2.3 Transport Modelling over Sea Areas

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Summary

A high-resolution mesoscale model has been developed to study the deposition and concentration fields of acidifying compounds over specific regions of Finland. The model uses EMEP advected concentrations as boundary conditions at the model domain (part of Finland or the Baltic Sea Basin). The effect of the presence of the sea along the southern coast of Finland was particularly investigated. Results show that the different stability and consequently mixing conditions over the sea lead to measured and modelled concentration and deposition values not expected from the overland conditions prevailing at the source.

Aims of the research

Pollutant dispersion conditions in the atmospheric boundary layer (ABL) over sea areas differ in many respects to the overland situation due to dramatic differences in surface roughness, heat content and humidity, leading for instance to a reversal in thermal stability, a step-like change in the ABL height and enhanced humidity catalysed chemical reactions. Finland is surrounded from the west to the southeast by the shallow and very sensitive Baltic Sea which separates it from the major European pollution source areas. The goal of the work was to study the origin and processes driving the fate of acidifying air pollutants over specific areas of the Baltic Sea, and get understanding, how the marine environment affects the pollutant transport in the long-term and during specific episodes. For this purpose we have developed a mesoscale model with a resolution of 0.1°–0.5°, enough to be able to simulate mesoscale patterns in the meteorological and pollutant fields, as well as to identify the relevant processes influencing the observed variability, and to trace back the source of pollution episodes. Special attention was put on studying the impacts of large bodies of water as well as inhomogeneities at the coastline on pollution transport.

Principal scientific results

Model developments

During this project two models with the same general structure, but differing in their meteorological inputs and preprocessors, have been developed. The domain of the models includes either parts of Finland with its surroundings or the Baltic Sea Basin. Vertically the models reach a height of about 3 km with 7–10 layers.

The Finnish limited area model for oxidised nitrogen compounds FINOX, and its sister FISOX for acidifying compounds, use synoptic measurements and soundings as meteorological input data. Their vertical distributions are calculated using boundary layer profile functions. The models have been used to study the acid load and pollutant concentrations with a 30 km grid over southern and central Finland.

On the other hand, the HILATAR model uses 6-hour weather predictions from HIRLAM (High Resolution Limited Area Model) with a 0.5° horizontal resolution. The simulation area is flexible and subareas with a resolution of *e.g.* 0.1° (11 km) can be used for regional and local air quality studies. The meteorological parameters are interpolated from the original data incorporating a rather exact sea-land distribution. Thus, the abrupt changes in the meteorological conditions at the coast would be simulated. The boundary layer height, cloudiness, and turbulence and stability parameters are calculated from the vertical temperature, specific humidity and wind profiles, and using surface values. The long-range transported (LRT) share of pollutants is simulated by adding daily average concentrations to the air inflow at the boundaries as calculated by the EMEP-MSC W model.

Three chemical submodels are connected to the transport module: 1) the oxidised nitrogen chemistry used in the FINOX model; 2) the EMEP acid model chemistry (Iversen *et al.*, 1990) with modified sink terms, and 3) a simple sulfur transformation scheme. A 1-D cloud module for the oxidation of sulfur in water droplets with pH dependent conversion rates has been constructed for testing the conversion rates during the transport. Cloudiness and the liquid water content are estimated from the specific humidity profile and rain. Close to the sea surface the sulfur conversion rate is modified with an additional friction velocity-weighted parameter.

Dry deposition over the different types of surface is calculated using actual meteorology and the resistance analogy. The surface resistance of the canopies depends on solar radiation, temperature, relative humidity, the rain amount, dew or fog, and the pollutant exposure time. The scavenging rate coefficients have specific formulae for particulates and gaseous substances, for below-cloud and in-cloud scavenging and for rain and snow. Close to the emission SO_2 is assumed to be transformed from an insoluble to a soluble form with a 2-hour half-life. The precipitation rate is assumed to decrease with height following a seasonally varying vertical weighting factor.

The model and methodology of Joffre (1988) and Lindfors *et al.* (1993) have been used to estimate the dry deposition velocities and stability conditions over sea areas in the FINOX model. For the HILATAR simulations the HIRLAM surface roughness over the sea and coastal areas has in some applications been redetermined in order to better characterise the marine conditions.

For the Baltic Sea drainage basin, the gridded EMEP emissions with a 150 × 150 km^2 resolution are available but without specific technical characterisation of

the sources. For some parts of the area (Finland, north-west Russia and Estonia), a data base with exact location and stack parameter information of the largest sources, has been collected. For the rest of the area it is planned to use the EC CORINAIR emission inventory.

Model simulations

Determination of the nitrogen deposition over Finland

The load of oxidised nitrogen compounds over Finland and its adjacent marine areas has been calculated with the FINOX model. The deposition for 1985 and 1988 was generally higher than previous estimates by the one-layer EMEP model, and the domestic share of the deposition was higher. The origin of the deposition calculated in the FINOX simulations is different since not all sources contribute to the deposition: *e.g.*, during wintertime and at night when an inversion isolated the high emission sources from the ground. In the EMEP model, on the other hand, all the emissions are mixed inside the boundary layer. Comparison with measurements shows that the precipitation weighted deposition of nitrate was underestimated. However, it was strongly influenced by the added LRT share from the EMEP model.

