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Enviro-HIRLAM online integrated meteorology-chemistry modelling system: strategy, methodology, developments and applications

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Abstract: The Environment - High Resolution Limited Area Model (Enviro-HIRLAM) is developed as a fully online integrated numerical weather prediction (NWP) and atmospheric chemical transport (ACT) model for research and forecasting of joint meteorological, chemical and biological weather. The integrated modelling system is developed by DMI in collaboration with several European universities. It is the baseline system in the HIRLAM Chemical Branch and used in several countries and different applications. The development was initiated at DMI more than 15 years ago. The first version was based on the DMI-HIRLAM NWP model with online integrated passive pollutant transport and dispersion, chemistry, aerosol dynamics, deposition and indirect effects. To make the model suitable for chemical weather forecasting (CWF) in urban areas the meteorological part was improved by implementation of urban parameterizations. The dynamical core was improved by implementing a locally mass conserving semi-Lagrangian numerical advection scheme, which improves forecast accuracy and model performance. The latest developing version is based on HIRLAM reference v7.2 with a more advanced and effective chemistry, aerosol multi-compound approach, aerosol feedbacks (direct and semi-direct) on radiation and (first and second indirect effects) on cloud microphysics. Since 2004 the Enviro-HIRLAM is used for different studies, including operational pollen forecasting for Denmark since 2009. Following main research and development strategy the further model developments will be extended towards the new NWP platform - HARMONIE. Different aspects of online coupling methodology, research strategy and possible applications of the modelling system, and 'fit-for-purpose' model configurations for the meteorological and air quality communities are discussed.

1. Methodology of the online coupled /seamless meteorology-chemistry modelling and aims of Enviro-HIRLAM model development

During the last decades 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 stil considered in a simplified concept of the off-line running of atmospheric chemical transport (ACT) 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 recently suggested (Grell et al., 2005; Baklanov and Korsholm, 2008; Baklanov, 2010; Grell and Baklanov, 2011). The current experience in the online integrated meteorology-chemistry modelling, importance of different chains of feedback mechanisms for meteorological and atmospheric composition processes are discussed for USA (Zhang, 2008) and European (Baklanov et al., 2014) models. The on-line integration of meso-meteorological models (MetM) and atmospheric aerosols and 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 seamless coupled models for meteorological, chemical and biochemical weather forecasting. Seamless approach for 'one atmosphere' integrated meteorology-chemistry/aerosols forecasting systems is analysed by the COST Action ES1004 EuMetChem (see e.g. Baklanov et al., 2015) and overview of the current state of online coupled chemistry-meteorology models and needs for further developments were published in (Zhang, 2008; WMO, 2016; Baklanov et al., 2017).

123456789012345678901234567890123456789012345678901234567890123456789012345678901234567890123456789012345 The methodology on how to realize the suggested integrated concept was demonstrated on an European example of the Enviro-HIRLAM (Environment - High Resolution Limited Area Model) integrated modeling system (Baklanov et al., 2008a; Korsholm, 2009). Experience from first HIRLAM community attempts to include pollutants into the NWP model (Ekman, 2000) and from pioneering online coupled meteorology-pollution model developments of the Novosibirsk science school (Marchuk, 1986; Penenko and Aloyan, 1985; Baklanov, 1988) was actively used for developments of the Enviro-HIRLAM modelling system.

The Enviro-HIRLAM is developed as a fully online integrated NWP and ACT modelling system for research and forecasting of meteorological, chemical and biological weather. The integrated modelling system is developed by DMI and other collaborators (Chenevez et al., 2004; Baklanov et al., 2008a, 2011b; Korsholm et al., 2008, 2009; Korsholm, 2009) and included as the baseline system of the Chemical Branch of the HIRLAM consortium (Figure 1).







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'.

The model development was initiated at DMI more than 15 years ago and it is used now in several countries. The modelling system is being used for different research projects (FP6 FUMAPEX; FP7 MEGAPOLI, PEGASOS, MACC, TRANSPHORM, MarcoPolo; NordForsk NetFAM, MUSCATEN, CarboNord; COST Actions – 728, 732, ES0602 ENCWF, ES1004 EuMetChem), and for operational pollen forecasting in Denmark since 2009 (Rasmussen et al., 2006; Mahura et al., 2006b) and planned for atmospheric composition (with focus on aerosols) for China since 2016 (Mahura et al., 2016). Following main strategic plans (Baklanov, 2008; Baklanov et al., 2011a) within HIRLAM-B,-C projects further developments of the modelling system will be shifting to new NWP platform (from HIRLAM to HARMONIE) and a close collaboration with the ALADIN (Aire Limitée Adaptation dynamique Développement InterNational) community was initiated in 2014.

In this paper an overall description of the Enviro-HIRLAM coupled modelling system and examples in different application areas are considered.

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2. Enviro-HIRLAM modelling system description

2.1. Modelling system structure

The Enviro-HIRLAM is a fully on-line coupled (integrated) NWP and ACT modelling system for research and forecasting of meteorological, chemical and biological weather (see schematics in Figure 2). The modelling system was originally developed by DMI 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). It was the first meso-scale on-line coupled model in Europe that considered two-way indirect feedbacks between meteorology and chemistry/aerosols (WMO-COST; 2008)

The following main steps of the model development were realised such as: (i) model nesting for high resolutions, (ii) improved resolving PBL and surface layer structure, (iii) urbanisation of the NWP model, (iv) improvement of advection schemes, (v) emission inventories and models, (vi) implementation of gas-phase chemistry mechanisms, (vii) implementation of aerosol dynamics. (viii) realisation of aerosol feedback mechanisms.

The first version was based on the DMI-HIRLAM NWP model with online integrated pollutant transport and dispersion 101 102 (Chenevez et al., 2004), chemistry, deposition and indirect effects (Korsholm et al., 2008; Korsholm, 2009) and later aerosol (only for sulphur particles) dynamics (Baklanov, 2003; Gross and Baklanov, 2004). To make the model suitable for chemical 103 104 weather forecasting in urban areas the meteorological part was improved by implementation of urban sub-layer parametrisations (Baklanov et al., 2008b; Mahura et al., 2008a; González-Aparicio et al., 2013). The model's dynamic core 105 106 107 was improved by adding a locally mass conserving semi-Lagrangian numerical advection scheme (Kaas, 2008; Sørensen, 2012; Sørensen et al., 2013), which improves forecast accuracy and enables performing longer runs. More details of the system development history is presented in the Annex 1.

The current new version of Enviro-HIRLAM (Nuterman et al., 2013; Nuterman et al., 2015) is based on the reference 109 HIRLAM v7.2 with a more advanced and effective chemistry scheme, multi-compound modal approach aerosol dynamics 110 modules, aerosol feedbacks on radiation (direct and semi-direct effects) and on cloud microphysics (first and second indirect effects). This version is continuously under development and evaluation for various weather and air quality related 112 applications. 113





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Figure 2: Schematics of the Enviro-HIRLAM modelling system.

2.2. Meteorological core of the system

The first version of Enviro-HIRLAM was based on a previous version HIRLAM-tracer and at its meteorological core lies DMI-HIRLAM, version 6.3.7 employed for limited area short range operational weather forecasting at DMI (Chenevez et al., 2004). The current model version used in studies is based on the reference version of the HIRLAM community meteorological NWP model HIRLAM version 7.2 and online-coupled environmental block (so-called, the Enviro-) allowing

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125to take into account spatial-temporal evolution of atmospheric chemical and biological aerosols driven by meteorology from 126 127 128 NWP block.

