Enviro-HIRLAM online integrated meteorology-chemistry modelling system: strategy, methodology, developments and applications (v. 7.2)

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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 ~~(Klein et al., 2012)~~. 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~~model is based on the ~~DMI~~ HIRLAM NWP model with online integrated ~~passive~~ pollutant transport and dispersion, chemistry, aerosol dynamics, deposition and ~~indirect effects~~atmospheric composition feedbacks. 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~~current version 7.2, ~~is based on HIRLAM reference v7.2 with~~in comparison with previous versions, has a more advanced and cost-efficient~~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, and operational forecasting atmospheric composition with downscaling for China since 2017. 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~~Introduction

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 still 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). First attempts at building online coupled meteorology and air pollution models for environmental applications were done in the 1980s, cf. Baklanov (1988), Schlünzen and Pahl (1992), Jacobson (1994). For climate applications the first coupled chemistry-climate models were developed and used in the 1990s, cf. Jacobson (1999, 2002), de Grandpré et al. (2000), Steil et al. (2003), Austin and Butchart (2003). More detailed overview of t~~T~~he history and 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. Klein et al. (2012) extended applications of coupled models for "biological weather", defined as "the short-term state and variation of concentrations of bioaerosols", in particular for pollen modelling and forecasting.

The ~~on-line~~online 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; Sokhi et al. 2017).

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~~modelling 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~~online 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 completed and ongoing research projects (FP6 FUMAPEX; FP7 MEGAPOLI, PEGASOS, MACC, TRANSPHORM, MarcoPolo, CRICIAL; NordForsk NetFAM, MUSCATEN, CarboNord, CRAICC-PEEX, CRUCIAL; 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 operational for atmospheric composition (with focus on aerosols) for China since Nov 2016 (Mahura et al., 2016; 2017). 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 current version of the Enviro-HIRLAM coupled modelling system with improved parameterisations of meteorology-composition two-way interactions, main steps in its development and examples in different application areas for air quality, weather and pollen forecasting are considered for the first time. Section 2 provides a detailed description of the Enviro-HIRLAM modelling system and its key developments in the meteorological core, chemistry and aerosol dynamics parts, aerosol-meteorology interactions, models urbanisation and improvements of numerical algorithms. Section 3 describes a few types of Enviro-HIRLAM applications for meteorological and environmental forecasting and assessment studies. Sections 4 and 5 continue discussions and summarise the model applicability and provide recommendations for future research. Annex 1 includes brief information about the Enviro-HIRLAM model development history. A list of acronyms is provided in Annex 2.In this paper an overall description of the Enviro-HIRLAM coupled modelling system and examples in different application areas are considered.

2. Enviro-HIRLAM modelling system description

2.1. Modelling system structure

The Enviro-HIRLAM is a fully on-lineonline 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 (http://hirlam.org/index.php/projects/chemistry-branchhttps://hirlam.org/trac/wiki). It was the first meso-scale on-lineonline 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 (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 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 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 HIRLAM v7.2 with a more advanced and effective chemistry 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). This version is continuously under development and evaluation for various weather and air quality related applications (in particular, within the COST Action ES1004 where the above mentioned effects were extensively discussed, see, e.g., in Baklanov et al. 2014).

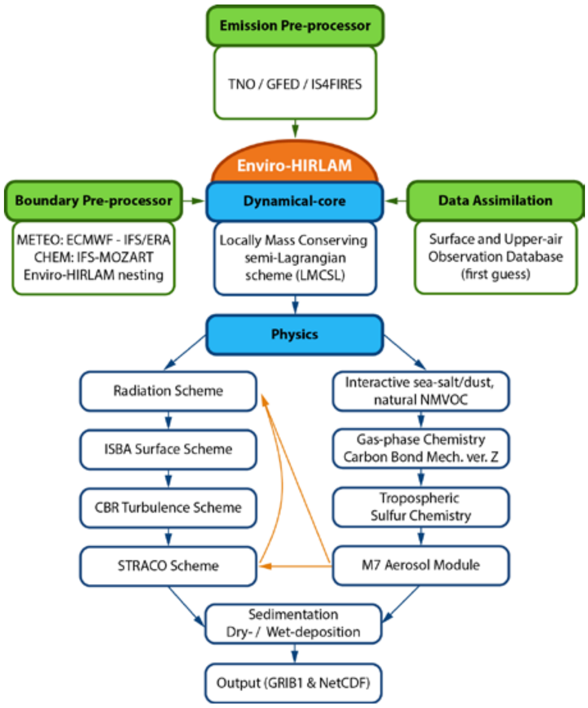


Figure 2: Schematics of the Enviro-HIRLAM modelling system.

Vertical structure and horizontal resolutions of the model are flexible. Limitations, e.g. due to the hydrostatic approximation, are provided (min 1,5 km of the horizontal resolution for flat terrains, e.g. for Copenhagen).

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 to take into account spatial-temporal evolution of atmospheric chemical and biological aerosols driven by meteorology from 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, including: air temperature and specific humidity, atmospheric pressure, wind speed and direction, 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 core).

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

The hydrostatic approximation of the model can be a limitation to increase the resolution for urban simulations. However, sensitivity tests for a medium size city demonstrated that the 2.5 km was the optimal resolution, allowing at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at local scale (González-Aparicio et al. 2013). For other metropolitan areas such as Paris, Rotterdam, St. Petersburg, Shanghai - a similar resolution was chosen, whereas for Copenhagen (with its flat terrain) the highest suitable resolution tested was 1.5 km and provided reasonable verification results (Mahura et al., 2006a, 2008bc, 2016). Within a selected metropolitan area there could be only a few grid cells having 100% representation of the urban fraction, but taking into account all urban grid cells, the boundaries of the cities (number of cells) could be substantially larger. Moreover, most of existing parameterizations in the physics core of any NWP model might need a revision when resolutions of 1 km and finer are used.

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).

The new non-hydrostatic version under HARMONIE is under development and only some elements are realised so far. The non-hydrostatic HARMONIE-AROME model includes only some aerosol effects. The physics included in this version of HARMONIE has recently been detailed by Bengtsson et al. (2017). HARMONIE-AROME is based partly on Meso-NH (Mesoscale Non-Hydrostatic atmospheric model), which is a cloud resolving model that includes state-of-the-art chemistry and aerosol interactions (e.g. Berger et al. 2016). However, Meso-NH cannot be run as a near real time NWP model, as it is possible with Enviro-HIRLAM.

2.3. Atmospheric chemistry

a) Tropospheric Sulfur Cycle

The simple tropospheric sulphur cycle chemistry module in Enviro-HIRLAM, used for long-term runs (up to one year), 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-}). The mechanism includes DMS and SO_2 oxidation by hydroxyl (OH) and DMS reactions with nitrate radicals (NO_3) in the gas-phase part. The heterogeneous aqueous phases chemistry comprises of SO_2 oxidation reactions by H_2O_2 and O_3. Accounting for dissolution effects of SO_2 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, H_2O_2, NO_2, and O_3.

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.

The tropospheric sulfur cycle chemistry is used together with M7 aerosol microphysics module because of its relative simplicity and low computational cost. The CBM-Z gas-phase chemistry (see the next section) is not interfaced with the M7 aerosol module because of several reasons: 1) the aerosol microphysics module does not include Secondary Organic Aerosols, therefore, there is no need of complex gas-phase mechanism with Volatile Organic Compounds related reactions and 2) it is too computationally expensive to use CBM-Z together with M7 for both weather and atmospheric composition prediction.

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.

Six environmental/smog chamber experiments were used to validate the gas-phase schemes as box models and within a regional climate model (Shalaby, 2012; Shalaby et al., 2012). The Tennessee Valley Authority (TVA) and the EPA chamber experiments were used to evaluate the different gas-phase schemes and different chemical solvers. Namely, TVA005 and TVA006 are designed to test the simple system of NOx; TVA068 is designed to test a simple mixture of VOC with very high NOx. EPA069A, EPA073A and EPA150A are used to validate the schemes with low NOx concentration and high VOC concentration.

c) Chemical Solvers

Calculating the time evolution of gas-phase chemistry requires a numerical integration of a set of stiff ordinary differential equations (ODE) and is among the most computationally expensive operations performed in a photochemical grid model. 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 to solve the tendency equation for photochemical production and loss: (1) the Rosenbrock (ROS) solver (Sandu et al., 1997 and 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). RBM utilizes the fact that much of the complexity of tropospheric chemistry stems from the HO_x radical family (OH, HO_2 and RO_2), which has a limited set of sources and sinks. The method solves reverse-Euler equations for OH and HO_2 based on the balance between sources, sinks and (if applicable) prior concentrations at the start of the time step. Reverse Euler equations for other species are solved in a reactant-to-product order, in some cases involving pairs of rapidly interacting species, and with some modifications to increase accuracy in exponential decay situations. The procedure is equivalent to a reverse Euler solution using sparse-matrix techniques, but with the matrix inversion linked specifically to the behaviour of OH and other species in the troposphere. Prior work tested several atmospheric chemistry mechanisms in the model taken into account different chemical solvers, we select the photochemical mechanism CBM-Z because it affords a reasonable trade-off between accuracy and computational efficiency. During the prior work including the validation stages of the gas-phase schemes (results not shown) we used KPP to generate the Fortran code of three different gas-phase schemes CBM-Z (Zaveri and Peters, 1999), GEOS-CHEM (Evans et al. 2003) and the Regional Atmospheric Chemistry Model "RACM" (Stockwell et al,1997). In order to fit within our main aim of the chemical weather predication, we didn't use both of GEOS-CHEM and RACM because they are very computationally expensive schemes due to their extensive number of chemical reactions. ~~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 due to its ability as a fast computational solver (Sandu et al 1997).

d) Photolysis Rates

Photolysis rates are determined as a function of various meteorological and conditional inputs~~: altitude, solar zenith angle, column densities for O_3, SO_2 and NO_2, 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~~latter is 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 3D~~-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).~~

For simplicity, photolysis rates are estimated as the following. At first, ~~:~~

~~1. F~~for the simple reactions the photolysis rates are estimated as a function of number of parameters such as meteorological and chemical inputs including altitude, solar zenith angle, overhead column densities for O_3, SO_2 and NO_2, surface albedo, aerosol optical depth, aerosol single scattering albedo, cloud optical depth and cloud altitude. At second,

~~2. F~~for the complex reactions, the photolysis rates are estimated as lookup table using the TUV model. TUV is run offline and calculated a lookup table of the photolysis rates, and then this lookup table is implemented under different weather conditions inside the model.

Photolysis rates can be significantly affected by the presence of clouds. 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). The method used to correct for cloud cover is based on Chang et al. (1987), which requires information on cloud optical depth for each model grid cell. Optical depth is used to reduce photolysis rates for layers within or below clouds to account for UV attenuation or to increase photolysis rates due to above-cloud scattering. In general, below cloud photolysis rates will be lower than the clear sky value due to the reduced transmission of radiation through the cloud. Similarly, photolysis rates are enhanced above the cloud due to the reflected radiation from the cloud. Cloud optical depths and cloud altitudes from Enviro-HIRLAM are used in the photolysis calculations, thereby directly coupling the photolysis rates and chemical reactions to meteorological conditions at each model time step.

e) Heterogeneous~~ic~~ 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 equilibria is included in NWP-Chem-Liquid. The "NWP-Chem-Liquid" is a thermodynamic equilibrium model, described in Korsholm et al. (2008). ~~At present t~~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 sulfur-type 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. Different aerosol types mentioned in above (as well as others, e.g. pollen particles) are provided as separate species in the model outputs along with lumped PM_{10} and $PM_{2.5}$.

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 SO_2 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) Wet-deposition

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

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, CH_4, NO_x, SO_2, NH_3, 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^o x 0.12^o 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 $PM_{2.5}$, PM_{10} emissions splitting into 6 aerosol species (BC, OC, Na, SO_4, 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 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 (Giglio, 2007). 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 addition to the more standard nucleation, accumulation and coarse aerosol size modes we consider, according to Köpke et al. (1997), the transported size mode to describe aerosols that have been transported over a long distance, for instance Saharan aerosols that have been blown to the Atlantic ocean. 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.

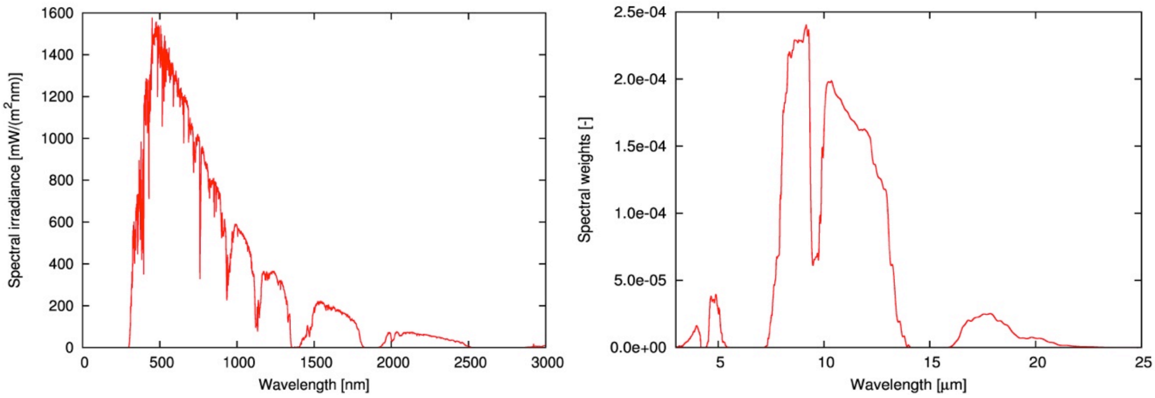


Figure 4: Left: The typical SW spectrum used for calculating average SW aerosol optical properties. Right: the spectral weights used for calculating average LW aerosol optical properties.

