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Meteorological and climatological factors affecting transport and deposition of nitrogen compounds over the Baltic Sea

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We studied the variation of the nitrogen deposition to the Baltic Sea with the Eulerian transport-chemistry model Hilatar. We analysed the effects of the climatological and meteorological conditions on the temporal variations of the pollution load to the sea over the period 1993–1998. The nitrogen deposition to the Baltic Sea and the concentrations at coastal stations showed strong monthly, seasonal and inter-annual variations and, apparently, no decrease with declining emissions over the period. We discuss the representativeness of coastal measurements for inferring the deposition flux to the open sea, as well as the episodicity affecting this pollution load. We also compare our results for the nitrogen deposition to the Baltic Sea with the previous estimates and discuss the implications for ecological modelling.

Introduction

The Baltic Sea is a young, shallow, brackish water basin with a relatively small volume and a long residence time for its water. Its aquatic biota is very poor in species, and the food chains are vulnerable to external disturbances. The high nutrient input can fuel excessive algae growth and threaten the oxygen content of the bottom waters (HELCOM 2001). Nitrogen (N) is the limiting factor for primary production in the Baltic Sea, except in its northernmost part (the Bothnian Bay) (Kuparinen and Tuominen 2001). As plankton ecosystems are strongly regulated by hydrodynamics, coupled atmospheric scales (diurnal, synoptic, seasonal) are also very important (Kononen 2001) for an understanding of algal bloom/eutrophication processes. Though the main sources of excess nutrients to the Baltic Sea have, by and large, been identified, neither the overall dynamics, nor the relative importance of the processes and pathways involved, such as internal loading, run-off and airborne deposition, have been adequately quantified. It is thus relevant to refine assessments of the distribution and variability of the atmospheric share of the N-budget of the Baltic Sea.

The airborne nitrogen input to the Baltic Sea is significant: it has been estimated to be between 30% and 50% of the corresponding load via rivers and direct coastal discharges (HELCOM 1996, 2001). The latter load has large inter-annual variations and uncertainties depending on riverine run-off and winter temperatures. Current annual atmospheric load estimates vary between 225 and 325 kt (N). The relative importance of the air pollution load increases towards the open sea, since the nitrogen load from rivers mostly impacts coastal areas, and half of it is released via five main rivers (HELCOM 2001).

Average load estimates hide a lot of a variability that is partly systematic (linked to climatology and related to the position of the source areas with respect to receptors), and partly random (due to meteorological variability). All these estimates also include several uncertainties depending on the estimation methods used (models, measurements, hybrid). Monthly averages smooth out strong episodic loading, while models using coarse grid sizes do not resolve well the strong gradients in emissions, concentration and meteorology in the vicinity of coasts. It is thus relevant to assess the importance of the sources of variability and whether a high temporal resolution leads to distinctive results.

Some of these issues were partly investigated during the EU-BASYS (Baltic Sea System Study) subproject "Air pollution load to the Baltic Sea". The primary objective of this subproject was to calculate and simultaneously measure the fluxes of airborne pollutants (nitrogen and heavy metals) to the Baltic Sea on a multi-annual, monthly and daily basis using a high spatial resolution. This entailed performing nested modelling studies in combination with dedicated experimental campaigns on two ships, at four land stations and by separate process study measurements. Furthermore, we tested the accuracy of some of the physico-chemical parameterisation schemes used in the models.

The purpose of this paper is to report on the modelled 1993–1998 N-flux estimates generated for the BASYS-project, and to study the various types of variability, how they contribute relatively to the overall load, what favours their occurrence and how they should be accounted for in monitoring and assessment efforts. Such results are relevant for ecological modelling and for impact studies for environmental policy measures.

Tool: the Hilatar model

The atmospheric load was estimated with the Eulerian chemistry-transport model Hilatar, which provides gridded estimates of the fluxes of sulphur, nitrogen and heavy metals in a domain covering either the whole of Europe (*see* Fig. 1a; ca. 56 km grid) or the Baltic Sea region (Fig. 1b; ca. 27 km grid). Here we only outline the main characteristics of Hilatar, since it has been extensively described in Hongisto (1998, 2003a, 2003b). The meteorological input consists of the six-hour predictions of the operational numerical weather model HIRLAM (HIgh Resolution Limited Area Model) of the Finnish Meteorological Institute (FMI), interpolated to one-hourly values. The Hilatar model uses the rotated spherical HIRLAM grid.

The dry deposition module uses the resistance analogy with specific aerodynamic, surface and molecular resistance schemes. Over the sea, the dry deposition velocities v_d are calculated using the parameterization of Joffre (1988) and Lindfors *et al.* (1993), and for particle deposition using the scheme of Williams (1982) with the empirical size-distributions of Hillamo *et al.* (1992) and Pakkanen *et al.* (1996). Over land, v_d is mainly based on the formulation of Voldner *et al.* (1986) and Wesely (1989). The non-linear, height-dependent wet scavenging coefficients follow principally the expressions of Chang (1984, 1986) and Asman and Janssen (1987). Turbulence parameters and the mixing height are calculated from the HIRLAM temperature, wind and specific humidity profiles using separate atmospheric boundary-layer pre-processors. The chemistry scheme is a modified and updated EMEP MSC-W acid model scheme (Iversen *et al.* 1989). It resolves gaseous (SO_2, NO_2, HNO_3, NH_3, PAN) and particulate (SO_4^{2-}, NO_3^-, NH_4^+) compounds.

The Baltic Hilatar version was used for the entire period 1993–1998, using the long-range transported concentrations at the model boundary from the EMEP MSC/W model for the years 1993–1995, and those from the European Hilatar version for 1996–1998. The model has ten vertical layers below three kilometres, while since June 1995 three additional levels between three and ten kilometres have been included. We use gridded EMEP emissions, complemented in the Baltic area by detailed stack emission inventories for which the effective plume height is calculated with Briggs' formulae. The monthly emissions are country-specific (Friedrich 1997) and traffic emission indices have weekly and diurnal cycles.

The basic verification and validation against experimental data have been reported in Hongisto (1998, 2003a). Hongisto *et al.* (2003) performed a more extensive validation against over 90 EMEP station measurements of 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