HIRLAM is a hydrostatic NWP model which is used for both research and operational purposes. The model provides forecast of the main meteorological fields: air temperature and specific humidity, atmospheric pressure, wind speed and direction, 129 130 cloud cover and turbulent kinetic energy (TKE) based on forward in time integration of the primitive equations (dynamical core) (Holton, 2004) and physical processes such as radiation, vertical diffusion, convection, condensation, etc. (physical 131 132 core).

The detailed NWP HIRLAM description can be found in the HIRLAM reference guide science documentation (Unden et al., 133 2002) and its following upgrades and modifications (see more details at http://www.hirlam.org).

134 135 136 137 138 139 140 141 142 143 Following the main strategic development within HIRLAM (HIRLAM-B and -C projects), there are plans for further developments of Enviro-HIRLAM shifting to new non-hydrostatic NWP platform (e.g. HARMONIE model) and incorporating chemistry modules and aerosol-radiation-cloud interactions into the future integrated system (Baklanov, 2008; Baklanov et al., 2011a).

2.3. Atmospheric chemistry

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a) Tropospheric Sulfur Cycle

The simple tropospheric sulphur cycle chemistry module in Enviro-HIRLAM, used for long-term runs, is based on the sulfur cycle mechanism developed by Feichter et al. (1996) treating three prognostic species dimethyl sulfide (DMS), sulfur dioxide (SO_2) and sulfate $(SO_4^{2^2})$. The mechanism includes DMS and SO₂ oxidation by hydroxyl (OH) and DMS reactions with nitrate radicals (NO₃) in the gas-phase part. The heterogeneous aqueous phases chemistry comprises of SO₂ oxidation reactions by H2O2 and O3. Accounting for dissolution effects of SO2 in the aqueous phase is performed according to Henry's law. An output of global chemistry transport model MOZART (Horowitz et al., 2003) is used to prescribe three dimensional oxidant fields of OH, H2O2, NO2, and O3.

The sulfate produced in the gas-phase is referred to the gases and can be condensed on pre-existing aerosols or to nucleate by the aerosol microphysics M7 module (see Sect. 2.4). Moreover, in-cloud produced sulfate is accumulated on the pre-existing accumulation and coarse mode aerosols.

b) Gas-phase chemistry

The gas-phase chemistry scheme consists of sets of chemical schemes running from simple schemes for Chemical Weather Forecasts (CWF) to highly complex schemes for research and case studies. In order to make the model computationally efficient for different applications and operational forecasting several condensed atmospheric chemical schemes have been tested into Enviro-HIRMAM since the first version of the model system was realised (Korsholm, 2009; Gross and Baklanov 2004). In the current version of Enviro-HIRLAM the tropospheric condensed Carbon-Bond Mechanism version Z (CBM-Z) (Zaveri and Peters, 1999), a variant of CBM-IV gas-phase chemistry scheme (Gery et al., 1989), with a fast solver based on radical balances (Sandu et al., 2006) has been implemented in the model. CBM-Z uses lumped species that represent broad categories of organics based on carbon bond structure. It is closely related to CBM-IV which is widely used in air pollution evaluations, but with expansions to include reactions that are important in the remote troposphere. It also uses the most general organic category (PAR for paraffin) to represent miscellaneous carbon content so that carbon mass is conserved.

c) Chemical Solvers

The equations for photochemical production and loss are computationally expensive because they form a stiff numerical system. The photochemical mechanisms described above were implemented using two different chemical solvers: (1) the Rosenbrock (ROS) solver (Hairer and Wanner, 1996) as implemented by the Kinetic Preprocessor (KPP) (Sandu et al., 2006); and (2) the computationally rapid radical balance method (RBM) of (Sillman, 1991). Each of these provides a solution to the tendency equation for photochemical production and loss. The KPP provides a flexible tool to generate a well coded chemical mechanism according to the user choice of a given Ordinary Differential Equation (ODE) solver. We use KPP tools to create the gas-phase chemical mechanisms including the solvers for three chemical mechanisms. Usually, the Rosenbrock solver is selected for most of simulations.

d) Photolysis Rates

Photolysis rates are determined as a function of various meteorological and conditional inputs: altitude, solar zenith angle, column densities for O₃, SO₂ and NO₂, surface albedo, aerosol optical depth, aerosol single scattering albedo, cloud optical depths and cloud altitudes. Rates for specific conditions are determined by interpolating from an array of pre-determined values. The pre-determined values are based on the Tropospheric Ultraviolet-Visible Model (TUV) developed by Madronich and Flocke (1999), using a pseudo-spherical discrete ordinates method (Stamnes et al., 1988) with 8 streams. The 8-stream TUV is the most accurate method for determining photolysis rates but is computationally too expensive for use in 3-d models. Photolysis rate constants are calculated using the Fast-J radiative transfer model (Wild et al., 2000) with O(1D) quantum yields updated to JPL2003 (Sander et al., 2003). Cloud optical depths are determined using the random overlap treatment described by Feng et al. (2004), which assumes that cloudy and cloud-free sub-regions in each model grid box randomly overlap with cloudy and cloud-free sub-regions in grid boxes located above or below (Briegleb, 1992).

e) Heterogenic chemistry

Many gas-phase species are water soluble and sulphate and ammonia together with water take part in binary/ternary nucleation. In order to consider these processes a simplified liquid-phase equilibrium mechanism with the most basic

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equilibria is included in NWP-Chem-Liquid. At present this equilibrium module is solved using the analytical equilibrium iteration method (Jacobson, 1999). The reactions are summarized in Korsholm et al. (2009) and the module will be updated to include the impact of organic compounds from anthropogenic and biogenic sources.

2.4. Aerosol formation, dynamics and deposition

a) Aerosol dynamics module

The first aerosol module in Enviro-HIRLAM was based on the CAC (Chemistry-Aerosol-Cloud) model with the modal approach for description of aerosol size distribution (Baklanov, 2003; Gross and Baklanov, 2004) and considered only sulfurtype aerosols (Korsholm, 2009).

The current version of the Enviro-HIRLAM model has M7 aerosol microphysics module (Vignati et al., 2004) together with aerosol removal processes ported from ECHAM5-HAM climate model (Stier et al., 2005). There are two types of particles considered: insoluble and mixed (water-soluble) particles. The particles are split into seven classes depending on particle size and solubility by means of "pseudomodal" approach. Four classes are used to represent mixed particles, i.e., nucleation, Aitken, accumulation, and coarse modes, and another three classes are for the insoluble (Aitken, accumulation, and coarse modes). Four predominant aerosol types are included - black carbon (BC) and primary organic carbon (OC), sulfate, mineral dust and sea salt. The M7 aerosol dynamics includes nucleation, coagulation, and sulfuric acid condensation processes. Coagulation and condensation lead to formation of mixed particles from the insoluble ones.

b) Dry-deposition and Sedimentation

The dry deposition fluxes of gases and aerosols (for both number and mass concentrations) are calculated from the aerodynamic, quasi-laminar boundary layer as the product of the surface layer concentration and the dry deposition velocity (Stier et al., 2005). The fluxes are used as the lower boundary condition in the semi-implicit vertical diffusion TKE-CBR scheme (Cuxart et al., 2000). The calculation of the dry deposition velocities is performed by means of serial resistance approach. And the "big-leaf" method is used to calculate surface resistance (Ganzeveld and Lelieveld, 1995; Ganzeveld et al., 1998) per each grid-cell for the snow/ice, water, bare soil, low-vegetation and forest surface types. The SO₂ soil resistance is a function of soil pH, relative humidity, surface temperature, and the canopy resistance, while surface resistances for other gases are prescribed. The canopy resistance is computed from stomatal resistance and monthly mean Leaf Area Index (LAI) values from the Enviro-HIRLAM Interaction-Soil-Biosphere-Atmosphere scheme (Noilhan and Planton, 1989).

