7.1 **PHYSICAL AND CHEMICAL WEATHER AS A JOINT PROBLEM: TWO-WAY INTERACTING INTEGRATED MODELLING** *(Invited Speaker)*

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1. INTRODUCTION INTO THE PROBLEM

During the last decade a new field of atmospheric modelling - the chemical weather forecasting (CWF) - is quickly developing and growing. However, in most of the current studies this field is considered in a simplified concept of the off-line running of chemical transport models with operational numerical weather prediction (NWP) data as a driver (Lawrence at al., 2005). A new concept and methodology considering the "chemical weather" as two-way interacting nonlinear meteorological and chemical/aerosol dynamics processes of the atmosphere have been suggested (Grell et al., 2005; Baklanov and Korsholm, 2007; Baklanov, 2010; Grell and Baklanov, 2011). The on-line integration of meso-meteorological models (MetM) and atmospheric aerosol and atmospheric chemical transport (ACT) models gives a possibility to utilize all meteorological 3D fields in the ACT model at each time step and to consider nonlinear feedbacks of air pollution (e.g. atmospheric aerosols) on meteorological processes / climate forcing and then on the chemical composition of the atmosphere. This very promising way for future atmospheric modelling systems (as a part of and a step toward the Earth System Modelling, ESM) will lead to a new generation of models for meteorological, environmental, chemical and bio-chemical weather forecasting. The methodology on how to realize the suggested integrated concept is demonstrated on an example of the European Enviro-HIRLAM (Environment – HIgh Resolution Limited Area Model) integrated modeling system (Baklanov et al., 2008a; Korsholm, 2009). European experience in the on-line integrated meteorology-chemistry modelling, importance of different chains of feedback mechanisms for meteorological and atmospheric chemistry processes and their strong nonlinearities are also discussed.

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2. OVERVIEW OF EUROPEAN EXPERIENCE

2.1 *Overview of European on-line CWF models*

Although most of the European operational CWF systems are currently fully off-line (Kukkonen et al., 2011), several modelling systems have also some on-line capabilities. Within the 18 CWF modelling systems considered there, only two (Enviro-HIRLAM and WRF-Chem) are realised as on-line integrated models with two-way interactions. The COST-728 model overview (COST-WMO, 2007) shows a surprisingly large number (at least 10) of on-line coupled MetM and ACT model systems already being used in Europe. This list of on-line models in Europe is continuously growing. Table 1 summarizes current list and characteristics of the on-line coupled or on-line access MetM and ACT models developed or applied in Europe. In 2007 there were only two models considered indirect aerosol feedbacks, nowadays, there are several models considering direct and indirect aerosol feedbacks.

However, these developments are realized on different platforms separately by local/national research groups in different countries, and there is no any coordination of these efforts within Europe (compared with USA, for example). Historically, Europe has not adopted a community approach to modelling, and this has led to a large number of model development programmers, usually carried out independently. However, a strategic framework would definitely help to provide a common goal and direction to European research in this field while having various multiple models.

2.2 *Main chemistry feedbacks on meteorology*

Chemical species influencing weather and atmospheric processes include greenhouse gases (GHGs) which warm near-surface air and aerosols such as sea salt, dust, primary and secondary particles of anthropogenic and natural origin. Moreover, some aerosol particle components (black carbon, iron, aluminium, polycyclic and

Table 1. On-line coupled or on-line access Atmospheric Chemistry-Meteorology models developed or applied in Europe (EuMetChem, 2010)**.**