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.
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.
Abdul-Razzak and Ghan (2000) parameterization for aerosol activation has been extensively tested in many online-coupled weather and climate models. However, the STRACO cloud microphysics scheme with parameterizations of aerosol activation, cloud droplets nucleation, sedimentation, evaporation, self-collection, has been evaluated only with 1D column HIRLAM, so it needs to be further thoroughly evaluated.

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 high-resolution 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 nesting technics and downscaling methods are actively and successfully used for urban areas to reach the necessary resolution for resolving or parameterisation of urban features and effects. The details of this approach with the Enviro-HIRLAM model were described e.g. in Baklanov and Nuterman (2009). With respect to metropolitan areas, the downscaling for finer resolution allows to reproduce smaller scale meteorological patterns, and then these patterns are further modified through running urban parameterization modules only for grid cells where the cities are presented.

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, 2006a, 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. The details of implementations of different urban modules, own developments and comparisons of different approaches and modules were published in previous papers~~Further information and results of testing and evaluation of the schemes may be found in~~ (Mahura et al., 2005ab; 2006a; 2008abc; 2010b; Baklanov et al., 2005, 2008b). The main approach includes an integration of the urban modules into the ISBA (Interaction Soil- Biosphere- Atmosphere) land surface scheme of the NWP / HIRLAM model. The urban modules are activated only on those grid cells of the model domain where the urban fraction is presented.

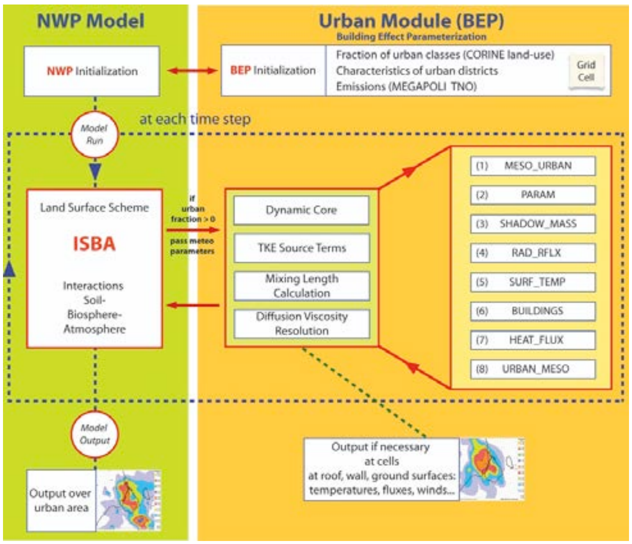


Figure 5: General scheme of the Building Effect Parameterisation (BEP) module for the Enviro-HIRLAM model urbanization with a structure of the subroutine conception (adapted from Mahura et al., 2010b).

The urban boundary layer is very inhomogeneous and plays an important role in forming urban meteorological fields and especially in dispersion of atmospheric pollutants, Therefore, for calculation of the urban mixing height, additionally to the common diagnostic approaches, prognostic equations were used according to Zilitinkevich et al. (2002) and Zilitinkevich and Baklanov (2002).

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 (Robert, 1981) 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, used in the present version of Enviro-HIRLAM, 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.

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

$$\frac{dq_i}{dt} = 0 \tag{1}$$

The traditional SL numerical integration of Eq (1) reads

$$(q_i)_k^{n+1} = (q_i)_{*k}^{n} \tag{2}$$

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

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

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 summing over the integration area the traditional SL scheme is not mass conserving. Therefore in LMCSL (Kaas, 2008) a different approach is followed, namely, as in most other mass conserving transport schemes, to solve the complete continuity equation

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

still omitting sources/sinks and turbulent mixing an then evaluating the mixing ratio from $(q_i)_k^{n+1} = (\rho_i)_k^{n+1} / (\rho_d)_k^{n+1}$. 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 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. In other words LMCSL is based on simple partition of unity. The modified weights become:

$$\hat{w}_{k,l} = \frac{V_l}{V_k} \frac{w_{k,l}}{\sum_{m=1}^{K} w_{m,l}} \tag{5}$$

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

$$(\rho_i)_k^{n+1} = \sum_{l=1}^{K} \hat{w}_{k,l} (\rho_i)_l^n \quad (\rho_i)_k^{n+1} = \sum_{l=1}^{K} w_{k,l} (\rho_i)_l^n \tag{6}$$

As the traditional SL scheme the LMCSL is not inherently 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-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 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 $PM_{2.5}$ 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 reproduce sharp gradients can be found in Sørensen et al. (2013), in particular Figure 3 and the accompanying discussion in that paper.

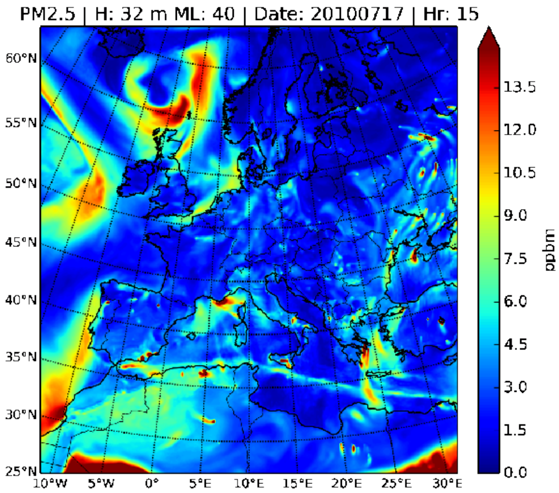


Figure 6: Example of the simulated $PM_{2.5}$ concentration over Europe on July 17 in 2010 with horizontal resolution of 16 km.

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 (Eq. ~~TR~~4), i.e. not the LMCSL. Therefore, the Enviro-HIRLAM is not formally wind-mass consistent regarding tracer transport. However, the large scale precipitation fields in the traditional HIRLAM and Enviro-HIRLAM are very similar (see, e.g., Figure 4 in Sørensen et al. (2013)), which suggests that wind-mass inconsistency is of minor importance. In principle no monotonic transport schemes can be mass-wind consistent since the monotonic limiters formally destroy the consistency (see discussions on the issue of mass-wind inconsistence in atmospheric models in Jöckel et al. (2001)).

3. Modelling system applications

Possible applications of the online integrated Enviro-HIRLAM ~~modeling~~modelling system include the following:

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

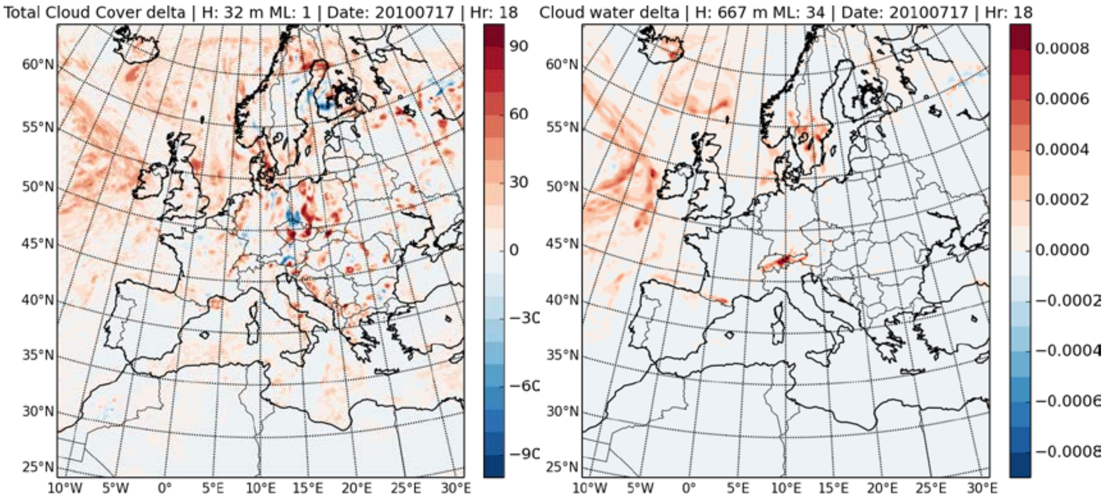
Several realised/tested types of applications of the Enviro-HIRLAM for meteorological, environmental and climate forecasting and assessment studies are highlighted in Figure 1 and will be demonstrated below.

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

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 NO_2 in the domain, primarily as a result of the second indirect effect.

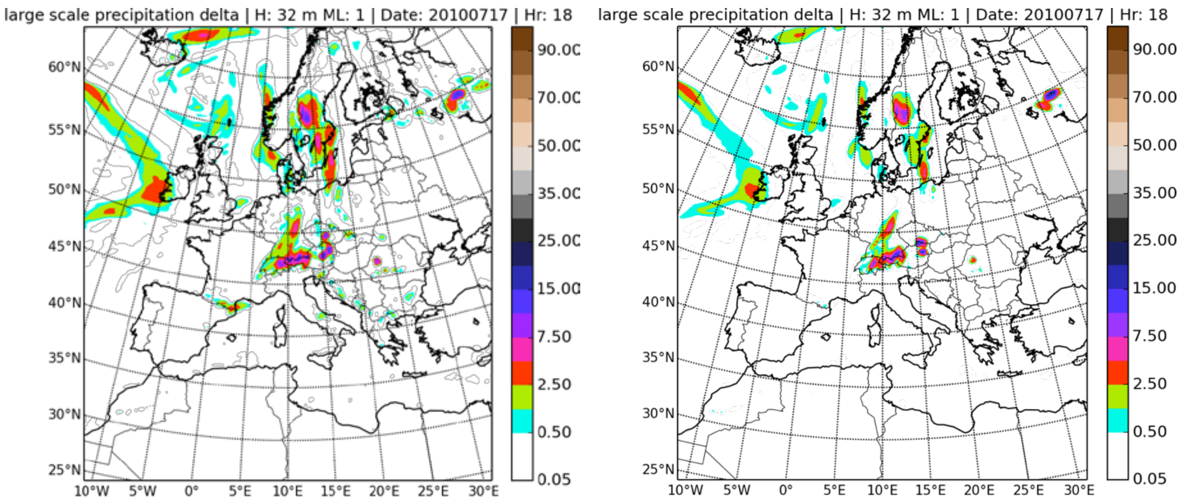
To perform analysis of atmospheric aerosol effects on clouds and precipitation, the year 2010 was selected for Enviro-HIRLAM simulations. That year, especially summer, was characterized by severe weather events such as floods, heat waves and droughts across Middle East, most of Europe and European Russia. The model was forced by boundary and initial 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° having 310 x 310 grid cells, and 40 vertical hybrid sigma levels extending to pressures less than 10 hPa, covers Europe, North of Sahara, and European Russia. The modelling domain was partitioned into 120 CPU cores and the model was run with time step of 300 seconds. 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 emissions (for details see Sect. 2.5).

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 frequency than on its amount. The REF model run tends to over-predict both frequency and amount of 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).



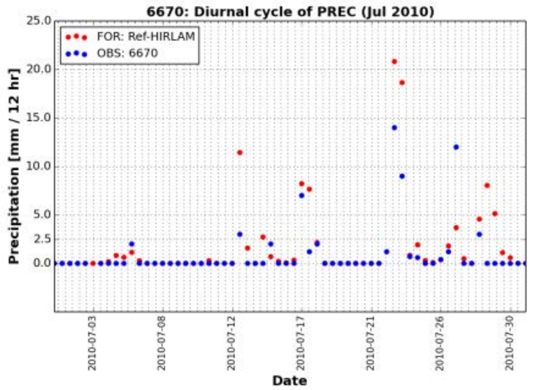
(a) (b)

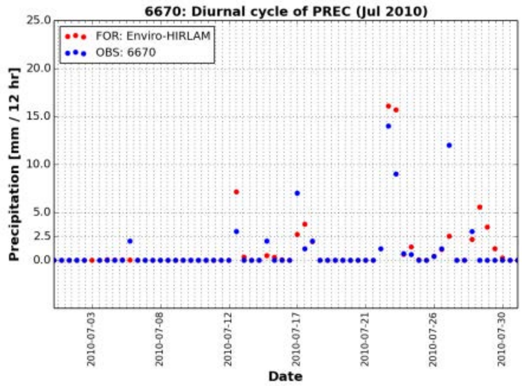
Figure 7: Delta (Enviro-HIRLAM – Reference-HIRLAM) of (a) vertically integrated total cloud cover [%] and (b) cloud water content [kg/kg] at average cloud base (667 m) on 17 Jul 2010, 18 UTC.



(a) (b)

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





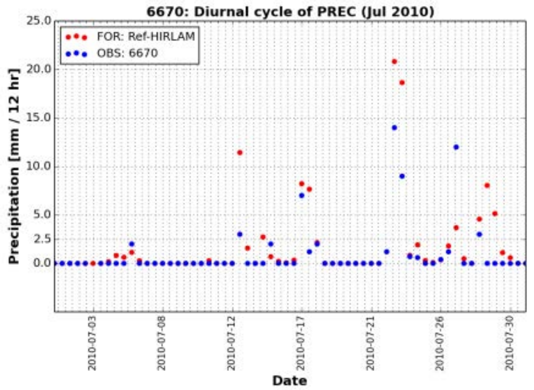


Figure 9: Precipitation amount (12 hours accumulated) of reference HIRLAM (left) and Enviro-HIRLAM with aerosol–cloud interactions (right) vs. surface synoptic observations at WMO station 6670 at Zurich, Switzerland (lat: 47.47; lon: 8.53) during Jul 2010.