The sedimentation of the aerosol particles is calculated throughout the atmospheric column. The calculation of the sedimentation velocity is based on the Stokes velocity with the Cunningham slip-flow correction factor accounting for non-continuum effects (Seinfeld and Pandis, 2006). In order to satisfy the Courant-Friedrich-Lewy stability criterion, the sedimentation velocity is limited by ratio of the model layer thickness and the time-step.

c) <u>Wet-deposition</u>

There are several options for the wet deposition in the model. The first version used the aerosol size dependent parameterisation of Baklanov and Sørensen (2001). In the latest version fixed size- and composition-dependent scavenging parameters are also applied for wet deposition calculation and are different for stratiform and convective clouds (Stier et al., 2005). They were derived from measurements of interstitial and in-cloud aerosol contents. These scavenging coefficients depend on the aerosol modes, total cloud water and fraction (liquid- and ice), and the conversion rates of cloud liquid water and cloud ice to precipitation through auto-conversion, aggregation, and accretion processes. The precipitation re-evaporation before it reaches the ground is also included. The STRACO cloud scheme (Sass, 2002) provides water- and ice- precipitation fluxes, normalized by the precipitation rates, to wet-deposition scheme, which uses prescribed size-dependent collection efficiencies for rain and snow (Seinfeld and Pandis, 1998).

2.5. Emission modules and pre-processor

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The model includes anthropogenic, biomass burning (wildfires) and natural emission fluxes of both gases and aerosols. These emissions are processed in different ways; because some of them are pure datasets derived from ground-based and satellite observations. The others are interactively developing during the model integration and depend on the meteorological conditions at current time-step and land-use, -cover or water surfaces types. The anthropogenic emission inventory developed by TNO (Kuenen et al., 2014) and linked to the model is a dataset of yearly-accumulated fluxes of gases, such as CO, CH₄, NO_x, SO₂, NH₃, Non-Methane Volatile Organic Compounds (NMVOC), and particulate matter (PM) in two size bins - 2.5 µm and 10 µm, which are attributed to 10 source-sectors, e.g., energy industries, residential combustion, industry, etc., denoted by SNAP (Selected Nomenclature for sources of Air Pollution) codes. The inventory has resolution of 0.06° x 0.12° and covers the entire Europe, European part of Russia, North of Sahara and a part of Middle East. Total NMVOC emissions are split into 25 VOC compound groups by source-sectors by country (Kuenen et al., 2010). The PM2.5, PM10 emissions splitting into 6 aerosol species (BC, OC, Na, SO₄, Coarse Other Primary and Fine Other Primary particles) is applied following TNO recommendation (Kuenen et al., 2010). Because the dataset contains accumulated surface fluxes, one needs to redistribute them in order to reproduce diurnal, weekly and monthly emissions variability. The emissions can also occur at different heights, e.g., emissions from power plants are elevated and from traffic are at the surface; so, vertical redistribution is applied within first 8 model hybrid levels. Therefore, temporal and vertical profiles developed by TNO for different gaseous and aerosol species and SNAP codes are used in the emission pre-processor. The global biomass burning (wildfires) so-called the IS4FIRES (Sofiev et al., 2012) emission inventory developed by FMI has similar structure except a number and kinds of available gaseous and aerosol species as well as the resolution. The inventory data is total PM flux. The flux is

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279 280 splitted into $PM_{2.5}$ and coarse PM consisting of ash. The $PM_{2.5}$ primarily consists of Organic and Black Carbon (OC and BC) and a remainder of organic matter that is not carbon; for details see (Andreae and Merlet, 2001). The biomass burning emissions typically show a diurnal cycle variability, and therefore, corresponding coefficients are applied. The wildfires emissions are also redistributed vertically having different proportions in lowest 200 m and the highest up to 1 km over the ground.

The natural emissions of gases and aerosols are fully interactive and calculated online. There is dimethyl sulfide (DMS; Nightingale et al., 2000) emission from oceans, which depends on the wind speed and seasonal variability of DMS solution in the water. Soluble sea-salt aerosol emissions (Zakey et al., 2008) are driven by wind speed and temperature and insoluble mineral dust aerosol emissions (Zakey et al., 2006) also depend on meteorology as well as hydrological parameters. Both sea-salt and dust aerosols are emitted in accumulation and coarse modes.

2.6. Aerosol feedback mechanisms

a) Direct and semi-direct effects

Enviro-HIRLAM contains parameterisations of the direct and semi-direct effects of aerosols. Direct and semi-direct effects are realised by modification of the Savijärvi radiation scheme (Savijärvi, 1990; Wyser et al. 1999) with implementation of a new fast analytical SW and LW aerosol transmittances, reflectances and absorptances. The 2-stream approximation equations for anisotropic non-conservative scattering described by Thomas and Stamnes (2002) are used for these calculations. The GADS/OPAC aerosols of Köpke et al. (1997) are used as input to the routine. 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" aerosols. In order to make the calculations fast, optical properties that are spectrally averaged over the entire SW and LW spectra are used. The spectra used are shown in Figure 4. The short wave spectrum is a clear sky spectrum from 2 km height in a standard atmosphere (Anderson et al. 1986) calculated with the DISORT algorithm (Stamnes et al. 1988) run in the LibRadtran framework (Mayer and Kylling 2005). The long wave spectrum is calculated similarly and is based on the overall atmospheric LW transmittance of a standard atmosphere.



Figure 4: Left: The typical SW spectrum used for calculating average SW aerosol optical properties. Right: the spectral weights used for calculating average LW aerosol optical properties.

b) First and second indirect effects

For cloud-aerosol interactions a modified version of the Soft TRAnsition COndensation (STRACO) cloud scheme (Sass, 2002) is used in Enviro-HIRLAM. This scheme developed for operational NWP has recently been upgraded using new efficient methods to account for aerosol effects on cloud formation and microphysics. The scheme is able to account for convective transports of new variables. The prognostic aerosol fields are coupled directly to the cloud physical and microphysical properties. Liquid cloud droplet number is calculated based on aerosol size, number and solubility and the STRACO subgrid super saturation field is used as basis for the droplet nucleation calculation. This ensures consistency with the cloud water mass.

The modelled liquid droplet number evolves in time according to the following processes: droplet nucleation, self-collection, sedimentation and evaporation. In order to close the tendency calculations the liquid cloud droplet distribution is assumed to follow a gamma distribution where the shape parameter is calculated online using Geoffroy et al. (2010). Several schemes have been implemented for nucleation comprising Twomey (1959), Cohard et al. (1998), Cohard et al. (2000) and Abdul-Razzak et al. (1998), Abdul-Razzak and Ghan (2000). Self-collection is the process whereby droplets collide and stick together, but do not become rain-drops. The parameterization of self-collection processes follow Seifert and Beheng (2006). Sedimentation is calculated to be consistent with the mass of rain water in a given model time step under the basic assumption that the largest droplets are removed first from the cloud. Similarly, evaporation of a droplet below activation radius is calculated to be consistent with the total evaporated cloud water under the assumption that the smallest droplets evaporate first.