The interannual seasonal variation was clear, depending partly on the varying emissions, meteorological conditions and role of the LRT contribution. Over parts of the Gulf of Finland and of the Bothnian Sea area the modelled deposition was 0.07–0.3 g (N) m^{-2} in 1985 and 0.1–0.35 g m^{-2} in 1988, where the average wet deposition share was 82 % in 1985 and 74 % in 1988. The LRT share was 73–83 % of the total deposition over the model sea area in 1988, and being more than 90 % along the south coast of Finland, while the LRT share of wet deposition was higher. Lindfors *et al.* (1993) found as a mean for the period 1980–1986 that the total nitrogen deposition over the Baltic Sea area was about 0.7 g m^{-2} in the northern part of the Baltic Sea (being about twice that value in the southern parts). Lindfors *et al.* (1993) also found that their empirical assessment overestimated EMEP model results by about 60 %.

The year 1988 was warmer, with more clouds and rain, while during 1985 winter months the surface wind were weaker and the mixing heights correspondingly lower. In 1985 and 1988, both models and measurements at the coastal station of Virolahti, showed an increasing trend in the nitrate content of precipitation in winter and a decreasing trend in the other seasons. At the marine station of Utö the measured deposition increased in winter and spring, and decreased during the other seasons, while the modelled deposition increased only in springtime.

Episodes over northern Finland

The HILATAR model was first tested by simulating the dispersion of the huge sulfur emissions from the Kola peninsula. Results were compared with measurements in Northern Finland and Russia during two periods: March–June 1991 and the end of November 1991 (Hongisto, 1992). The model produced quite realistic surface concentration distributions at stations near the sources. For longer

transport distances, the 55 km horizontal resolution seemed to be too scarce for describing northern dispersion conditions during stable situations. Measured concentrations in Lapland generally consist of high short-duration peaks even at more than 250 km away from the Kola sources since the plumes seem to persist for long periods of time.

Transport from Russian and Estonian sources over the Bay of Finland

The 11 km resolution, 10-layer HILATAR model was used for evaluating how much of the sulfur pollution in 1993 originated from north-western Russian and Estonian sources. For this study a special emission inventory has been prepared in co-operation with the Russian and Estonian authorities. (Häkkinen *et al.,* 1995). The time-variation was that used by the EMEP MSC-W acid deposition-model, with additionally a diurnal weighting factor.

The operational HIRLAM used in 1993 too few actual marine grid points for Lake Ladoga and the Gulf of Finland east of 25° E. Thus, the ABL stability conditions over the coastal and even marine areas resembled those over land. Recalculating the surface roughness with the wind dependent Charnock formula, and keeping the rest of the meteorological vertical profiles unaffected, yielded the monthly average of the inverse Monin-Obukhov length 1/L, of the friction velocity u_* and of the number of occurences of stability classes to be representative of marine dispersion conditions. The simulated surface concentrations were verified with the measured average concentrations and wet deposition at the coastal EMEP station of Virolahti.

The origin of the simulated concentration and deposition events was sought by calculating the backward trajectories to Virolahti for the whole year at 6 h intervals. The identification of individual sources using one-layer trajectories was not always successful since the highest concentrations corresponded generally to stable situations with a strong vertical veering of the wind direction. Also, since trajectories had to cross the sea before reaching the Virolahti station, situations with enhanced convection above the sea mixed pollutants originating from different places.

Even with a local scavenging correction, wet deposition represented about half of the modelled deposition. The identification of sources for the wet deposited sulfur cannot be made exactly since during most of the year rain is connected with fronts. These fronts appear in the baroclinically instable zone of cold polar air and warmer, often moist and marine, air advected to the north-east direction, and which are even amplified over the warm sea during late summer and autumn. If a low pressure centre passes north of Virolahti over Finland, westerlies prevail. During the overpassing of the front wind direction changes rapidly, even by 180–360°, and trajectories sweep swiftly over all source areas. Consequently, all Baltic and Russian sources can contribute to the sulfate amount in rain. On the other hand, if the pressure centre follows the northern route, warm and cold fronts bring rain when passing over Finland from the west. Even when surface winds turn to the South before the frontal passage, the upper layers blow from the west or south-

west bringing pollutants from western and central Europe. Bearing this in mind, and by studying the calcium concentration and acidity of the wet deposition sample at Virolahti, it can be concluded, that at Virolahti sulfate in rain has several origins: an Estonian one with calcium - and an acidifying central-European one.

