Online coupled chemical weather forecasting based on HIRLAM – overview and prospective of Enviro-HIRLAM

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1 Introduction

Chemical weather is a field of increasing popularity and several institutes, such as Environment Canada and NOAA, currently forecast both chemical and meteorological weather. Following (Lawrence et al., 2005) a definition of chemical weather may be given as local, regional and global distributions of trace gases and aerosols with corresponding variability ranging from minutes to days. Two modeling paradigms exists for forecasting chemical weather; the online approach and the offline approach (Baklanov and Korsholm, 2008, Baklanov, et al., 2008). The use of the terms online and offline in this context has been ambiguous, but a formal definition may be given as follows: offline models are separate chemical transport models (CTM's) forced by output from meteorological models, analyzed or forecasted meteorological data from archives or data sets, pre-processed meteorological data, measurements or output from diagnostic models; online models comprise online access models in which the meteorological forcing fields are available at each time step of the meteorological driver and online coupled models in which the CTM has been integrated into the meteorological driver and feedbacks between meteorological and chemical and aerosol fields are possible. In this definition it is possible to generate an online access model by employing an offline model with a very high coupling frequency e.g every 10 minutes, for example using a coupler such as OASIS4 or US-ESMF (COST-NetFAM, 2007, Dickinson, 2002), to exchange chemical and meteorological fields each time step. However, the handling of high resolution data in this manner is highly cumbersome and time consuming and therefore, not suited for operational chemical weather forecasting. Alternatively, an online coupled model may be generated by integrating a CTM in the meteorological driver, thereby, reducing the external data handling to a minimum. This should be done in such a way that all transport, dispersion and transformation of the chemical and aerosol species are done on the grid used by the meteorological model using the same numerical solution methods. We believe that this approach contains a number of advantages that makes it convenient. These include the use of the same parameterizations and numerical solution schemes for the chemical and aerosol species and the meteorological driving fields eliminating this type of inconsistency as well as the removal of spatial and temporal interpolation of the meteorological forcing fields. Comparing to offline models the online approach takes full advantage of the variability of the meteorological fields, it may include feedbacks and has shorter execution time. One might say that when using an offline model the computational resources are spread out over a longer time span, since, the meteorological forecast, the pre-processing (spatial and temporal interpolation) of the driving fields and the execution of the CTM are done separately, whereas with an online model, as advocated above, it is all done in during one run. It should be noted that, if it is assumed that feedbacks are not of importance, offline models have advantages in other disciplines such as sensitivity studies, including air quality impact studies, where meteorology is kept constant and emissions are varied. According to the definition given above chemical weather forecasting is not a new discipline, since offline air quality models have been used for several decades. However, if one wish to simulate the short term variability (below one hour), present in chemical and aerosol fields, which is important for feedbacks (see Feedback section later) online coupled models are needed.

Previously, the development of chemical weather and meteorological models progressed independently; a further advantage when developing an online coupled model following the integration approach is that the close relationship between air pollution and meteorology may be utilized for the greater good of both communities. Since the meteorological input is one of the main uncertainties in air pollution modeling an online coupled model, producing both meteorological and chemical weather forecasts, is expected to forecast chemical species and aerosols more precisely, especially in high resolution applications where much of the variability of the meteorological fields is explicitly resolved and has short time scales, i.e less than one hour. In a recent study it was shown that offline models are susceptible to large errors when variability in the vertical wind field is large. During a particular frontal passage the coupling interval of an offline model had to be as low as 10 minutes to capture 85% of the variability in the vertical velocity (Grell et al., 2004). On the other hand the detailed descriptions of trace gases and aerosols (natural as well as anthropogenic) will lead to improved forecasts for precipitation, cloud cover (through cloud and aerosol microphysical interactions) and surface energy balance (through direct aerosol interactions with both short- and long -wave radiation, trace gas radiation interactions and through changes in cloud optical properties). Furthermore, focus on common interest points, other than the feedbacks mentioned above, such as boundary layer parameterization, advection, convection and vertical diffusion will greatly increase, leading to increased model development capabilities.

Enviro-HIRLAM is an online coupled model and constitutes an important step towards a model which can produce both meteorological and chemical weather. Several issues must be addressed when integrating a chemical transport model in a numerical weather prediction (NWP) model. First of all the choice of NWP model is of importance. It seems reasonable to use a model which is well known to us and where we have expertise in problem solving at hand. The transformation of HIRLAM into HARMONIE will in fact include the possibility of running with chemistry (the Regional Atmospheric Chemistry Mechanism (RACM) (Stockwell et al., 1997) scheme has been implemented in AROME). This scheme, however, is mainly oriented towards research purposes and is to computationally intensive for operational efforts. Hence, a new scheme must be implemented in any case. The HARMONIE system does not contain any descriptions of aerosol dynamics or heterogeneous chemistry and does consequently not contain aerosol-meteorology feedbacks. In addition to this HIRLAM will still be employed at DMI for many years to come, at least for nesting purposes, and all new features implemented in Enviro-HIRLAM are modular and done using FORTRAMN90 memory allocation features. Currently, four different institutions: the Danish Meteorological Institute, University of Copenhagen, Tomsk University and the Russian Hydro-Meteorological University are working on Enviro-HIRLAM development and the University of Tartu are joining the Enviro-HIRLAM development team within a few months. A detailed account of the strategy is given in a separate paper (Baklanov, 2008), here we present the model developments (section 2) and show some examples of output (section 3).