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333 334 Cloud droplet effective radius controls the liquid phase absorptivity and transmissivity and is calculated from liquid water mass and droplet number and is here also dependent on the shape of the droplet distribution which evolves in time. Autoconversion follows Rasch and Kristjansson (1998), and is directly dependent on the calculated droplet number.

2.7. Urban parameterisations and models urbanisation

The representation of urban areas in Enviro-HIRLAM contains the following aspects and processes (Baklanov et al., 2005: (i) model down-scaling, including increasing vertical and horizontal resolution and nesting techniques; (ii) modified highresolution urban land-use classifications, parameterizations and algorithms for roughness parameters in urban areas based on the morphologic method; (iii) specific parameterization of the urban fluxes in the meso-scale model; (iv) modelling/ parameterization of meteorological fields in the urban sublayer; (v) calculation of the urban mixing height based on prognostic approaches.

The urban parameterizations in the model contain three different approaches which may be combined. The first - simplest implementation contains modifications of the surface roughness, the anthropogenic heat flux, the storage heat flux and the albedo over urban areas. These are identified in the model using urban fractions extracted from the land-use database (CORINE) employed at DMI (Mahura et al., 2005b 2006, 2007a; Baklanov et al., 2005, 2008). The first module is the computationally cheapest way of "urbanising" the model and it can be used for operational NWP as well as for regional climate modelling. The second - Building Effect Parameterization (BEP) (Martilli et al., 2002) - module gives a possibility to consider the energy budget components and fluxes inside the urban canopy although it is a relatively more expensive (5-10% computational time increase) (Mahura et al., 2008bc; 2010b; Figure 5). However, this approach is sensitive to the vertical resolution of NWP models and is not very effective if the first model level is higher than 30 m. Therefore, the increasing of the vertical resolution of current NWP models is required. The third - Soil Model for SubMeso Urbanized (SM2-U) version (Dupont and Mestayer, 2006; Dupont et al., 2006) - module is considerably more expensive computationally than the first two modules (Mahura et al., 2005a Baklanov et al., 2008b). However, the third one provides the possibility to accurately study the urban soil and canopy energy exchange including the water budget. Therefore, the BEP scheme is considered as the baseline option and third SM2-U module is recommended only for use in advanced urban-scale NWP and meso-meteorological research models. Further information and results of testing and evaluation of the schemes may be found in (Mahura et al., 2005ab; 2006a; 2008abc; 2010b).



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344 345 Figure 5: General scheme of the Building Effect Parameterisation (BEP) module for the Enviro-HIRLAM model urbanization with a structure of the subroutine conception (adapted from Mahura et al., 2010b).

2.8. Transport schemes

Until 2012 there were basically two options for transport schemes in Enviro-HIRLAM (Chenevez et al., 2004): a) the traditional non-conserving but highly efficient semi-Lagrangian (SL) scheme in HIRLAM, b) the much less efficient flux based and positive definite finite volume scheme by Bott (1989) with updates by Easter (1993). In 2012 the default transport scheme was updated to a new monotonic version of the locally mass conserving semi-Lagrangian (LMCSL) scheme (Kaas 2008, Sørensen et al. 2013). This scheme to be described briefly below is almost as efficient as the traditional SL scheme but now with the attractive properties of inherent mass conservation, plus being monotonic and positive definite.

348 In HIRLAM and former versions of Enviro-HIRLAM a traditional SL scheme is used for advecting the specific concentration of water constituents or the mixing ratio q_i of any tracer *i*. Considering mixing ratio this means that when ignoring any sources/sinks and turbulent mixing the prognostic transport equation to be solved is simply





(1)

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352 The traditional SL numerical integration of Eq (1) reads 353

$(\mathbf{q}_i)_k^{n+1} = (\mathbf{q}_i)_{*k}^n$ (2)

354 where subscript k is the grid point/cell index and superscripts n and n+1 represent two consecutive time steps, respectively. 355 The subscript *k indicates the tricubic interpolation to the location of the departure point of the upstream trajectory, which 356 arrives in grid point k at time level n+1. The tricubic interpolation in (2) can also be represented as a sum of interpolation 357 weights involving 64 grid points surrounding the departure point. Formally this can be expressed

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$$(q_i)_k^{n+1} = \sum_{l=1}^{N} w_{k,l} (q_i)_l^n$$
 (3)

359 where K is the total number of grid points in the entire integration domain. Note that for each k only 64 $w_{k,l}$ weights are different from zero. When converting mixing ratio into volume density, i.e., $(\rho_i)_k^{n+1} = (\rho_d)_k^{n+1} (q_i)_k^{n+1}$, and subsequently 360 361 summing over the integration area the traditional SL scheme is not mass conserving. Therefore in LMCSL (Kaas, 2008) a 362 363 different approach is followed, namely, as in most other mass conserving transport schemes, to solve the complete continuity equation

$$\frac{\partial \rho_i}{\partial t} = -\nabla \cdot (\rho_i \boldsymbol{u}) \quad \text{or} \quad \frac{\boldsymbol{d} \rho_i}{\boldsymbol{d} t} = -\rho_i \nabla \cdot \boldsymbol{u} \tag{4}$$

365 still omitting sources/sinks and turbulent mixing an then evaluating the mixing ratio from $(\mathbf{q}_i)_k^{n+1} = (\rho_i)_k^{n+1} / (\rho_d)_k^{n+1}$. In LMSCL (4) is solved in a rather unusual way by modifying the interpolation weights in (3) in such a way that the sum of 366 367 mass given off at time step n by a Eulerian grid cell l to all departure points that it influences is exactly equal to its own mass. 368 In other words LMCSL is based on simple partition of unity. The modified weights become:

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$$\hat{W}_{k,l} = \frac{V_l}{V_k} \frac{W_{k,l}}{\sum_{m=1}^{K} W_{m,l}}$$
 (5)

370 where V_k is the volume of Eulerian grid cell k. Using the modified weights the basic LMCSL forecast reads:

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$$(\rho_i)_k^{n+1} = \sum_{l=1}^n W_{k,l}(\rho_i)_l^n$$
 (6)

372 As the traditional SL scheme the LMCSL is not monotonic or positive definite. Therefore an a posteriori iterative locally mass-conserving (ILMC) filter was developed, Sørensen et al. (2013). This filter ensures that the mixing ratio of the forecast will never be larger/smaller than the largest/smallest mixing ratio of the eight grid cells surrounding the upstream trajectory departure point at time level n. The ILMC filter designed to be as local as possible since non-local filters will generate non-376 377 physical chemical reactions. This is ensured by an iterative approach where the mass discrepancy is re-distributed among the neighbouring cells in the first iteration, and increasing the distribution radius, in case there is are remaining mass 378 discrepancy, for the next iteration(s) In general one or two iteration(s) are sufficient.