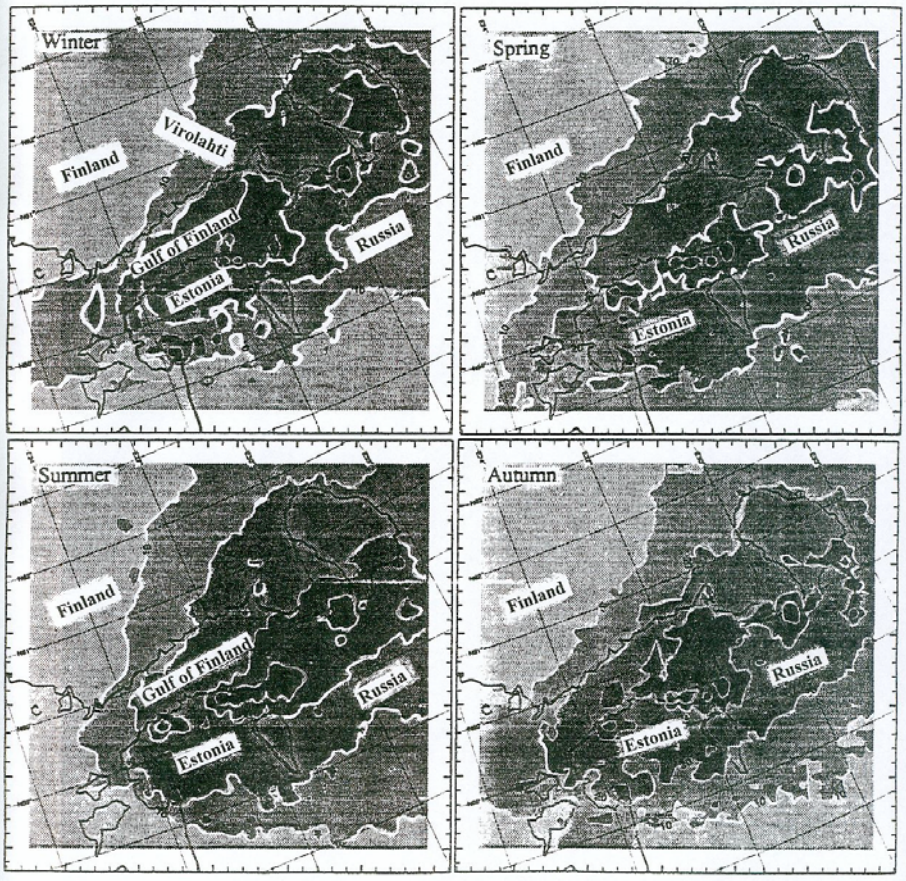

Fig. 2.3.1: Seasonal variation of the total sulfur deposition over the Gulf of Finland in 1993; contribution of the Estonian and north-western Russian sources (Contours of 10, 20, 50 and 100 mg(S) m^{-2}.

The sea surface roughness correction seemed to be physically sound and has provided good correlation with measurements. However, when analysing the surface concentrations of late autumn 1993, it appeared that the correction should not be used. After 4 November the daily average temperature falls below zero in south-eastern Finland, the sea starts to freeze after 11 November near St. Petersburg, and the ice cover grows to cover a sector more than 1° wide on 23 November. HIRLAM contains climatological sea parameters such as the ice extent. With the original high roughness, stable conditions over land prevailed also over actual sea ice and a good correlation with measurements could be detected for

trajectories originating from St. Petersburg. On the other hand, when the modified sea parameters were used without knowing the exact ice cover, the correlation was lost and the modelled concentrations were underestimated due to the mixing over the sea.

The calculated seasonal deposition distributions for 1993 are presented in Fig. 2.3.1 The annual deposition in 1993 on the Finnish side of the border did not exceed 0.1 g (S) m^{-2}, except near the Svetogorsk industrial complex. The calculated deposition ranging between 0.1 and 0.7 g (S) m^{-2} for 1993, is comparable with the average 1985–1993 deposition estimated by EMEP/MSC-W (Tuovinen *et al.*, 1994) for the two EMEP squares in south-eastern Finland.

The Estonian oil shale power plant emissions dispersions were calculated separately, because they are thought to have a major influence to the south of Finland. The simulated concentration distribution in 1993 showed that especially during springtime polluted air is transported on higher layers over Finland. During most of the time the mixing over the sea was not efficient enough to transport down the emitted materials.

Conclusions

In evaluating the FINOX model results with respect to the specificity of sea-areas, one should note that the comparison with marine measurements was made with the average values of the nearest grid square. At those locations, deposition is strongly dependent in offshore winds on the advected concentrations from the EMEP model. The grid-array was too sparse to simulate the sea-breeze circulation, but the stability differences between land and sea areas as well as the vertical exchange in the coastal region due to flow velocity changes are, however, described.

It can be concluded that the Estonian sulfur emissions do not have as much contribution to the wet deposition over Southern Finland as previously estimated (Plancenter, 1991). The reason lies in the wind direction frequency prevailing under rainy periods. Additionally, sulfur is neutralised by the emitted alcalic particles and these particles dropped down not far away from the sources. Thus, the measured acidity of rain can decrease by even several pH units between the Estonian and Finnish South coast.

Sea areas have a seasonal effect on the transport and mixing. For instance, in autumn (unstable conditions with a warm sea and cool air) high stack emissions on the Estonian side of the Gulf of Finland can be much easily observed in Finland than in winter, when mostly only surface/ low stack source contributions are detected.

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