Sensitivity studies on the model response to aerosol effects indicate strong "signals", but it doesn't guaranty improvements. E.g., Korsholm (2009) considered evaluations only for some elements (e.g., the coupling interval) in the previous analysis and made corresponding conclusions about the improvements. Other feedback mechanisms, especially for aerosol-cloud interactions, were analysed mostly as sensitivity studies or evaluations for short-term episodes.
The model formulations have only been tested on a case basis and although strong signals have been found, this does not imply improved meteorological performance of the model. In particular, testing over longer periods including all seasons was not conducted that time. Furthermore, the interactions between aerosols and the cloud ice-phase are not in a state where improvements would be expected. Therefore~~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 cloud-aerosol 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. Recently such evaluation studies are realised within the CarboNord project for monthly and annual validation studies and will be published separately.

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 González-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.

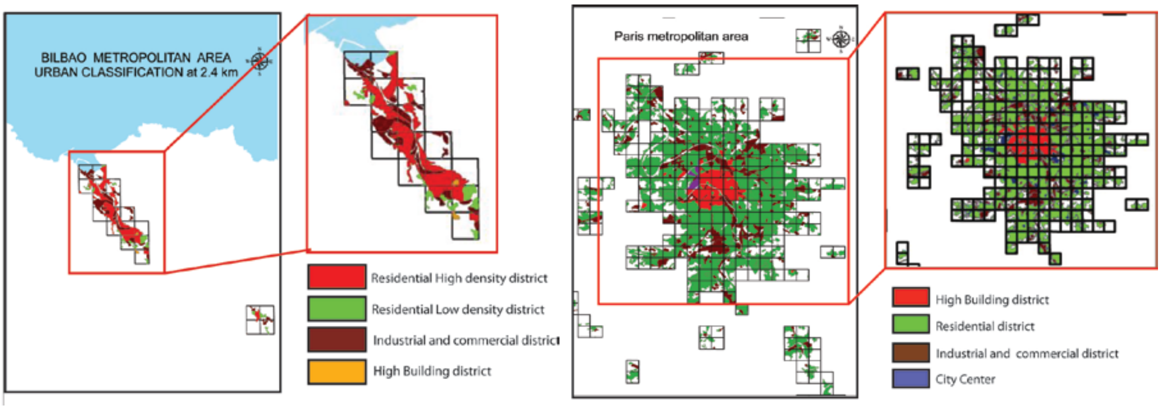


Figure 10: Urban district classification based on urban zoning data for the a) Bilbao and b) Paris metropolitan area, including the residential area (ReD), low and high building districts (LBD and HBD, respectively) and industrial and commercial districts (ICD). Spatial distribution of urban districts (HBD – high buildings, RD – residential, ICD – industrial commercial, and CC – city center districts) for the Paris metropolitan area within the P01 modelling domain (partly adopted from González-Aparicio et al. 2014).

The statistical analysis showed that the urban simulation had a reduced bias with respect to observations than the control simulations. ~~Statistical analysis showed that f~~For Paris, on a monthly basis, the correlations for air temperature were higher ~~(e.g. closer to observations)~~ for the URB compared to CTRL run, and results improved up to 10% on a diurnal cycle (with a maximum of 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) during afternoon and night-time. Moreover, correlations at suburban and urban stations were similar to correlations at rural stations (see Figure 11a). Analysis for Bilbao (González-Aparicio et al. 2013) showed similar performance of the model for 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 (0.4) near the coast (inland) stations.
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 2.2ºC (LHVP) and 0.6ºC (SIRTA). It started at mid-night and expanded covering area of about 2000 km^2. 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 (0.2ºC) 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 1ºC, and heat island expanded covering area of about 130 km^2. In addition, González-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 (González-Aparicio et al., 2014). Enviro-HIRLAM model runs showed that under calm conditions during summer and winter, the UHI could reach up to 2.2ºC covering area of about 400 km^2 when city is doubled in size or doubled in AHF. When city is tripled in size, the UHI could reach up to 3ºC with urban island expansion up to 550 km^2 (Figure 11c). Analysis of UHI for Bilbao (e.g. triple size city scenario) vs. current UHI over Paris showed similar intensity of up to 3ºC, 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.

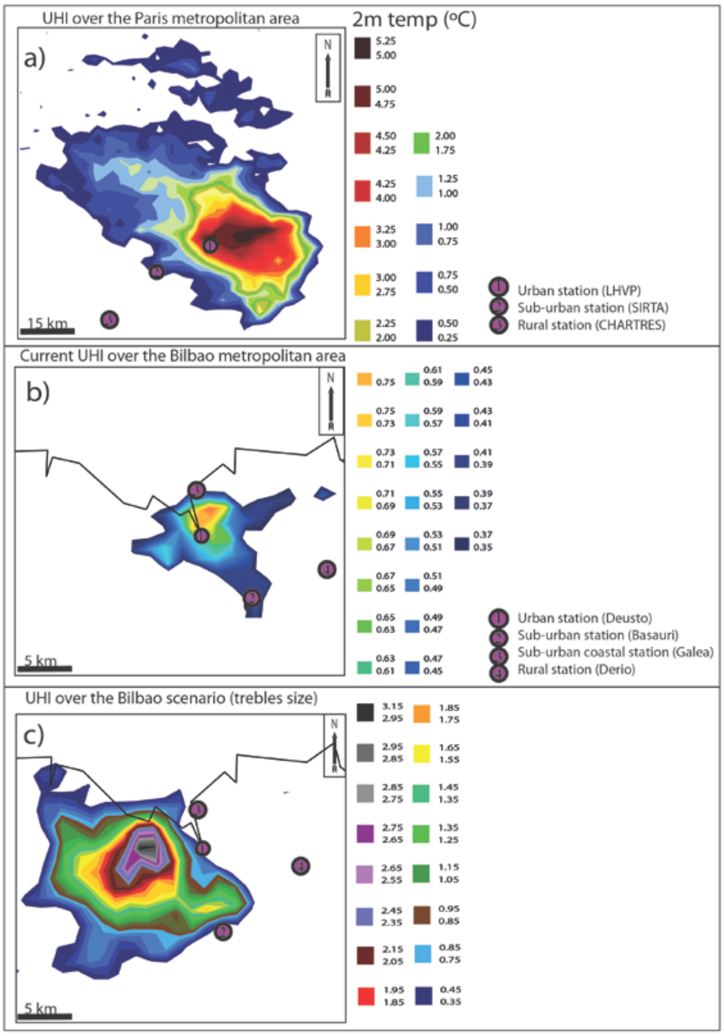


Figure 11: Difference plots for the air temperature at 2m between outputs of the URB (urbanized -BEP + AHF-) and CTRL (non-urbanized) Enviro-HIRLAM model under calm conditions during summer 2009 for the a) Paris metropolitan area and for the Bilbao metropolitan area b) in its current size of the city 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~~ important 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, Switzerland, Austria), CHIMERE (France), Enviro-HIRLAM, DEHM (Denmark) and others. The pollen emissions ~~is~~ are strongly dependent on meteorology, so it is advantageous~~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.

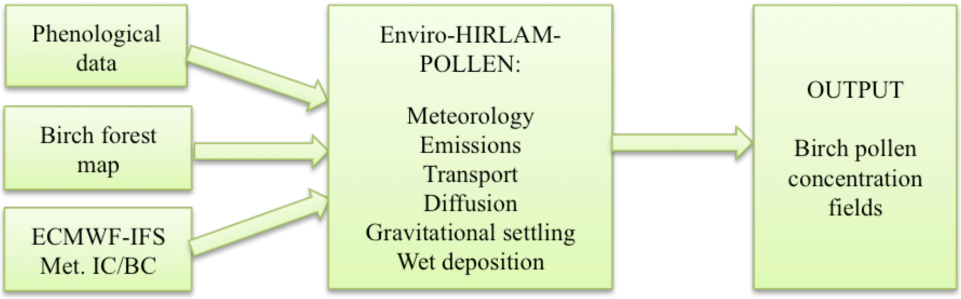
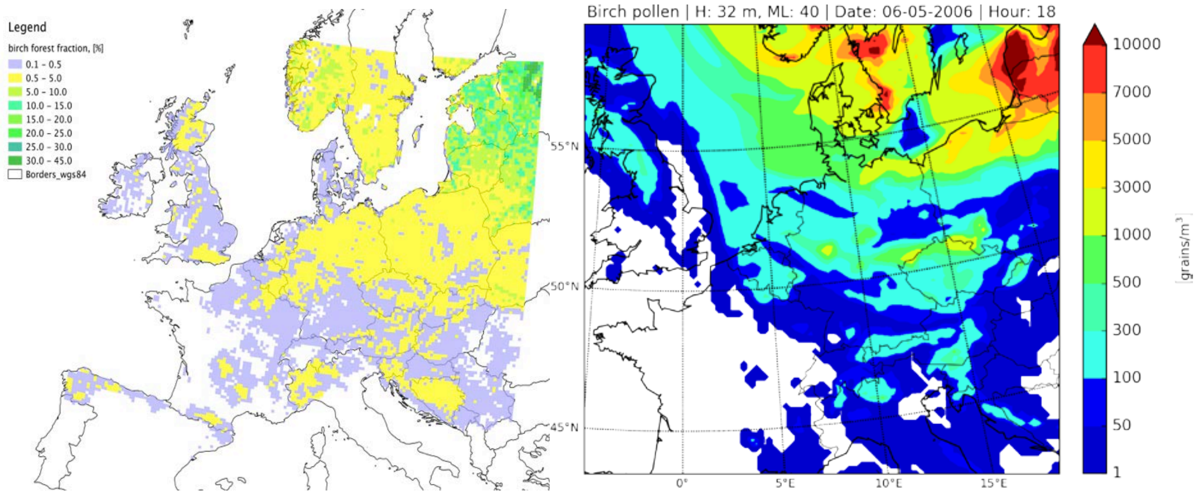


Figure 12: General scheme of Enviro-HIRLAM birch pollen forecasting.

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 emissions are 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. TThe modelingmodelling domain has 15 km horizontal resolution with 154 and 148 grid points along longitude and latitude, correspondeingntly. The domain covers the main European part and is centered around Denmark.

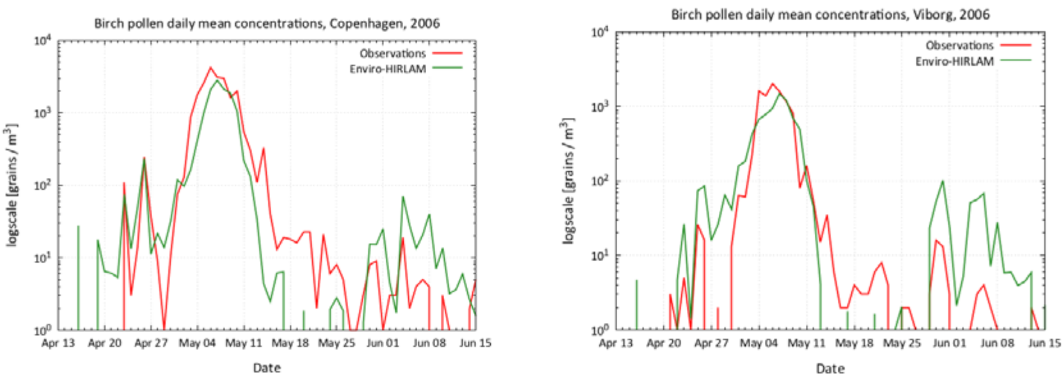


(a) *(b)*

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 modelingmodelling domain. The map (Fig. 13a) was obtained fromshows birch forest fraction in each model grid cell. Three GIS based databases were used in the derivation procedure: 1) Global Land Cover CharacterizationCharacterisation (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 modeledmodelled and observed birch pollen concentrations showed extremely high values (daily averages about and even more than 1000 grains/m^3) 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.



(a) *(b)*

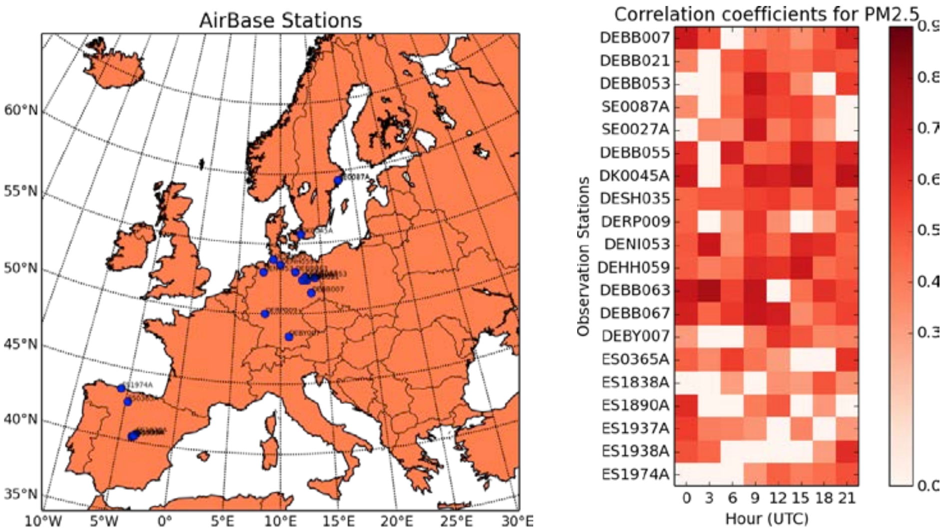
Figure 14: Birch pollen concentrations observed (red) vs. ~~modeled~~modelled (green) at Danish sites: Copenhagen (a) and Viborg (b).

