Variability of the marine boundary layer parameters over Baltic Sea sub-basins and their impact on nitrogen deposition

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Abstract

The variability of the marine boundary layer parameters over the Baltic Sea and its sub-basins and their impact on the 6 h, monthly or seasonal deposition of oxidized nitrogen compounds was studied using results of the Hilatar chemistry-transport model, the 6th hour forecasts of the HIRLAM weather prediction model and meteorological measurement data. The monthly load of oxidized nitrogen was highest in the winters of 1993–1995 and 2000, and lowest in 1996–1997 and 2005; no trend was detected. Short-time correlations were low, but a significant correlation of the monthly deposition with the NAO index and ice-season was found over northern sub-basins.

1. Introduction

Global anthropogenic reactive nitrogen N_r emissions increased from 23 Tg(N) yr^{-1} in 1860 to 93 Tg (N) yr^{-1} in the early 1990s, and it is estimated that they will grow further to 189 Tg N yr^{-1} in 2050 (Galloway et al. 2004). The increase of N_r in the environment has given rise to concern in recent years as a result of increasing emissions in developing countries. In Asia, reactive nitrogen N_r emissions grew from 14.4 Tg (N) yr^{-1} in 1961 to 67.7 Tg (N) yr^{-1} in 2000 (Zheng et al. 2002). The globalized reactive nitrogen problem has an influence on the carbon cycle and on biological production in marine and terrestrial areas. Our understanding of the rate of nitrogen accumulation in environmental reservoirs is still poor (Galloway & Cowling 2002, Matson et al. 2002, Wenig et al. 2003, Galloway et al. 2008, Gruber & Galloway 2008). The deposition of atmospheric inorganic nitrogen to the oceans increased from the pre-industrial value of 22 Tg (N) yr^{-1} to 39 Tg (N) yr^{-1} in the 1990s, and is predicted by IPCC (2007) to grow to 69 Tg (N) yr^{-1} by 2100 (Krishnamurthy et al. 2007).

The 1979 UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) has been implemented through eight emission reduction Protocols, two of which deal with reactive nitrogen. The Task Force on Reactive Nitrogen was established under the Working Group on Strategies and Review in December 2007. The task force on the Hemispheric Transport of Air Pollution, created in December 2004, has provided annual assessment reports of the hemispheric transport of air pollutants and their precursors (UNECE 2010).

The Baltic Sea (BS) is the world's largest brackish water area. Its average depth is 52 m and, over most areas, the water column has temperature and salinity stratification the whole year round (BACC 2008). The BS is semi-enclosed: the small water exchange over the sill separating the BS from the oceans makes the sea very vulnerable to external loads of pollutants and nutrients. Primary production in the Baltic's open sea areas is nitrogen-limited (Eilola & Stigebrandt 1999, Thomas et al. 2003), except in the Gulf of Bothnia. One third of the nitrogen load is assumed to be deposited from the air (Elmgren & Larsson 2001, HELCOM 2009a,b,c). The accumulated nutrients, as well as further input of nitrogen from the air, rivers and diffuse sources expose the small number of species comprising the food chain to the harmful consequences of eutrophication (HELCOM 2009a,b,c). The frequency of saline water pulses from the North Sea is important for oxygen availability in bottom areas. If bottom areas become anoxic, nutrients in the bottom sediments can be, and have been, released as an internal load. Since 1976 major inflows have been rather rare events, occurring maybe once in ten years (Nehring et al. 1995, Feistel et al. (eds.) 2009).

BS consists of sill-separated sub-basins, each with a characteristic climatological and ecological status. The differences in salinity, fluvial runoff, temperature, precipitation, wind and light conditions make the different sub-basins unique: the external nutrient load from the air has a different impact on their ecosystems (Rönnberg 2001, 2005, HELCOM 2010).

The climatology of the Baltic Sea is strongly influenced by the large-scale atmospheric circulation. We can describe this variability by imagining the Earth as a rotating ball covered with stratified fluid layers. The flow is disturbed by the surface structure and its response to radiation in the presence of several physical forces. These disturbances can generate vortices and waves, which have a low-frequency interdecadal or shorter period variability. Rossby waves – long ridges and troughs in the westerly flow of the upper troposphere with a wavelength of around 2000 km – were discovered in 1939. The Arctic Oscillation (AO) (Thompson & Wallace 1998) is the main component of sea-level pressure variability over the northern hemisphere. It is characterized by a deep, zonally-symmetric variation of geopotential height perturbations of opposite signs in the polar cap region and in the surrounding zonal ring centred near latitude 45°N. The corresponding Southern Oscillation (SO) had already been detected from the seasonal mean values of rainfall, surface temperature, and sea-level pressure by Walker & Bliss (1932). Over the Atlantic Ocean, AO is highly correlated with the patterns of the North-Atlantic Oscillation (NAO), and a teleconnection between the SO and AO has been discussed, e.g. in Horel & Wallace (1981).

Over the BS the modes of oscillation of the NAO determine, e.g. the severity of winter weather, the frequency and latitude of winter storms and cyclone tracks, as well as the geographical variation in precipitation and volume of river runoff; these have consequences for all human activities. The variability of the strength of continental high and low pressure areas over Russia and blocking event frequency are also manifestations of the NAO. During the negative phases of the AO and NAO, as in winter 2009–2010 (NOAA 2010), higher than normal pressure existed over Scandinavia and the surroundings of the BS, and the winter was cold. During the positive phase of the AO, zonal winds are stronger and oceanic storms follow northerly routes, bringing warmer and wetter weather to Scandinavia and drier conditions to the Mediterranean area.