The LMCSL transport scheme in combination with the ILMC produces accurate monotonic and positive definite forecasts for water vapour, liquid/ice water and chemical constituents. As an example the simulated PM2.5 concentration on July 17 in 2010 with horizontal resolution of approximately 16 km's is shown in Figure 6. It can be seen that the model is able to reproduce, e.g., sharp transitions related to fronts over the North Atlantic. A more in depth analysis of the ability of ILMC to 383 reproduce sharp gradients can be found in Sørensen et al. (2013), in particular Figure 3 and the accompanying discussion in 384 that paper.







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Figure 6: Example of the simulated PM2.5 concentration over Europe on July 17 in 2010 with horizontal resolution of 16 km

It should be noted that the dynamical core in Enviro-HIRLAM is identical to that of HIRLAM. Thus, the dry-air density for dynamics is calculated using a traditional SL approximation to (TR4), i.e. not the LMCSL. Therefore, the Enviro-HIRLAM is not formally wind-mass consistent regarding tracer transport.

3. Modelling system applications

Possible applications of the online integrated Enviro-HIRLAM modeling system include the following: chemical weather forecasting,

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- 396 air quality and chemical composition longer-term assessment,
- 397 weather forecast (e.g., in urban areas, severe weather events, etc.),
- 398 pollen and bio-aerosols transport forecasting,
- 399 climate change modelling (EnvCLIMA, Enviro-HIRHAM),
- 400 studies of climate change effects on atmospheric pollution on different scales,
- 401 anthropogenic impacts on atmospheric processes, weather modifications, geo-engineering,
- 402 volcano eruptions, dust storms, nuclear explosion consequences,
- 403 other emergency preparedness modelling.

Several realised/tested types of applications of the Enviro-HIRLAM for meteorological, environmental and climate 405 forecasting and assessment studies are highlighted in Figure 1 and will be demonstrated below. 406 407

3.1. Improvements of Numerical Weather Prediction for High Impact Weather events

409 410 411 412 413 414 415 416 417 Several Enviro-HIRLAM sensitivity and validation studies of aerosol feedbacks on meteorological processes were done previously (see e.g., Korsholm, 2009; Korsholm et al., 2010; Baklanov et al., 2011ab; Sokhi et al., 2016). For example, 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). Further studies (Korsholm et al., 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 (see Section 3.4), 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 analysed 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 can reach 0.5°C (Figure 2.2b in Korsholm et al., 2010). Korsholm (2009) studied the impact of aerosol indirect effects on surface temperatures and air pollutant concentrations for a 24 h simulation over a domain in northern France including Paris in a convective case with low precipitation. He found a marginally improved agreement with observed 2m temperatures and a marked redistribution of NO2 in the domain, primarily as a result of the second indirect effect.

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423 424 To perform analysis of atmospheric aerosol effects on clouds and precipitation, the year 2010 was selected for Enviro-HIRLAM simulations. That year, especially summer, was characterized by severe weather events such as floods, heat waves 425 and droughts across Middle East, most of Europe and European Russia. The model was forced by boundary and initial 426 427 conditions produced by ECMWF IFS (IFS-CY40r1) and MOZART (Horowitz et al., 2003) models for meteorology and atmospheric composition, respectively. The Enviro-HIRLAM modelling domain with horizontal resolution of 0.15° x 0.15° 428 and 40 vertical hybrid levels covers Europe, North of Sahara, and European Russia. The model includes emissions from anthropogenic sources developed by TNO and from wildfires produced by FMI as well as interactive DMS, sea-salt and dust 430 emissions (for details see Sect. 2.5). 431

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For aerosol-cloud interactions, these were estimated also for July 2010 by means of delta function, i.e., difference between outputs of models: Enviro-HIRLAM with aerosol-cloud interactions (ENV) and Reference-HIRLAM (REF). Fig. 7a shows deltas (ENV-REF) of total cloud cover over model domain, which is mainly increased (with local maxima up to 90%) except several inland areas, such as Finland, borders of Germany, Poland and Austria, where cloud cover decreased by almost 10 fold. The ENV runs revealed the increase of average cloud top height by approximately 2%. The delta function of cloud water content at average cloud base shows (Fig. 7b) its increase compared to REF and local maxima over North Atlantic, North Sea, Sweden, Switzerland, and Austria. These areas are occupied by precipitating clouds as seen in Fig. 8.

The absolute frequencies of stratiform and convective precipitation over computational domain are decreased compared to the REF model, while the amount of convective precipitation during heavy precipitation events is increased. Hence, the wet deposition of particles decreases in summer because it rather depends on precipitation. But the inclusion of aerosol-cloud interactions can improve general model performance, i.e., the ENV run bias for precipitation with respect to its frequency and amount has been decreased compared to the REF model run (Fig. 9).











Figure 8: Accumulated (3 hour) precipitation patterns from Reference-HIRLAM (REF) and Enviro-HIRLAM with aerosolcloud interactions (ENV) on 17 Jul 2010, 18 UTC: stratiform precipitation: (a) – REF, (b) – ENV.





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Figure 9: Precipitation amount (12 hours accumulated) of reference HIRLAM (left) and Enviro-HIRLAM with aerosolcloud interactions (right) vs. surface synoptic observations at WMO station 6670 at Zurich, Switzerland (lat: 47.47; lon: 8.53) during Jul 2010.

However, it is necessary to mention that it is too early to make conclusions about the improvement of precipitation forecasting by implementation of the indirect aerosol effects, because of large uncertainties in parameterisation of the cloudaerosol microphysics processes (especially for ice-nucleation) and due to adjustments of such effects indirectly in NWP model parameters and constants (retuning of them after implementation of the aerosol feedbacks is needed). More investigations, further improvements and evaluations are needed for aerosol indirect effects and aerosol-cloud microphysics schemes in the model.

3.2. Urban meteorology and environment prediction and assessments

The analysis of urban boundary layer (UBL) for metropolitan areas of megacity Paris (more than 10 mil population) and growing medium-size Bilbao (1 mil) placed over a semi-flat and coastal-complex terrains, respectively, was performed employing the Enviro-HIRLAM model. In particular, the 1) evaluation of the model performance coupled with urban module for different types of terrain and size of cities; and 2) estimation of urban heat island (UHI) development over selected urban areas and surroundings were done.

The Enviro-HIRLAM simulations were performed for nested domains with horizontal resolutions of 15, 5 and 2.5 km and for selected periods in July 2009. The meteorological boundary conditions were provided by the European Centre for Medium Range Weather Forecast (ECMWF) every 3 hour. The model was employed in 2 modes. The 1st mode is *control (CTRL)* run. The 2nd mode is *urban (URB)* run – e.g. coupled with the Building Effect Parameterization (BEP, Martilli et al., 2002) module and anthropogenic heat fluxes (AHF) from the Large scale Urban Consumption of energY (LUCY) model (*Allen et al., 2010*). Extracted AHFs were 60 and 40 W m⁻² for the Paris and Bilbao metropolitan areas, respectively. For the URB run at the finest resolution, the Paris and Bilbao urban areas were represented by 220 and 16 urban cells, respectively (Figure 10; adapted from Gonzalez-Aparicio et al. 2010). In each grid-cell, BEP parameterizes the flux exchange between the urban surface and the atmosphere depending on combination of different urban districts, e.g. residential, low and high buildings, industrial and commercial.