2　　Description of Enviro-HIRLAM

The basic idea when developing an online coupled model is to make the meteorological model solve the governing equation describing transport, dispersion and transformations of chemical and aerosol species in its native grid, using the native parameterizations. Following this idea a first version of Enviro-HIRLAM, including passive tracer transport, has previously been developed (Chenevez, et al., 2004). The governing equation can be written as:

$$\frac{\partial \psi_i}{\partial t} = \left(\frac{\partial \psi_i}{\partial t}\right)_{emis} + \left(\frac{\partial \psi_i}{\partial t}\right)_{adv} + \left(\frac{\partial \psi_i}{\partial t}\right)_{diff} + \left(\frac{\partial \psi_i}{\partial t}\right)_{conv} + \left(\frac{\partial \psi_i}{\partial t}\right)_{dep} + \left(\frac{\partial \psi_i}{\partial t}\right)_{chem}$$

where ψ_i represents the mass concentration of either a gas-phase or an aerosol specie. The terms on the right hand side represents primary emissions, advection, vertical and horizontal diffusion, convection, dry and wet deposition of ψ_i respectively, while the last term represents transformations of ψ_i due to gas-phase and hetrogeneous chemistry as well as aerosol dynamics. Each of the terms in the above equation originates from different physical processes acting on different spatial and temporal scales. For this reason it is a very difficult

task to solve the equation in one common numerical scheme. Therefore, a splitting procedure has been applied (Marchuk, 1986, McRae, 1982). The solution is achieved by splitting the equation into the following six processes: emission, advection, horizontal diffusion, vertical diffusion and dry deposition, convection and wet deposition, chemistry and aerosols. Intermediate solutions of each process is used as starting point for the solution of the next process etc. (Fig. 1 displays a call tree displaying the main new and modified sub-routines). An advantage of this procedure is that it is possible to use different numerical schemes for the different physical processes. Precise numerical solution schemes are more important for some processes than for others (for example deposition versus advection) and if a simple parameterization is used for some process then there is no need for it to be represented by a very precise, but computationally heavy numerical scheme. Below follows a description of the solution method for each of the terms on the right hand side of the equation.
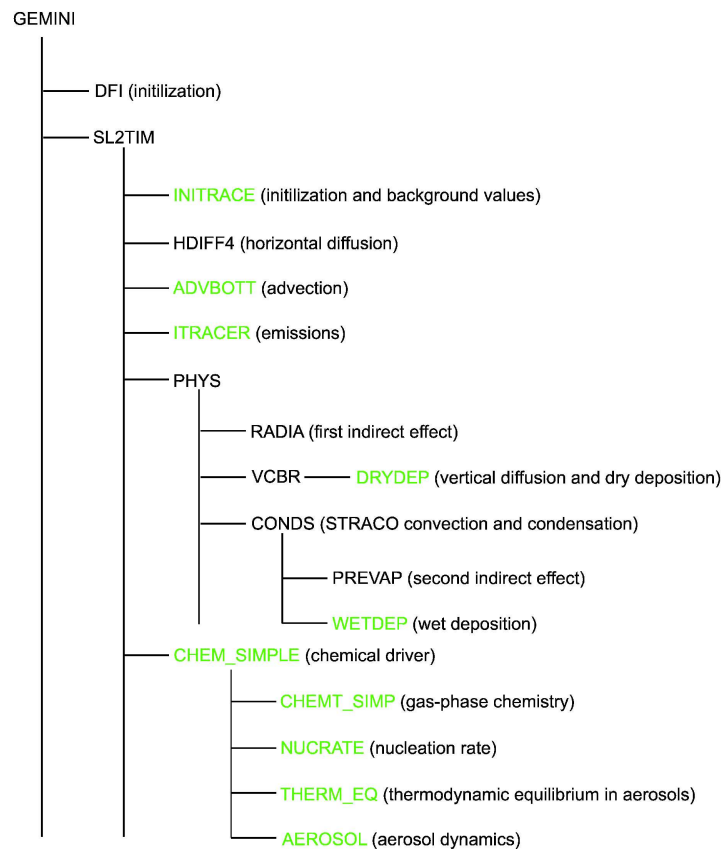

Figure 1: Partial call-tree displaying the main routines which have been modified as well as new additions (in green) with a short description.

2.1 Emissions

Emissions are currently handled as Eulerian point sources located within the lowest model layer, extending approximately 32 meters above the surface. It is necessary to assume that the emitted specie is well mixed within the grid box containing the emission point. Given the emission rate, emission time interval and the location of the emission point, the grid box volume may be calculated to give the emitted mass concentration Q_i per time interval. The modification of ψ_i then follows the Leapfrog scheme:

$$\psi_i^{t+1} = \psi_i^{t-1} + 2\Delta t Q_i$$

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The grid point closest to emission point is used to locate the emission grid box. The volume of this, and therefore the emitted concentration, is depended on the current resolution of the model. Hence, the higher the resolution the higher the emitted concentration. This influence of the resolution on the specie concentration is mainly visible close to the source (neighboring grid boxes) and disappears as the concentration field spreads. The emission rate, location and temporal development of the emission may be prescribed directly in the code for point source releases. For areal sources, which are treated as a sequence of point sources, this information may be supplied by emission files generated from emission inventories. A corresponding emission pre-processor has been developed in order to generate the necessary emission files. The pre-processor reads the GEMS-TNO or EMEP inventory, unpacks the temporal variations of the species, implements splitting of the volatile organic compounds, handles unit conversions and prepare the data on the model grid, accounting for grid rotations. A Birch pollen emission module has also been developed in collaboration with the Finish Meteorological Institute, to be used for pollen forecasting at DMI.