A stronger winter AO indicates a strengthening of the winter polar vortex from sea level to the lower stratosphere (Thompson & Wallace 1998) and changes in upper-air jet streams, driving factors for weather in the northern hemisphere (Ambaum et al. 2001, Archer & Caldeira 2008). The AO/NAO also affect the latitude of the polar front and cyclone tracks, cyclone intensity (depth and radius), and cyclone number (Simmonds & Keay 2009). The winter (JFM) NAO was positive during the period 1987–2007 except in 1996, 2001 and 2005–2006, and negative in 2009–2010, whereas the summer (JJA) NAO has been negative or close to zero since 1998 (NAO 2011).

Nitrogen deposition to the BS is highly episodic, a feature that can be detected from measurements (available, e.g. from the EMEP/NILU measurement data base) or using model simulations (Hongisto & Joffre

2005). Dry deposition is also episodic (Hongisto 2003). The changes in large-scale weather systems may affect the frequency of the nitrogen deposition episodes.

This paper examines whether any of the changes in the large-scale circulation can be detected in the forecast meteorological and marine boundary layer (MBL) parameters, most important for nitrogen deposition processes over the Baltic Sea, and whether they have an effect on nitrogen deposition to the Baltic Sea. Numerical time series for trends are investigated in an attempt to discover the frequency of occurrence of certain peak values in the MBL variables. In addition, the dependence of deposition episodes on regional weather phenomena, such as storm frequency, storm track latitude and variability of precipitation are studied.

2. Material and methods

Variation in nitrogen deposition over the BS is studied using the results of the Hilatar chemistry-transport model (Hongisto 2003), the forecasts of the HIRLAM hydrostatic weather prediction model (High Resolution Limited Area Model, HIRLAM 2002, Undén et al. 2002) and measurements at certain Finnish meteorological stations over the period 1959–2010. HIRLAM has been in operational use at the Finnish Meteorological Institute (FMI) since 1990. The current European model has 60 vertical layers and a horizontal grid of 0.15° resolution; the model covering the Baltic Sea has a finer, 0.068° resolution.

The Hilatar chemistry-transport model, a nested dynamic Eulerian model covering Europe and the Baltic Sea area, provides gridded estimates of the fluxes and concentrations of oxidized and reduced nitrogen and sulphur compounds. Gaseous (g) and particle (p) concentrations are calculated for the following compounds: NO_x(g), HNO_3(g), NO_3(p), PAN(g), NH_4NO_3(p), NH_3(g), SO_2(g), SO_4(p) and $(NH_4)_{1.5}SO4$(p), where PAN is peroxyacetyl nitrate and $NO_x = NO + NO_2$. The chemistry module comprises the EMEP-MSC-W chemistry code (Iversen et al. 1989) with some modifications (Hongisto 2003).

Hilatar uses as its meteorological input the gridded 6th hour predictions of HIRLAM, developed as a research co-operation project between various European meteorological institutes since 1985. The model is updated regularly and there are several new releases of the HIRLAM code each year with different models of physical parameterization. A reference version of HIRLAM and the operational data archive are maintained at the European Centre for Medium-range Weather Forecasts (ECMWF: http://www.ecmwf.int).

Model documentation has been provided for the user community in scientific reports, newsletters and on-line documents (the earliest being HIRLAM 1990, Kållberg 1992, Källan (ed.) 1996, Eerola 2000, 2002, 2003, Undén et al. 2002, 2003). At the FMI, major changes occurred in 1991 and June 1995. During 1996–1997 the operational version of HIRLAM 2 was used with an improved radiation scheme, a 0.5° horizontal resolution and 31 vertical levels. From November 1999 until May 2003 HIRLAM 4.6.2 was used with a 0.4° grid and 40 vertical levels. The ECMWF lateral boundary conditions were introduced in July 2001. Since March 2003 HIRLAM 5.1.4 with a 0.3° grid and the 3DVAR analysis scheme has been used, and this was followed in February 2004 by HIRLAM 6.2.1 (0.2° grid). HIRLAM 7.1 with 0.15° resolution and 60 vertical layers was brought into use on 28.3.2007, HIRLAM 7.2 on 2.9.2008 and HIRLAM 7.3 on 2.11.2010. Over northern Europe additional forecast runs with a finer horizontal resolution have been produced.

In Hilatar, the horizontal advection is solved numerically with the positive definite, area-preserving flux-form advection algorithm of Bott (1989), the chemistry with the QSSA (quasi steady-state approximation) method of Hesstvedt et al. (1978), and the vertical diffusion with the Crank-Nicholson differentiation algorithm (Tuovinen 1992). Dry deposition velocities are used as the lower boundary condition of the vertical diffusion equation, these being calculated using the resistance analogy. The boundary-layer schemes of Lindfors et al. (1991, 1993) are used for calculating the MBL parameters for dry deposition velocities over sea areas. Wet deposition is calculated separately for in-cloud and below-cloud conditions for particles and gases, the scavenging rates being based on, for example, the work of Chang (1984, 1986), Scott (1982), Jonsen & Berge (1995) and Asman & Janssen (1987).