Figure 10: Urban district classification based on urban zoning data for the a) Bilbao and b) Paris metropolitan area, including the residential area (ReD), low and high building districts (LBD and HBD, respectively) and industrial and commercial districts (ICD). Spatial distribution of urban districts (HBD – high buildings, RD – residential, ICD – industrial commercial, and CC – city center districts) for the Paris metropolitan area within the P01 modelling domain.

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497 Statistical analysis showed that for Paris, on a monthly basis, the correlations for air temperature were higher (e.g. closer to 498 observations) for the URB compared to CTRL run, and results improved up to 10% on a diurnal cycle (with a maximum of 499 0.83 at 08 UTC). The correlations were slightly lower (down to 0.5) at early morning hours and slightly higher (up to 0.8) 500 during afternoon and night-time. Moreover, correlations at suburban and urban stations were similar to correlations at rural 501 stations (see Figure 11a). Analysis for Bilbao (Gonzalez-Aparicio et al. 2013) showed similar performance of the model for 502 503 both runs: with correlation for air temperature about 0.85 and 0.88 for summer and winter, respectively. For the specific humidity it was 0.75 and 0.92. For the wind speed, the highest value (0.8) is in summer, and during winter it decreased to 0.6 504 (0.4) near the coast (inland) stations. 505 506

The results of simulations for two selected cities showed that the model reproduced well the meso-scale processes at regional scale, inland winds over Paris and land-sea breeze interactions over Bilbao. For selected locations (e.g. coastal vs. inland sites), the bias between the observations and simulations was higher over Bilbao (maritime) than over Paris (continental) cities. Although hydrostaticity of the model over a complex terrain is a limitation, but sensitivity test over Bilbao showed that at 2.5 km optimal resolution it is possible at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at finer scales.

The UHI development was also for short-term periods (here, for Paris - 28 Jul 2009; for Bilbao - 15 Jul 2009) with calm and anticyclonic conditions. For Paris, three different locations were considered: urban (LHVP), suburban (SIRTA) and rural (CHARTRES) stations (Figure 11a). As seen, the UHI was fully developed at 04 UTC with air temperature anomaly of 2.2°C (LHVP) and 0.6°C (SIRTA). It started at mid-night and expanded covering area of about 2000 km². The heat island was retained until 11 UTC, but during the daytime (e.g. 11-17 UTCs) the effect disappeared due to contribution of incoming solar radiation. At CHARTRES this effect (0.2°C) was almost negligible. Both the wind speed and relative humidity were also affected by the urban area: at LHVP the wind speed reduced by maximum 3.5 m s⁻¹ at 06 UTC, and the relative humidity down to 15% under developing UHI. At SIRTA the change in wind speed was down to 0.7 m s⁻¹ and at CHARTRES the changes in wind speed and relative humidity were almost negligible.

For Bilbao, model showed that for breezes from northern directions, the impact of urban area on local flow dynamics is inhibited; however, for breezes from southern directions - the urban effect had appeared. For example, on 15 Jul 2009, the UHI was developed during night-morning hours (e.g. 23-09 UTCs) with maximum up to 1°C, and heat island expanded covering area of about 130 km². In addition, Gonzalez-Aparicio et al. (2013) showed that the UHI intensity is lower in winter compared with summer, underlying that dominating factor is the surface heating during daytime, which is higher in summer than in winter.

As medium size cities are under continuous development, future impacts of urbanization are expected to become more significant. Several different scenarios of urban development were tested for Bilbao (Gonzalez-Aparicio et al., 2014). Enviro-HIRLAM model runs showed that under calm conditions during summer and winter, the UHI could reach up to 2.2°C covering area of about 400 km² when city is doubled in size or doubled in AHF. When city is tripled in size, the UHI could reach up to 3°C with urban island expansion up to 550 km² (Figure 11c). Analysis of UHI for Bilbao (e.g. triple size city scenario) vs. current UHI over Paris showed similar intensity of up to 3°C, and UHI boundaries are different, e.g. for Paris it was 4 times larger. Such differences can be explained by different cities' sizes, morphologies and characteristic AHFs.







Figure 11: Difference plots for the air temperature at 2m between outputs of the URB (urbanized -BEP + AHF-) and CTRL (non-urbanized) Enviro-HIRLAM model under calm conditions during summer 2009 for the a) Paris metropolitan area and for the Bilbao metropolitan area b) in its current size of the city and c) under a scenario tripling the size of the city.

3.3. Pollen forecasting

Among air-pollinated allergens, birch pollen is one of the most dangerous for the population group suffering allergic diseases. The number of allergic patients sensitive to birch pollen is assessed as 20% of European population (WHO, 2003; Linneberg, 2011) and this number is constantly increasing. In particular, in Denmark the number of allergic patients has increased twice over the past few decades (Linneberg, 2011). These facts demonstrate the importance of operational birch pollen forecasting for the European population especially during the spring season. Currently, birch pollen is presented as biological air pollutant in different NWP and ACT models such as SILAM (Finland), COSMO-ART (Germany), CHIMERE (France), Enviro-HIRLAM, DEHM (Denmark) and others. The pollen emission is strongly dependent on meteorology, so it is easy to simulate and forecast pollen pollution episodes by online-coupled meteorology-air pollution models since all necessary meteorological fields are available at each model time step.

Original developments of the dynamical Enviro-HIRLAM based operational modelling system for the birch pollen forecasting in Denmark (called Env-POLL) were started in 2006 (Rasmussen et al., 2006; Mahura et al., 2006b) including previously developed statistical methods (Rasmussen, 2002), modelling of elevated concentrations episodes, analysis of spatio-temporal and diurnal cycle variabilities, contribution of remote source regions into pollen levels, improvements in emissions and parameterizations, etc. (Mahura et al., 2007b, 2009, 2010a). The most recent developments are shown in Kurganskiy et al. (2015) with revised general scheme of input and output of the Enviro-HIRLAM birch pollen forecasting system presented in Figure 12. The input includes the meteorological initial/ boundary conditions (IC/BC) obtained from the IFS model system, birch forest fraction map, phenological data, i.e. temperature sum thresholds for start of flowering (Sofiev et al., 2013), accumulated total number of birch pollen particles emitted from a unit area during the pollinating season.

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Figure 12: General scheme of Enviro-HIRLAM birch pollen forecasting.

The forecasting of birch pollen concentrations requires information/data on the spatial birch tree distributions, characteristics of pollen release, its atmospheric transport and dispersion, its deposition due to gravitational settling and wet deposition, i.e. scavenging by precipitation. Birch pollen emission is fully dependent on temporal and spatial variability of meteorological conditions. The emission module (Sofiev et al., 2013) includes the following parameters affecting the pollen release: 2-meter air temperature and relative humidity, 10-meter wind speed, and accumulated precipitation. The atmospheric transport is handled in the same way as for aerosols (see section 2.8). Dry deposition of birch pollen particles in the atmosphere is represented by gravitational settling (Seinfeld and Pandis, 2006) whereas dry deposition due-to interactions of particles with the surface can be neglected according to Sofiev et al., (2006). The wet deposition scheme distinguishes between in-cloud (Stier et al., 2005) and below-cloud scavenging (Baklanov and Sørensen, 2001). The output in terms of birch pollen forecasting, and for analysis, contains 2D fields of the birch pollen concentration at the lowest vertical model level. The modeling domain has 15 km horizontal resolution with 154 and 148 grid points along longitude and latitude correspondently. The domain covers the main European part and is centered around Denmark.