2.2   Advection and the continuity equation

The native Semi-Lagrangian and Eulerian advection schemes may be used to transport the species, however, mass conservation of chemical species is expected to be of great importance to chemistry and therefore these schemes are not sufficient, when developing an online coupled model (Chenevez, 2004). In order to improve mass conservation the Bott advection scheme is used (Bott, 1989a, 1989b) for chemical species and aerosols, while the Semi-Lagrangian scheme is used for meteorological quantities, thereby, facilitating fast simulation times. This inconsistency does not seem to cause loss of mass conservation on the short time-scales considered here (Chenevez, 2000). The Bott scheme is an Eulerian flux based scheme, implemented using fourth order polynomials in the horizontal directions and second order polynomials in the vertical. The scheme has been extended to three dimensions by using a splitting procedure, in which the one dimensional solution scheme is applied alternately in each direction (McRae, 1982). In order to assure that the CFL criterion is fulfilled in each direction at each time step the scheme is typically called three times inside a dynamical time step (at a horizontal resolution of 15 km). The main reason as to why the Bott scheme is not used to advect both meteorological and chemical species is that it requires to much cpu-time. Currently, a new scheme for solving the continuity equation, which is locally mass conserving, is being implemented in Enviro-HIRLAM, thereby removing the inconsistency. Full consistency is only achieved if the algorithms of the underlying dynamical core are fully consistent with the solution of the continuity equation for each specie. The new scheme consists of a simple and cost effective method to ensure local (inherent) mass conservation in traditional semi-implicit (SI), semi-Lagrangian (SL) models and an efficient locally mass conserving spatial filter. The locally mass conserving SL method (LMCSL) is relatively simple to construct from existing SL general circulation models (Kaas, 2008). The trick is to introduce weights at the upstream departure points ensuring that the total mass given off by a given Eulerian grid point to the surrounding SL departure points is equal to the volume represented by that grid point. Based on the Eulerian grid point weights it is possible to obtain departure point weights representing the divergence associated with the SL trajectories as in the so-called cell-integrated semi-Lagrangian (CISL) models. The LMCSL based dynamical core in Enviro-HIRLAM has come quite far and the model seems to be able to run stably with realistic results. The main obstacle in the implementation has been the inclusion of Lagrangian vertical coordinate. Most of the work carried out for building a CISL based dynamical core of the HIRLAM model (Lauritzen et al., 2008) could fortunetaly be re-used.

The scheme has only been tested in traditional two dimensional advection tests and in a semi-Lagrangian, semi-implicit shallow water model in plain geometry but with topography. Without the filter the scheme is at least as accurate as the traditional SL schemes. The filter enhances the accuracy for all tests carried out. However, it becomes particularly impressive near sharp gradients and discontinuities. The increase in numerical cost of the new scheme relative to traditional SI-SL models is small, particularly when there are several passive tracers.

2.3   Turbulent diffusion

Vertical diffusion is done with the native TKE-l scheme, CBR, which has been modified to include diffusion of extra scalars (other than TKE) using the same coefficients as for cloud water (Cuxart, 2000). For horizontal diffusion the native extra scalar system has been utilized. The native implicit fourth order diffusion scheme may be used to diffuse the extra scalars, it should, however, be noted that this scheme is not mass conservative, and therefore may not improve results when used (Chenevez, 2000).

2.4   Convection

The Tiedtke mass-flux scheme may be used to convect aerosols and gases The mass-flux calculation is the same as for air, hence, the density difference between air and the gases is not accounted for and it is assumed that the settling velocity is negligible compared to the convective updraught velocity. Another option is to use the STRACO convection and condensation scheme. This scheme is currently being modified to account for the convection of extra tracers.

2.5   Urbanization

The representation of urban areas in Enviro-HIRLAM contains three different approaches which may be combined. The 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 (Baklanov et al., 2005). The effects of buildings and street canyons have been implemented via the building effect parameterization, termed BEP (Martilli et al., 2002). Re-classification of land-use with respect to urban surface sub-types, such as buildings, artificial surfaces with and without vegetation and urban districts with detailed morphological characteristics can be included through the urban version of the soil model for sub-meso scales (SM2-U) (Dupont et al., 2004, 2006). Further information and results of preliminary testing may be found in (Mahura et al., 2005ab; 2008).

2.6   Dry deposition

Dry deposition refers to the processes by which gases and aerosols are removed from the atmosphere, in the absence of precipitation, at the atmosphere - surface interfaces (such as canopy, leafs, trees, water surfaces, grass surfaces etc.). There are two aspects which needs must be considered when modeling dry deposition; the transport of the aerosols or gases from the atmosphere to the reacting surface and the interaction with the surface. The small scale transport from the near surface environment to the surface can conceptually be viewed as occurring in two steps. During step one the particles are transported across the surface layer and into the laminar sub-layer. The governing processes are vertical mixing and sedimentation. Step two accounts for transport across the laminar sub-layer onto the surface. Brownian diffusion and sedimentation are governing processes but also phoretic effects, impaction and interception are relevant in this region. Once at the surface specie specific interactions may take place. The dry deposition velocity is defined as:
$v_d^i(x, y, t) = -F_i(x, y, t)/\psi_i(x, y, z = z_r, t)$ where F_i is the flux of specie i from the atmosphere to the surface (downward flux is defined to be negative) at position x, y with $z = z_r$ a reference height, taken to be the lowest full model level and t a time variable. Defined in this manner parameterizations of v_d^i will yield the requested flux. Following (Wesely, 1989) v_d^i is parameterized using an an electrical analog; the resistance approach. The dry deposition velocity may then be written as $v_d^i = 1/r_{total}^i$ where r_{total}^i is the total resistance of specie i to dry deposition. This parameterization is convenient because the total resistance may be split into component resistances, representing different parts of the atmosphere, which are coupled in series. By coupling the resistances in parallel, mass transport can occur through parallel paths, such as through plant stomata and leaf surfaces.

The dry deposition flux to the surface provides the lower boundary condition for the vertical diffusion of aerosol number and mass concentration (zeroth and third moment). Deposition is done in each aerosol mode separately (see aerosol section below) using the deposition velocity for the mass concentration as described below and using the geometric mean radius of each aerosol mode as the aerosol size.

Considering the fluxes of aerosols through each of the layers one may derive an expression for r_{total}, where i is now omitted, since, aerosol species are only distinguished by their size, it follows:

$$v_d = \frac{1}{r_{total}} = (r_a + r_b + r_a r_b v_g)^{-1} + v_g$$

Hence, the deposition velocity of the aerosols may be viewed as the reciprocal of three resistances in series and one in parallel. In this expression r_a is termed the aerodynamic resistance describing the turbulent transfer of particles to the laminar sub-layer, r_b is the resistance to Brownian diffusion across the laminar sub-layer, $r_a r_b v_g$ is an artifact of the equation manipulation and v_g is the gravitational settling velocity. According to the size of the aerosols v_g is calculated using Stoke's law in the laminar regime or an iterative procedure in the intermediate or turbulent regime (Näslund and Thaning, 1991). The Cunningham correction factor accounts for non-continuum effects, while parameterizations of r_a and r_b follows (Wesely, 1989, Zanetti, 1990) and is generally depended on atmospheric stability, land-use, friction velocity, wind speed, kinematic viscosity and Brownian diffusivity.

