6.15 THREE-DIMENSIONAL CHEMISTRY-TRASPORT MODELLING: UNCERTAINTIES CONNECTED TO THE METEOROLOGICAL INPUT

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INTRODUCTION

Chemistry-transport models (CTM:s) need meteorological forcing for calculating the transport and mixing of pollutants in the lower troposphere. The accuracy of these data is crucial for the air pollution forecasts to be reliable enough. The determining parameters for the life-time of air pollutants emitted into the atmospheric boundary layer ABL are the mixing height (hmix), precipitation intensity (P), wind velocity (uabs) and direction (udir), friction velocity (u_*), Monin-Obukhov length (1/L), surface roughness (z_o), temperature scale (t_*), moisture, temperature (T) and the total cloudiness.

The emitted pollutants are split partly inside the ABL, partly into free atmosphere, depending e.g. on the heat content and exhaust velocity of the stack gases, thermal state of the atmosphere and hmix value. While the wind direction veers with the height, the effective height of the plume determines the initial transport direction and speed. Most of the ABL schemes use vertical temperature difference near the surface to estimate the stability state, thus the accuracy of the 2m temperature is very important for dispersion modelling. Most pollutant episodes occur or get their origin in stable conditions with a low mixing height, so the estimate of the inversion height, strength and dynamics have often been named to be the most important parameter for the air quality studies. The mixing of the contaminants to the upper troposphere and to the surface is also controlled by the stability and the height of the ABL.

Temperature and humidity determine the chemical conversion and the in-cloud-scavenging rates, while the below-cloud scavenging depends on the vertical distribution, intensity and type of the precipitation. Dry deposition is a rather complicated process, normally described by the resistance analogy. The aerodynamic resistance, determining how fast the pollutants are transported down to the surface, is a function of stability state of the atmosphere, (z_o, u_* and 1/L being the most important control parameters). The resistance to penetration across the atmospheric near-surface layer, where molecular transport dominates over turbulent transport and the resistance associated with pollutant-surface interaction, depend on the state and type of the surface, on solar radiation, surface T, relative humidity, amount of rain, dew or fog and pollutant exposure time.

MODELS AND MEASUREMENTS

HIRLAM (HIgh Resolution Limited Area Model) (*HIRLAM* 1990, 2002) is the hydrostatic weather prediction model used since 1990 operationally at the FMI. The forecast area extends from North America to Ural, from Northern Sahara to North Pole. It uses rotated spherical grid coordinates as its frame of reference in the horizontal direction: the pole has rotated to get the equator at the 60th latitude. Vertically the model has hybrid η terrain-following coordinates. The model versions have been updated and improved continuously since 1990, and the grid resolution has been changed several times. The version used in 1996-1997 was HIRLAM 2 with an improved radiation scheme, a parallelized version of it was used since the end of 1997. The current operational version run since February 1994 at the FMI is HIRLAM

6.2.1. A full scientific documentation of version 5.1 is available on-line in http://hirlam.knmi.nl/

Hilatar (*Hongisto* 1998, 2003) is of Eulerian type grid model for calculating and forecasting air quality situations at the background areas. The instant concentrations of the compounds are calculated by numerically solving the transport equation containing terms for advection, vertical diffusion, emissions, chemical transformation and dry and wet deposition. Vertical mixing is solved using the gradient transport theory: the turbulent fluxes are assumed to be proportional to local mean concentration gradients and the proportionality factor, the eddy diffusivity is analogical to molecular diffusion but 10^4-10^5 times stronger. The meteorological input parameters are taken from the 6-hour predictions of the HIRLAM model.

The Hilatar model has been applied in calculating in European scale the concentrations and depositions of nitrogen and sulphur compounds in background areas since 1995 June – simulations over Scandinavia cover a longer period. It has been used e.g. in estimating the nutrient flux to the Baltic Sea, factors that affect it and gradients from coastal areas to the open sea, in analyzing some dust episodes and for simulating heavy metal transport over the Nordic countries.

The model has been verified by comparing the daily concentrations of SO_2, NO_2, NH_3, SO_4^{2-}, NO_3^-, NH_4^+, HNO_3^+, NO_3^- and NH_3+NH_4^+ in air, and monthly mean wet depositions of $SO_4^=$, NO_3^- and NH_4^+, with EMEP measurements extracted from the EMEP/NILU database. Over the period June 1995 – September 1999 around 90 European EMEP stations and over the years 1993 and 1996-1998 data from 29 Nordic EMEP stations were used. Additionally the model has been verified against national and field campaign measurements. For verification of the wet deposition, un-accuracy of the measured precipitation common at the EMEP-stations, has some effect to the results. Differences in the monthly precipitation collected by close to the each other situated meteorological and an air quality gauges at the same station can be 50 %. (Hongisto et al., 2003, *Sofiev et al*., 2001, *Zlatev et al.*, 2001, *Schulz et al.,* 1999)

Jokioinen sounding station is located in South Finland at location 60° 49` 30.0"; 23° 30` at an elevation of 103 m from the sea level. The wind sensor is at a height of 30 m in a tower located in an edge of a pine forest with 20-25 m height trees. The temperature sensor is at 2 m.