Model/Country/References	**On-line coupled gas phase chemistry and aerosol**	**Feedback of pollutants to meteorology**	**Applications**	**Scale**
BOLCHEM, Italy http://bolchem.isac.cnr.it/	SAPRC90 gas phase chemistry, AERO3 aerosol module	Direct aerosol effect on radiation	CWF; climate; Episodes	Conti-nental to regional
COSMO-ART, Germany Vogel et al., 2009	Extended RADM gas phase chemistry, modal aerosol, soot, pollen, mineral dust	Direct aerosol effect on radiation	Episodes	Conti-nental to regional
COSMO-LM-MUSCAT, Germany Wolke et al., 2004; Heinold et al., 2007	RACM gas phase chemistry, 2 modal aerosol models, mineral dust module	Direct aerosol effect on radiation for mineral dust	Episodes	Conti-nental to regional
ECHAM5/6-HAMMOZ, Germany Pozzoli et al., 2008	MOZART gas phase chemistry, HAM aerosol scheme	Direct aerosol effect, indirect aerosol effect	Episodes, long term	Global
Enviro-HIRLAM, Denmark and HIRLAM countries Baklanov et al, 2008a; Korsholm et al., 2008, Korsholm, 2009	NWP gas phase chemistry, modal and sectional aerosol modules, liquid phase chemistry	Direct and indirect aerosol effects	Episodes, chemical weather forecast	Hemis-pheric to regional and urban
IFS-MOZART (MACC/ ECMWF) Flemming et al., 2009, Kinnison et al., 2007, http://www.gmes-atmosphere.eu	MOZART gas phase chemistry with updates to JPL-06, MACC aerosol scheme	Direct aerosol effect, indirect aerosol effect	Forecasts, Reanalysis, Episodes	Global
MCCM, Germany Grell et al., 2000; Forkel & Knoche, 2006	RADM, RACM or RACM-MIM with modal aerosol module	Direct aerosol effect	Episodes, climate-chemistry	Regional
MC2-AQ, Canada (use in Polen) Kaminski et al., 2007	ADOM gas phase chemistry	none, but possible	Episodes	Regional to urban
Meso-NH, France http://mesonh.aero.obs-mip.fr/mesonh/	RACM or ReLACS gas phase chemistry, modal aerosol module	Direct aerosol effect	Episodes	Continent al to regional
MESSy(-ECHAM5) , Germany Jöckel et al., 2005; http://www.messy-interface.org/	Various gas phase chemistry modules, modal aerosol module	Direct aerosol effect, indirect aerosol effect	Episodes, long term	Global
MetUM (Met Office Unified Model), UK Morgernstern et al, 2009; O' Connor et al 2010	2 tropospheric chemistry schemes, 1 stratospheric chemistry scheme. 2 alternative aerosol schemes.	Direct & indirect aerosol effects, radiative impacts of N_2O, O3, CH_4	Episodes, CWF, climate-chem. studies, poll. transport	Regional to Global
M-SYS (on-line version), Germany von Salzen & Schlünzen, 1999	RADM Gas phase chemistry, sectional aerosol module	none, but possible	Episodes	Regional to local
RegCM-Chem, Italy Zakey et al., 2006, Solmon et al., 2006	Updated GEOS-CHEM RACM, CBMZ, uni-modal aerosol, sectional mineral dust, sulfur chemistry	Direct aerosol effect	Climate-chemistry	Continent al to regional
RAMS/ICLAMS, USA/Greece http://forecast.uoa.gr/ICLAMS/index.php, Kallos et al. 2009, Solomos et al. 2011	On-line photolysis rates. Coupled SAPRC99 gas phase, modal aerosol, ISORROPIA equilibrium and SOA, cloud chemistry.	Direct and indirect aerosol effect	Episodes, CWF, meteo-chemistry interactions	Continent al to urban
WRF/Chem, US (used in UK, Spain, etc.) Grell et al., 2005; Fast et al., 2006, refs see in Zhang, 2008	RADM, RACM, RACM-MIM with modal aerosol module or CBM-Z with sectional aerosol module, liquid phase chemistry	Direct aerosol effect, indirect aerosol effect	Episodes, CWF, climate-chemistry	Continent al to regional
WRF-CMAQ Coupled System, USA (used in UK) Pleim et al., 2008; Mathur et al., 2010	Gas-phase mechanisms: CB05, SAPRC-99; Modal aerosols based on the AERO5 CMAQ module	Direct aerosol effects on radiation and photolysis	Episodes to annual	Urban to Hemisphe ric

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 back-scattering incident short-wave radiation to space. Therefore, it is necessary to highlight that those effects of aerosols and other chemical species on meteorological parameters have many different pathways, e.g.:

- Direct effect decreases solar/ thermal-IR radiation and visibility;
  - warming: GHGs, BC, OC, Fe, Al, polycyclic/ nitrated aromatic compounds
  - cooling: water, sulfate, nitrate, most OC (scattering, absorption, refraction, etc.)
- Semi-direct effects affect atmospheric boundary layer (ABL) meteorology and photochemistry;
- First indirect effect influences cloud drop size, number, reflectivity, and optical depth via CCN;
- Second indirect effect influences cloud liquid water content, lifetime, and precipitation;
- Chain of all aerosol effects (nonlinear interactions).