For gas-phase species $v_g = 0$ and the term $r_a r_b v_g$ is replaced by r_s which describes the surface resistance due to direct specie-surface interaction, this term is zero for aerosols since they are assumed to stick to the ground once impacted. The resistance to diffusion, r_b, follows (Maryon and Ryall, 1996), while r_a is the same as for aerosols. The surface resistance should represent the major serial and parallel deposition pathways. Over land these include the stomata and mesophylic pathways into active plants (r_{plant}), the canopy pathway (r_{canopy}) and the pathway directly into the ground (r_{ground}), therefore:

$$r_s = \frac{1}{r_{plant} + r_{canopy} + r_{ground}}$$

The resistance of deposition in plants is considered to consist of two parallel pathways, represented by the stomatal and mesophyllic resistances (r_{st} and r_m respectively), so that $r_{plant} = 1/(r_{st} + r_m)$. The canopy resistance is splitted into the upper canopy (r_{uc}) and the lower canopy which includes two parallel paths, the resistance to buoyant convection (r_{dc}) and the resistance to uptake in leaves, twigs and other exposed surfaces (r_{cl}). For the canopy we therefore use $r_{canopy} = 1/r_{up} + 1/(r_{dc} + r_{cl})$. The ground resistance comprises two parallel paths, a transfer resistance due to processes which is only dependent on canopy height (r_{ac}) and a resistance against uptake in soil, leaf litter, etc. on the ground (r_{gs}). The ground resistance may then be written $r_{ground} = 1/(r_{ac} + r_{gs})$. Following (Wesely, 1989) these resistances are typically dependent on season and land-use, in Enviro-HIRLAM they are adjusted for solar insulation and surface wetness (Binkowski, 1999).

Following (Sehmel, 1980) the surface resistance over water is given as:

$$r_s = \frac{1}{3.9 \times 10^{-5} H T_s u^*}$$

where T_s is the surface temperature, H is the effective Henry's law constant and u^* is the friction velocity.

2.7   Wet deposition

Wet deposition refers to the processes by which gases and aerosols are removed from the atmosphere due to precipitation release. Below-cloud scavenging is the process whereby ambient gases and aerosols receding below cloud base are removed by rain or snow fall while in-cloud scavenging is the processes by which gases and aerosols receding inside the cloudy environments are removed. Wet deposition processes are

parameterized via the scavenging coefficient $\Lambda^i(x, y, z, t)$:

$$\frac{\partial \psi_i}{\partial t} = -\Lambda^i \psi_i$$

and is carried out for gases and in each aerosol mode separately, using corresponding scavenging coefficients. Dissolution of gases in aqueous aerosols, involves the iteration of a thermodynamic equilibrium between gases and aerosol water as is described in the aerosol section below.

In-cloud and below-cloud scavenging of aerosols by rain is dependent on the aerosol radius and rain rate and follows the extensions in (Baklanov and Sørensen, 2001). For gases there are two main mechanisms regarding the scavenging of ambient gases; they may be removed by uptake in falling precipitation (both in-cloud and below-cloud) and they may be removed by dissolution in cloud water which is sub-subsequently scavenged by precipitation (in-cloud). The parameterization of Λ for these processes follows (Seinfeld and Pandis, 1998) and generally depends on the temperature corrected Henry's law constant for the specific gases, the mass transfer coefficients, rain rate, aerosol size and fall speed. Note that here deposition is three dimensional, i.e Λ^i may be calculated in each grid point, since the rain rate produced by HIRLAM is three dimensional.

2.8   Chemical solvers

The ability of a chemical weather model to forecast trace gas mixing ratios or deposition fields greatly depends on the accuracy of the gas-phase chemistry scheme. The rate constants, product yields, quantum yields and absorption cross sections for the photolysis reactions must all be of high accuracy. A large variety of lumped gas-phase chemical mechanisms based on the knowledge of the atmospheric chemical composition, reactions and corresponding rate constants have been developed in order to describe the chemical composition of the troposphere. Examples of such mechanisms include the EMEP mechanism (Simpson et al., 1997), the RACM, the Carbon Bond IV and V mechanisms (Gery, et al., 1989, Yarwood, et al., 2005) and the SAPRC-99 mechanism (Carter, 2000).

Currently, four mechanisms for gas-phase chemistry has been implemented in Enviro-HIRLAM, the Numerical Weather Prediction Chemistry scheme (NWP-Chem-Gas), the Regional Acid Deposition Model version 2 (RADM2) (Stockwell, et al., 1990), the RACM mechanism and an extended version (includes isoprene and DMS chemistry) of the Carbon Bond IV mechanism (CBMZ). Cpu-time and memory requirements of chemistry schemes may pose a large bottleneck for online coupled models. The NWP-Chem-Gas mechanism, developed at DMI, has been constructed for operational usage in online coupled models and is economical with respect to cpu-time and memory usage. It currently employs 17 advected chemical species, whereas the RADM2 mechanisms use 41 species, the RACM mechanism 49 and the CBMZ mechanism 72. It covers the most important chemical mechanisms for air pollution and aerosol formation and has been compared to more detailed schemes in zero dimensional simulations of both the free troposphere and the clean and polluted boundary layer producing satisfactory results. The NWP-Chem-Gas scheme is the default chemistry scheme in Enviro-HIRLAM.