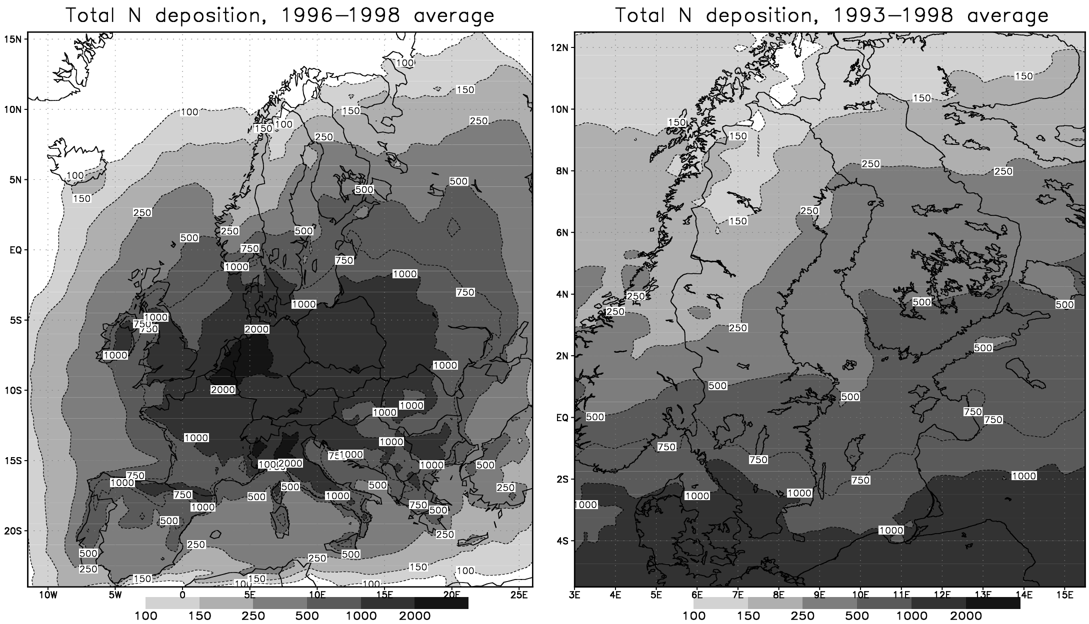

Fig. 1. Average total N deposition over the European and Baltic Sea model domain for three and six years (mg (N) m^{-2}), presented in the HIRLAM grid (rotated spherical coordinates where the equator goes along the 60 latitude).

depositions of SO_4^{2-}, NO_3^- and NH_4^+, extracted from the EMEP/NILU data base (www.emep.int). Furthermore, an inter-comparison of various measured and modelled nitrogen species as well as Pb, Cd and Zn concentrations in aerosol and in precipitation during the BASYS field campaigns at four coastal stations and two research vessels is reported in Sofiev *et al*. (2001), Schulz *et al*. (1999) and Schneider *et al*. (1999). An intercomparison between other Scandinavian Eulerian long-range transport models and Hilatar is presented in Zlatev *et al*. (2001). All these simulations showed that by and large Hilatar yields satisfactory results that are generally within a factor of 1.5–2, though some larger discrepancies may occur locally and in southern Europe.

The model tends to underestimate NO_2 and overestimate NO_3 and HNO_3 concentrations. During episodes, this is a consequence of the initial dilution of the emitted plumes to grid averages and the subsequent horizontal dilution of sharp peaks during advection, as well as chemical conversion rates that are possibly too rapid. For background values, this is the result of neglecting non-European contributions and missing some natural emissions.

The technical testing of Hilatar (Hongisto 1998, 2003a, 2003b) showed that the main uncertainties were connected with both horizontal and vertical grid resolutions, the emission inventory, diagonal advection and the simplified description of complicated air chemistry. The uncertainties in the weather forecast used as meteorological input are a rather significant part of the error sources (Hongisto 2004). The slight over-prediction of weak precipitation coupled with efficient scavenging at low rain rates yields rather high deposition background values. A comparison of HIRLAM-modelled profiles to radiosoundings during the BASYS ship experiment showed that, over the sea, the model yielded lower mixing height values.

Results: Simulated nitrogen deposition to the Baltic Sea

Inter-annual variability and comparison with other estimates

The six-year average total sum of oxidised (NO_x) and reduced nitrogen (NH_x) deposition over the

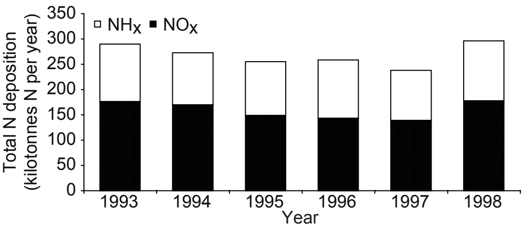

Fig. 2. Annual deposition to the Baltic Sea in kt(N), 1993–1998, for oxidised nitrogen NO_x, and reduced nitrogen NH_x.

northern parts of the Baltic Sea had a slight southeast–northwest decreasing gradient, while it increased towards Denmark in the southern parts (Fig. 1). The deposition decreased from 1200 mg (N) m^{-2} along the south-western coast of the Baltic to 400 mg (N) m^{-2} over the Finnish archipelago, falling to below 300 mg (N) m^{-2} over most of the northern part of the Gulf of Bothnia.

The annual total N deposition to the open Baltic Sea area (391 000 km^2) was highest in 1998 (Fig. 2). Over 50% of this load fell on the northern HELCOM sub-areas: the Gulf of Bothnia (B1), the Gulf of Finland (B2) and the northern Baltic Proper (B3), the values being 16%, 6% and 31%, respectively, as compared with 35% for the southern Baltic Proper (B4) and 12% for the Kattegatt–Belt Sea (B5). The inter-annual variation of the geographical distribution is not very high.

The total N deposition comprised mainly wet deposition (around 80% for NO_x, and 65% for NH_x compounds). Nevertheless, dry deposition could be dominant during dry summer months, as in July 1994 (75% for NH_X and 71% for NO_X), although the lowest v_d values generally occur during spring and summer over the sea. During the study period, the dry deposition of oxidised nitrogen declined from its 1993–1994 levels, while the wet deposition increased. Due to the inherent deposition gradient occurring over coastal grids, in this work the nitrogen load was calculated only over those grids where the open-water share was at least 65%.

Our estimates of the total N deposition to the Baltic Sea are compared in Table 1 with empirical estimates (HELCOM 1991, 1996, Lindfors *et al*. 1993), with EMEP MSC-W model calculations (Bartnicki *et al*. 1998, 2002, 2003) as well as with the model results of Hertel *et al*. (2003). It is noteworthy, however, that all these models use a different Baltic Sea area mask.

The inter-annual variation of the total N deposition is high, but these estimates give a generally decreasing trend for the nutrient load over the last 20 years. However, all models report a rather high N deposition in 1998–2000, close to the values occurring at the end of the 1980s. Moreover, neither the N deposition to the Baltic Sea nor coastal concentrations have decreased linearly with reported European N emissions (www.emep.int) during the period studied. NO_x emissions in the EMEP area declined by 6.2% between 1993 and 1996, but the decline was strongest (–6.3% just between 1995 to 1996) in the area most influencing the Baltic Sea (i.e., the riparian countries, UK, The Netherlands and France).

Table 1. Model (M) and empirical (E) estimates for the total N deposition (in kt yr^{-1}) and flux density (mg m^{-2}) to the Baltic Sea.

Total deposition	Unit	1980–1986	1988–1990	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002
EMEP	kt yr^{-1}	329(1986)[1)]	324–311[1)]	273[1)]	268[1)]	248[1)]	261[1)] –290[2)]	266[2)]	325[2)]	300[2)]	317[2)]	224[3)]	
This work[4)]	kt yr^{-1}			290	272	255	258	238	296		274[4b)]	263[4b)]	224[4b)]
HELCOM	kt yr^{-1}	395[5)]	314–330[6)]										
Others	kt yr^{-1}	414[7)]	298–343[8)]							318[9)]			
EMEP[1–3)]	mg m^{-2}	826	821–780	685	718	622	655–728	668	816	753	796	562	
This work[4)]	mg m^{-2}			742	696	652	660	609[4a)]	757[4a)]		701[4b)]	673[4b)]	573[4b)]
Other	mg m^{-2}									684[9)]			

[1)]Bartnicki *et al*. (1998), 420 603 km^2, (M) and HELCOM 1997. [2)]Bartnicki *et al*. (2002), (M). [3)]Bartnicki *et al*. (2003), (M). [4)] This work, open water area, 391 000 km^2, (M). [4a)]1996 emissions. [4b)]1998 emissions. [5)]HELCOM 1991 (E, only wet). [6)]HELCOM 1996 (E, only wet). [7)]Lindfors *et al*. (1993) (wet E, dry M). [8)]Dedkova *et al*. (1999) (range for 1987–1991). [9)]Hertel *et al*. (2003) 464 406 km^2, (M).