According to Sofiev et al., (2011) and Siljamo et al. (2013) 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~~modelled vs. observed birch pollen concentrations (C) relatively to a threshold value N_{th} = 50 grains/m^3 (i.e. $C \geq N_{th}$ 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, $C \geq N_{th}$ 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 likelihood for getting "high" day concentration instead of a "low" ~~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~~modelled 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~~modelled 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~~online 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., 201~~6~~7).
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~~online 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).

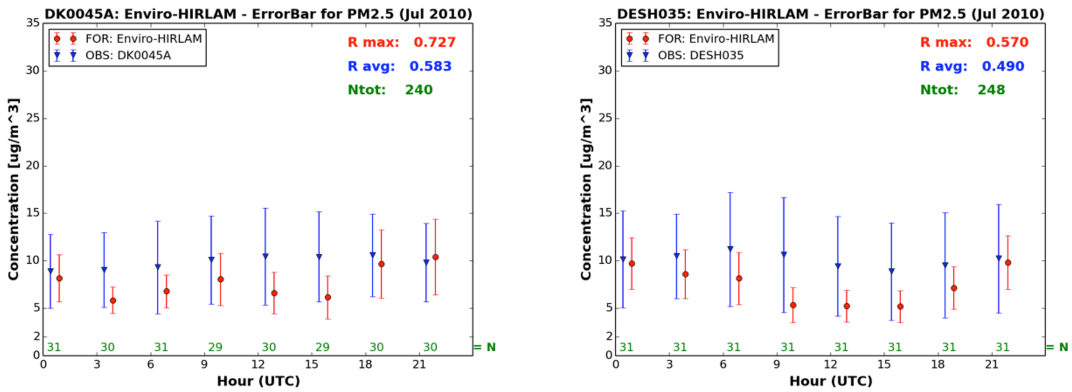
To perform further analysis of online coupling and feedback effects on atmospheric pollution forecasting, the year 2010 was selected (for details see Sect. 2.5). Nuterman et al. (2013) evaluated the Enviro-HIRLAM model for July 2010 vs. ground-based observations of $PM_{2.5}$ from EU AirBase air-quality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



(a) (b)

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.

On the monthly based evaluation t~~T~~he 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) rougher~~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 rough ~~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.



(a) (b)

Figure 16: Error-bar concentrations [ug/m^3] on diurnal cycle for AirBase observations vs. Enviro-HIRLAM ~~modeling~~modelling 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 downsc~~l~~aling from regional-to-urban 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 in a long-term mode is realized in CarboNord with focus on evaluation of black carbon as well as higher 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 f~~Further ~~research and /developments needs~~discussions

The Environment – HIgh Resolution Limited Area Model (Enviro-HIRLAM) is developed as a fully online coupled/integrated numerical weather prediction (NWP) and atmospheric chemical transport (ACT) modelling system (CTM) for research and forecasting of joint meteorological, chemical and biological weather. Possible applications of the modellling system include: chemical weather forecasting (CWF), air quality (AQ) and chemical composition for short- and longer-term assessments on population and environment, weather forecast (e.g., in urban areas, severe weather events, etc.), pollen forecasting, climate change forcing modelling, studies of climate change effects on atmospheric pollution on different scales, weather modification and geoengineering methods, volcano eruptions, dust storms, nuclear explosion consequences, other emergency preparedness modelling. Several types of the above mentioned applications of the Enviro-HIRLAM for meteorological, environmental and climate forecasting and assessment studies are were tested and demonstrated. Different applications of Enviro-HIRLAM (with downscaling from hemispheric - regional – subregional – urban scales) were realised for different geographical regions and countries including European countries such as Denmark, Lithuania, France, Spain, Ukraine, Russia, Turkey and well as for other regions – China and Arctic.

It is clear that the seamless/ online integrated modelling approach realised in Enviro-HIRLAM is a prospective and state-of-the-art way for future single- atmosphere modelling systems, providing advantages for all three communities: Meteorological modelling including NWP, AQ modelling including CWF, and climate modelling. However, there is not necessarily one configuration of the integrated online modelling approach/ system suitable for all communities, and that should be further investigated with practical needs for areas applications, approaches to coupling and computing resources usage. 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 physical-chemical 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 versionsconfigurations 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 heavy polluted episodes and in urban areas (although statistically these effects are not so strong on long-term runs).
- For pollen forecast: iImprove pollen emission parameterization simulation and correspondingly modelling of concentration and depositions. Feedbacks are not important. Chemistry is not considered yet, but interaction with allergens would be interesting to study in future (not done yet).
- For climate studies: suitable only for understanding the feedback mechanisms, it is too expensive for climate runs (Enviro-HIRLAMthe model had been used maximum for 1l year period runs). Chemistry is important, the modelre is a 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 cancelling effects); and (iii) model evaluation for online models needs more (process) data and long-term measurements – and a test-bed. It should also be mentionstressed that Tthe considered evaluations were done only for some elements (e.g., the coupling interval) in the previous analysis and main conclusions about the improvements were doneprovided just for thesem. Other feedback mechanisms, especially for aerosol-cloud interactions, were analysed mostly as sensitivity studies or evaluated for short-term episodes. In particular,
Tthe STRACO cloud scheme contains fairly simplified cloud microphysics (heavily parameterized). Hence, tuning is essential for the overall performance of the model, when it comes to precipitation and cloud physical properties.

5. Conclusions

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 physical-chemical processes and interactions and to create specific model configurations that are tailored for their respective purposes.

So, it could be concluded that the 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 using the Enviro-HIRLAM model demonstrated the importance of including the meteorology and chemistry (especially aerosols) interactions in online-coupled models. However, there is no one unique integrated online modelling system configuration, which is best suitable for all communities, and hence, different configurations should feed for different purposes. In particular, the following could be recommended for the considered applications:

For AQ: online coupling improves 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 heavy polluted episodes and in urban areas.

For pollen forecast: the online coupling improves pollen emission parameterization and correspondingly modelling of concentration and deposition. However, the feedbacks are not so important. Chemistry is not considered yet, but interaction with allergens would be interesting to study in future (not done yet).

For climate studies: it is suitable only for understanding the feedback mechanisms, but too expensive for climate time-scale runs (the model had been used usually for one year period runs). Chemistry is important, there is a need 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 cancelling effects); and (iii) model evaluation for online models needs more (process) data and long-term measurements – and a test-bed.

Code and/or data availability

The Enviro-HIRLAM modelling system is a community model, the source code is available for non-commercial use (research, development, and science education) 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 http://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.)
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)
2003: Aerosol dynamics model developed and tested first as 0D module in offline CAC (A. Baklanov, A. Gross)
2004: Test of different formulations for advection of tracers incl. cloud water (K. Lindberg)
2005: Urbanisation of the model (funded by FP5 FUMAPEX) (A. Baklanov, A. Mahura, C. Peterson)
2005: COGCI grant for PhD study of aerosol feedbacks in Enviro-HIRLAM (U. Korsholm, supervised by A. Baklanov, E. Kaas)
2006: Test of CISL scheme in Enviro-HIRLAM (P. Lauritzen, K. Lindberg)
2007: First version of Enviro-HIRLAM for pollen studies (A. Mahura, U. Korsholm, A. Rasmussen, A. Baklanov)
2008: New economical chemical solver NWP-Chem (A. Gross)
2008: First version of Enviro-HIRLAM with indirect aerosol feedbacks (U. Korsholm PhD)
2008: Testing new advection schemes in Enviro-HIRLAM (UC: E. Kaas, A. Christensen, B. Sørensen, J.R. Nielsen)
2008: Decision to build HIRLAM Chemical Brunch (HCB) with Enviro-HIRLAM as baseline system, Enviro-HIRLAM becomes an international project
2008: 1st International Young Scientist Summer School (YSSS) on "Integrated Modelling of Meteorological and Chemical Transport Processes" (based on Enviro-HIRLAM) in St. Petersburg, Russia: http://netfam.fmi.fi/YSSS08/
2009: Integrated version of Enviro-HIRLAM based on reference version 7.2 and HCB start
2011: New chemistry (A. Zakey), direct and semi-direct aerosols effect (K.P. Nielsen) schemes
2011: 2nd International YSSS (based on Enviro-HIRLAM/HARMONIE) in Odessa, Ukraine: http://www.ysss.osenu.org.ua/
2012: New effective aerosol scheme for multi-compound aerosols (R. Nuterman)
2012: New mass conserving and monotonic semi-Lagrangian transport (B. Sørensen et al. 2013)
2013: New STRACO scheme with aerosol-clouds interaction (U. Korsholm and B. Sass)
2013: Model evaluation study within the AQMEII, phase 2 exercise (R. Nuterman)
2014: Moving to the HARMONIE platform and building a joint strategy with ALADIN community
2014: 3rd International YSSS (based on 5 online coupled models including Enviro-HIRLAM/HARMONIE) in Aveiro, Portugal: http://aveirosummerschool2014.web.ua.pt/
2014-2016: Enviro-HIRLAM birch pollen forecasting system (A. Kurganskiy et al.)
2015: New radiation scheme with aerosol direct and semi-direct effects for SW and LW radiation (K.P. Nielsen et al.)
2016: Application of Enviro-HIRLAM for China (Marco-Polo project, A. Mahura et al.)

Annex 2: Abbreviations and acronyms used in this article:

ACT	Atmospheric chemical transport
AHF	Anthropogenic heat flux
ALADIN	Aire Limitée (pour l') Adaptation dynamique (par un) Développement InterNational (model and consortium)
AOD	Aerosol Optical Depth
AQ	Air Quality
AQMEII	Air Quality Model Evaluation International Initiative
AROME	Application of Research to Operations at Mesoscale-model (Météo-France) ARW. The Advanced Research WRF solver (dynamical core)
BC	Black Carbon
BEP	Building Effect Parameterization
CAC	Chemistry-Aerosol-Cloud model (tropospheric box model)
CarboNord	Nordic project "Impact of Black Carbon on Air Quality and Climate in Northern Europe and Arctic"
CBM-IV	The modified implementation of the Carbon Bond Mechanism version IV
CBM-Z	CBM-Z extends the CBM-IV to include reactive long-lived species and their intermediates, isoprene chemistry, optional DMS chemistry
CHIMERE	A multi-scale CTM for air quality forecasting and simulation
CISL	Cell-integrated semi-Lagrangian (transport scheme)
COGCI	Copenhagen Global Climate Initiative
CORINE	European land-use database
COST	European Cooperation in Science and Technology (http://www.cost.eu/)
COSMO	Consortium for Small-Scale Modelling
COSMO-ART	COSMO + Aerosols and Reactive Trace gases
CPU	Central Processing Unit
CRAICC-PEEX	CRyosphere-Atmosphere Interactions in a Changing Arctic Climate - Pan Eurasian EXperiment
CRUCIAL	Nordic project "Critical steps in understanding land surface atmosphere interactions: from improved knowledge to socioeconomic solutions
CTM	Chemistry-Transport Model
CWF	Chemical Weather Forecasting
DMI	Danish Meteorological Institute
DMS	Dimethyl sulphide
DEHM	Danish Eulerian Hemispheric Model
ECMWF	European Centre of Medium-Range Weather Forecasts
EFI	European Forest Institute
ECHAM5-HAM	Global aerosol–climate model: Global GCM ECHAM (version 5) + Aerosol chemistry and microphysics package HAM (MPI for Meteorology, Hamburg)
ENCWF	European Network on Chemical Weather Forecasting
Enviro-HIRLAM	HIgh Resolution Limited Area Model HIRLAM with chemistry (DMI and collaborators)
EnviroRISKS	EU FP6 project: 'Environmental Risks: Monitoring, Management and Remediation of Man-made Changes in Siberia'
EPA	USA Environmental Protection Agency
ESM	Earth System Modelling
EuMetChem	The COST Action ES1004 – European framework for online integrated air quality and meteorology modelling (eumetchem.info)
ETEX	European Tracer Experiment
FAR	False alarm ratio
FMI	Finnish Meteorological Institute
FP5,6,7	European Union Framework Programs
FUMAPEX	EU FP5 project "Integrated Systems for Forecasting Urban Meteorology, Air Pollution and Population Exposure"