COMPARISON OF ABL PARAMETERS, 1996-1998

The reliability of the HIRLAM ABL parameters is assessed by comparing 2 m temperature T(2m), relative humidity RH, precipitation, hmix, P, u*, 1/L and t* measured at Jokioinen or calculated from the Jokioinen soundings with those from the HIRLAM profile at the corresponding grid. The data covers all soundings in the years 1996-1998. Since for the years 1996-1998 the ABL parameters were not directly available from HIRLAM, they were estimated for both data sets with the same method, FMI meteorological preprocessor adapted from the local dispersion model system, as described in *Hongisto* (1998) p. 22-23. Profiles of wind u(z) and virtual potential temperature $\theta_v(z)$, instead of the dry potential temperature, and 6 hours time interval was used.

As annual averages the differences were very small as can be seen from Table 1, because the instant deviations, under- or overestimations cancel each other. When measures near zero are ignored, the annual relatively differences were highest for u* and T(2m), uabs, udir and precipitation, respectively. E.g. During the rather cold year 1996 the predicted temperature

was too warm, the ABL stability state was too label, the wind blow 20° more from southwest than measured and precipitation was underestimated.

Table 1. HIRLAM estimate minus the sounding value

	T(2m) oC	RH %	PREC mm	UDIR O	uabs M s^{-1}	1/L 1/m	t* oC	u* m s^{-1}	hmix m
1996	1.17	2.71	-169.85	19.19	0.51	-0.08	-0.05	0.16	8.33
1997	-0.10	2.63	-141.45	32.65	0.60	-0.13	-0.03	0.14	-18.55
1998	0.56	4.82	84.83	30.14	0.90	-0.08	-0.02	0.19	-32.66

Comparison of the measured and modelled monthly averages is seen in Fig. 1. HIRLAM surface seems to be too cold in summer, too warm in winter. HIRLAM generates slightly too high winds at 30 m, and too much mechanical turbulence. Hmix differences are most important for the model results. Fig. 1 illustrates, that the HIRLAM monthly average hmix was during some spring months down to -40 % smaller as that estimated from the sounding profile, while in June and July the anomaly can be +40-+30 %. In 1996 the respective monthly absolute positive and negative anomalies were +100 ...150 m and -250... -100 m in spring, +400...+350...and -300...-200 m in June and July 1996. The night-time difference was in 1996 +- 150 m throughout the year, while during daytime the instant difference could exceed 2 km.