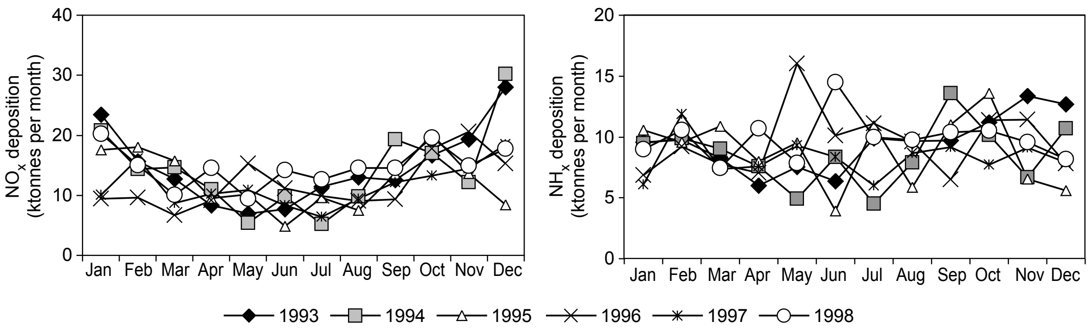

Fig. 3. Monthly deposition of oxidised and reduced nitrogen to the Baltic Sea, kt(N) per month, 1993–1998.

Our results are within the range of the EMEP MSC-W calculations that indicate a decline in the total deposition load between 1986 and 1995 and an increase to 325 kt(N) in 1998. The EMEP MSC-E estimate for the 1987–1991 load is of the same magnitude (Dedkova *et al*. 1993). The deposition estimates for the southern Baltic using Hilatar were approximately the same as those derived from the BASYS field campaign measurements for both the summer and winter periods (Schulz *et al*. 1999). Before 1995, the empirical estimate of Lindfors *et al*. (1993) and the Hilatar model, both with high-temporal resolution data, yield slightly higher deposition values than estimates based on either gross empirical values (HELCOM) or the larger grid-size model (EMEP). The total N deposition density calculated with Hilatar after 1996 is slightly smaller than the respective EMEP estimates. In the Hertel *et al*. (2003) study, the load density for the year 1999, 684 mg m^{-2}, is 9% lower than the EMEP estimate.

The largest differences between the Hilatar and EMEP model results occur in the southern Baltic Proper. Ammonia deposition was higher in the EMEP model over the Gulf of Finland. In 1996, Hilatar calculated slightly smaller dry deposition and slightly higher wet deposition values than the EMEP model. The models were of different type having, e.g., different horizontal resolutions (EMEP: 150–50 km, Hilatar: 27 km), as well as different vertical structures, physical parameterizations, sink rates, meteorological input and temporal variations of the emissions. In the Europe version of Hilatar, compounds were rather effectively lifted up into higher model layers over coastal or mountainous areas in specific wind directions, and also over the open sea in winter and during convective events. This resulted in lower surface concentrations and dry fluxes than in the one-dimensional EMEP model, where all emitted compounds are initially evenly mixed inside the mixing layer.

Seasonal variability

The intensity of algal blooms varies seasonally in the Baltic Sea, with the phytoplankton biomass being highest during the spring bloom (e.g., Stipa *et al*. 2002). The derivation of representative estimates of the N flux to the water on this temporal scale is therefore essential to evaluate the importance of the external load for eutrophication of a given sea basin.

NO_x deposition was highest during the winter months, although the inter-annual variation was large (Fig. 3). Wintertime NO_x deposition had a clear maximum in 1993–1994 and in 1998. Summer deposition was smallest in 1994–1995. Three individual months (Jan. 1993, Dec. 1993 and Dec. 1994) displayed NO_x deposition about twice as high as the winter average. Monthly NH_x deposition is rather evenly distributed throughout the year, with a similar large variation between individual months.

Towards the end of the period, wintertime dry deposition of oxidised N decreased from the 1993–1994 level, while there was no clear change in the summertime deposition. Wet deposition was higher during the last two years of the period, because the summers of 1993 and 1994 and the winter of 1996 were rather dry, while precipitation was heavier in spring 1995, 1997 and 1998 and the summer months of 1997–1998, when emissions were lower.

The difference between the time variations of the NO_x and NH_x deposition is due to the monthly and shorter-period variation of the emissions. In wintertime, when weak solar radiation is the limiting factor for algae blooms, nutrients accumulate in the ecosystem, because NO_x emission factors for January–February are 33% higher than the yearly average. On the other hand, NH_3 emission intensity grows from 0.7 of the yearly average in January to 1.3 in June and July, and thus ammonium contributes more to the summer bloom, when NO_x emissions are only 67% of their yearly average. Moreover, NO_x emissions have a diurnal cycle with a maximum in the morning and afternoon traffic rush hours, while NH_3 emissions do not have such a diurnal variation.

Since algal bloom processes can be triggered on scales comparable to meteorological variability (Kononen 2001), our results show that, in order to adequately simulate algal bloom events, marine hydrodynamic-ecological models should be forced by the outputs from meteorological-chemistry transport models with high temporal resolutions.

Gradients to the open sea

Marine and land areas follow a reversed meteorological cycle, particularly around the shallow Baltic Sea. The atmospheric boundary layer is more stable over land during the winter, but over water during the spring and early summer, if warm air spreads over the cold sea. Over land convective turbulence is strong in summertime, but over the sea in the autumn and early winter.

Coastal measurements are used to represent the deposition over Baltic Sea sub-area, e.g., in HELCOM (2002). The extrapolation of the coastal data to the open sea is not always sound, due to the stepwise change of all meteorological parameters and their gradients along the shoreline, with a seasonally-varying direction. High-resolution simulations provide one approach for assessing the gradients from land to sea, as these simulations can cope with the changing meteorological conditions over this discontinuity.

Quantitative estimates of coastal gradients by the Hilatar model (Hongisto and Sofiev 2001) indeed show that short-term gradients are very site-specific, depending on the prevailing wind direction with respect to coastal emissions, and on the season and the chemical form of the substance. These gradients are highest in summer. In winter, over the area from the Baltic Proper up to the Bothnian Bay, the six-year average total N deposition increased by 20%–80% from the Swedish coast eastwards towards Finland. Over the southern Baltic Proper, winter deposition decreased northwards from the emission-intensive coastline of Germany. Over the Gulf of Finland, winter deposition decreased southwards from the Finnish coast towards Estonia. Along the Norwegian Atlantic coast, the coastal deposition in winter was smaller than both that over the nearby sea and that over mountain areas. Thus, the land–sea gradient can be in any direction and may be opposite in direction at different locations because the climatological conditions are so different in the southern, northern, eastern and western parts of the sea.

During summer months, the deposition was generally everywhere smaller over the sea. At the latitude of the northern Baltic Proper, the total N deposition over the Baltic Sea was around half of that at the Swedish coast, and 70% of the deposition at the Estonian coast. Thus, during both seasons, deposition measured at the coast line does not always represent the conditions over the water.