GADS	Global Aerosol Data Set
GAW	Global Atmosphere Watch (WMO Programme)
GEOS-Chem	GEOS–Chem is a global 3-D chemical transport model (CTM) for atmospheric composition driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office
GIS	Geographical Information System
GLCC	Global Land Cover Characterization
HAM	Simplified global primary aerosol mechanism model
HARMONIE	Hirlam Aladin Research on Meso-scale Operational NWP in Europe (model)
HCB	HIRLAM Chemical Branch
HIRLAM	HIgh Resolution Limited Area Model (http://hirlam.org/)
HR	Hit rate
IC/BC	Initial / Boundary Conditions
IFS	Integrated Forecast System (ECMWF)
ILMC	Posteriori iterative locally mass-conserving filter
ISBA	Interaction Soil- Biosphere- Atmosphere land surface scheme
IS4FIRES	Global biomass burning (wildfires) emission inventory developed by FMI
KPP	Kinetic Pre-Processors
LAI	Leaf Area Index
LMCSL	Locally Mass Conserving Semi-Lagrangian scheme
LUCY	Large scale Urban Consumption of energY model
LW	Long-wave radiation
MA	Model accuracy
M7	Modal aerosol model
MACC	Monitoring Atmospheric Composition and Climate (EU project)
MEGAPOLI	EU FP7 project 'Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation' (http://megapoli.info/)
MESO-NH	Non-hydrostatic mesoscale atmospheric model (French research community)
MetM	Meteorological prediction model
MOZART	Model for Ozone And Related Tracers (global CTM)
NetFAM	Nordic Research Network on Fine-scale Atmospheric Modelling
NMVOC	Non-Methane Volatile Organic Compounds
NWP	Numerical Weather Prediction
OC	Organic Carbon
ODE	Ordinary Differential Equation
OR	Odds ratio
PBL	Planetary Boundary Layer
POFD	Probability of false detection
OPAC	Optical Properties of Aerosols and Clouds (software library module)
PEGASOS	EU FP7 project: Pan-European Gas-Aerosol-Climate interaction study (http://pegasos.iceht.forth.gr/)
PM	Particulate Matter in two size bins – 2.5 µm and 10 µm ($PM_{2.5}$ and PM_{10})
RBM	Radical balance method
RACM	Regional Atmospheric Chemistry Mechanism
SL	Semi-Lagrangian scheme
SMHI	Swedish Meteorological and Hydrological Institute
SM2-U	Soil Model for SubMeso - Urbanized version
STRACO	Soft TRAnsition and Condensation (Cloud scheme)
SW	Short Wave radiation
TKE-CBR	Turbulent Kinetic Energy Cuxart, Bougeault and Redelsperger scheme
TNO	the Netherlands Organisation for Applied Scientific Research
TRANSPHORM	EU FP7 project: 'Transport related Air Pollution and Health impacts - Integrated Methodologies for Assessing Particulate Matter'
TSI	Tree Species Inventory
TUV	Tropospheric Ultraviolet-Visible Model
TVA	Tennessee Valley Authority
UBL	Urban Boundary Layer
UHI	Urban Heat Island
YSSS	Young Scientist Summer School
VOC	Volatile Organic Compounds
WHO	World Health Organization
WMO	World Meteorological Organization

Reply to the Topical Editor:

Dear Dr. Jason Williams,

Thanks a lot for your careful and very useful comments. We have revised the MS correspondingly. See some replies below in your text.

I have read the three independent referee reviews of your manuscript and, although there have been significant revisions, the current format does not adhere to the guidelines provided for authors. Please look at the instructions provided and follow the manuscript composition guidelines. To address these issues a revision is required in order to allow the manuscript to be published. In that the new document contains no line numbers makes commenting difficult. Please submit your revised manuscript with line numbers.

Sorry, it is done now.

Abstract: The abstract needs to be re-written removing redundant information concerning the history of the model development. Please only include details related to version 7.2 i.e. what has been updated in this version compared to that in previous publications. Also summarise the improvements compared to previous versions using the examples which are included in Sect.3. This should agree somewhat with the conclusions section.

The abstract is rewritten correspondingly.

Section 1 should be simply entitled "Introduction". At the end of introduction there should be a paragraph to lead the readers into the manuscript. Please refer to example papers previously published in GMD.

The Section 1 is rewritten correspondingly.

Section 3: Remove the bullet points and convert details into a paragraph.

Done.

Section 4 needs to be split into (4) Further Discussion and (5) Conclusions. Remove the bullet points and convey details in normal text.

Done.

The website https://hirlam.org.trac/wiki is currently available when I tried. Please replace the reference with something in the literature which is readily accessible. Refer to the author guidelines regarding the use of webpages in publications.

The current one is: http://hirlam.org/index.php/projects/chemistry-branch. It is corrected.

Specific comments: Please pay attention to adopting correct English grammer in order to avoid extensive modifications which will be imposed before going to print.
"First attempts of … " → "First attempts at …"
"For climate applications the …"

Thanks a lot. Corrected.

Authors response to comments of the Referee #1 Prof. N. Moussiopoulos

We thank the Referee #1 Prof. N. Moussiopoulos for the interesting and important comments on our manuscript. All the individual comments are addressed below in red.

This manuscript provides a thorough presentation of Enviro-HIRLAM representing one of the first serious development efforts towards implementing a fully online coupled meteorological and chemical weather model. It contains detailed descriptions of methodology selected and implementation followed, including some coverage of less welldefined aspects of online coupling and performance evaluation. The paper is well written and contains a large amount of information. A section on model applications provides additional insight on the extremely important aspects of evaluation and validation.
As the overall assessment of the present referee, the paper successfully describes the remarkable effort that has been devoted to the development of a state-of-the-art online meteorological and chemical weather model. It is adequately referenced and contains detailed explanations of the main physical mechanisms and selected parameterisations. It also highlights some of the more promising aspects of the coupling idea, both in the area of aerosol-radiation treatment and in cloud microphysics.
The only weak point in the manuscript is the rather sketchy discussion of the extent to which the explicit introduction of all effects will lead to improvements in model performance.

Response:
The paper is focusing mostly on the model description and its applications, therefore it was not much space for detailed discussion of specific effects of different model improvements. Some aspects were published in previous papers, some are still in new specific papers to be submitted (e.g. for pollen applications, operational air quality forecasting).
We have extended this part in the revised version.

Section 4 of the manuscript represents of course an honest attempt to summarise what we know on the effect of coupling in model performance for different applications. The authors are encouraged to provide more explicit comments in this respect. This should be combined with a more thorough discussion on how all parameters required in the various process parameterisations could be fine-tuned (for instance, expanding the comments made in the last four lines of the paper).

Response: The Section 4 is extended correspondingly in the revised version.

In the below listed specific comments references are made to specific lines in the text.

1. Methodology and modelling system structure

a. The model coupling implements aerosol impacts on radiation (direct and semidirect effects) and on clouds (first and second indirect effects), l. 110-111. It appears appropriate to include an explicit reference to COST action ES1004 in the framework of which these effects were extensively discussed.

Response: Thanks, agree. The corresponding reference is included.

b. The cloud feedback module includes some rather advanced approximations (l. 287-293); the reader would welcome more remarks on the extent to which this complex cloud model has been validated.

Response:

Abdul-Razzak and Ghan parameterization for aerosol activation has been extensively tested in many online-coupled weather and climate models. However, the STRACO cloud microphysics scheme with parameterizations of aerosol activation, cloud droplets nucleation, sedimentation, evaporation, self-collection, has never been thoroughly evaluated, only with 1D column HIRLAM. These evaluation results are not ready to be published yet, but will be analysed and published on further steps.

c. The present HIRLAM NWP model core is based on the hydrostatic approximation (l. 127) which could be a serious limitation over complex terrain (l. 508) and/or in cases of nesting down to urban areas. A plan for a transition to a new, non-hydrostatic platform (e.g. HARMONIE, l. 135) is mentioned, but more information in this respect would be helpful.

Response:

Yes, we agree about the limitations and write openly about them. The new version under HARMONIE is only under development and only some elements are realised so far, so it is too early to describe it extensively it in more details at this stage.

The non-hydrostatic HARMONIE-AROME model includes only some aerosol effects. The physics included in this version of HARMONIE has recently been detailed by Bengtsson et al. (2017). HARMONIE-AROME is based partly on Meso-NH (Mesoscale Non-Hydrostatic atmospheric model), which is a cloud resolving model that includes state-of-the-art chemistry and aerosol interactions (e.g. Berger et al. 2016). Meso-NH can, however, not be run as a near real time NWP model, which is possible with Enviro-HIRLAM.

Corresponding extension text is included in the revised version.

The following additional references are included:

Bengtsson, L., U. Andrae, T. Aspelien, Y. Batrak, J. Calvo, W. de Rooy, E. Gleeson, B. Hansen-Sass, M. Homleid, M. Hortal, K. Ivarsson, G. Lenderink, S. Niemelä, K. P. Nielsen, J. Onvlee, L. Rontu, P. Samuelsson, D. Santos Muñoz, A. Subias, S. Tijm, V. Toll, X. Yang, and M. Ødegaard Køltzow, 2017: The HARMONIE-AROME model configuration in the ALADIN-HIRLAM NWP system. Mon. Wea. Rev. doi:10.1175/MWR-D-16-0417.1, in press.

Berger A., M. Leriche, L. Deguillaume, C. Mari, P. Tulet, D. Gazen and J. Escobar, 2016: Modeling Formation of SOA from Cloud Chemistry with the Meso-NH Model: Sensitivity Studies of Cloud Events Formed at the Puy de Dôme Station. In: Steyn D., Chaumerliac N. (eds) Air Pollution Modeling and its Application XXIV. Springer Proceedings in Complexity. Springer, Cham

d. The atmospheric chemistry modules implement a wide array of new parameterisations and numerical schemes (page 4, l. 141-192). Although these were obviously validated separately, their combined implementation in a coupled model definitely needs further validation. Did the authors take already actions in this direction, and if not, what are their plans?

Response:
Yes, these chemical schemes/solvers were tested and validated as standalone versions (Reference:_Shalaby, A., Zakey, A. S., F., Giorgi, and M.M. AbdelWahab "Coupling of Regional Climate Chem Aerosol Model", Ph.D. thesis, Faculty of Science, Cairo University-Egypt, 2012). Six environmental/smog chamber experiments were used to validate the gas-phase schemes and different chemical solvers as box models.
The Tennessee Valley Authority (TVA) and the EPA chamber experiments were used to evaluate the different gas-phase schemes and different chemical solvers. Namely, TVA005 and TVA006 are designed to test the simple system of NOx; TVA068 is designed to test a simple mixture of VOC with very high NOx. EPA069A, EPA073A and EPA150A are used to validate the schemes with low NOx concentration and high VOC concentration.
Also, the same chemistry schemes/solvers are coupled with the Regional Climate Model (Reference: Shalaby, A., Zakey, A. S., Tawfik, A. B., Solmon, F., Giorgi, F., Stordal, F., Sillman, S., Zaveri, R. A., and Steiner, A. L.: Implementation and evaluation of online gas-phase chemistry within a regional climate model (RegCM-CHEM4), Geosci. Model Dev., 5, 741-760, doi:10.5194/gmd-5-741-2012, 2012.)
However such validations need to be further continued and completed, this is the issue for further analysis.
Corresponding extension text and the above mentioned additional references are included in the revised version.

e. The aerosol dynamics model introduces a very interesting classification of particles depending both on particle size and particle composition per emission source. This could allow, in theory, a separate per-source type treatment of particles throughout the chemical mechanism. But is there such a procedure (with potential applications in source apportionment) really implemented or planned?

Response:
The particles classification with respect to their size and composition is based on aerosols classification in M7 microphysics module. As for total particulate matter the splitting is in species, the procedure follows guidelines and recommendations associated with different emission inventories. For now, there is no procedure for source apportionment in the model or plans to do that in the nearest future.

f. Specific emission models for anthropogenic biomass burning (e.g. wildfires) are included (section 2.5). These are based on satellite or other inventory-estimates of yearly fluxes that are temporally disaggregated using pre-defined temporal profiles. Are the latter site dependent, and which is the origin of the coefficients used by the authors?

Response:
The Finish Meteorological Institute developed the IS4FIRES (http://is4fires.fmi.fi) wildfires emission inventory. The IS4FIRES inventory provides temporal profiles for emissions disaggregation with site dependency. However, the profiles used in Enviro-HIRLAM runs for this paper are site independent (mean) and are the functions of a local time only. They were also provided by FMI for AQMEII-2 (COST Action ES1004) initiative.
Changes in manuscript:
L257: The biomass burning emissions typically show a diurnal cycle variability, and therefore, corresponding coefficients are applied (*Giglio, 2007*).
Added to reference list: Giglio, L., 2007: Characterization of the tropical diurnal fire cycle using VIRS and MODIS observations, Remote Sensing of Environment, 108, 4, pp. 407-421.

g. The model contains several "urbanisation" features (section 2.7), including a subset of previously proposed urban parameterisations (Martilli, Dupont, Masson, Grimmond et al.). This is an interesting and original approach, but there are several concerns on how it is implemented (please see comment 2c below).

Response:
The details of implementations of different urban modules, our own developments and comparisons of different approaches and modules were published in previous papers (Mahura et al., 2005b, 2006a, 2007a, 2008bc;

Baklanov et al., 2005, 2008), so we don't describe them in this paper. The main approach includes an integration of the urban modules into the ISBA (Interaction Soil- Biosphere- Atmosphere) land surface scheme of the NWP / HIRLAM model. The urban modules are activated only on those grid cells of the model domain where the urban fraction is presented.
More explanations and corresponding references on the above papers are included in the revised version.

h. In l. 372-378 some aspects of the so-called locally mass conserving semi-Lagrangian (LMCSL) transport scheme are described. The description emphasizes the approximate mass-conserving properties of the algorithm for 1st neighbour cells, but one could ask whether and how is mass consistency ensured in the larger scale. In l. 389-390 it is stated "[: : :]Enviro-HIRLAM is not formally wind-mass consistent regarding tracer transport". The authors should discuss possible consequences of this failure.