The reactions included in the scheme is summarized in table 1, the Di-Methyl-Sulphide (DMS) and biogenic chemistry will be implemented in the future, thereby extending the number of advected chemical species. The scheme describes the basic chemistry of the photo-oxidation of VOC to peroxy radicals, the most important NO_x reactions, the most important ozone formation reactions, sulphur chemistry where DMS is included (since the emission of DMS is estimated to account for approximately 25% of the total global gaseous sulphur emissions released into the atmosphere), isoprene chemistry (it is well known that biogenic emissions of isoprene α-pinene and other terpenes affects gas-phase chemistry (such as ozone) as well as aerosol formation). In the present version of NWP-Chem-Gas the ordinary differential equations are solved using the Quasi-Steady-State Approximation (Hesstvedt, et al., 1978).

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Table 1: Gas-phase chemistry in NWP-Chem (NWP-Chem-Gas)

Photolysis reactions

1	$NO_2 + hv \rightarrow O(^3P) + NO$
2	$O_3 + hv \rightarrow O(^1D) + O_2$
3	$HCHO + hv \rightarrow 2\ HO_2 + CO$
4	$HCHO + hv \rightarrow H_2 + CO$

Inorganic chemistry

5	$O(^3P) + O_2 \rightarrow O_3$
6	$O(^1D) + N_2 \rightarrow O(^3P) + N_2$ and $O(^1D) + O_2 \rightarrow O(^3P) + O_2$
7	$O(^1D) + H_2O \rightarrow HO + HO$
8	$HO_2 + NO \rightarrow NO_2 + HO$
9	$O_3 + NO \rightarrow NO_2 + O_2$
10	$CO + HO \rightarrow HO_2 + CO_2$
11	$HO + NO_2 \rightarrow HNO_3$
12	$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$
13	$HO_2 + HO_2 + H_2O \rightarrow H_2O_2 + H_2O + O_2$

Organic chemistry

14	$HC + HO \rightarrow RO_2 + H_2O$
15	$RO_2 + NO \rightarrow HCHO + HO_2 + NO_2$
16	$HCHO + HO \rightarrow HO_2 + CO + H_2O$
17	$RO_2 + HO_2 \rightarrow ROOH + O_2$
18	$RO_2 + RO_2 \rightarrow prod$

Sulphur chemistry

19	$HO + SO_2 \rightarrow H_2SO_4 + HO_2$
20	$DMS + HO \rightarrow 0.85\ SO_2 + 0.15\ MSA$
21	$DMS + HO \rightarrow MSIA$
22	$MSIA + HO \rightarrow 0.85\ SO_2 + 0.15\ MSA$

Biogenic chemistry

23	$BIO + HO \rightarrow BIOO_2$
24	$BIO + O_3 \rightarrow 1.23\ HCHO + 0.14\ CO + 0.28\ ROOH + 0.25\ HO_2 +$ $0.25\ HO + 0.18\ RO_2 + 0.09\ H_2O_2$
25	$BIOO_2 + NO \rightarrow 2.0\ HCHO + 1.0\ HO_2 + 1.0\ NO_2$
26	$BIOO_2 + HO_2 \rightarrow RO_2$
27	$BIOO_2 + BIOO_2 \rightarrow 2.0\ HCHO + HO_2$

2.9   Aerosol solvers

The formation and transformations of atmospheric aerosols are known to be influenced by the dissolution of trace gases (if wetted), nucleation, condensation, evaporation and coagulation. Therefore, the aerosol module in Enviro-HIRLAM comprises two parts, a thermodynamic equilibrium model (NWP-Chem-Liquid) and an aerosol dynamics model. For simulating aerosol dynamics three models has been implemented, covering both sectional and modal approaches. These comprise the modal aerosol module from the Chemistry-Aerosol-Cloud (CAC) model developed at DMI (Gross and Baklanov, 2002, Baklanov, 2003, Gross, et al., 2005), the sectional Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2007) and the Modal Aerosol Dynamics Model for Europe (MADE) (Ackermann et al., 1998) with the secondary organic aerosol model (SORGAM) (Schell et al., 2001) (referred to as MADE/SORGAM) (Note that only the implementation of the CAC aerosol model has been tested so far).
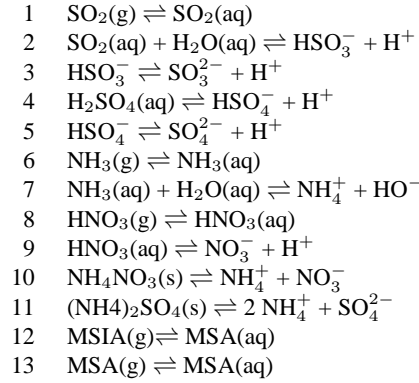
2.9.1   Thermodynamic equilibrium

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. At present this equilibrium module is solved using the analytical equilibrium iteration method (Jacobson, 1999). The reactions are summarized in table 2 and in the future the module will be updated to include the impact of organic

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compounds from anthropogenic and biogenic sources.

Table 2: Liquid-phase equilibrium mechanism in NWP-Chem (NWP-Chem-liquid)

1	$SO_2(g) \rightleftharpoons SO_2(aq)$
2	$SO_2(aq) + H_2O(aq) \rightleftharpoons HSO_3^- + H^+$
3	$HSO_3^- \rightleftharpoons SO_3^{2-} + H^+$
4	$H_2SO_4(aq) \rightleftharpoons HSO_4^- + H^+$
5	$HSO_4^- \rightleftharpoons SO_4^{2-} + H^+$
6	$NH_3(g) \rightleftharpoons NH_3(aq)$
7	$NH_3(aq) + H_2O(aq) \rightleftharpoons NH_4^+ + HO^-$
8	$HNO_3(g) \rightleftharpoons HNO_3(aq)$
9	$HNO_3(aq) \rightleftharpoons NO_3^- + H^+$
10	$NH_4NO_3(s) \rightleftharpoons NH_4^+ + NO_3^-$
11	$(NH4)_2SO_4(s) \rightleftharpoons 2\,NH_4^+ + SO_4^{2-}$
12	$MSIA(g) \rightleftharpoons MSA(aq)$
13	$MSA(g) \rightleftharpoons MSA(aq)$