The development of ship emissions

The annual international ship traffic emissions in the Baltic Sea were estimated twice by Lloyd's Register for the EMEP MSC/W, for the 1980s and for 1990. The new ship emission inventory is now 14 years old. Since 1990, following the political and economical changes in the Russian Federation and the Baltic States, a rapid increase in the ship traffic volume, especially in the Gulf of Finland, has occurred. The oil pipeline from Russia (currently the world's largest oil producer) to Ventspils in Latvia has been closed and Russian container traffic to the Baltic States' harbours has been reduced. The new Russian harbours of

Primorsk and Vysotsk have been established in the Gulf of Finland. Simultaneously, the export volume from St. Petersburg and Kaliningrad as well as from the Estonian harbours of Tallinn, Muuga and Paldinski have been expanded significantly. In Estonia and Russia, eight new oil export harbours along the shores of the Gulf of Finland are under construction or in the planning stage. Additionally, the passenger traffic between Finland and Estonia, which started in 1965, has increased by a factor of ten since the 1980s.

The most recent estimates of the EMEP ship emissions in the Baltic Sea, for the base year 1990, amount to 353 kt NO_2 (*vs*. 80 kt previously) and 229 kt SO_2 (72 kt SO_2 previously) (Bartnicki *et al*. 1998, 2001). Simulating the year 1993 with these higher ship emissions yielded NO_x deposition to the Baltic Sea that increased by 8% (total), 12% (dry) and 7% (wet) as compared with previous simulations. The maximum increase (19%) in the monthly dry deposition occurred in June. Due to the increased traffic volume from Russia and the other Baltic States through the Danish Straits, we can roughly estimate that the calculated deposition to the Baltic Sea could be underestimated by at least 5%–10%, with this underestimation being stronger over the Gulf of Finland.

Influence of meteorological conditions

Flow direction and pressure anomalies

Atmospheric circulation patterns determine the flow direction over the different sub-basins of the Baltic Sea, thus strongly influencing the time variation of nitrogen deposition during rainy periods. This influence was studied using the weather maps of the German Weather Service (DWD 1993–1998), in which meteorological situations are classified according to the surface pressure distribution over Europe, western Russia and the Atlantic using the Hess-Brezowski weather type classification.

The most relevant weather types for our study are the zonal circulation types Wa, Wz and Ws (Western anticyclonal, cyclonal and southern flow, respectively). South-westerly flow over the Baltic Sea is strongest when cyclones move across the Atlantic towards the northern Norwegian coast. If they take a more southerly route, the flow direction over each sub-basin does not remain constant for long: these local pressure anomalies cause changes in the wind direction. During these events, the trajectory analysis fails to identify the source area, while rapidly-turning trajectories sweep over all the nearby areas. After the event has passed, the wind blows from the opposite direction.

Using the weather types listed in Table 2, we studied the main flow sectors prevailing over sub-basins B3–B5. The northern sub-basins B1–B2 were generally more affected by north-easterly flow than the Baltic Sea proper, and were thus omitted.

A zonal circulation pattern prevailed from 1993 to spring 1995 and also in 1998, while the large-scale circulation changed to a meridional type (high west-east pressure difference) for almost the whole of the two intermediate years, being strongest in 1996 (Fig. 4). The seasonal variation of the duration of the grouped weather types is presented in Fig. 5. During the winter, the share of primarily

Table 2. Main flow direction over sub-basins B3–B5 classified according to the large-scale weather types of Hess-Brezovski (1993) with the following abbreviations: N, NE, E, SE, S, SW, W, NW: compass points; HM/BM/TrM: high pressure centre/ridge/trough over central Europe; HN/HB/HF/HNF: high pressure over North Sea/British Isles/Fennoscandia/North Sea-Fennoscandia; TM/TB: low over central Europe/British Isles; the subscripts refer to: z = cyclonal, a = anticyclonal, s = southern, and w = angular. (http://www.pik-potsdam.de/~uwerner/gwl/welcome.7.htm).

Main wind direction	Weather types
Primarily W-SW types.	Wz, SWa, SWz, Wa,
Sometimes W-SW types or mainly SW, S	Ws, Ww, NWz, HM, BM, TrW, Sa, Sz
Mainly N, NE, E, SE	TM, NEa, NEz, HFz, HNFa, HNFz, SEa, SEz, TB
Mainly NW, N, NE	NWa, Na, Nz, HNa, HNz, HB, Nea
Mainly E, SE, S	TrM, Hfa

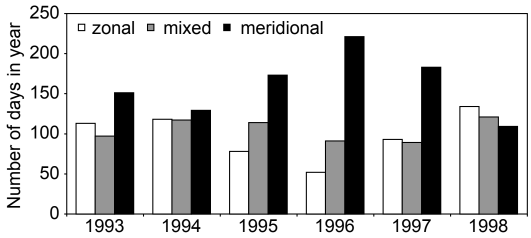

Fig. 4. Occurrence of large-scale weather types during 1993–1998. No. of occurrence days in the year.

south-westerly (SW-S) flows was 76%–82% of the time during 1993–1995 and again in 1997–1998, but dropped to 27% during winter 1996. The period from spring 1995 to summer 1996 was exceptional, with a high number of northerly and north-easterly flows and cooler air outbreaks, which were usually also rather dry.

In winter 1996 the share of the clean air sectors, NW–N–NE–E, was 57% of all days. A strong, almost permanent high-pressure centre remained over Russia in January and February, also extending several times over Scandinavia and the Baltic Sea. This northerly flow share reached 59% in summer 1995, being lowest in winter 1995 (7%).

The correlation between deposition and the air mass origin is illustrated in Fig. 6. For the whole six-year period there is a significant correlation of monthly NO_x deposition with the primary southern flow in all Baltic Sea sub-areas except the southern Baltic Proper; there is also anti-correlation with northerly flow. These correlations were significant at the 1% level according to both the Spearman-Kendall *t*-distribution test and Pearson's two-directional critical correlation coefficient test. For the NH_x deposition the correlation was significant in the southern and northern sub-areas. The daily correlation coefficients of the primarily S–SW wind direction over the whole Baltic Sea with dry NO_x deposition was 0.66/0.48/0.36/0.02 in winter, spring, summer and autumn, respectively, being significant (1% level) only in winter.

The North Atlantic Oscillation

The North Atlantic Oscillation (NAO) index is positive when the sea-level pressure difference between the Icelandic low and the Azores high is above normal. The NAO index determines the strength and direction of the westerlies and the temperature field across the North Atlantic, especially during winter and early spring. A geographically more general index for the same phenomena is the Zonal Index (ZI), i.e., the pressure difference between latitudes 35° and 55° averaged over longitudes 20°W–40°E. Both are encompassed by the Arctic Oscillation (AO) reflecting the variability of the sea-level pressure structure poleward of 20°N. The wintertime AO has had an upward trend during the past several decades, indicating a strengthening of the wintertime polar vortex from sea level to the lower stratosphere (Thompson and Wallace 1998).

The correlation of N deposition with these indices was computed to find out whether the deposition in milder years, reflecting a more zonal flow (high NAO index), is higher than during colder years, characterised by more meridional flow. The six-year average correlations of the monthly NAO index with NO_x and NH_x depositions were 0.43 and 0.22, respectively. In winter the correlation with NO_x deposition was significant (1%–5%, Pearson test) except over the Gulf of Bothnia, while in springtime the correlation was significant over the northern sub-areas B1, B2 and B3 (Fig. 7). During other seasons there was no correlation.

The correlation with ZI was somewhat higher (Fig. 7). The annual ZI correlated with NO_x deposition to the Baltic Sea at the 1% significance level, but the correlation was not significant for the ammonium deposition. The seasonal variation was the same as for the NAO. One fact that decreased the correlation is that the main location of the cyclone tracks during events characterised by the same NAO index can occur at slightly different latitudes over the Baltic Sea. The NAO index can also be in conflict with the length of the Baltic ice winter, e.g., during winter 1993/1994 it was positive from November to April, although the ice winter was of medium strength.

