between chemistry and meteorology includes the treatment of cloud-aerosol interactions. Most current cloud physics schemes neglect the linkages of CCN to predicted aerosol distributions. In order to couple the chemistry part with cloud physics involves modifying existing chemistry and cloud subroutines that normally do not interact. This is a very complicated process that requires a lot of man power.

One example not related to the aerosol direct or indirect effects is the current treatments of boundary layer mixing which greatly affects near-surface concentrations. Quite often chemical models (if running online) rely on eddy coefficients from the meteorological model for the vertical mixing of trace-gas and particulate scalars. This only works since the mixing is determined by eddy coefficients that are calculated in the meteorological part and can be saved for use in the chemical part to mix the tracers. It can not work if the meteorological parametrization uses a different method to mix tracers. It would be much more desirable if designers of the meteorological parametrizations would consider the mixing of a scalar in

their scheme from the beginning. This would lead to more general and more accurate vertical mixing algorithms that handle both meteorological and chemical scalars. In addition it would provide meteorological modelers with an additional independent source for possible evaluation. Similar arguments can be made for the treatment of parameterized convection, where only very few convective parametrizations exist in meteorological models that allow for transport and modification of tracers.

Implementation of the feedbacks into integrated ACP-NWP models could be realized in different ways with varying complexity. The following variants serve as examples:

- Simplest offline coupling: The chemical composition fields from atmospheric CTMs may be red by MetM/NWP at a limited time period and used as driver for aerosol forcing on meteorological processes.
- Online access coupling: Driver and partly aerosol feedbacks, for ACTMs or for NWP (data exchange on each time step) with or without the following iterations with corrected fields.
- Fully online coupling/integration: ACTM and feedbacks included inside MetM on each time step.

The above examples represent the different levels of online integration that have been achieved within Europe. Many of the systems are currently either fully off-line or have some online capabilities. A few systems now exist that are fully online coupled (eg ENVIRO-HIRLAM and the UKCA systems). Historically Europe has not adopted a community approach to modelling and this has led to a largen number of model development programmes, usually working independently. However, a strategic framework will help to provide a common goal and direction to Europen research in this field.

There are a number of key elements need to be part of the overall strategy framework. These include:

(i) Scientific questions to be addressed by online systems

A major reason for developing online systems is to take account of feedback mechanisms for accurate modelling of NWP/MM-ACTM and quantifying direct and indirect effects of aerosols. Several questions can be identified in this regard:

• What are the effects of climate/meteorology on the abundance and properties (chemical, microphysical, and radiative) of aerosols on urban/regional scales?
• What are the effects of aerosols on urban/regional climate/meteorology and their relative importance (e.g., anthropogenic vs. natural)?
• How important the two-way/chain feedbacks among meteorology, climate, and air quality are in the estimated effects?
• What is the relative importance of aerosol direct and indirect effects in the estimates?
• What are the key uncertainties associated with model predictions of those effects?
• How can simulated feedbacks be verified with available datasets?

(ii) Processes/feedbacks to be considered

A detailed treatment of the main processes is required in the models in ordet to answer the above questions. These processes include:

• Direct effect - Decrease solar/thermal-IR radiation and visibility
– Processes needed: radiation (scattering, absorption, refraction, etc.)
– Key variables: refractive indices, ext. coeff., SSA, asymmetry factor, AOD, visual range
– Key species: cooling: water, sulfate, nitrate, most OC warming: BC, OC, Fe, Al, polycyclic/nitrated aromatic compounds
• Semi-direct effect - Affect PBL meteorology and photochemistry
– Processes needed: PBL/LS, photolysis, met-dependent processes
– Key variables: T, P, RH, Qv, WSP, WDR, Cld Frac, stability, PBL height, photolysis rates, emission rates of met-dependent primary species (dust, sea-salt, biogenic)
• First indirect effect – Affect cld drop size, number, reflectivity, and optical depth via CCN
– Processes needed: aero. activation/resuspension, cld. microphysics, hydrometeor dynamics
– Key variables: int./act. frac, CCN size/composition., cld drop size/number/LWC, COD, updraft velocity
• Second indirect effect - Affect cloud LWC, lifetime, and precipitation
– Processes needed: in-/below-cloud scavenging, droplet sedimentation
– Key variables: scavenging efficiency, precipitation rate, sedimentation rate
• All aerosol effects
– Processes needed: aerosol thermodynamics/dynamics, aqueous chemistry, precursor emissions, water uptake
– Key variables: aerosol mass, number, size, composition, hygroscopicity, mixing state

3. Implementation Strategies and Milestones
The roundtable discussion focused on workable objectives that span short and long term time frames. Some of the elements that will need to be considered include
• Strategic
– Support both offline and online coupled model frameworks
– Identification of the capabilities of online models for air quality applications

- Strategy should be implemented in a phased manner to allow groups to opt in depending on their requirements, capabilities and needs

– Development of benchmarks or guidelines should be considered reflecting the main uses and applications for Europe
– In light of these developments computational resources available in Europe for intense computational studies should be re-assessed
– Where possible common model evaluation including design of process based field and laboratory studies (e.g., closure experiments) should be suggested
– An important aspect would be to initiate and encourage the training of graduate and post-doctoral researchers via formal courses/summer schools

5. Coordination Plan and Logistitics
• COST728 has been instrumental in laying the first foundations of such as strategy. However, for the strategy to have wide acceptance, close cooperation and interaction will be required with the main groups in Europe. The following international coordination activities will need to be established:
– Progress review meetings annually
– Establish working subgroup focusing on specific areas

– Annual workshop alternating in Europe and North America

– Working group meeting in conjunction with annual workshop
– Special journal issues dedicated to workshop papers
- Plan for securing resources and funding to implement the strategy that is ambitous, forward-looking and sustainable.