The Hilatar model uses the HIRLAM grid: horizontally-rotated spherical coordinates and vertically hybrid sigma coordinates with selected (10–21) vertical layers up to 5–10 km in height. The long-range transport of compounds to the fine-resolution model area is estimated by nesting the different scale models: for the period 1993–June 1995 the EMEP acid model (Iversen et al. 1989) background was used.

Tests of the numerical schemes are documented in Hongisto (1998). Validation of the model through comparison with EMEP-network measurements covering four years are reported in Hongisto et al. (2003). The model was additionally validated in the subproject 'Air Pollution Load' of the EU-MAST project BASYS (Baltic Sea System Study) against the summer and winter observations with four coastal stations and two research ships (Schulz et al. 1999, Plate 2000).

The model uses the 50-km EMEP-emission inventory for European emissions, a specific Baltic Sea ship emission inventory (Stipa et al. 2007, Jalkanen & Stipa 2009), and the FMI inventory for Finnish and north-western Russian sources. The time variation is based on the GENEMIS project 1990 for country-specific emissions and on diurnal and weekly traffic indices. The initial vertical mixing was estimated by emission height profiles or using a plume rise algorithm.

According to EMEP data, the NO_x emissions of the 19 countries and sea areas contributing the most to the Baltic Sea deposition (Russia being excluded due to a change in the EMEP area in 1997) dropped by 19% from 1990 to 1995, by 14% 1995–2000 and by 14.6% between 2000 and 2008.

3. Results

In this article, only the variation in oxidized nitrogen ($NO_y = NO_x$, NO_3-particles, HNO_3 and PAN) deposition is studied, because although the NH_3 emission intensity is very high in areas to the south and south-west of the Baltic Sea, NH_3 has a shorter transport distance in the atmosphere, and the majority of NH_3 and NH_4 particles are deposited in the southern Baltic Sea.

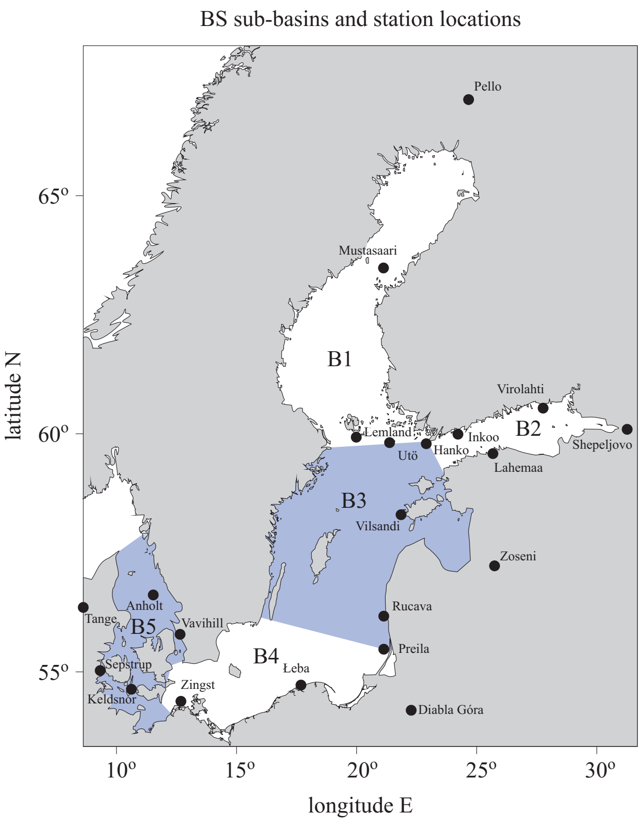

Figure 1. Location of the measurement stations and the Baltic Sea sub-basins considered in the study. B1 – Gulf of Bothnia, B2 – Gulf of Finland, B3 – northern Baltic Proper, B4 – southern Baltic Proper, B5 – Kattegatt and Belt Sea. The stations at Westerland (EMEP-code DE1), Birkenes (NO1), Faroerne (DK1) and Utsjoki Kevo are located beyond the area shown on the map

The studies are performed separately over the five BS sub-basins defined in Figure 1: the Gulf of Bothnia (B1), the Gulf of Finland (B2), the northern Baltic Proper (B3), the southern Baltic Proper (B4), and the Kattegatt

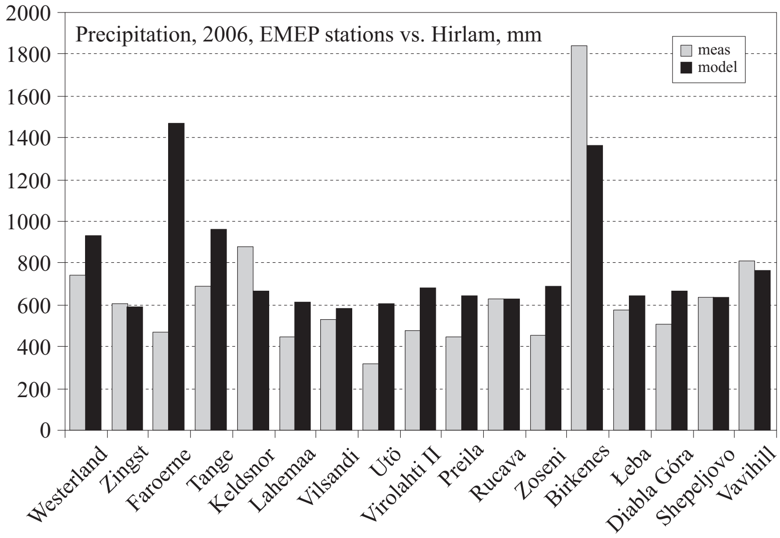

Figure 2. Comparison of HIRLAM modelled and measured annual accumulated precipitation at EMEP stations in 2006 [mm]