The reason for the higher correlation between circulation indices and NO_x deposition than with NH_x deposition is due to the fact that oxidised nitrogen compounds have a longer life-time because they are partly released from elevated sources and their chemical conversion and deposition rates are slower. Reduced ammonium is

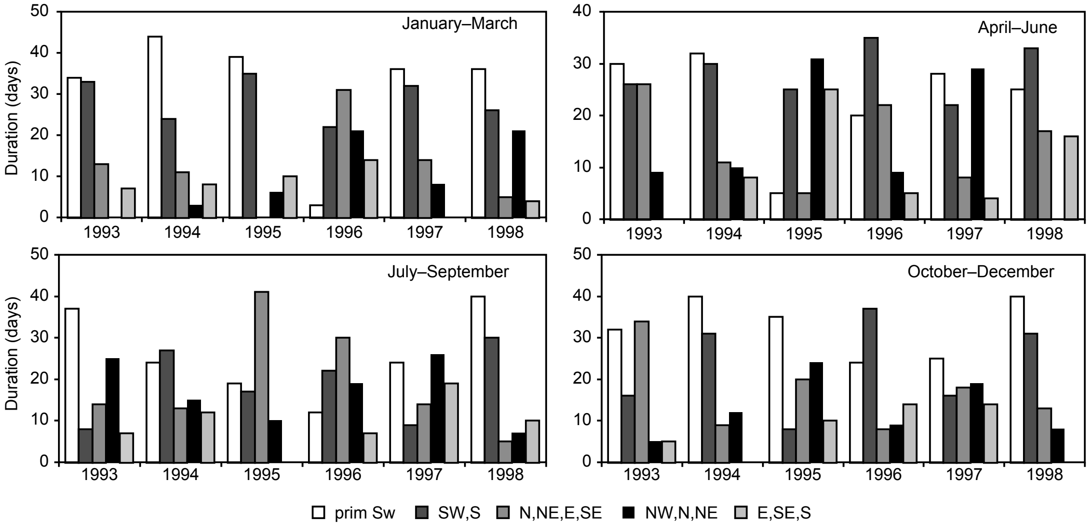

Fig. 5. Duration of large-scale flow regimes over the Baltic Sea sub-areas B3–B5, 1993–1998.

mainly emitted from the surface (agriculture) and in summertime. NH_x compounds, having a shorter residence time, are deposited relatively quickly and thus are not so readily incorporated into large-scale circulation patterns.

In conclusion, the deposition to the Baltic Sea is indeed higher during milder winters. According to Tuomenvirta (2004), the observed increase in mean daily minimum and maximum temperatures in Finland between 1950 and 1995 together with the simultaneous cooling in West Greenland are manifestations of the strengthening of the NAO. Consequently, these phenomena should strengthen westerlies and indirectly yield increased N deposition to the Baltic Sea.

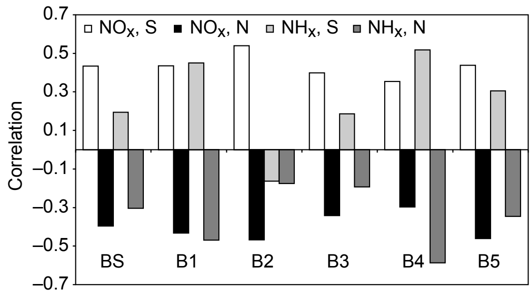

Fig. 6. Correlation of NO_x and NH_x total deposition with combined northerly (N = N, NW, NE) and southerly (S = S, SE, SW) wind directions for the whole Baltic Sea and its individual sub-basins.

Precipitation

The monthly precipitation sums over the Baltic Sea provided by the HIRLAM model vary over the five sub-basins (Fig. 8 and Table 3). Most of the precipitation fell during autumn and summer, with a rather high monthly variation, standard deviations being usually more than 50% of the average monthly value.

The HIRLAM model generally overestimates the observed precipitation at all EMEP sites in the Baltic Sea region by 10%–20%, but at a few sites specific local conditions (coast, orography) alter this general trend. Consequently, NO_x wet deposition is also overestimated by Hilatar (Hongisto *et al.* 2003) but by a smaller amount (5%–10%). On the other hand, reduced nitrogen is under-predicted by 25%, especially in summertime. This indicates that the scavenging efficiencies for these species are probably too low. These biases compensated each other in total N-deposition estimates.

During the winters 1992/1993 to 1994/1995, when south-westerly flows prevailed and NO_x deposition was high, precipitation was also higher than normal, except during the cold February of 1994. Maximum rain amounts over the Baltic Sea occurred in 1998, in autumn 1997 and in 1994. As compared with the 1961–1990 normal, the period spring 1995–spring 1996 was dry over

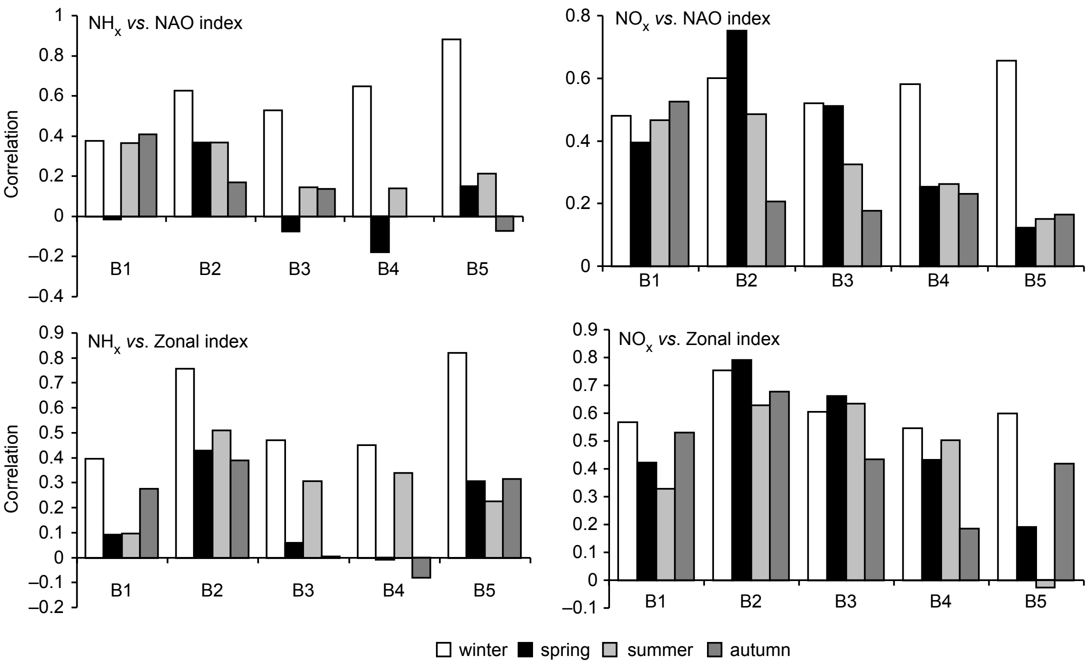

Fig. 7. Seasonal correlation of NO_x and NH_x deposition to the Baltic Sea sub-areas with NAO and ZI indices.

sub-basins B3–B5, the accumulated precipitation over most of the Baltic Sea being in January 1996 below 25%, and, during October–December 1995, 20%–50% of the normal amount. The cold summers of 1993 and 1998 were dry, precipitation being 58% and 75% of the mean seasonal value, respectively. Ammonium deposition reached its maximum during the wet months of November 1996 and October 1998.