5.3. Advanced Interfaces between NWP and ACTM Models

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There are a number of points that need to be considered when developing new integrated model systems. The basic recognition is for the importance of dealing with the requirements of the chemical model at the same time as with the meteorological ones. In the past, this was normally neglected and the system designed on meteorological grounds while only adding in the chemistry later. With modern supercomputers, on-line coupled systems are decidedly preferable, guaranteeing high temporal resolution of all coupled processes and feedbacks. There are several advantages of including many models as modules into 1 system framework for maximum information, flexibility and applicability. At the same time, computer resources should allow modelers to be free of needs for parallelization or other cumbersome technical requirements.

Interantionally there several online systems such as that employed in at CSIRO for Australian applications. It works well on the smaller scales (1-1000km), e.g. for the import/export of bush or forest fire smoke, while the system is used at the UK Hadley Centre for the larger, global scale.
In Japan an adapted version of WRF/Chem model is being used. Off-line models used in Japan are mainly US models for chemistry while the meteorological models are provided by the Japanese meteorological agency.

Within Europe examples include of the online coupling of the COSMO-EU (formerly Lokalmodell) to chemistry model. The German Weather Service DWD and the MPI for Meteorology in Hamburg are developing the ICON model as a combined NWP and climate model on a scale-adaptive icosahedral grid including chemistry to become the operational NWP model at the DWD in 2012. **T**here was a prime interest in precipitation forecasting at ECMWF, and the current introduction of chemistry into the models (in GEMS) in order to build an integrated forecast system. The HIRLAM model now has on-line chemistry features at DMI.

As outlined before a single system approach will not be a viable option for Europe at this stage but an open system platform for partners to add modules with harmonized interfaces and parameters could be considered, but will still require improved communication and cooperation between the European partners.

Taking into account the large number of small institutes and administrations across Europe engaged inair quality modelling the need for practical, easy-to-handle solutions on small computers with few staff and restricted modelling experience was emphasized by several partners. These systems are of necessity off-line often using measurement NWP forecasts of

ECMWF or national met offices. Therefore, these systems often strongly depend on interfaces connecting the chemistry modules to measurement postprocessing and/or meteorological modules.

The need for unification of interface modules at least concerning high modularity and I/O format specifications, but possibly also parametrizations, and for according guidelines was also apparant. ECMWF practice was proposed as a European guideline by Johannes Flemming (ECMWF) who also saw the necessity to test OASIS standards for more of these systems.

5.4. Model Validation Studies, including Air Quality Related Episode Cases

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A problem of coupling the NWP and ACTM models does not exactly follow the line between on- and off-line methods of coupling. Numerous compromises accepted in both models, different histories and tasks have created several niches for various combinations of these systems and for various interfaces between them.

For example, primarily offline interface between the meteorological and dispersion models is used for real-time emergency applications. Huge uncertainties in information about the source term in any real emergency situation and strong time limitations for analysis make cheaper and faster off-line models the only acceptable choice.

Another extreme is climate-related studies where missing the interaction between atmospheric composition and dynamics is hardly acceptable.

Most other applications are somewhere in-between the above extremes and a particular choice of modelling tools and their interactions varies from case to case.
It is worth mentioning that the reasons for moving towards more complicated but also more comprehensive online coupled systems can also be different, as well as the actual means of the online coupling. For instance, the access to NWP fields at each time step is clearly an advantage providing that the dynamic core is unique for both meteorological and chemical parts. Otherwise, differences in features of the advection and diffusion schemes of the coupled models will create tough problems instead of solving them. The meteorological fields computed within the NWP sub-model with own advection implementation will be inconsistent from the point of view of chemical transport sub-model. Most modern ACTMs have special meteorological pre-processors that deal with this inconsistency, which is then treated as a feature of the NWP fields. To overcome this very serious obstacle, the NWP and ACTM models have to actually unified, so that a single dynamic core handles both chemical and meteorological transport. A pre-requisite for this is a decent dynamics, which satisfies both sides – a luxury rarely available in existing modelling systems, especially if they were created separately.
The other strong motivation to move towards online coupling is a possibility to arrange feedbacks from chemical side to meteorological one. Being indeed simpler in such a model configuration, the feedbacks themselves can be comparatively easily arranged also with off-line systems with very limited overhead. Admittedly, the off-line feedbacks can hardly reach

the frequency of every model time step – but in most cases this is not needed either. The feedback impact even in strongly polluted areas is still a small perturbation for the main processes. Nearly the only exception is the dust storm events where the aerosol concentration is so high that it cannot be considered as a tracer any more. A simplification, however, is that it is enough to add just a couple of aerosol modes into the NWP variables still keeping the bulky chemistry as an external model.

Concluding, it looks like the advantages and disadvantages of the online and offline coupling of the meteorological and chemical transport models are relative and depend on the specific task. Diversity and specifics of the tasks, in their turn, will dictate the existence of both approaches for the foreseeable future.

Finally, it is important to highlight that this initiative to develop a European strategy for online coupled mesoscale models for air quality applications should fully benefit from all relevant groups and projects including COST 728 and new Actions such as COST 602 (Towards a European Network on Chemical Weather Forecasting and Information Systems) as well as users and policy makers.