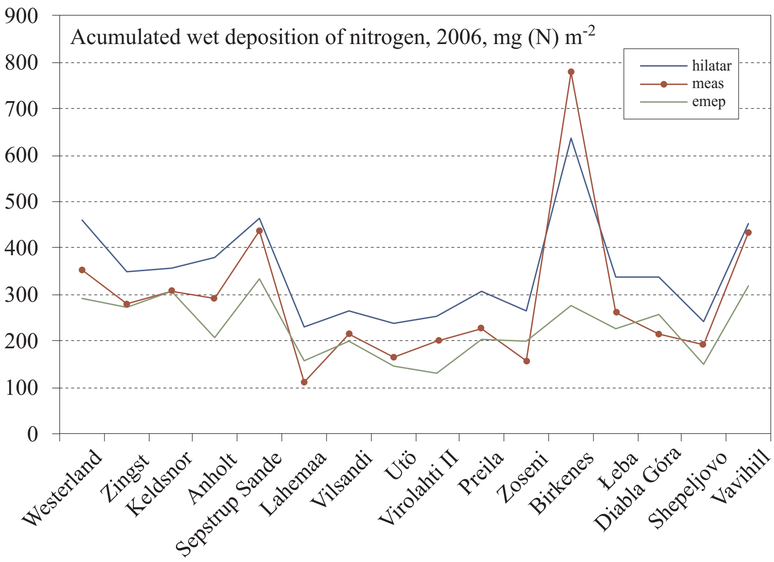

Figure 3. Comparison of measured and modelled (Hilatar and EMEP models) annual accumulated wet deposition of inorganic nitrogen, mg (N) m^{-2}, at stations surrounding the Baltic Sea in 2006

and Belt Sea (B5). To obtain an estimate of the inaccuracies contained in the simulation results, intercomparison of wet deposition with measurements at stations surrounding the Baltic Sea for the year 2006 are presented in Figures 2–3. At stations surrounding the BS, the HIRLAM grid average precipitation differs from the EMEP station measurements by $-30\%... +60\%$. The calculated depositions exceed those measured; however, it should be noted that for measurements the precipitation amount of the air quality gauge is used: in winter this is usually lower at windy coastal stations than the corresponding result of an official meteorological gauge fulfilling the WMO criteria for arrangements at precipitation measurement stations.

The annual variation in the area-scaled deposition of oxidized nitrogen to the Baltic Sea over the period 1973-2009 is presented in Figure 4. Around 55–70% of the deposition is received during the winter and autumn (defined here as January–March, JFM and October–December, OND) owing to the higher emissions, stronger cyclone activity and greater amounts of precipitation in these seasons, and this proportion was highest during 1993–1995. The percentage of wet deposition was highest over the northern sub-basins, around 65% over B1 and B2 in winter and autumn.

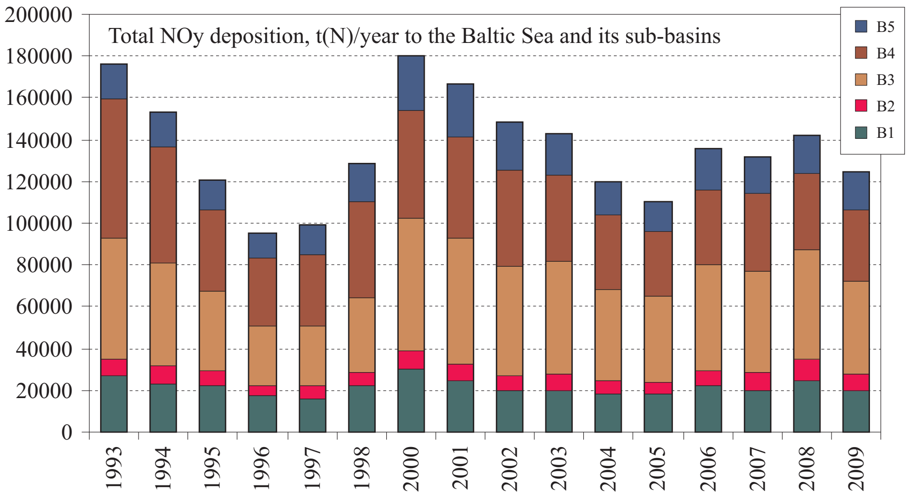

Figure 4. Total annual accumulated deposition of oxidized nitrogen to BS sub-basins B1–B5, as defined in Figure 1, t (N) year^{-1}, 1993–2009

Nitrogen deposition to the Baltic Sea is very episodic. The number of high deposition events in 1993–1998 (Hongisto & Joffre 2005, Figure 13) shows clear differences in the annual variation of the oxidized and reduced nitrogen depositions. The annual and seasonal numbers of wet episodes