Monthly NO_x deposition is non-linearly related to the precipitation amount (Fig. 9). The highest monthly deposition in each sub-area was reached with, on average, 50% of the monthly precipitation maximum. During the rainiest months, the deposition was usually around 60% of the maximum monthly value. The frequency of deposition episodes is highest in winter, while 31%–33% of rain falls in autumn. Over individual sub-basins, the monthly NO_x-deposition correlation with the precipitation sum was significant (1% level) with, however, a coefficient ranging between only 0.46 and 0.71.

Meteorological variability is not reflected as clearly in the ammonium deposition. For reduced N, the transport distance is shorter and the highest emissions occur during summertime, when precipitation is not correlated with wind direction so strongly.

Table 3. Monthly accumulated precipitation (Mt) to the Baltic Sea sub-basins.

	B1	B2	B3	B4	B5	Total
Average	5468	1545	7236	6428	2402	23078
S.D.	3097	818	3461	3231	1325	10937
Min.	553	102	299	37	513	2070
Max	13366	3575	15786	17388	7245	56950

Ice winters

The sea ice cover has an effect on the strength of both wet and dry fluxes: deposition is higher over open sea than over ice. The winters of 1992/1993 and 1994/1995 were extremely mild, the maximum extent of the ice cover being around 70 000 km^2 and the ice season about eight weeks shorter as compared with the climatologically average winter (1960/1961–1989/1990). The winters of 1996/1997 and 1997/1998 were also mild (128 000 km^2), while in 1993/1994

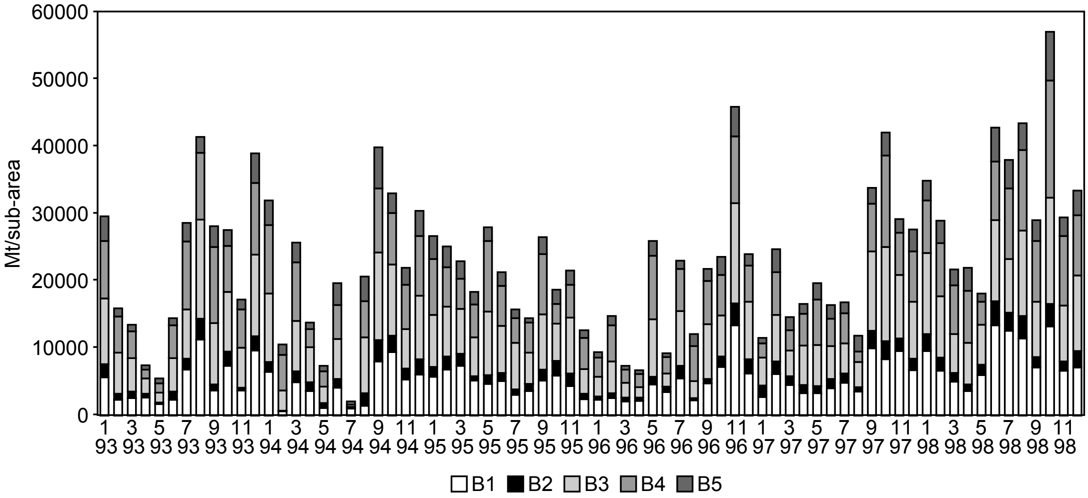

Fig. 8. Monthly precipitation sums (10^6 m^3 = Mt/sub-area) for the Baltic Sea sub-areas during 1993–1998.

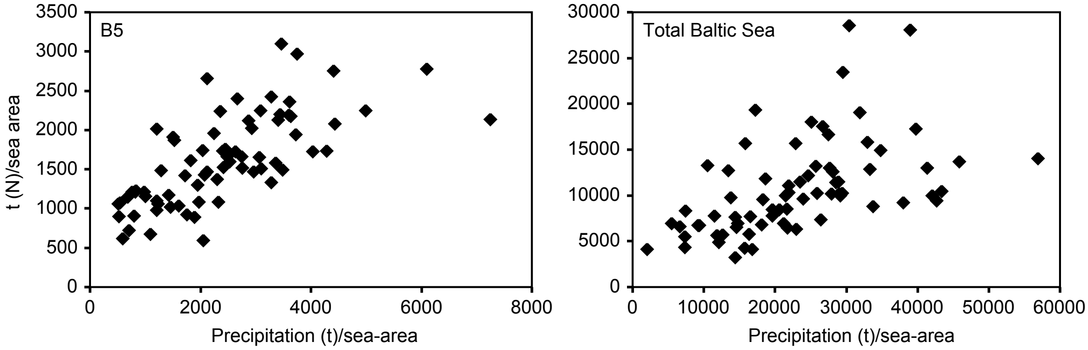

Fig. 9. Dependency of monthly NO_x deposition on monthly accumulated precipitation for the whole Baltic Sea and for the south-west sub-basin B5.

and 1995/1996 the ice cover extent was average (206 000–262 000 km^2) and the ice season was 2–4 weeks longer than normal (Seinä *et al.* 1996). The winter of 1994/1995 was the warmest; the winter 1993/1994 was also warm until the very cold February of 1994. The winter months of 1996 were the coldest of the whole 1993–1998 period; for 57% of the time, winds blew from the sector between north and south-east, precipitation being half and NO_x deposition below 50% of the average winter.

The anti-correlation between NO_x deposition and the monthly average fractional ice cover was significant (1% confidence level): –0.54 over the Gulf of Finland B2 and –0.48 over the Gulf of Bothnia B1 (Fig. 10), but higher if April is ignored (–0.62 for B2 and –0.52 for B1). In spring, when the marine atmospheric boundary layer is stably stratified, the ice cover retreat does not increase the deposition as much as in early winter, when convective conditions often prevail over the open sea. The anti-correlation between NO_x dry deposition and the ice cover was significant only over B2 (–0.48, –0.4 and –0.34 over B2, northern B1 and southern B1, respectively). The frequency of deposition episodes decreased as the ice cover increased.

Boundary-layer parameters

The variability of the atmospheric boundary layer height, wind strength, mixing conditions and relative humidity affect the vertical dilution, transport

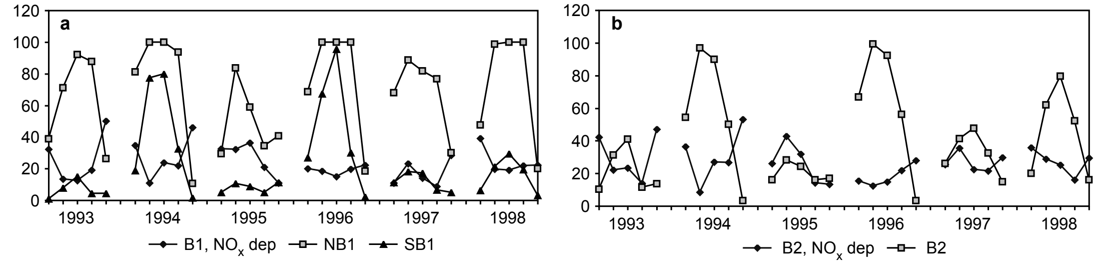

Fig. 10. — **a**: temporal variation of scaled NO_x deposition to B1 (10 t N/sea area B1) and average monthly ice cover (%) in the southern (SB1, $r = -0.34$) and northern (NB1, $r = -0.47$) Gulf of Bothnia. — **b**: same for the Gulf of Finland, B2.