2.9.2 Aerosol dynamics

The CAC aerosol model is the default used in Enviro-HIRLAM. It is based on the modal description of the particle distributions as developed by (Whitby and McMurry, 1998). Here a brief overview of the model formulation is given. The aerosols are divided into three size modes, the nuclei mode (i) consisting of aerosols generated by nucleation (typical diameter 0.01 μ m), the accumulation mode (j) consisting of aged aerosol and cloud condensation nuclei (typical diameter 0.1 μ m), and the mechanically generated coarse mode (c) (typical diameter 10 μ m). The definition of the k'th moment, M_k, of the distribution is:

$$M_k = \int d^k n(\ln d) d(\ln d)$$

where n is the aerosol size distribution and d is the aerosol diameter. Following (Whitby and McMurry, 1998) we assume that the size distribution in each mode is log-normal:

$$n(\ln d) = \frac{N}{\sqrt{2\pi} \ln \sigma_g} \exp\left[-\frac{1}{2}\left(\frac{\ln \frac{d}{d_g}}{\ln \sigma_g}\right)^2\right]$$

where N is the number concentration, d_g is the geometric mean diameter and σ_g is the geometric mean standard deviation for the modes. This assumption closes the equations and the k'th moment of each mode may be written:

$$M_k = N d_g^k \exp\left(\frac{k^2}{2} \ln^2 \sigma_g\right)$$

M_0 represents the total number concentration in a given mode, M_2 is proportional to the total aerosol surface area and M_3 is proportional to the total aerosol volume and therefore to the total aerosol mass concentration. If we assume that σ_g is constant in time, prediction of two moments, M_0 and M_3, allows the number and mass concentration as well as geometric mean diameter to be diagnosed. The model may now be formulated in terms of the effect of the dynamical processes on the moments of the size distribution. For accumulation mode we can write the rate of change if the k'th moment as:

$$\frac{\partial M_{kj}}{\partial t} = G_{kj} - C_{kjj} + C_{kij} + E_{kj}$$

where G_{kj} is the condensation growth term, C_{kjj} represents the moment loss due to intra-mode coagulation, C_{kij} is the gain in moment due to inter-mode coagulation and E_{kj} represents primarily emitted accumulation

mode aerosols. A similar equation governs the rate of change of the k'th moment in nuclei mode:

$$\frac{\partial M_{ki}}{\partial t} = \dot{M}_{ki} + G_{ki} - C_{kii} - C_{kij} + E_{ki}$$

where $\dot{M}_{ki}$ is the production rate of the k'th moment due to nucleation. For the coarse mode we get:

$$\frac{\partial M_{kc}}{\partial t} = E_{kc}$$

Under the assumption of lognormal size distributions analytical expressions may be found for the terms on the right hand side of the above equations and we can solve for the evolution of the number concentration and mass concentration for each mode. Advection, convection, diffusion and deposition of these modes proceeds as explained in the previous sections.

The water mole fraction in sulfuric acid - water aerosols is calculated using the parameterization by (Wilck, 1999), while binary nucleation follows either the parameterization by (Kerminen and Wexler, 1994) or the one by (Kulmala, et al., 1998).

2.10 Feedbacks

There are many pathways for pollutant-meteorology interactions, including the aerosol direct, indirect and semi direct effects, the self-feedback effect, the photochemistry effect, the smudge-pot effect, the day-time stability effect and the black carbon low cloud positive feedback loop (Jacobson, 2002, Twomey, 1974, Albrecht, 1989). These effects have characteristic time-scales below one hour, e.g it takes in the order a half to one hour for cloud and precipitation development. Interactions between pollutants (aerosols and gases) and meteorology will inevitably lead to variations in the meteorological fields, thereby, feeding back on the pollutant concentration fields. The importance of such feedbacks on weather, climate and pollutant dispersion is, however, unknown and models such as Enviro-HIRLAM, WRF-chem (Grell, et al., 2005) or GATOR (Jacobson, 1996) are needed to prioritize the importance of these effects. In the current version of Enviro-HIRLAM two feedback pathways has been implemented.

2.10.1 The direct aerosol effect

The presence of aerosols in the atmosphere may absorb, scatter and re-emit incoming shortwave radiation. These effects have not been implemented into the model yet and the radiation scheme used in HIRLAM (Savijärvi, 1990) is to simplified to account for these effects. Following (Seinfeld and Pandis, 1998) it is possible to estimate the effect of a layer of scattering aerosol accounting for surface reflections, by modifying the surface albedo accordingly. Another approach would be to use look-up tables for the complex index of refraction for various aerosol compositions, assuming that the aerosol is in the Rayleigh scattering regime.

2.10.2 The first indirect aerosol effect

As anthropogenic aerosols enter cloud environments the number concentration of cloud condensation nuclei (CCN) is modified, generally, resulting in more numerous and smaller CCN (decreased mean diameter). The cloud radiation characteristics depend on bulk cloud properties and a decrease in droplet mean diameter results in a modification (whitening) of the cloud albedo. The HIRLAM radiation scheme is based on (Savijärvi, 1990) and all water cloud radiation is parameterized via the cloud droplet effective radius, r_e, which may be written as:

$$r_e = \left(\frac{3L_c}{4\pi \rho_w k N} \right)$$

where L_c is the cloud condensate content, ρ_w is the density of water, k is a fitting parameter which distinguishes between land and water surfaces and N is the cloud droplet number concentration (Wyser, et al., 1999). N may be decomposed into a natural background and an anthropogenic contribution: $N = N_{back} + N_{anthr}$, where N_{back} is a constant for clean air supplied in HIRLAM depending only on the surface type (land or water), while N_{anthr} is calculated in the aerosol module assuming that all accumulation mode aerosols may act as CCN.

2.10.3   The second indirect aerosol effect

As the number concentration of aerosols increase more aerosols compeed for the available water vapor, the growth of droplets into precipitation drops and thereby the initiation of precipitation is delayed. Hence, cloud life-time is prolonged and the second indirect effect refers to the influence of this on the optical depth.