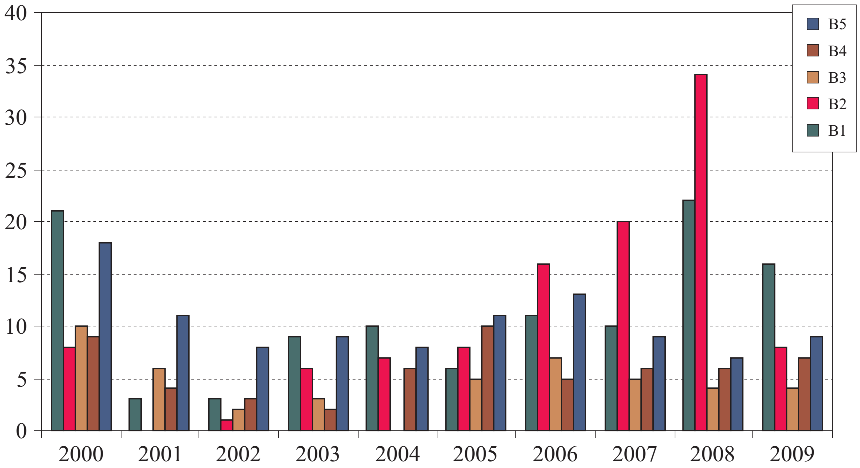

Figure 5. Frequency of annual wet episodes in 2000–2009: annual number of monthly 6 h deposition events of sub-basin B1 exceeding 10-fold the 10-year average 6 h deposition of the month for this sub-basin

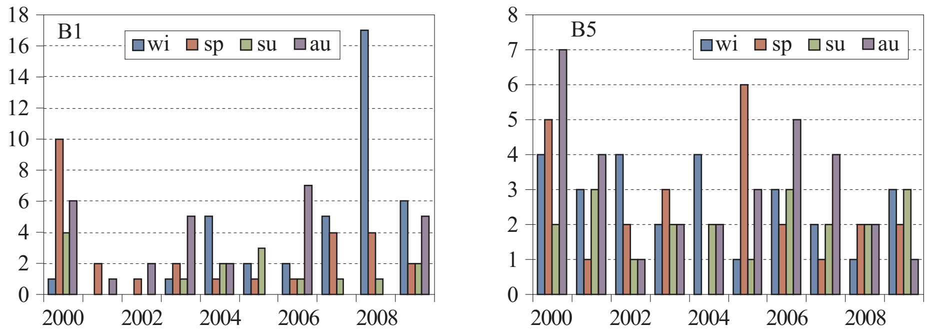

Figure 6. Number of wet episodes/season: 3-monthly sums of 6 h deposition events over sub-basins B1 (left) and B5 (right) exceeding 10-fold the 10-year average 6 h deposition of the month for the respective sub-basin, wi=JFM, sp=AMJ, su=JAS, au=OND

(defined here as the 6 h deposition over a sub-basin exceeding 10-fold the 10-year average 6 h deposition of the month for that sub-basin) in 2000–2009 are presented in Figures 5 and 6. The frequency of NO_y deposition episodes had distinct minima in the periods 1995–1997 and 2001–2005, and there was another decrease in 2009. The correlation coefficient R of the number of episodes with the total annual NO_y deposition was $R > 0.55$ over B1–B3, the index of determination R^2 was 31–33% but the P-value was higher than 0.05, indicating only a statistically suggestive correlation. The winter episodes depend on the ice conditions: in 2008, when the Gulf of Bothnia and the Gulf of Finland were ice-free most of the time, the episode frequency increased, whereas in the more southerly sea areas seasonal differences in the number of episodes were not so much in evidence.

The average MBL conditions have interannual, seasonal, diurnal and very short term variations, different in different BS sub-basins. Over all the sub-basins, precipitation was most intensive in the winters of 2007–2008 and 2001–2002, as well as in summer 2007 and autumn 2000–2001; during these seasons, the pressure was lower than the periodic average. The wind velocity was lowest over the narrower gulf areas. One can notice a rather high interannual variation in the seasonal averages. The MBL height has a north-south gradient, and there is generally a rather high annual variation in seasonal average MBL heights.

The correlations R of the 6 h values of wet and dry deposition of NO_y over B3 and B1 with wind speed, precipitation, surface pressure, mixing height, friction velocity and temperature in 2000–2009 are presented as seasonal averages in Figure 7, while the corresponding explanation factors (R^2) are shown in Figure 8. The annual correlations are small because opposing stability conditions prevail over BS in spring and autumn: there are > 14 000 time periods, and dispersion of all parameters was high, especially during the peak deposition events. The correlation coefficients indicate only if a linear regression between the variables exists. However, from the scatterplots one can conclude that deposition is nonlinearly dependent on most of the meteorological parameters, and this seems to be the case even for the dependence of wet deposition on precipitation. If we study 6 h correlation averages over shorter periods, e.g. monthly values over selected years, the correlations are higher, but the years are different.