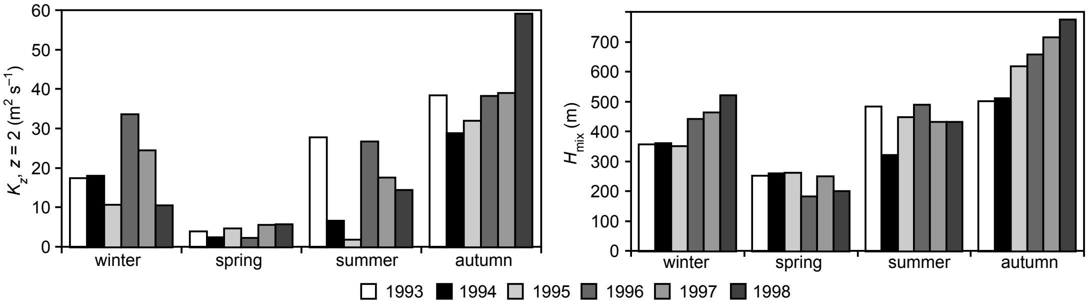

Fig. 11. Seasonal and interannual variability of eddy viscosity K_z (at model level $z = 2$, ca. 114 m) and the atmospheric boundary layer H_{mix} at the centre of the northern Baltic Proper, sub-basin B3.

speed and chemical conversion rates of contaminants transported over the sea. The annual differences at the centre of sub-basin B3, the northern Baltic Proper, in the seasonal averages of the atmospheric boundary layer height and eddy diffusivity K_z at model level 2 (around 114 m) are illustrated in Fig. 11. During late autumn or winter, when a northerly flow occurred over a warmer open sea, the mixing height was large and vertical mixing efficient, so that pollutant concentrations were diluted more efficiently. Concomitantly, dry deposition velocities were also high. However, on average, the dry deposition rate to the surface is slower than the mixing rate to the upper layers. During winter 1995, characterised by frequent southerly flows, the vertical mixing efficiency was only a third of that of the cold winter 1996, when the three-month average wind velocity at a height of about 30 m was almost 3 m s^{-1} lower than in the previous or the following winters. Figure 11 shows that spring is meteorologically the most stable period over the Baltic Proper and that summer values of K_z have a high inter-annual variability.

This indicates that any sensitivity study is very complex, because all the meteorological measures are inter-related in various ways. For instance, in autumn, stability conditions over the Baltic Sea are mainly unstable/convective, which makes the dry deposition velocity higher and thus, provided the downwind source is the same, would be expected to produce enhanced deposition. However, the convective boundary layer then is much deeper, bringing about a strong dilution of concentrations and thus a decreased deposition. Considering the very simple scenario of an advected column, Joffre (1988) showed that the height of the boundary layer had an appreciable effect on the dry scavenging of pollutants.

Episodicity

The episodic nature of most atmospheric processes is a well-recognised feature that can be detected by measurements with sufficient resolution. Most of the monthly wet deposition is collected over a few days, which usually are not those of maximum accumulated precipitation.

The simulated daily deposition over the Baltic Sea in 1993–1998 (e.g. Fig. 12) displays the typi-

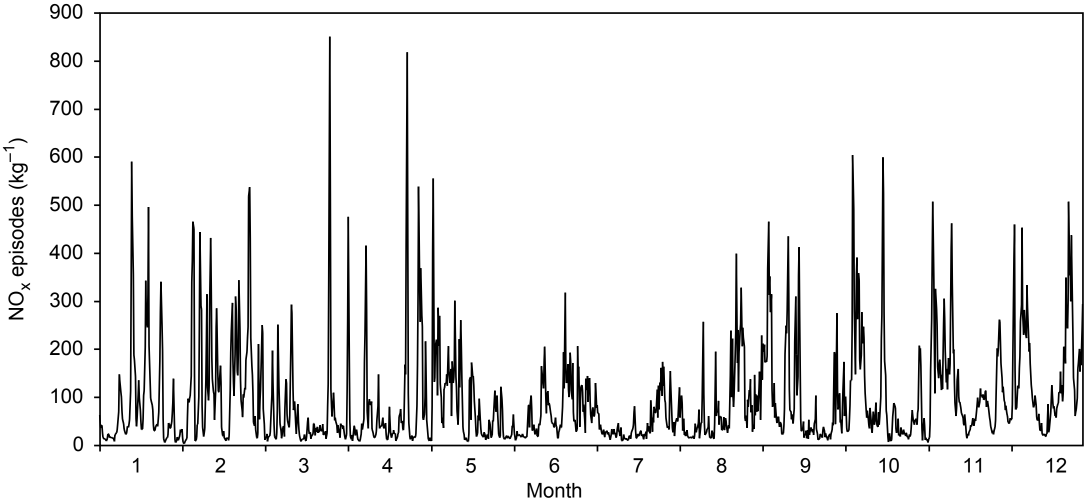

Fig. 12. Simulated daily NO_x deposition to the Baltic Sea area in 1997.

cal peaks characterising episodes of air pollution. There are no long dry periods that cover the whole sea. The Baltic Sea is large and situated in a mid-latitude zone of alternating polar, marine and continental air masses; meteorologically opposite situations can thus prevail over any sub-basin, with a west–east or north–south divide.

During 1996–1998, 30% of the annual NO_x deposition was received within 30 days, 20% in 17 days, 10% in seven and 5% in three days. The number of episodes varies from year to year (Fig. 13), as illustrated by the frequency of exceeding a six-hour load of 400 tonnes for oxidised and reduced N to the whole Baltic Sea. This diagram also partly provides an answer to the question of the inter-annual variation of NO_x deposition. Dry deposition also had a rather episodic nature.

The frequency with which a given share of the monthly deposition is exceeded is higher for single marine points than over the entire Baltic Sea. The modelled episodicity of wet deposition is also weaker as compared with daily measurements (Fig. 14). However, it is common for the meteorological rain gauges at EMEP stations to measure much more precipitation than the chemistry rain collectors. In 1997 for instance, at Utö Island, in southern Finland, the official meteorological gauge measured rain during 179 days, while wet deposition only was analysed for 94 days (Leinonen 1999). The main reasons for the discrepancy in this case were the higher collection efficiency and different location of the meteorological gauge; in some cases the precipitation was also below the threshold level or the samples were suspected to be contaminated. Nevertheless, omitting so many rainy days leads to an underestimation of the measured deposition.

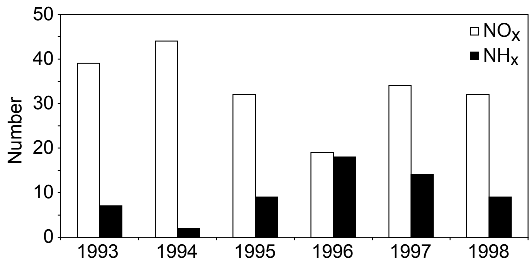

Fig. 13. Number of high deposition episodes, i.e., exceeding 400 t (6 h)$^{-1}$ for NO_x and NH_x over the whole Baltic Sea.