The above mentioned effects have to be prioritized and considered in on-line coupled models for different space and time scales. High-resolution on-line integrated models with a detailed description of the ABL structure are necessary to simulate such chains of two-way feedback mechanisms.

2.3 *EU COST Action ES1004 EuMetChem*

New EU COST Action ES1004 EuMetChem: 'European framework for on-line integrated air quality and meteorology modelling' (2011-2015) was recently accepted (web-site: http://w3.cost.eu/index.php?id=206&action_number=ES1004). It will focus on a new generation of on-line integrated ACT and Meteorology (Numerical Weather Prediction and Climate, NWP-CLIM) modelling with two-way interactions between different atmospheric processes including chemistry (both gases and aerosols), clouds, radiation, boundary layer, emissions, meteorology and climate. The overall objective of the Action is to set up a multi-disciplinary forum for on-line integrated air quality/meteorology modelling and elaboration of the European strategy for a new-generation integrated ACT/NWP-CLIM modelling capability/ framework. The main topics are:

1. On-line versus off-line modelling: advantages and disadvantages,
2. Analysis of priorities focusing on interaction/ feedback mechanisms,
3. Chemical data assimilation in integrated models,
4. European strategy/ framework/ centre for on-line integrated modelling,
5. Evaluation and validation framework of on-line ACT/NWP-CLIM models,
6. Collection of suitable datasets for model development, testing and evaluation.

At least, two application areas of the integrated modelling are aimed to be considered:
(i) improved NWP and CWF with short-term feedbacks of aerosols and chemistry on meteorological variables, and
(ii) two-way interactions between atmospheric pollution/ composition and climate variability/ change.

The framework will consist of 4 Working Groups namely: 1) Strategy and framework for on-line integrated modelling; 2) Interactions, parameterisations and feedback mechanisms; 3) Chemical data assimilation in integrated models; and finally 4) Evaluation, validation, and applications.

Establishment of such a European framework (involving also key American experts) will enable to develop world class capabilities in integrated ACT/NWP-CLIM modelling systems, including research, forecasting and education.

More than 20 teams from 14 European countries, ECMWF, WMO, US EPA, NOAA, etc. are already involved into the Action.

The COST Action initiated also a new session AS4.25: 'Integrated physical and chemical weather modelling with two-way interactions' at the EGU Vienna, Austria, 3-8 April 2011 (see: http://meetingorganizer.copernicus.org/EGU2011/session/7498).

3. ENVIRO-HIRLAM ON-LINE INTEGRATED ACT-NWP MODELLING SYSTEM

3.1 *Enviro-HIRLAM on-line integrated model with two-way interactions*

The Enviro-HIRLAM is an on-line coupled numerical weather prediction and atmospheric chemical transport modelling system for research and forecasting of joint meteorological and chemical weather (Figure 1). Originally this integrated modelling system was developed by DMI (Chenevez et al., 2004; Baklanov et al., 2004, 2008a; Korsholm et al., 2008, Korsholm, 2009) and further with other collaborators, and now it is included by the European HIRLAM consortium as a baseline system in the HIRLAM Chemical

Branch (https://hirlam.org/trac/wiki). The model development was initiated at DMI more than 10 years ago. It was the first meso-scale on-line coupled model in Europe that considers two-way feedbacks between meteorology and chemistry/aerosols.

The first version was based on the DMI-HIRLAM NWP model with fully on-line integrated pollutant transport, dispersion and deposition (Chenevez et al., 2004), chemical and aerosol (only for sulfur particles) dynamics models (Gross and Baklanov, 2004) and indirect effects of aerosols (Korsholm et al., 2008; Korsholm, 2009). For urban areas, where most of population is concentrated, the meteorological part was improved by implementation of urban sublayer modules and parameterisations (Baklanov et al., 2008b; Mahura et al., 2007; 2009).

The current version of Enviro-HIRLAM is based on the reference HIRLAM version 7.2 with new developed more effective chemical lumped scheme, multi-compound modal approach aerosol dynamics modules, aerosol feedbacks on radiation (direct and semi-direct effects) and on cloud microphysics (first and second indirect effects).