The description of cloud microphysics in the STRACO scheme is based on the Sundqvist parameterization (Sundqvist, 1988, Sundqvist, et al., 1989, Sundqvist, 1993). STRACO has been extended to include the effects of cloud drop number concentration and characteristic droplet radius r, by combining the autoconversion term for cloud water from (Rasch and Kristjansson, 1998) with the existing formulation in the STRACO scheme (Sass, 2002). In STRACO precipitation release is written $G_p = \Phi q_c(1 - \exp(-X^2))$ where q_c is the cloud condensate, $X = \widehat{q_c}/\mu$ where $\widehat{q_c} = q_c/f$ is the in-cloud specific cloud condensate and f is the grid box fractional cloud cover. The Φ term is defined as: $\Phi = \Phi_1\Phi_2\Phi_3\Phi_4$ where Φ_2 describes the effect of collision/coalescense and the Bergeron-Findeisen mechanism, Φ_3 expresses a temperature dependency at cold temperatures, Φ_4 is height dependent and describes an enhanced sedimentation of cloud droplets from fog (clouds at very low levels) and Φ_1 is the autoconversion term which is now defined as:

$$\Phi_1 = C_{l,out}\widehat{q_c}\frac{\rho_a}{\rho_w}\left(\widehat{q_c}\frac{\rho_a}{\rho_w}N\right)^{\frac{1}{3}}H(r - r_0)$$

Here ρ_a represents air density, H is the Heavy-side step function, $C_{l,aut}$ is a constant (Rasch and Kristjansson, 1998), $r = [(3\rho_a q_c)/(4\pi N\rho_w)]^{1/3}$ is the mean cloud droplet radius and r_0 is a constant threshold drop radius (5μm).

Rasch and Kristjansson (Rasch and Kristjansson, 2008) refer to a need of reducing the term Φ_1 by an order of magnitude to obtain a sufficiently small autoconversion when precipitation release is not well developed (small). Here the Sundqvist term (X) as given above has been introduced in order to achieve the same effect in a more physically based way. The effect of the X term is to make precipitation release less efficient at small amounts of in cloud condensate. The parameter is affected by the denominator μ which implies increasing X with increasing precipitation release entering the grid-box from above and depends on the temperature

As before $N = N_{back} + N_{anthr}$ where N_{back} depends only on surface type and N_{anthr} is calculated in the aerosol module. The modifications made to the STRACO scheme is currently being tested, but preliminary runs show that it gives results similar to the latest STRACO version.

2.11   Initial and lateral boundary conditions

Boundary conditions generally follows the HIRLAM set-up. Boundaries are over-specified by externally imposed fields. In order to counteract sharp differences between external and internal fields the mass and number concentrations are relaxed towards the imposed fields, which includes clean or dirty background values. In this way it is possible to run nesting scenarios, whereby downscaling over a certain area is performed. The background values are taken from (Poppe, et al., 1996). The Bott scheme applies constant values for inflow and linear extrapolation, to calculate the needed grid point values outside of the domain, for outflow conditions. The vertical diffusion scheme uses the dry deposition flux as lower boundary condition. Initial conditions are either first guess fields from a previous forecast or prescribed fields describing clean or

polluted background atmospheres. The initial values may be filtered during initialization to reduce the influence of noise.

2.12   Optimization

Enviro-HIRLAM currently runs on the NEC-SX6 super-computer at DMI, but will be ported and optimized to the new CRAY-XT5 at DMI within a few months. The chemical scheme is the main bottleneck regarding both cpu-time and memory usage, hence, it is important to limit the number of species as much as possible, as is done with the NWP-Chem mechanisms. Except for the Bott advection scheme, all new implementations follow the single program multiple data parallelization strategy used in HIRLAM. The Bott scheme is not parallelized and advection is currently carried out on a single MPI-task, hindering the full effect of the parallelization and reducing scalability. No controlled timing experiments exists yet, but an estimate based on runs on a non-dedicated system can be made. A 24 hour forecast will take about two hours if 24 NEC-SX6 processors are used, with 26 advected species in the NWP-Chem and the CAC aerosol model, on a domain with $0.05° \times 0.05°$ horizontal resolution and 40 levels ($90 \times 74 \times 40$ grid-points).

2.13   Outstanding issues

Two updates are imminent, the model will be updated to HIRLAM version 7.1 and a representation of the direct aerosol effect will be implemented. Currently, there are a number of outstanding issues which will be dealt with in the future. The most important issue regards the implementation and parallelization of a new advection scheme by the University of Copenhagen. At DMI we are currently working on updates for the gas-phase chemistry and aerosol modules. In the 7FP project MEGAPOLI it is planed to test EnviroHIRLAM with a range of aerosol model. On a perspective a mass conservative horizontal diffusion scheme will be implemented. Currently, simple heterogeneous chemistry exists in the CAC model. This will be expanded and implemented in Enviro-HIRLAM. Plans also exist for an expansion of the HARMONIE system to include the Enviro-HIRLAM chemistry and aerosol features, this would be a joint effort for the Enviro-HIRLAM developers and the HIRLAM consortium.