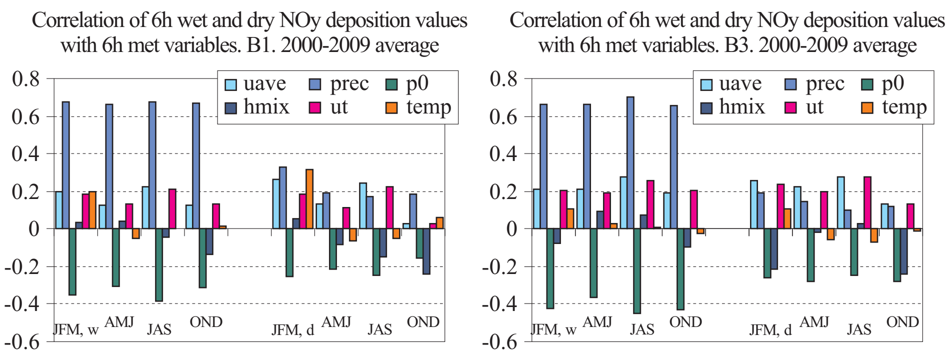

Figure 7. Correlation coefficient of 6 h values of wet and dry deposition of NO_y over sub-basins B1 (Gulf of Bothnia) and B3 (northern Baltic Proper) with wind speed, precipitation, surface pressure, mixing height, friction velocity and temperature

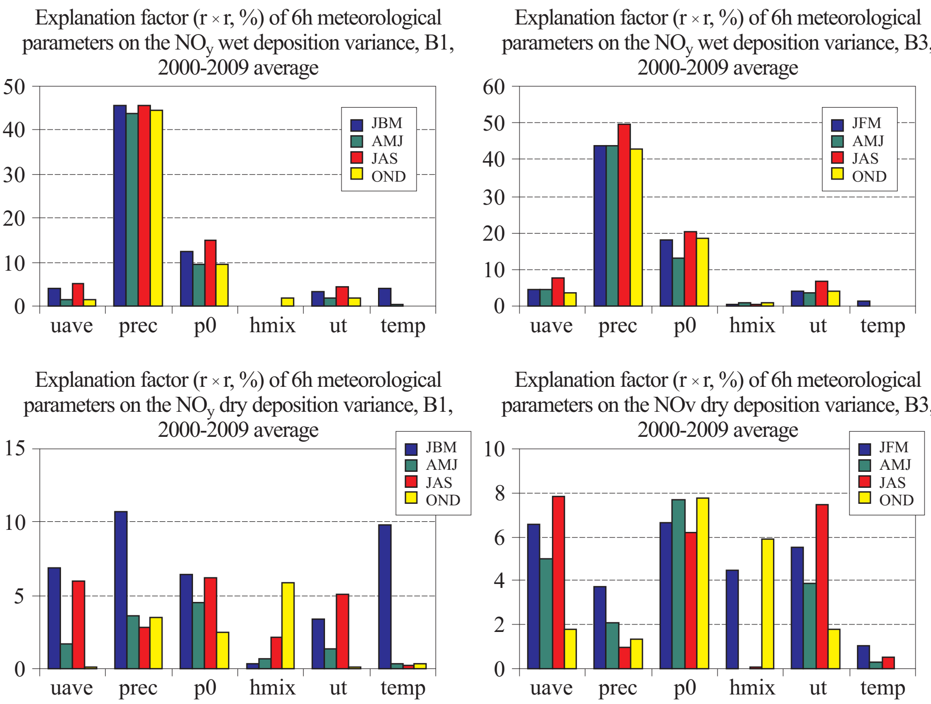

Figure 8. Explanation factor (correlation coefficient squared) of 6 h values of wet and dry deposition of NO_y over sub-basins B1 (Gulf of Bothnia) and B3 (northern Baltic Proper) with wind speed, precipitation, surface pressure, mixing height, friction velocity and temperature

The correlation depends on the stability of the sea area; it is always negative with p0, linking deposition events with cyclone activity. In autumn, if the MBL is deep, cold air from northern sectors is advected over the warmer sea, and the pollutants, if transported into the area, are diluted into a large volume; dry deposition is thus weak. In winter and early spring, most of the B1 and B2 are ice-covered and neutrally stratified. In later spring, the correlation of dry deposition with temperature can be negative, because if warm air is advected over a cold sea, the stratification is very stable.

In both winter and summer, high dry deposition events over B1 seem to occur in warm and windy weather. However, this deposition is from long-range pollution transportation; the Gulf of Bothnia is located rather far from the most intensive emission areas. Thus, even if highly turbulent conditions persist over the water area, for a deposition event to occur, there also has to be advected inorganic nitrogen of anthropogenic origin in the air. For wet deposition the dependences are more evident. Winter cyclones usually arrive from the Atlantic, and the main wind direction ahead of these low-pressure areas is from the most intensive emission areas. Thus, precipitation connected to fronts that cross the BS from SW to NE washes the pollutants down, and correlations are higher.

Wet deposition depends non-linearly on the amount of precipitation; high deposition events can also occur with light rain. If we look at the dependence of total NO_y deposition on wind direction, most of the deposition is seen to occur when the wind blows from the W-SW sector. Even so, some high deposition events also occur when the instantaneous wind direction is northerly. Because the wind direction may change by 180° when a cyclone or front is passing through the area, there is no point in studying the dependence of instantaneous wind direction values any further.