The *GasChem* module consists of:

- The condensed CBM gas-phase mechanism based on CBMZ (Zaveri et al., 1999*),* which is *s*implified lumped structure photochemical mechanism and most fast chemical solver; the radical balance solution technique (Sillman, 1991); the chemical module has 120 reactions and 23 advected species.
- Photolysis rates are setup as a function of altitude, solar zenith angle, cloud optical depth; J-values were calculated based on Madronich algorithm

The *AeroChem* module consists of:

- Thermodynamic equilibrium module HETV (Makar et al., 2003),
- Simple aqueous-phase module,
- Aerosol dynamics module M7 (Vignati et al., 2004).

These modules in the latest version of the model are currently under the testing and validation stage.

3.2 *Aerosol feedback parameterisations in Enviro-HIRLAM*

Enviro-HIRLAM contains parameterisations of the direct, semi-direct, first and second indirect effects of aerosols. Direct and semi-direct effects are realised by modification of Savijarvi radiation scheme (Savijärvi, 1990) with implementation of a new fast analytical SW and LW (2-stream approximation) transmittances, reflectances and absorptances (Nielsen et al., 2011). Simplified analytical parametrization for inclusion of direct aerosol effect on short-wave radiation was developed based on Koepke et al. (1997) using the DISORT model and considering the full spectral radiance field.

The species include BC (soot), minerals (nucleus, accumulation, coarse and transported modes), sulphuric acid, sea salt (accumulation and coarse modes), "water soluble", and "water insoluble".

Condensation, evaporation and autoconversion in warm clouds are considered to be fast relative to the model time step and are not treated prognostically.

The bulk convection and cloud microphysics scheme STRACO (Sass, 2002) and the autoconversion scheme by Rasch and Kristjansson (1998) form the basis of the

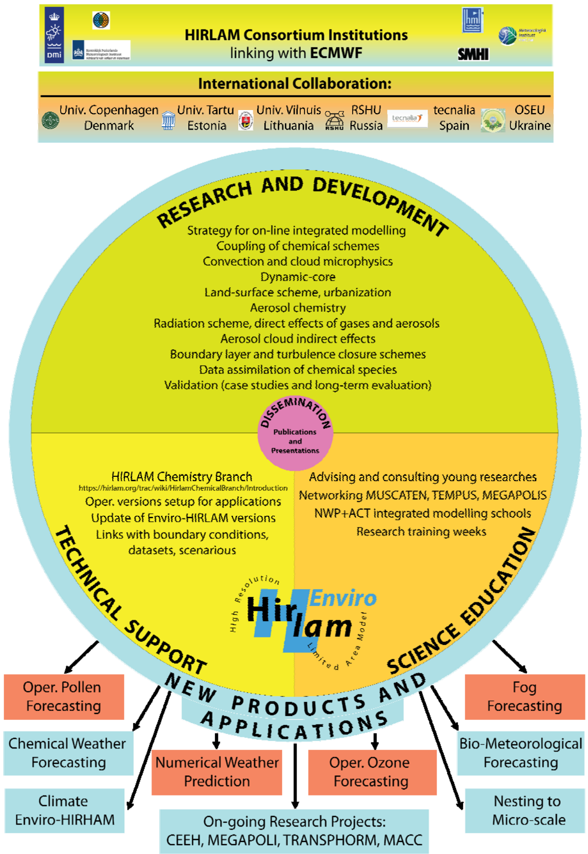

Figure 1. General scheme of international collaboration, research and development, technical support and science education for the on-line integrated Enviro-HIRLAM: 'Environment – HIgh Resolution Limited Area Model'.

parameterisation of the second aerosol indirect effect.
As aerosols are convected they may activate and contribute to the cloud droplet number concentration, thereby, decreasing the cloud droplet effective radius affecting autoconversion of warm cloud droplets into rain drops.
Cloud radiation interactions are based on the cloud droplet effective radius (Wyser et al., 1998).
As it decreases warm cloud droplet size and reflects more incoming short wave radiation, thereby, we parameterise the first aerosol indirect effect.
A clean background cloud droplet number concentration is assumed and the anthropogenic contribution is calculated via the aerosol scheme.