3   Examples of usage

Passive tracer transport has been tested by simulating the release made during the first European Tracer Experiment (ETEX-1)(Graziani, et, al., 1998) (Fig. 2). Dry and wet deposition were tested in simulations of the Cs^{136} deposition after the Chernobyl accident (Fig. 3). In both cases model output was compared to measurements and the statistical scores showed that the model reproduced the tracer and deposition fields satisfactorily.
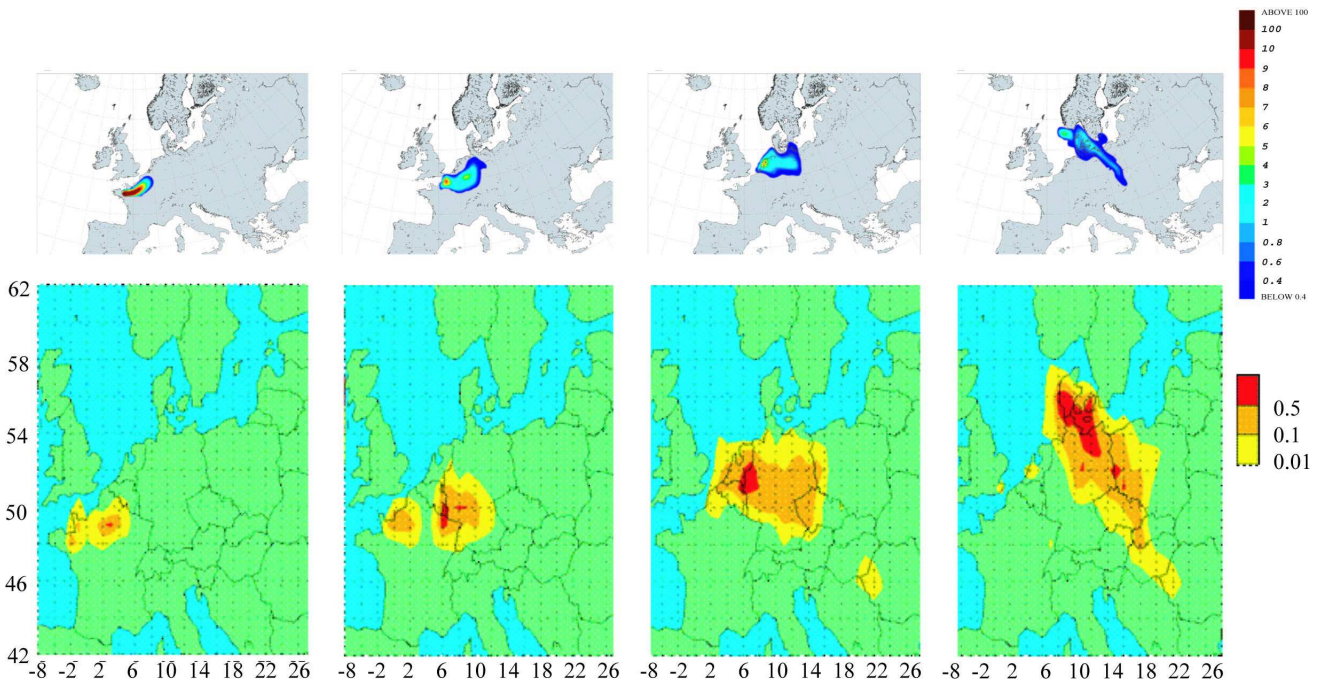
Figure 2: Top panel: evolution of modeled tracer surface concentration (ngm^{-3}) at 12, 24, 36 and 48 hours after start of the release. The bottom panel shows the corresponding measurements in the same units, note that the interval is not the same in the two plots. Average correlation and normalized mean square error relative to stations selected during ETEX-1 to represent short and long range transport is 0.49 and 4.25, while the average bias is 0.18 $\mathrm{ng\,m^{-3}}$

Some simple tests regarding the effects of feedbacks on the spread of pollutants has also been carried out (Baklanov and Korsholm, 2008) and as well as a comparison study between offline and online modeling (Korsholm, et al., 2008).
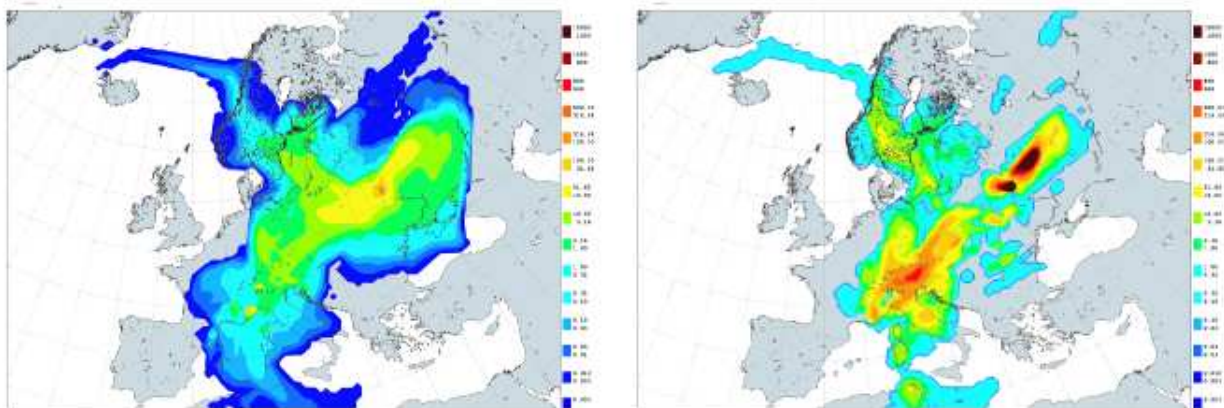

Figure 3: Right: accumulated wet deposition on May 1 1986 (kBqm^{-2}). Left: accumulated dry depositio n on May 1 1986 (kBqm^{-2}). Statistical comparison to data gathered after the accident gives a global correlation of 0.59 , a normalized mean square error of 6.34 and a bias of 38.7 kBqm^{-2}, there were 316 measurements available for the stati stics.

4   Summary and concluding remarks

Enviro-HIRLAM is an online coupled meteorological and chemical weather model. The handling of the chemical species and aerosols are done on the native HIRLAM grid using the native parameterizations (currently the advection equation is solved using the Bott scheme). This have been achieved by utilizing the 'extra tracer' capabilities of HIRLAM. Feedbacks between aerosol and meteorological fields are mediated through the first and second indirect aerosol effects, but the direct aerosol effect will be implemented. Passive tracer transport and deposition has been tested against the ETEX-1 measurements and in a simulation of the Chernobyl accident with satisfying results. Currently, tests of the gas-phase chemistry is being carried out. The default chemistry and aerosol schemes have been chosen to facilitate operational activities, while a series of other chemical schemes has been implemented for research purposes. Model development proceeds at several institutes and the model will be employed in multiple national and international projects through which testing and improvements will also be carried out. It has recently been decided to open a HIRLAM chemistry branch. In this context Enviro-HIRLAM will be used as a baseline system for further testing of the chemistry and aerosol schemes and for future developments.

5   Acknowledgments

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