4. Storm frequency

During the summer storm of August 2001, very high instantaneous deposition values were modelled (Hongisto 2001). The episode began with a strong inversion over central and north-western European areas with intensive NO_y-emissions. The pollutants accumulated in the air were transported north-westwards by a cyclone crossing the Baltic Sea: they circulated around the cyclone in a front over the Baltic States and were eventually washed down to the surface over the northern Baltic Proper and adjacent areas. The deposition maximum did not occur geographically along the track of the storm centre: rain is connected to fronts that can extend far from the cyclone centre. Thus, when checking whether any connection between extreme weather events and deposition exists, the location of the cyclone centre itself is not especially significant.

Because this unique case did show a connection between storminess and deposition episodes, the trends of the frequency and latitude of storms were estimated from the HIRLAM weather predictions in the 2000s. Because the period was rather short and the annual variation too high to allow any conclusions on real trends to be drawn, the storm frequency was also estimated from measurements made at selected marine meteorological stations.

Figure 9 shows the frequency (number of 6 h periods in a year averaged over the BS-sub-basin) and the average latitude of the grid-points for which the instantaneous maximum wind speed over the sub-basin exceeded 15 m s^{-1} in 2000–2009. Most of these cases occurred during the cold season. The monthly frequencies in winter and autumn indicate that there seems to have been a rather quiet period in 2002–2004 and that in the northern part of the BS (B1, B2, B3) the high wind episodes occurred over slightly higher latitudes at the end of the period. The years 2003–2006 were less windy

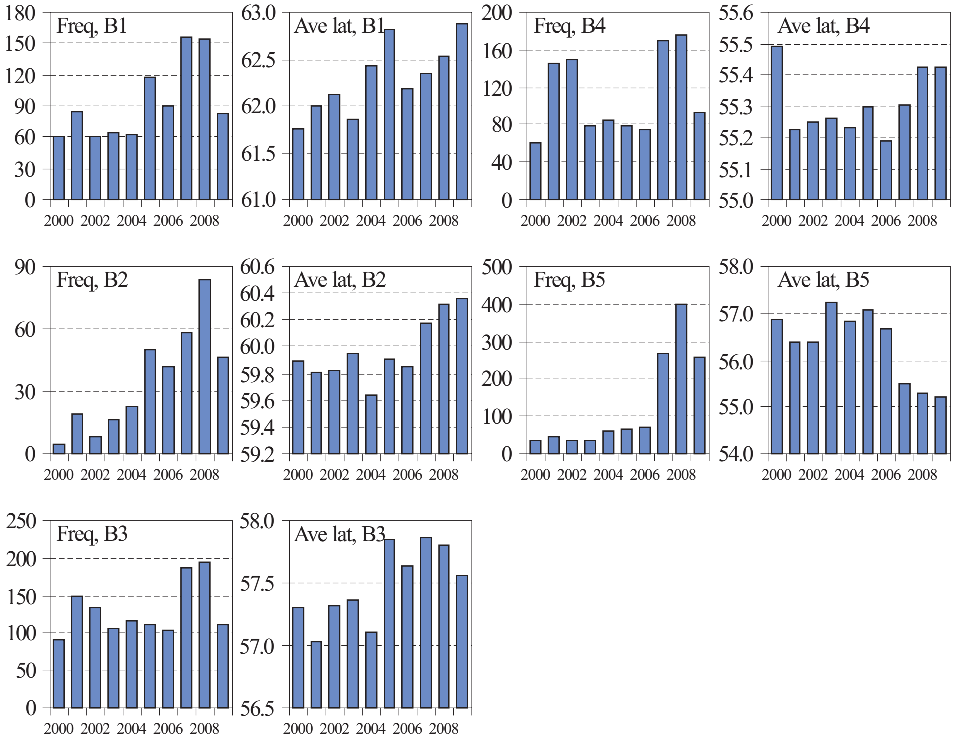

Figure 9. Frequency (number of 6 h periods/year, left-hand column) and average latitude (right-hand column) of grids for which the maximum 10 m wind speed (umax) of HIRLAM over the sub-basin is umax > 15 m s^{-1}, 2000–2009

over B3 and B4. The cyclones over the sub-basin B5, the Belt Sea and the Kattegat, followed a slightly more southerly route at the end of this period.

The 6 h gridded data series over the different BS basins were also filtered to pick out cases when the surface pressure was below 980 hPa. The latitude of the grid-point with the minimum pressure over the BS sub-basins fulfilling the criteria in 2000–2010 does not show any clear trend, and differences exists between sub-basins.

When the same criterion, p0 < 980 hPa, is applied to some marine and northerly meteorological station measurements (at 3 h time intervals) over the period from January 1993 to August 2010, the results (Figure 10) show a minimum storm frequency in 1996, 2000–01, 2003–06 and 2009–10 for the marine BS stations. The northern stations are influenced more by easterly and northerly air masses.