Response:
We have added a sentence to clarify that mass-wind inconsistency is a minor problem. The traditional HIRLAM is (at least in principle) wind-mass consistent. In Enviro-HIRLAM where all moisture fields are transported with the LMCSL scheme, there is no formal consistency, yet, since precipitation is very similar to that in HIRLAM (except for individual convective systems that are chaotic/unpredictable in their nature), the mass-wind inconsistency is small in practice.

Suggested new version of lines 372-378 in the original text (changes in bold)
As the traditional SL scheme, the LMCSL is not **inherently** monotonic or positive definite. Therefore a posteriori iterative locally mass-conserving (ILMC) filter was developed, Sørensen et al. (2013).
……
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 (4), i.e. not the LMCSL. Therefore, the Enviro-HIRLAM is not formally wind-mass consistent regarding tracer transport. **However, the large scale precipitation fields in the traditional HIRLAM and Enviro-HIRLAM are very similar (see, e.g., Figure 4 in Sørensen et al. (2013)), which suggests that wind-mass inconsistency is of minor importance.**

2. Model applications and validation
a. Sensitivity studies on the model response to aerosol effects do indicate some strong "signals" (difference between coupled and uncoupled runs), e.g. l. 418. But these do not necessarily imply an improved model performance, and the authors should state this clearly in the manuscript, cf. l. 420 "[Korsholm (2009)] found a marginally improved agreement[: : :]", and l. 464-467 "However: : :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 : : : and due to adjustments of such effects: : :and constants".

Response:
Yes, we agree with the reviewer but don't see contradictions between these statements. Sensitivity studies on the model response to aerosol effects indicate strong "signals", but it doesn't guaranty improvements. E.g., Korsholm (2009) considered evaluations only for some elements (e.g., the coupling interval) in the previous analysis and made corresponding conclusions about the improvements. Other feedback mechanisms effects, especially for aerosol-cloud interactions, studied mostly as sensitivity studies or evaluations for short-term case studies.
The model formulations have only been tested on a case basis and although strong signals have been found this does not imply improved meteorological performance of the model. In particular, testing over longer periods including all seasons was not conducted that time. Furthermore, the interactions between aerosols and the cloud ice-phase are not in a state where improvements would be expected. Therefore we wrote that conclusions about the improvement of precipitation cannot be done at this stage and need more analysis.
Recently similar evaluation studies are realised within the CarboNord project for monthly and annual validation studies. However, they are recently started.

b. This referee believes that careful tuning is needed in view of the large number of parameters in the complex feedback modules, especially with regard to cloud effects. It is not obvious how and to what extent this could be achieved only by comparing final simulation results (i.e., without a further quantitative study of the cloud physical mechanisms themselves).

Response:
We fully agree with the reviewer. The STRACO cloud scheme contains fairly simplified cloud microphysics (heavily parameterized). Hence, tuning is essential for the overall performance of the model, when it comes to precipitation and cloud physical properties. Further work to improve aerosol-cloud interaction and precipitation forecast is needed.

c. An evaluation application for the urbanisation modules was performed for the cities of Paris and Bilbao. There are several issues regarding this application that are neither explained in the text nor in the referenced publications:
i. A domain spatial resolution of 2.5 km appears to be insufficient for such an application.

Response:
Sensitivity tests demonstrated that the 2.5 km was the optimal resolution allowing at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at local scale without being diminished due to a coarse resolution, taking into account the limitations of the hydrostaticity of the NWP model.

ii. The resolution of the BEP dense sub-grid is not mentioned. Is it also 2.5 km?

Response: Yes, the BEP is also computed at 2.5 km resolution.

iii. The authors seem having assumed only four urban classes, cf. Figure 10. Such a classification would ignore the important role of green urban areas in UHI evolution.

Response:
Although we assumed four types of urban areas, the urban grid is not fully covered by urbanisation; it also contains a fraction of the green area, defined in the CORINE 2000. The classification and the percentage of urban/non-urban grid can be found in Gonzalez-Aparicio et al. (2010), page 17 Table 4.

iv. Is the 2.52.5 arc-minute resolution (5km) of the AHF data adequate for assessing UHI effects in an urban scale? In the Bilbao case it appears that the entire urban area is covered (and classified) in only 16 cells!
v. Are AHF data constant during the day, or do the authors assume an intrinsic diurnal profile?
vi. Values of 40 or 60 Wm-2 for the AHF are mentioned. Is this a mean annual value or a daily estimation following a seasonal profile?

Response (for iv-vi):
For the Bilbao study, Enviro-HIRLAM didn't implement any AHF parameterization and therefore, the AHF factors were estimated from the LUCY model, as a value for summer and winter without including any daily profile. The value was constant for the urban fraction in the 16 grid cells (it is multiplied – e.g. depends on urban fraction: if 100% -> then max value) covering the area of Bilbao (92 km^2). Although it is not as big as the Paris metropolitan area, the effects of the AHF and the UHI on the atmospheric boundary layer could be visible. A sensitivity analysis of the effects of the AHF and the UHI on the atmospheric boundary layer can be found in Gonzalez-Aparicio et al. (2014).

vii. Concerning the validation process, it is unclear whether a combination of statistical indicators is used or just the correlation coefficient. Not much evidence is presented (e.g. in form of figures or tables) that the model reproduces satisfactorily the mesoscale features.

Response:
The full validation process can be found in Gonzalez-Aparicio et al. (2013) and Gonzalez-Aparicio et al. (2014). The text summarised the overall performance over the two episodes analysed.

viii.It is well documented in the literature that the Paris UHI is expanding just after midnight, but not that this expansion lasts until 11 UTC, especially during a summer period. Comments by the authors would be welcome.

Response:
We agree that the Paris UHI is generally expanding just after the midnight and this is very well documented. In this paper we present the evolution of the UHI on the single day of the 28th July 2009. The UHI was expanding after midnight and the effect was visible up to 11 UTC, not meaning that the expansion lasted until 11 UTC.

ix. Confusion is caused by the fact that in the second paragraph of section 3.2 the authors claim that the model was applied for July 2009, while in the last paragraph of the same section they write ": : :showed that under calm conditions during summer and winter: : :".

Response:
The analysis described in the second and third paragraph of the section refers to July 2009, as the text describes. The last paragraphs indicate the outcomes presented in Gonzalez-Aparicio et al. (2013) and Gonzalez-Aparicio et al. (2014) and focused on winter and summer episodes.

d. Enviro-HIRLAM is operationally used for birch pollen forecasting in Denmark. This appears to be one of the more mature applications of the model, with rather advanced emission, deposition and scavenging modules. However, no mention is made on the effect of online coupling (and the relevant feedbacks) on these simulations. In the conclusions it is mentioned that feedbacks are not important in pollen forecast (l. 711-712). How did the authors reach this conclusion?

Response:
The current version of the birch pollen model presented in the paper has not been used in operational mode yet. Online coupling is important for birch pollen simulations due to dependency of the birch pollen emissions on meteorology. It is also specified in lines 550-552 of the paper.
The current version of Enviro-HIRLAM considers birch pollen as a passive tracer with no pollen feedbacks on meteorology. Online coupling (i.e. impact of meteorology on the emissions) is of main importance in the birch pollen study.

e. Section 3.4 attempts an evaluation of the feedback effects on air pollution forecasting. It is mentioned that online coupling improves the forecast skill, however without referring to specific applications, as for instance the MEGAPOLI Paris campaign.

Response:
As we mentioned, the considered evaluations were done only for some elements (e.g., the coupling interval) in the previous analysis and main conclusions about the improvements were done just for them. Other feedback mechanisms effects, especially for aerosol-cloud interactions, analysed mostly as sensitivity studies or evaluations for short-term episodes. Unfortunately, during the MEGAPOLI Paris measurement campaign we were not able to include measurement studies of aerosol-cloud interactions, so it was not possible to make evaluations of aerosol feedbacks vs the MEGAPOLI Paris data. So, we wrote only about a general reasonable performance of the model vs. measurement data.
Corresponding corrections and explanations are made in the revised version of the paper.

From a technical point of view, the paper is excellent. Yet, the authors should check it again for inconsistencies (e.g., both "online" and "on-line" are found in the manuscript).

Response: Thanks a lot. It is corrected in the revised version.

Authors response to comments of the Referee #2

We thank the Anonymous Referee #2 for the interesting and important comments on our manuscript. All the individual comments are addressed below in red.

General comments:
It would appear that the primary objectives of the presented manuscript were to introduce, document and promote a 'fit-for-purpose' application of the Enviro-HILRAM model.
The Eviro-HILRAM model is well established in the community. It is being used and developed trough a broad international collaboration. It is important that a proper reference to this valuable tool is provided. The Authors made an effort to present the origin and evolution of the model over the years. Also, a short description of model components and applications was provided. Specific comments and suggestions are given in the next section.
In the manuscript, the Authors advance terms and concepts of "online coupling", "fully online integration", "seamless meteorology-chemistry modelling", "two-way interacting", "on-line integration". The use of these terms is not consistent and confusing.

Response: Thanks. The terminology is harmonised/corrected in the revised version.

Also, the concept of a meteorological/NWP model with chemistry was proposed, implemented and published earlier than the provided reference to Grell at al. (2005).
Coupled chemistry-climate models were developed and used in the 1990s, cf. Steil et al., 2003 (doi:10.1029/2002JD002971), Austin and Butchart, 2003 (Q. J. R. Meteorol. Soc., 129, 3225–3249), de Grandpré et al., 2000 (J. Geophys. Res, 105, 26,475–26,492), among other publications. Thus, a proper historical and scientific perspective should be preserved, especially in a paper that presents "strategy and methodology" and dedicates several paragraphs to model evolution and origin.

Response:
Thank you for the comments. The references are included in the revised version. However, more comprehensive historical overviews of coupled chemistry-meteorology models were done e.g. by Zhang (2008), Kukkonen et al. (2011), Baklanov et al. (2014).

The Authors introduced the term "biological weather". It is the understanding of the reviewer that this term refers to birch pollen modelling. However, the meaning of the term is unclear and probably misleading.

Response:
The "biological weather" term is defined in Klein et al. 2012 as "the short-term state and variation of concentrations of bioaerosols".
Thus in the current paper, biological weather refers to birch pollen modelling.
The reference to Klein et al. 2012 is included for clarification.

It is not evident, from the presented model description that it is a multiscale or a wideband atmospheric model. In most of the presented examples, the model domain covers the European continent. Application of the model to urban scale with a resolution of 2.5 km in a hydrostatic mode is rather problematic. The Authors should further comment and justify its use at the said resolution (cf. Lines 508-509).

Response:
Yes, the hydrostatic approximation of the model was a limitation to increase the resolution to perform the urban simulations. However, sensitivity tests demonstrated that the 2.5 km was the optimal resolution allowing at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at local scale without being diminished due to a coarse resolution, for a medium size city (even possibly can be considered for a small size city). For other metropolitan areas such as Paris, Rotterdam, St. Petersburg, Shanghai - a similar resolution was chosen, although for Copenhagen (with its flat terrain) the highest possible/ suitable resolution tested was 1.5 km and provided reasonable verification results. Within a selected metropolitan area there could be only a few grid cells having 100% representation of the urban fraction, but taking into account all urban grid cells, the boundaries of the cities (number of cells) could be substantially larger. Moreover, it should

be noted that most of existing developed parameterizations in the physics core of any existing NWP model might need a revision when resolutions of 1 km and finer are used.

The Authors provided references to all model components and applications. However, this paper should explicitly provide all 'vital model information' such as vertical structure, horizontal resolutions (with clearly stated limitations), numerical methods and approximations employed in different modules (components), modularity and scalability of components, examples of integration time and computer topology used for benchmarking.

Response:
Vertical structure and horizontal resolutions of the model are flexible. Limitations, e.g. due to the hydrostatic approximation, are provided (min 1,5 km for flat terrains, e.g for Copenhagen). Corresponding information, as requested, is included in the revised version.

What is the required computer power, maximum number of computational cores, can the model be run on a heterogeneous architecture with GPUs? All these characteristics should be addressed and tabulated with appropriate references and notes.

Response: The model is parallelized with both OpenMP and MPI technics, but it cannot be run on heterogeneous architectures with GPUs. The parallelization algorithm performs 2D decomposition of a modeling domain. The Enviro-HIRLAM can be run on Linux/Unix clusters and CRAY XT5/XC30 high performance computers.
We have not heard of tests where effect on scalability of introducing chemistry, aerosols etc. have been made.
Changes in manuscript:
L427: The Enviro-HIRLAM modelling domain with horizontal resolution of 0.15^o x 0.15^o having 310 x 310 grid cells, and 40 vertical hybrid sigma levels extending to pressures less than 10 hPa, covers Europe, North of Sahara, and European Russia. The modeling domain was partitioned into 120 CPU cores and the model was run with time step of 300 seconds.

In several sub-sections, the Authors included a description of earlier versions of the model. Thus, it is not clear to the reader which parameterizations are used in the current version of the Enviro-HIRLAM model. It would be advisable to move these paragraphs to an appendix presenting development stages and perspective of the Enviro-HIRLAM model.

Response:
More concrete info about parameterizations used in the considered case studies and in the current version of the Enviro-HIRLAM model is provided in the revised version.