Importance of the atmospheric load compared with other external and internal nitrogen sources

The Baltic Sea is shallow and vulnerable and pollutants have deteriorated its environmental state already now rather dramatically (Rönnberg 2001). The accumulation of external nutrients has continued for a long time, and most of these nutrients end up in internal sinks, with only 10%–15% of them being exported through the Danish Straits (Eilola and Stigebrandt 1999, Zuelicke

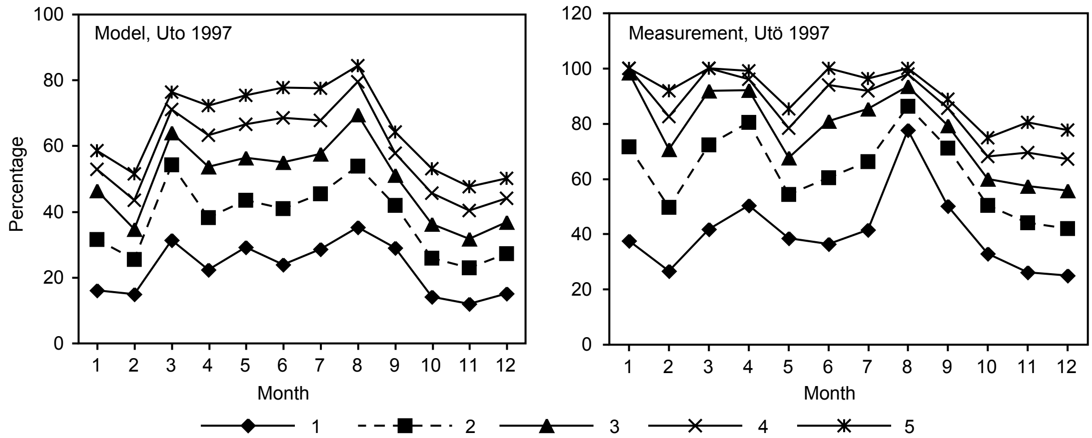

Fig. 14. Cumulative share of monthly wet deposition of oxidised N received during one to five days at Utö; 1997.

1999). Increased concentrations of nutrients in the water have already led to physical, chemical and biological changes in the water, fauna and flora, as well as to changes in oxygen conditions in the water, in the sediments and on the sea bed.

The variability in the exchange volume of salty water between the North Sea and the Baltic Sea, together with variations in haline and temperature stratification, have led to alternating anoxic/oxic conditions in the deeper waters throughout the 8000 years period of the brackish Baltic Sea (Winterhalter 1999). Salinity changes are in turn connected to the nutrient problem: when the bottom layer becomes anoxic, high amounts of the nutrients accumulated there can be released, as has occurred in the Gotland Deep and the Gulf of Finland in the 1990s. This internal loading can further accelerate the eutrophication processes.

Shallow sills divide the Baltic Sea into isolated basins with unique features, each basin having a different eutrophication state. The significance of the atmospheric nitrogen load to eutrophication is thus different for each basin, over open water and in coastal areas. In winter, for instance, light is the growth-limiting factor for algae, while in summer phosphorus becomes the limiting factor near the shores and in the Bothnian Bay.

The atmospheric load calculated with the Hilatar model is compared with other external N sources over distinct Baltic Sea sub-basins in Hongisto (2003c). As compared with the average river input, the atmospheric N load was around 50% over the Gulf of Bothnia, 70%–90% over the Baltic Proper, 10%–13% over the Gulf of Finland and 28%–35% in the southern parts. However, budget calculations for the Baltic Proper showed that the external N load was only a fraction of the total nitrogen needed to produce the total annual primary production (PP). Marine food chains are very effective, especially in summer, with 90% of the PP sustained by nutrient regeneration. Furthermore, N fixation by blue-green algae is also a significant external source — approximately as high as the river load over the Baltic Proper. Additionally, nutrients released from the bottom due to changes in the oxygen state can be transported upwards to the surface. Eutrophication is thus a very complicated chain of events and the significance of the atmospheric deposition to eutrophication has to be assessed using a hydrodynamic-ecosystem model.

Conclusions

We have simulated the dispersion, transport, transformation and deposition of atmospheric N compounds over Europe with the Eulerian high-resolution model Hilatar. In this paper we particularly addressed the variability of N deposition over the Baltic Sea and the factors affecting it with the intention of providing support for knowledge-based environmental policy measures aimed at protecting this large brackish water area that is beset by several grave environmental problems.

Neither nitrogen concentrations nor nitrogen deposition to the Baltic Sea decreased during the period studied. Episodic, seasonal and inter-annual variations were significant, and the load did not depend linearly on precipitation. Both wet and dry deposition are episodic. Total nitrogen deposition was found to be, to a large extent, long-range transported from central Europe, as demonstrated by the correlation with analogical synoptical situations favouring south-westerly flow. N deposition also strongly depended on the climatological conditions prevailing over central and northern Europe, as seen in its moderate correlation with the NAO and ZI indices. Inter-annual differences in wintertime deposition depended on the frequency and latitude of cyclone tracks. When low-pressure centres move along a northerly route, frontal precipitation occurs simultaneously with high NO_x deposition over the Baltic Sea.

The Hilatar model is capable of describing the dynamical transport and dispersion of contaminants in the atmosphere, the lifting of pollutants into upper layers in low pressure centres, fronts and thermally-unstable situations, as well as in-cloud or in-precipitation scavenging of pollutants and dry deposition. However, since models only treat grid averages, the modelled concentrations are generally lower than those measured.

Boundary-layer scale effects, such as mixing height and turbulence intensity, influence the deposition load. Most importantly, the turbulence conditions over the sea are often in an opposite phase with respect to corresponding conditions over land due to the strong annual cycle of the air–sea temperature difference over the Baltic Sea (no similar situation over other European seas). This has a strong influence on empirical deposition estimates made with coastal monitoring data, but is also challenging for modelling that should be able to resolve coastal gradients in emissions, meteorology, concentrations and deposition.

The various assessments of the total N deposition differ in their approach and in their results. There is, however, consensus that modelled estimates should nowadays be more accurate, since empirical assessments rely on too few stations, and these do not necessarily represent open water conditions.

Correct simulation of the anthropogenic load to the Baltic Sea over a longer period depends to a certain extent on the ability to properly account for the episodic character of marine phenomena and the sensitivity to climatic variations. Average meteorological conditions did not prevail over long periods during the simulated years. Nitrogen deposition was very sensitive to slight changes in the wind direction with respect to the main emission areas, and its monthly and inter-annual variations were large. Any increasing occurrence of blocking events, defined as large positive 500 hPa geopotential height deviations from the corresponding latitudinal mean, persisting for at least seven consecutive days at a given point, also decreases forecasting capabilities, because blocking is not a normal solution to a dynamical flow problem. The WMO time series for 1949–1993 indeed suggest a tendency for an increase in wintertime blocking intensity and duration in the Atlantic-European sector (Philips 1995). This is accompanied by warmer and drier winters in the latitudinal zone 40–70°N and colder and wetter-than-normal conditions on both the southern and northern sides of the blocking feature. Additionally, the concomitant observed strengthening of the NAO would yield increased N deposition to the Baltic Sea.

This continuum of scales, from turbulence to multi-year, influencing N deposition, also requires the ecological investigations (both modelling and observational) to cope with it and avoid use of monthly or annual averages. Furthermore, in marine ecosystem studies, the fluxes should be used with a module describing ice melting. The release of nutrients accumulated on the ice cover to the seawater is not the same as their instantaneous deposition rate, since they can be chemically converted and partly biologically consumed already on the snow/ice layer.

The modelled six-hour fluxes, concentrations and meteorological parameters have been compiled into a constantly-updated database that now also contains results for the years 2000–2002. These data are available for use in environmental research.

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