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Figure 13: (a): Birch forest fraction map; (b): Example of the simulated birch pollen concentration in the modelling domain on the 6th of May, 2006 at 18 UTC.

Birch forest habitat map has been derived by GIS (Geographic Information System) analysis (http://www.spatialanalysisonline.com) for the selected modeling domain. The map (Fig. 13a) was obtained from birch forest fraction in each model grid cell. Three GIS based databases were used in the derivation procedure: 1) Global Land Cover Characterization (GLCC, http://landcover.usgs.gov/glcc/), 2) European Forest Institute (EFI, Päivinen et al., (2001)) and 3) Tree Species Inventory (TSI, Skjøth et al., (2008)). Both GLCC and EFI have 1 km horizontal resolution, whereas TSI has 50 km resolution.

As examples for the birch pollen season 2006 the model results were compared with observations for two Danish sites: Copenhagen and Viborg (see in Fig. 14). This year was dominated by a relatively cold spring over large areas of Europe followed by rapid warming and little/no rain. It caused short but intensive birch pollen season with long range transport episodes before the local flowering start and thereby emissions. The evaluation for both modeled and observed birch pollen concentrations showed extremely high values (daily averages about and even more than 1000 grains/m³) during 5-10 May 2006 episode for Copenhagen and 5-8 May 2006 episode for Viborg. The extremely high birch pollen concentrations over Denmark are also visible in Fig. 13b.







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Figure 14: Birch pollen concentrations observed (red) vs. modeled (green) at Danish sites: Copenhagen (a) and Viborg (b).

According to Sofiev et al., (2011) the following criteria can be used for assessment of birch pollen concentration forecasting: model accuracy (MA), hit rate (HR), false alarm ratio (FAR), probability of false detection (POFD) and odds ratio (OR). All of the criteria are calculated using four parameters obtained by assessment of the number of low and high modeled vs. observed birch pollen concentrations (C) relatively to a threshold value $N_{th} = 50$ grains/m³ (i.e. $C \ge N_{th}$ for high and $C < N_{th}$ for low-concentration days). The threshold has been chosen since most of the pollen allergy sensitive population might start suffering from allergic reactions when daily mean birch pollen concentration, $\hat{C} \ge N_{th}$ in the air (Jantunen et al., 2012).

The results of statistical analysis showed high MA for both Danish stations (0.95 for Copenhagen and 0.84 for Viborg, 0.9 in average). Prediction of elevated/top concentrations (HR values) by the model was assessed as 0.93 for Copenhagen and 0.58 for Viborg. The FAR values indicated that the probability to get an incorrect top model concentration was 0.07 and 0.42 for Copenhagen and Viborg, respectively. The POFD criterion showed low probability to get high modelled concentrations for observed low-concentration days (0.02 for Copenhagen and 0.18 for Viborg). Finally, the OR indicated that the chances to get the "high" day than the "low" (if the model prediction is "high") were 42 and 3.26 times higher for Copenhagen and Viborg, respectively. In other words, the OR values show the ratio between HR and POFD. As it is seen from the OR values provided above, a fraction of the correct forecasts is prevailing for both Danish stations in this study.

It was found that comparing with observations, the modeled results reflected the general shape of changes in pollen concentration during the episode studied for both Danish stations: Copenhagen and Viborg. As it is also seen in Fig. 14 the model reproduces the magnitude of birch pollen concentrations for the peak period of the season in comparison with observations. However, some overestimation of the modeled concentration is visible for both stations at the end of the season. It can be explained by contribution due to long-range atmospheric transport of pollen from other remote regions, presumably from those located more northerly than Denmark and where the pollen season starts and ends later relatively to the Danish sites.

3.4. Chemical Weather Forecasting and air pollution applications

Validation and sensitivity tests (on examples of case studies and short-time episodes) of the online 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 MEGAPOLI campaigns (summer 2009) datasets and showed that the model had performed reasonably well (Korsholm, 2009; Korsholm et al., 2009; 2010; Sokhi et al., 2016).

630 631 632 633 634 635 636 636 637 638 On-line vs. off-line coupled simulations for the ETEX-1 release 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 (Korsholm et al., 2009). Further studies (Korsholm et al., 2010) of urban aerosol effects on the atmospheric composition showed that aerosol feedbacks through the first and second indirect effect induce large changes in chemical composition, in particular nitrogen dioxide, in a case of convective clouds and little precipitation. For the Paris campaign, on diurnal cycle variability the ozone concentration patterns showed dependencies on meteorological parameters, and especially seen at urban scale runs (Mahura et al., 2010b).

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To perform further analysis of online coupling and feedback effects on atmospheric pollution forecasting, the year 2010 was 642 643 selected (for details see Sect. 2.5). Nuterman et al. (2013) evaluated the Enviro-HIRLAM model for July 2010 vs. groundbased observations of PM25 from EU AirBase air-quality network (Guerreiro et al., 2014), with a number of stations located 644 in Denmark, Sweden, Germany and Spain (see Fig. 15a). The model runs were performed for the entire July 2010 with 7 645 days spin-up in June. Fig. 15b shows correlation coefficients on a diurnal cycle for PM2.5 concentrations at selected <u>646</u> measurement sites. In general it shows a fairly good positive correlations (more than +0.3), except for several Spanish 647 stations (such as ES1938A at daytime, and ES1974A - at nighttime). 648







Figure 15: (a) Map of selected AirBase air-quality monitoring stations (http://acm.eionet.europa.eu/databases/airbase/) across Europe; (b) PM_{2.5} correlation coefficient on diurnal cycle for selected AirBase observation stations.

The model predicts well $PM_{2.5}$ day-to-day variability, but always has negative bias (Fig. 16). This under-prediction is due to several reasons: i) aerosol microphysics without secondary organic aerosols; ii) lack of partitioning of ammonium nitrate; iii) crude model resolution, which still cannot capture small-scale effects like complex orography and urbanized regions (in particular, due to lack of fine-resolution emissions from anthropogenic sources, like urban traffic). For instance, the model shows negative bias of $PM_{2.5}$ during daytime at Danish urban station (Fig. 16a). It is apparently due to crude model resolution in the considered runs. It was also found that $PM_{2.5}$ values are very influenced by changes in atmospheric stability conditions, which difficult to predict accurately in many NWP models. This can be observed from correlation coefficient decrease at stations during night-time (at 03 UTC) or from underestimation of elevated concentrations. In spite of these issues, the model can well reproduce diurnal cycle of aerosols at different sites, e.g. urban (Fig. 16a), coastal and rural (Fig. 16b), and shows good overall performance.



Figure 16: Error-bar concentrations [ug/m³] on diurnal cycle for AirBase observations vs. Enviro-HIRLAM modeling results; (a) Danish urban station and (b) German rural station; Right top corner indicates maximum and average correlation coefficients for the station as well as total number of analysed observation samples; Green numbers along X axis indicate number of observation samples per time slice.

Further on-going developments of the Enviro-HIRLAM modelling system for atmospheric composition applications are realised within the FP7 MarcoPolo and NordForsk CarboNord projects. The Enviro-HIRLAM downsclaing from regional-tourban scale modelling is realised in MarcoPolo for the East China region and largest metropolitan agglomerations in China (Mahura et al., 2016) with a focus on providing services on meteorology and atmospheric composition (with focus on aerosols). The Northern Hemispheric low resolution modelling over European domain in a short-term mode with focus on feedbacks mechanisms evaluation (Nuterman et al., 2015; Kurganskiy et al., 2016).