In Section 3 (Modelling system applications), the Authors refer to several earlier publications. It is not clear if the presented manuscript contains any results that were not published. It would be advisable to add a table (in Section 3) with a list of presented experiments and model versions used for simulations together with appropriate references.

Response:
Most of results presented in the paper are new (used only in technical reports). We include more accurate references to appropriate papers, if some experiments were considered in previous publications, in the revised version. However, it is difficult to provide such information in a table form.

Also, if a figure is adopted from an earlier publication, a proper reference should be included in a figure caption.

Response: Thanks, checked and done.

Pollen module description should be moved from Section 3.3 (Pollen forecast) to Section 2 (system description).

Response:

Pollen applications require specific parameterisations of pollen emission sources and other characteristics, so it is more relevant to describe in the section 3.3.

Sub-section 3.4 should be moved and inserted as 3.1

Response:
Section 3.1 focuses on the effect of weather while 3.4 is about air-quality forecasting. Although these are two distinct subjects which seem reasonable to address individually.

Overall, the justification of advantages of the on-line approach is not sufficiently demonstrated.

Response:
The advantages of the on-line approach were discussed in details in the previous EuMetChem paper (Baklanov et al., 2014).

Verification aspects should be included in a more coherent way. Presented experiments refer to relatively short periods (one summer month). Results for the gas phase chemistry are not discussed.

Response:
Yes, we agree that many additional verification and sensitivity experiments are needed for different applications (long-term validation, chemistry mechanisms, etc.). We are working with some of them and they will be in following papers.

The Authors should restructure the manuscript to emphasise the overall modelling philosophy and future directions of the proposed model development and applications.

Response:
Thanks. We modified the concluding sections correspondingly in the revised version. However, the overall modelling philosophy and future directions of coupled meteorology-chemistry model development were subjects of our previous papers of EuMetChem, CCMM, etc. (see corresponding references in the paper). Here we focus on the Enviro-HIRLAM model description and its applications.
However, we'd prefer do not change the papers structure dramatically, especially keeping in mind that two other reviewers have found that "The manuscript is well structured and provides a comprehensive presentation of Enviro-HIRLAM development ….".

Specific comments:
The presented comments are in a sequential order and refer to the line numbering in the presented manuscript.
L22: "Online integrated passive pollutant transport" - the same term should not be used for the simplified approach.

Response: Thanks, agree. We mean the online consideration of tracer equations together with other equations at the same time steps (without feedbacks). We modified the sentence.

L27: What is "effective chemistry"?

Response: Thanks. Changed to 'cost-efficient'.

L35-36: The section title is too long and awkward.

Response: Thanks. The title is shortened.

L68: The style of Figure 1 does not conform to a convention used in scientific publications.

Response:
Yes, it might be not the standard/ most common way of the material presentation, but the Figure 1 presents the overall structure of the modelling system, its research development, technical realisation, science education and

potential application areas. All these elements are necessary main building blocks in elaboration and maintenance of the modelling system and it is important/useful to present them in such a graphical form.

L108: “current new version” – should be either “current” or “new”?

Response: Done.

L128: “main meteorological fields” – please define.

Response: It is specified in the text.

L142: How long are the “long-term runs”. Please explain and justify.

Response: Done: up to one year.

L175-185: The whole section on photolysis rates is confusing and misleading.

Response:
For the simplicity of photolysis rates estimation we used the following:

1. For the simple reactions, we estimated the Photolysis rates as a function of number of parameters such as meteorological and chemical inputs including altitude, solar zenith angle, overhead column densities for O3, SO2 and NO2, surface albedo, aerosol optical depth, aerosol single scattering albedo, cloud optical depth and cloud altitude.
2. For the complex reactions, we estimated the Photolysis rates as lookup table using the Tropospheric Ultraviolet-Visible Model (TUV) developed by Madronich and Flocke (1999) and a pseudo-spherical discrete ordinates method (Stamnes et al., 1988) with 8 streams. We run TUV offline and calculated a lookup table of the Photolysis rates, and then we implemented this lookup table under different weather conditions inside our model.

L177: Please explain how the ozone column is set above the model top.

Response:
We used the climatological chemical boundary conditions from MOZART chemical transport model using a monthly average of years 2000–2007 (Horowitz et al., 2003; Emmons et al., 2010). The model top (50 hPa, corresponding to the lower stratosphere) uses a climatological ozone concentration based on interpolated MOZART ozone fields.
Therefore, the model top layer contains ozone concentrations comparable to the stratosphere. Indeed, we implement the climatological values for computational efficiency during model development and test simulations.

L181: The assertion that the 8-stream method is “the most accurate” should be justified.

Response:
The 8-stream method is used and justified in TUV model system, developed by Madronich and Flocke (1999):
Reference: “Madronich, S. and Flocke, S.: The role of solar radiation in atmospheric chemistry; in: Handbook of Environmental Chemistry, edited by: Boule, P., Springer-Verlag, New York, 1–26, 1999”

L282: In Figure 4 X-axes have different units.

Response:
Both the left hand plot and the right hand plot in Fig. 4 have x-axes, showing the electromagnetic wavelength. Since the left hand plot shows SW wavelengths, these are given in units of nm, while the LW wavelengths in the right hand plot have units of µm. It is common practice to use these units for SW and LW wavelengths, respectively.

L343: What is “traditional” SL? Please provide a reference.

Response: Thanks. We provided the reference to the “traditional semi-Lagrangian” scheme:

Robert, A. 1981. A stable numerical integration scheme for the primitive meteorological equations. Atmos.-Ocean 19, 35–46.

L382: Figure 6: The presented figure alone does not prove that the model can deal with sharp gradients.

Response:
Detailed model tests of the ability of ILMC to reproduce sharp gradients are described in Sørensen et al. (2013), in particular Figure 3 and the accompanying discussion in that paper.
The text in the revised version is corrected to avoid confusions.

Line 389: What is TR4?

Response: Thanks. It is a mistyping. TR4 should be Eq. (4). Corrected.

Line 390: The mental jump referring to "formal conservation" should be explained.

Response:
We have already answered this question to Reviewer 1.
We have added a sentence to clarify that mass-wind inconsistency is a minor problem. The traditional HIRLAM is (at least in principle) wind-mass consistent. In Enviro-HIRLAM, where all moisture fields are transported with the LMCSL scheme, there is no formal consistency, yet, since precipitation is very similar to that in HIRLAM (except for individual convective systems that are chaotic/unpredictable in their nature), the mass-wind inconsistency is small in practice.
A more careful discussion on the issue of mass-wind inconsistence in atmospheric models would require a rather extensive addition. In principle, no monotonic transport schemes can be mass-wind consistent, since the monotonic limiters formally destroy the consistency.
We also add a reference to the paper: Jöckel, P., von Kuhlmann, R., Lawrence, M. G., Steil, B., Brenninkmeijer, C. A. M., Crutzen, P. J., Rasch, P. J., and Eaton, B.: On a fundamental problem in implementing flux-form advection schemes for tracer transport in 3-dimensional general circulation and chemistry transport models, Q. J. R. Meteorol. Soc., 127, 1035–1052, 2001.

L407: The title is confusing, and the whole section is too long. Half of the first paragraph refers to urban applications, which are discussed in the next section.

Response: In the revised version we modified the title to 'Applications for Numerical Weather Prediction' and slightly shortened the text.

L497: It is wrong to assert that higher correlation implies that the model is "closer to observations."

Response: We modified the text; the statistical analysis showed that the urban simulation had a reduced bias with respect to observations than the control simulations.

L505: The ability of a weather prediction model (i.e. HIRLAM) to reproduce meso-scale processes at the regional scale should not depend on the use of an urban parameterization. The presented conclusions do not belong in Section 3.2.

Response:
Yes, the ability of a weather prediction model to reproduce meso-scale processes does not depend on the use of an urban parameterization. However, since the hydrostaticity of the model was a limitation for increasing the resolution to study the urban impacts, several sensitivity tests demonstrated that the 2.5 km was the optimal resolution allowing at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at local scale without being diminished due to a coarse resolution (as fraction of urban areas in grid cells of coarser resolution became very diluted).

L654: The calculations were analysed for one month (July 2010) only. Thus, the sentence is too general.

Response: Thanks. The sentence is changed in the revised version.

L656: “crude model resolution” – what does it mean?
The use of the English language:
The Authors should pay particular attention to the use of articles, prepositions and tenses in the revised manuscript. Also, the Authors used words that do not exist i.e. Line 255: ’to split’ is an irregular verb – the simple past tense is ‘split’, or words in a wrong context i.e. Line 187 ‘Heterogenic chemistry’ should be ‘Heterogeneous chemistry’.

Response: Thanks. We checked and corrected the language in the revised version.

Recommendation:
In the opinion of this reviewer, the presented manuscript could constitute an important contribution documenting the Enviro-HILRAM model. The paper should be published after major revisions.

Response: Thanks a lot. We do our best for that.

Authors response to comments of the Referee #3

We thank the Anonymous Referee #3 for the interesting and important comments on our manuscript. All the individual comments are addressed below in red.

The manuscript presents the online coupled model Eviro-HIRLAM, which is well known in the atmospheric modelers community. The manuscript is well structured and provides a comprehensive presentation of Enviro-HIRLAM development with a description of the different approaches and physical schemes implemented during the model evolution. The computational schemes and parameterizations adopted by the models are properly introduced and referenced. A minor shortcoming of this approach is that it is somewhere not very clear to the reader which computational scheme is the one chosen for the present version of the model or what alternatives are provided to the user.

Response:
The revised version provides more information about computational schemes chosen for the present version of the model.
The LMCSL with monotonic filter is the scheme chosen for the present version. As of now one may be in doubt if this just an option.

A relevant number of applications are referenced for almost all the model development fields. Some of the items (e.g. pollen) are described providing explicit summary of the overall results that make the paper more readable and useful for a reader that is not willing to read the large number of referenced papers and documents. Other examples of application are mainly discussed through references and do not allow the reader to appreciate the model effectiveness and the improvement offered by the online modelling approach.

Response:
We agree with the reviewer: the pollen part was not described in previous publications, so we did it in more details. Other aspects, considered in specific previous papers, are only briefly described here with corresponding references.

If the general approach of online coupling is physically sound and it can be agreed that it will probably become the prevailing modelling approach in the next future, the manuscript does not clarify, through its application examples, to what extent the online coupling and the main parameterizations introduced (e.g. urbanization) provide an improvement of model capability to predict observed pollutant concentrations and key meteorological parameters. An improvement of the analysis of the online coupling effectiveness is desirable and would make the manuscript more complete, interesting and valuable.

Response:
These issues are really very important, but the previous EuMetChem paper (Baklanov et al., 2014) considered them more comprehensive and not only for the Enviro-HIRLAM model.

Text and figures include a large number of acronyms for project names, parameterization schemes, etc. Even if many of them are known, it is quite difficult for the reader to know and remind all their meaning. It would be helpful to add an acronym legend section.

Response: Thanks. Done.

Specific comments:
Section 1. Methodology
Lines 72-75 The authors say that Enviro-HIRLAM is being used for different research project, but most cited project have already concluded they activity. In the Figure 1 lowest box most project mentioned as ongoing are finished since a few years.

Response:
Many previous and recent projects are mentioned in the text (FUMAPEX, MEGAPOLI, MACC, PEGASOS, MarcoPolo, EuMetChem, CarboNord, CRAICC-PEEX, CRUCIAL, …).
We have adjusted the info in the Figure 1 lowest box correspondingly.

Section 2.1 Modelling system structure
Line 92 The URL http://hirlam.org/trac/wiki/ is password protected and therefore not accessible to the reader. It should be substituted with an open access web site.

Response:
This is the policy of the HIRLAM consortium. We are in contact with the HIRLAM web-master to open this link or to provide another open one.

Section 2.3 Atmospheric chemistry
It is not clear if the “tropospheric sulfur cycle” is a simple scheme alternative to the CBM-Z, that is presently maintained for simplified simulations (what is the specific interest?), or if it is an obsolete option which is going to be abandoned. It is not specified how the CBM-Z gas-phase chemistry scheme is interfaced with the M7 aerosol module. Due to the relevance of secondary particle production modelling, more details would be appreciable to provide a comprehensive model description.

Response:
The tropospheric sulfur cycle chemistry is used together with M7 aerosol microphysics module because of its relative simplicity and low computational cost. The CBM-Z gas-phase chemistry is not interfaced with the M7 aerosol module because of several reasons: 1) the aerosol microphysics module does not include Secondary Organic Aerosols, therefore, there is no need of complex gas-phase mechanism with Volatile Organic Compounds related reactions and 2) it is too computationally expensive to use CBM-Z together with M7 for both weather and atmospheric composition prediction.

Lines 171-172 The authors say they “use KPP tools to create the gas-phase chemical mechanisms including the solvers for three chemical mechanisms.” What are the three mentioned chemical mechanisms? Only two of them have been previously presented: a) Tropospheric Sulfur Cycle, b) Gas-phase chemistry (CBM-Z).

Response:

Indeed, during the validation stages of creating the gas-phase schemes we used the Kinetic Preprocessor (KPP) (Sandu et al., 2006); we used KPP to create the Fortran code of three gas-phase schemes CBM-Z (Zaveri et al. 1999), GEOS-CHEM (Evans et al. 2003) and the Regional Atmospheric Chemistry Model “RACM” (Stockwell et al,1997).