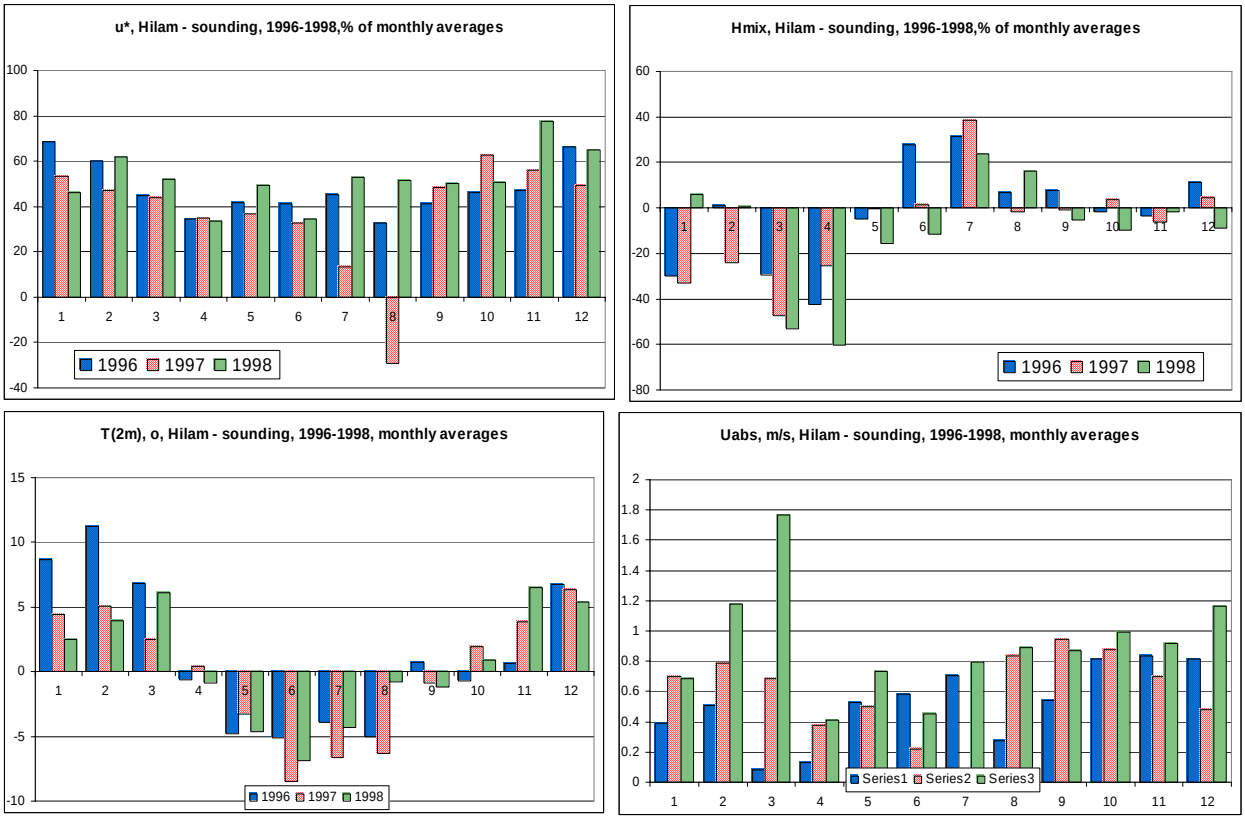

Figure 1. Monthly average difference of some predicted or measured ABL parameters, Jokioinen.

The correlation with the annual values was e.g. for the year 1998 0.64, 0.78, 0.72, 0.27 and 0.76 for hmix, uabs, T, 1/L and u*, respectively. The correlations were usually higher during the summer period.

COMPARISON OF ATMOSPHERIC BOUNDARY LAYER PARAMETERS CALCULATED BY DIFFERENT HIRLAM VERSIONS

Additionally, the 6th hour forecasted hmix, P, uabs, udir, T(2m) and q(2m), and calculated u*, 1/L and t*, estimated with two HIRLAM versions (ATA-, HIRLAM 4. with 0.4° grid and ATC-, HIRLAM 5.1 with a 0.3° grid) running operationally in parallel in winter and spring 2003, were compared over the Scandinavia. The differences vary geographically. In January 2003, hmix ATC-ATA was +50...+250 m over Norwegian Sea and all Scandinavian land-areas, but up to -100...-250 m negative over the Baltic Sea. Precipitation increased up to 50 % in Europe. All parameters changed, but the direction did depend on the area.

EFFECT TO THE MODELLED TRANSPORT DISTANCES

These two meteorological data sets: ATC- and ATA fields of different grid resolution were used to simulate the long-range transport and deposition of nitrogen and sulphur compounds over Europe in Spring 2003. While the emission inventory was updated for the new simulations, the generally declining emission trend produced slightly smaller depositions in ATC-runs. Increase in the model resolution also led to a finer scale structure in the deposition. E.g. in January, as a results of the change of the forcing fields it was found, that in Scandinavia the increase of the ABL height by 50-250 m resulted to smaller surface concentrations in dry situations. The precipitation increase in Europe and in the North-Eastern Scandinavia (by up to 50 %) increased wet deposition over the respective areas. Decrease in friction velocity u* in Scandinavia yielded to lower dry deposition flux. The local changes in temperature, moisture and wind parameters should have an effect to mixing and to chemical transformation rates, however the differences are instant, and while the effects partly compensate each other, the comparison should be made by detailed process analysis. In general, the use of ATC-meteorology leads to shorter transport distances in Europe and higher depositions near the sources. In the target areas, e.g. in Finland, use of the new data will reduce calculated deposition estimates in winter.

CONCLUSIONS

From the operational HIRLAM verification studies as well as this 3-year comparison of T(2m) it is clear that the diurnal cycle of the temperature is strongly underestimated in the forecasts. During the daytime the temperatures are too low and in the night-time too warm. The same is also seen in the two meter humidity values. This feature is most prominent in winter and in spring, however, well seen also in summer. This leads to a conclusion that strong inversions, important for air pollution studies, are difficult to forecast. In spring the difficulties are often during the day, when the predicted temperature is too cold. In this short study the highest discrepancies in the ABL height were found in summer.

HIRLAM parameterization is constantly under development to reach more reliable forecasts. For air pollution studies, it is very important to improve further the ABL scheme in it.

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