4. Conclusions and further research/development needs

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681 The Environment - HIgh Resolution Limited Area Model (Enviro-HIRLAM) is developed as a fully online 682 coupled/integrated numerical weather prediction (NWP) and chemical transport model (CTM) for research and forecasting of 683 joint meteorological, chemical and biological weather. Possible applications of the modeling system include: chemical 684 weather forecasting, air quality and chemical composition longer-term assessment, weather forecast (e.g., in urban areas, 685 severe weather events, etc.), pollen forecasting, climate change forcing modelling, studies of climate change effects on <u>686</u> atmospheric pollution on different scales, weather modification and geoengineering methods, volcano eruptions, dust storms, 687 nuclear explosion consequences, other emergency preparedness modelling. Several types of the above applications of the 688 Enviro-HIRLAM for meteorological, environmental and climate forecasting and assessment studies are tested and 689 demonstrated. Different applications of Enviro-HIRLAM were realised for different geographical regions and countries 690 including European countries such as Denmark, Lithuania, France, Spain, Ukraine, Russia, Turkey and well as for other 691 regions - China and Arctic.

692 693 It is clear that the online modelling approach realised in Enviro-HIRLAM is a prospective way for future single- atmosphere modelling systems, providing advantages for all three communities: Meteorological modelling including NWP, AQ 694 modelling including CWF, and climate modelling. However, there is not necessarily one configuration of the integrated 695 online modelling approach/system suitable for all communities.

Comprehensive online modelling systems, like Enviro-HIRLAM, built for research purposes and including all important mechanisms of interactions, will help to understand the importance of different processes and interactions and to create specific model configurations that are tailored for their respective purposes.

- Seamless online integration modelling approach is a prospective way for future single-atmosphere modelling systems with advantages for applications at all time scales of NWP, AQ and climate models.
- Episode studies demonstrated the importance of including the meteorology and chemistry (especially aerosols) interactions in online-coupled models.
- There is no one unique integrated online modelling system configuration, which is best suitable for all communities, and hence, different model versions should feed for different purposes.
- For AQ: online coupling improve air quality forecasts, and especially with full chemistry and aerosol feedbacks effects included.
- For NWP: gas chemistry is not critical and can be simplified (or omitted), but aerosol feedbacks important for radiation and precipitation and especially for very polluted episodes and in urban areas (although statistically these effects are not so strong on long-term runs).
- For pollen forecast: Improve pollen emission simulation and correspondingly concentrations. Feedbacks are not important. Chemistry is not considered, but interaction with allergens would be interesting to study in future (not done vet).
 - For climate studies: suitable only for understanding the feedback mechanisms, it is too expensive for climate runs (Enviro-HIRLAM had been used maximum for 11 year period runs). Chemistry is important, the model needs to be optimised and simplified.

It should be stressed that there are still main gaps remaining in understanding of several processes such as: (i) aerosol-cloud interactions (still poorly represented); (ii) data assimilation in online models (still to be developed to avoid over-specification and opposite effects); (iii) model evaluation for online models needs more (process) data and long-term measurements - and a test-bed.

Code and/or data availability

723 724 725 726 727 728 The Enviro-HIRLAM system is a community model, the source code is available for non-commercial use upon agreement through contact to Bent Hansen Sass (bhs@dmi.dk) and Roman Nuterman (nuterman@nbi.ku.dk). Documentation. educational materials and exercises are available from hirlam.org and YSSS training schools: http://netfam.fmi.fi/YSSS08/, http://www.ysss.osenu.org.ua/ and http://aveirosummerschool2014.web.ua.pt/.

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Annex 1: Enviro-HIRLAM model development history:

1999: Started at DMI as an unfunded initiative (A. Baklanov et al.)

- 1034
- 2000: Used previous experience of the Novosibirsk scientific school (A. Baklanov) and SMHI (A. Ekman PhD) 2001: Online passive pollutant transport and deposition in HIRLAM-Tracer (J. Chenevez, A. Baklanov, J.H. Sørensen) 1035
- 10<u>36</u> 2003: Aerosol dynamics model developed and tested first as 0D module in offline CAC (A. Baklanov, A. Gross)
- 1037 2004: Test of different formulations for advection of tracers incl. cloud water (K. Lindberg)
- 1038 2005: Urbanisation of the model (funded by FP5 FUMAPEX) (A. Baklanov, A. Mahura, C. Peterson)
- 1039 2005: COGCI grant for PhD study of aerosol feedbacks in Enviro-HIRLAM (U. Korsholm, supervised by A. Baklanov, E. 1040 Kaas)
- 1041 2006: Test of CISL scheme in Enviro-HIRLAM (P. Lauritzen, K. Lindberg)
- 1042 2007: First version of Enviro-HIRLAM for pollen studies (A. Mahura, U. Korsholm, A. Rasmussen, A. Baklanov)
- 1043 2008: New economical chemical solver NWP-Chem (A. Gross)
- 1044 2008: First version of Enviro-HIRLAM with indirect aerosol feedbacks (U. Korsholm PhD)
- 1045 2008: Testing new advection schemes in Enviro-HIRLAM (UC: E. Kaas, A. Christensen, B. Sørensen, J.R. Nielsen)
- 104<u>6</u> 2008: Decision to build HIRLAM Chemical Brunch (HCB) with Enviro-HIRLAM as baseline system, Enviro-HIRLAM 1047 becomes an international project

1048 2008: 1st International Young Scientist Summer School (YSSS) on "Integrated Modelling of Meteorological and Chemical 1049 Transport Processes" (based on Enviro-HIRLAM) in St. Petersburg, Russia: http://netfam.fmi.fi/YSSS08/

- 1050 2009: Integrated version of Enviro-HIRLAM based on reference version 7.2 and HCB start
- 1051 2011: New chemistry (A. Zakey), direct and semi-direct aerosols effect (K.P. Nielsen) schemes
- 1052 2011: 2nd International YSSS (based on Enviro-HIRLAM/HARMONIE) in Odessa, Ukraine: http://www.ysss.osenu.org.ua/
- 1053 2012: New effective aerosol scheme for multi-compound aerosols (R. Nuterman)
- 1054 2012: New mass conserving and monotonic semi-Lagrangian transport (B. Sørensen et al. 2013)
- 1055 2013: New STRACO scheme with aerosol-clouds interaction (U. Korsholm and B. Sass)
- 10<u>5</u>6 2013: Model evaluation study within the AQMEII, phase 2 exercise (R. Nuterman)
- 1057 2014: Moving to the HARMONIE platform and building a joint strategy with ALADIN community
- 1058 2014: 3rd International YSSS (based on 5 online coupled models including Enviro-HIRLAM/HARMONIE) in Aveiro, 1059 Portugal: http://aveirosummerschool2014.web.ua.pt/
- 1060
- 2014-2016: Enviro-HIRLAM birch pollen forecasting system (A. Kurganskiy et al.) 1061
- 2015: New radiation scheme with aerosol direct and semi-direct effects for SW and LW radiation (K.P. Nielsen et al.)
- 1062 2016: Application of Enviro-HIRLAM for China (Marco-Polo project, A. Mahura et al.)