3.3 *Model validation and application*

Possible applications of the Enviro-HIRLAM for meteorological, environmental and climate forecasting and assessment studies are highlighted in Figure 1.
Validation and sensitivity tests (on examples of case studies and short-time episodes) of the on-line vs. off-line integrated versions of Enviro-HIRLAM (Korsholm et al., 2008) showed that the on-line coupling improved the results. Different parts of the model were evaluated vs. the ETEX-1 experiment, Chernobyl accident and Paris campaigns (summer 2009) datasets and showed that the model had performed reasonably (Korsholm, 2009; Korsholm et al., 2009; MEGAPOLI, 20010).

On-line vs. off-line coupled simulations for the ETEX-1 release (Korsholm et al., 2009) showed that the off-line coupling interval increase leads to considerable error and a false peak (not found in the observations), which almost disappears in the on-line version that resolves meso-scale influences during atmospheric transport and plume development.
The effects of urban aerosols on the urban boundary layer height, can be comparable with the effects of the urban heat island (Δh is up to 100–200m for stable boundary layer) (Baklanov et al., 2008a). Current studies (Korsholm et al., in MEGAPOLI, 2010) of megacities effects on the meteorology/climate and atmospheric composition showed that aerosol feedbacks through the first and second indirect effect induce considerable changes in meteorological fields and large changes in chemical composition, in particular NO_2, in a case of convective clouds and little precipitation. The monthly averaged changes in surface temperature due to aerosol indirect effects of primary aerosol emissions in Western Europe were analyzed and validated vs. measurement data. It was found that a monthly averaged signal (difference between runs with and without the indirect effects) in surface temperature is about 0.5°C (Figure 2).

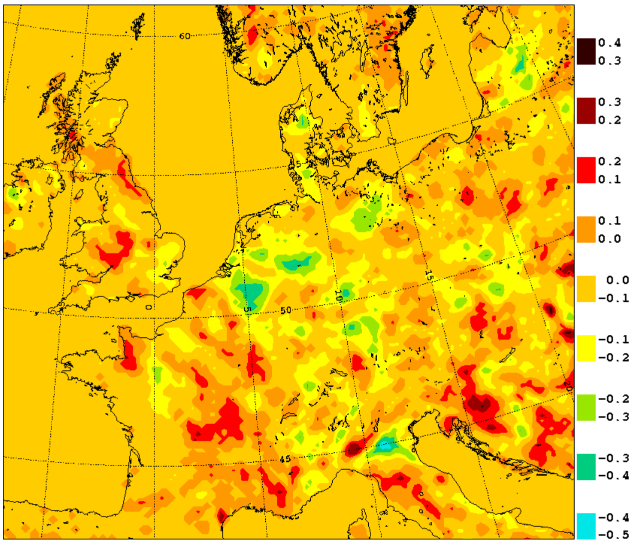

Figure 2. Averaged monthly (June 2009) difference in surface temperature T_s (°C) for the Enviro-HIRLAM runs with and without aerosol indirect effects (MEGAPOLI, 2010).

4. SUMMARY

- Suggested concept - chemical weather as two-way interacted meteorological weather and chemical composition of the atmosphere - is realised in the new COST Action ES1004 EuMetChem.

- On-line integration of MetMs and ACT models enables:
  - *utilisation of all meteorological 3D fields in ACT models at each time step;*
  - *consideration of feedbacks of air pollution on meteo-processes and climate forcing.*

- New generation of integrated models - not only for CWF, but also for climate change modelling, NWP (e.g., in urban areas, severe events, etc.), air quality and mitigations, long-term assessment of chemical composition, etc.

- Main advantages of on-line coupling:
  - *only one grid for MetM and ACT models, no interpolation in space and time;*

- ➢ *physical parameterizations are the same, no inconsistencies;*
- ➢ *all 3D meteorological variables are available at each time step;*
- ➢ *no restriction in variability of meteorological fields;*
- ➢ *possibility to consider two-way feedback mechanisms;*
- ➢ *does not need meteo- pre/ post-processors.*

- Feedback mechanisms can be important (supported by simulation results) in CWF modelling and quantifying direct and indirect effects of aerosols (and probably GHGs).
- Indirect aerosol feedbacks (based on the Paris case study): sensitivity of meteorology and chemistry, strong non-linearity, e.g. indirect effects induce large changes in NO_2.

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