For the chemical weather predication propose, GEOS-CHEM and RACM are very computational expensive schemes. GEOS-CHEM and RACM schemes include a large number of chemical reactions.

For more simplicity we cooperate with Dr. Rahul Zaveri (Personal communication with Dr. Ashraf Zakey) in order to simplify CBM-Z and online coupled it with the Enviro-HIRLAM Model.

“Tropospheric Sulfur Cycle scheme” is a very simple sulphur scheme (Easter et al., 2004). It was ported from HAM without use of the KPP tool. Reference: Easter, R. C., S. J. Ghan, Y. Zhang, R. D. Saylor, E. G. Chapman, N. S. Laulainen, H. Abdul-Razzak, L. R. Leung, X. Bian, and R. A. Zaveri (2004), MIRAGE: Model description and evaluation of aerosols and trace gases, J. Geophys. Res.,109, D20210,doi:10.1029/2004JD004571”

Lines 172-173 The authors say that Rosenbrock solver is usually selected. Why?

Response:

The Rosenbrock solver is mostly used within the air quality models communities because it is computational fast.

Line 190 What is “NWP-Chem-Liquid”?

Response: The “NWP-Chem-Liquid” is a thermodynamic equilibrium model, described in Korsholm et al. (2008). 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 equilibria is included in NWP-Chem-Liquid. This equilibrium module is solved using the analytical equilibrium iteration method (Jacobson, 1999).

Section 2.4. Aerosol formation, dynamics and deposition

Line 197 Is CAC still available in Enviro-HIRLAM or it is mentioned only for historical development reasons?

Response:

No, it is not used in the last reference version and in the described simulations, but can be called for specific studies. See e.g. Gross and Baklanov (2004), Korsholm (2009).

Lines 205-206 Is the aerosol type identity maintained through the model simulation and provided as separated output contribution to the total PM?

Response: Different aerosol types mentioned in the model description and simulations (as described in section 2.4) are provided as separate species in the model outputs along with lumped PM_{10} and $PM_{2.5}$.

Section 2.5. Emission modules and pre-processor

Line 254 Does wildfires emission module consider PM only or gas phase pollutants too?

Response:

The wildfires emissions were from the Finish Meteorological Institute - Fire Assimilation System v.1.1, which provides total lumped emissions. The total was split according to Andreae and Merlet, 2001 in organic and black carbon, and gaseous emissions of SO_2 only. The gas-phase pollutants like Nitrogen Oxide (NO) and Volatile Organic Compounds (VOCs) were not considered or processed.

Line 274 What are “transported modes”?

Response:

The "transported" mineral size mode in the GADS/OPAC data set (Köpke et al. 1997) is usual aerosol size mode that comes in addition to the more standard "nucleation", "accumulation" and "coarse" mineral size modes. Köpke et al. (1997) uses the "transported" size mode to describe aerosols that have been transported over a long distance, for instance Saharan aerosols that have been blown to the Atlantic ocean.

Section 2.7. Urban parameterizations and models urbanization
This section is relevant because it highlights the need for a mass conserving transport scheme in on-line coupled NWP and ACT models. For offline coupling this request is less strict because mass consistency is usually guaranteed by the coupler module.
Line 311 Bracket missing.

Response: Thanks. Done.

Line 312 Grid nesting is an effective technique to increase model resolution but it is rather confusing to consider it a method to represent urban areas.

Response:
The nesting technics and downscaling methods are actively and successfully used for urban areas to reach the necessary resolution for resolving or parameterisation of urban features and effects. The details of this approach was described e.g. in Baklanov and Nuterman (2010).
With respect to metropolitan areas, the downscaling for finer/ better resolution allows to reproduce smaller scale meteorological patterns, and then these patterns are further modified through running the urban modules such as BEP, SM2U, BEM, etc. only for grid cells where the cities are presented.
The text of this section is modified correspondingly.

Line 315 The "calculation of the urban mixing height based on prognostic approaches" is neither described nor commented in the following text.

Response:
Thanks. This issue was published previously in BLM papers Zilitinkevich et al. (2002) and Zilitinkevich and Baklanov (2002). Some clarifications were done: additional text and references on specific papers are included.
References:
Zilitinkevich, S. and A. Baklanov, 2002: Calculation of the height of stable boundary layers in practical applications. Boundary-Layer Meteorology, 105(3), pp. 389-409.
Zilitinkevich, S., A. Baklanov, J. Rost, A.-S. Smedman, V. Lykosov & P. Calanca, 2002: Diagnostic and prognostic equations for the depth of the stably stratified Ekman boundary layer. Quarterly Journal of the Royal Meteorological Society, 128, pp. 25-46.

Section 2.8. Transport schemes
Line 371 Is hat symbol missing on "modified weight" in equation 6 ?

Response: Thanks. Yes, it is. There should be a hat over the W. Corrected in the revised version.

Line 377 ""is are" should be corrected

Response: Thanks. Done.

Lines 388-390 This sentence concerning Enviro-HIRLAM mass consistency for tracer transport should be better explained and discussed. What are the possible limitations caused by this lack of mass conservation? What is TR4?

Response: Thanks. It is a mistyping. TR4 should be Eq. (4).
We have already answered this question to Reviewer 1.
We have added a sentence to clarify that mass-wind inconsistency is a minor problem. The traditional HIRLAM is (at least in principle) wind-mass consistent. In Enviro-HIRLAM where all moisture fields are transported with the LMCSL scheme there is no formal consistency, yet, since precipitation is very similar to that in HIRLAM (except for individual convective systems that are chaotic/unpredictable in their nature), the mass-wind inconsistency is small in practice.
A more careful discussion on the issue of mass-wind inconsistence in atmospheric models would require a rather extensive addition. In principle no monotonic transport schemes can be mass-wind consistent since the monotonic limiters formally destroy the consistency.
We also add a reference to the paper: Jöckel, P., von Kuhlmann, R., Lawrence, M. G., Steil, B., Brenninkmeijer, C. A. M., Crutzen, P. J., Rasch, P. J., and Eaton, B.: On a fundamental problem in implementing flux-form

advection schemes for tracer transport in 3-dimensional general circulation and chemistry transport models, Q. J. R. Meteorol. Soc., 127, 1035–1052, 2001.

Section 3 Modelling system applications
What are the mentioned "EnvCLIMA, Enviro-HIRHAM"?

Response: Thanks. It is clarified/modified in the revised version.

Lines 415-418 Do the mentioned temperature changes due to indirect effects improve model results? How relevant is the improvement? The reference given by the authors is to a Project report that can be hardly available, not to a journal publication. In the following sentence (lines 420-421) the authors mention a marginal improvement on surface temperature. They also mention a redistribution effect on NO2 concentration, but they do not specify is this effect improves model results.

Response:

Yes, these study results were described only in reports and proceeding papers. Corresponding journal paper is under preparation. The improvements due to the indirect effects exist (as shown e.g. in Fig 9), but the existing parameterisations of indirect effects need further improvement and evaluation. Several publications of different authors (e.g. Vogel et al., 2015) also stressed that these indirect mechanisms are the most uncertain and need further improvements.
We have answered in more details on the similar question to the Reviewer 1.

Lines 442-444 and Figure 9 The authors say "the ENV run bias for precipitation with respect to its frequency and amount has been decreased compared to the REF model run (Fig. 9)." Legends printed on the pictures seem opposite to what indicated in the caption (Enviro-HIRLAM on the left). Results showed in Figure 9 seem different during different parts of simulation: until July 21st the right side simulation seems better, while the left side one seems better during the last part of the simulation. What is the difference of the overall biases?

Response:
It is an unfortunate mistake; the left and the right figures must be swapped.
According to observations at WMO station 6670 at Zurich, Switzerland, the mean 12 hours accumulated precipitation in July 2010 was 0.97 mm, the median was 0 mm and the precipitation variance at the site was 7.52. As for the reference HIRLAM run, the modeled monthly mean, the median and the variance of 12 hours accumulated precipitation are equal to 1.83 mm, 0.14 mm and 16.90, respectively. The Enviro-HIRLAM model with aerosol-cloud interactions predicted the mean value of 1.16 mm, the median of 0 mm and the variance of 9.53 of 12 hours accumulated precipitation for the same month. That means the reference model tends to overpredict both the precipitation frequency and its amount, but the aerosol-cloud feedbacks in the Enviro-HIRLAM model reduce such over-prediction tendencies. Moreover, the values of Fractional Bias of Ref-HIRLAM (-0.61) and Enviro-HIRLAM (-0.18) along with Normal Mean Square Error values of Ref-HIRLAM (4.17) and Enviro-HIRLAM (3.45) show improvement of the Enviro-HIRLAM prediction score comparing to Ref-HIRLAM.

Lines 480-489 A grid size of 2.5 km seems quite crude to resolve Bilbao city. In x and y directions the city seems to be described by 2 to 4 grid cells which can be hardly considered sufficient to develop a "urban signal". Why has not been used a finer resolution? Is it due to the hydrostatic model limitations?

Response:
Yes, the hydrostatic approximation of the model was a limitation to increase the resolution to perform the urban simulations. However, sensitivity tests demonstrated that the 2.5 km was the optimal resolution allowing at the same time to obtain satisfactory reproducibility of the large scale processes and to explore the urban effects at local scale without being diminished due to a coarse resolution, for a medium size city (even possibly can be considered for a small size city). For other metropolitan areas such as Paris, Rotterdam, St. Petersburg, Shanghai - a similar resolution was chosen, although for Copenhagen (with a flat terrain) the highest possible/ suitable resolution tested was 1.5 km and provided reasonable verification results. Within a selected metropolitan area

there could be only a few grid cells having 100% representation of the urban fraction, but taking into account all urban grid cells, the boundaries of the cities (number of cells) could be substantially larger. Moreover, it should be noted that most of existing developed parameterizations in the physics core of any existing NWP model might be also needed to be revised when resolutions of 1 km and finer are used.

Figure 10 Why different land use classifications have been used for the two considered cities? What is the P01 modelling domain mentioned in the caption?

Response:
Depending on a country-by-country basis and national architectural specifics, different metropolitan areas could have different types of urban fabric with specific aerodynamical and morphological characteristics of urban districts. The size of the Bilbao metropolitan area is at least 10 times less than the Paris metropolitan area. Therefore, to harmonize the urban classification we considered that Bilbao had a Residential high and low density districts (RLD, RHD, respectively); while Paris metropolitan areas was characterised by a residential district (RD) and the city centre (CC). Also, note that for the land-use classification of the Bilbao metropolitan area, a local land-use database was used and for Paris, the land-use database CORINE 2000 was applied. (Gonzalez-Aparicio et al. 2010).
The P01 domain is just one of names for the modelling domains created for the Enviro-HIRLAM model runs with the focus on the Paris metropolitan area located in the centre of the domain. It has been removed from the caption.

Line 498 Does 10% improvement refer to the correlation value?

Response: Yes, it is referred to the overall correlation values.

Lines 499-500 It is not clear how the mentioned correlations have been computed. Time correlation for separated hours? How many stations have been used to compute the mentioned correlations?

Response:
The correlations were computed for the winter and summer months, simulated averaged over each hour of the day (e.g. considering the diurnal cycle), at each of the three types of locations considered (urban, suburban and rural – Figure 11a).

Lines 501-504 Where the mentioned results for Bilbao better than those obtained without urbanization? Was the improvement significant?

Response:
The results have been mentioned in Gonzalez-Aparicio et al. (2013). The results of those simulations were significant since we showed that the Enviro-HIRLAM model (urbanized version) was able to simulate the effect of the Urban Heat Island over a medium size city located in a coastal and complex terrain area characterized by land-sea breeze.

Lines 512-535 The authors show that model urbanization allows to describe UHI phenomenology in Paris and Bilbao, but they do not discuss if the urbanization improves results and reduces possible model bias with respect to urban observations.

Response:
Gonzalez-Aparicio et al. (2013) discussed the urban parameterization implementation in the Enviro-HIRLAM model and the improvement with respect to the control simulations for the Bilbao city. The urban effect and the results were compared with the results obtained in an experimental campaign over the city.

Lines 635-639 The mentioned effects of aerosol feedbacks on chemical composition are quite interesting. Did the mentioned changes on NO2 and O3 improve model results and increase its capability to reproduce measured values?

Response:
Unfortunately it was just a sensitivity study and a proper long-term validation was not realised yet. So, we prefer to avoid conclusions.

Figure 15 Right side color scale legend needs correction. How are correlations for separated hours computed?

Response:
We do not know how to change the legend scale, because the referee did not specify any required correction.
In order to compute correlation coefficients on diurnal cycle, the Enviro-HIRLAM model output was collected for separate time slices (00, 03, 06, … 21 UTC) and observation sites, and then the correlation coefficients were computed separately for each time-slice and site.

Lines 675-677 The authors mention new model applications without providing any detail about recent results potentially relevant and interesting for the readers. The mentioned feedback mechanisms evaluation is one of the key point of the paper.

Response:
Unfortunately we cannot answer on all the questions of online chemistry-meteorology modelling in one this paper. Some potential applications are just briefly mentioned in the paper and they are topics for further studies and analysis. In particular the results of the CarboNord project for Black Carbon feedbacks for the Arctic are now under analyses and will be published in a separate paper.

Section 4 Conclusions
Lines 692-702 These sentences contain repetitions of the same concepts that could be removed.

Response: Thanks. Done.