In Figure 11 the maximum BS ice extent (Schmelzer et al. 2008, Niskanen et al. 2009) is presented together with the number of 3 h periods when p0 < 980 hPa at Finnish meteorological stations during the period

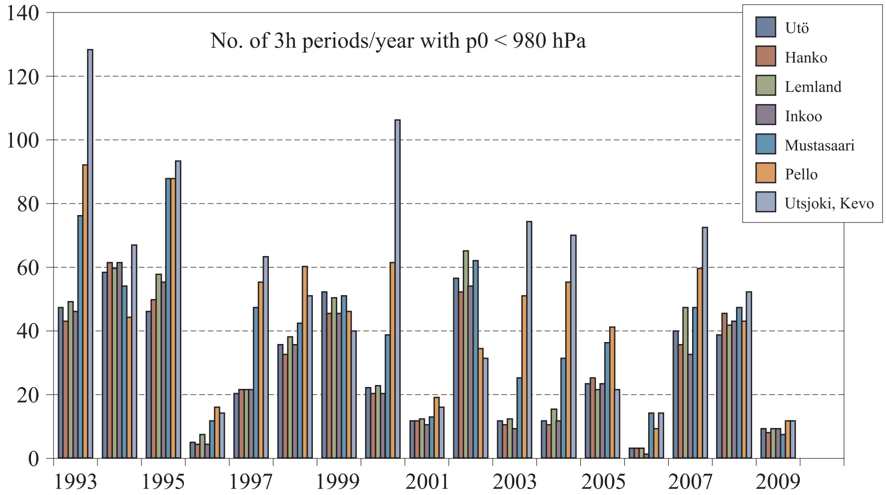

Figure 10. Number of 3 h periods with p0 < 980 hPa, Jan. 1993–Aug. 2010, at the Finnish meteorological stations on Figure 1. The stations at Utö, Hanko, Lemland, Inkoo and Mustasaari are marine ones; for comparison, data for stations in northern Finland (Pello and Utsjoki) are also presented

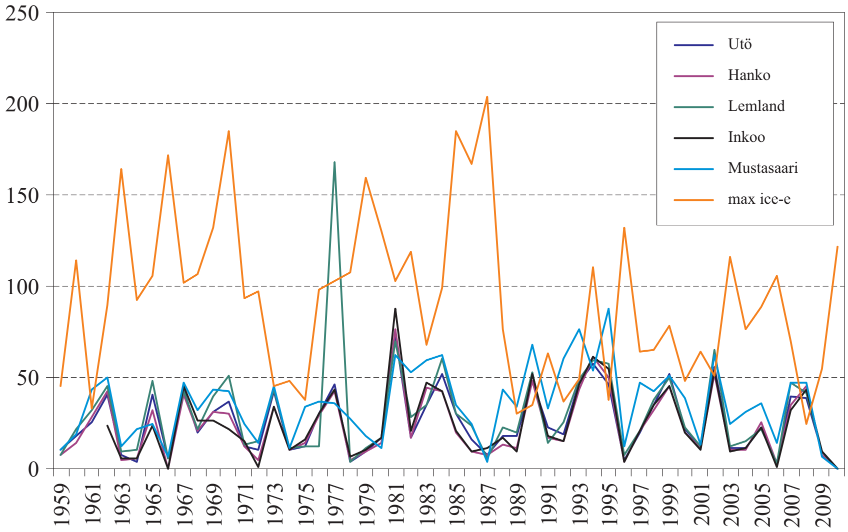

Figure 11. Maximum BS ice extent. 1000 km^2 multiplied by 0.5 (max ice-e) and the number of 3 h periods when p0 < 980 hPa, 1959–2010, at the Finnish meteorological stations presented in Figure 1

1959–2010. The anti-correlation of the maximum ice extent with the number of occasions of pressure < 980 mbar varied between −0.2 and −0.6, being highest in the north. All the marine stations are situated quite close to the coast and surrounded by ice every winter.

The number of 3 h periods/year in 1959–2010 when p0 < 980 hPa for different wind directions at the Utö station is presented in Figure 12. Most of these low-pressure cases occur in the winter months, but winters are different; over this 50-year period, winter low-pressure situations occurred at Utö most frequently in 1981. However, from Figure 13 (the monthly variation of cases when p0 < 980 hPa and the wind speed

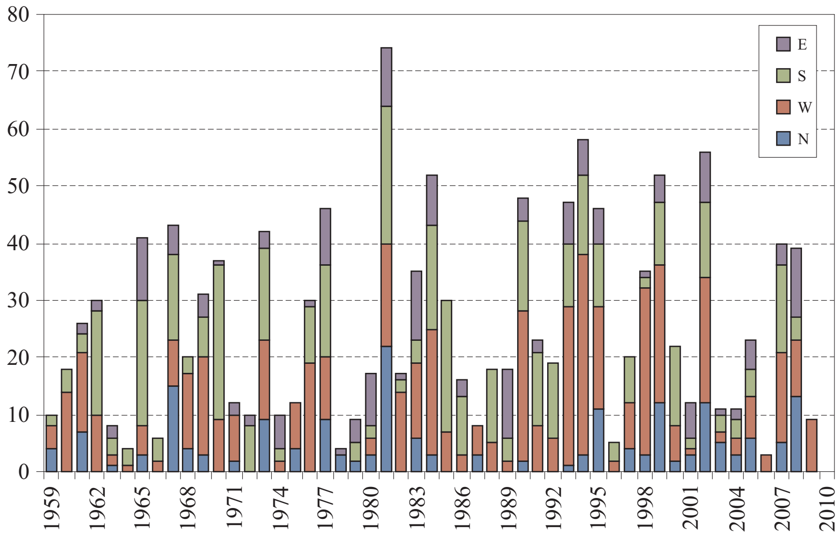

Figure 12. Number of 3 h periods/year when p0 < 980 hPa in different wind directions at Utö (station 2) in 1959–2010

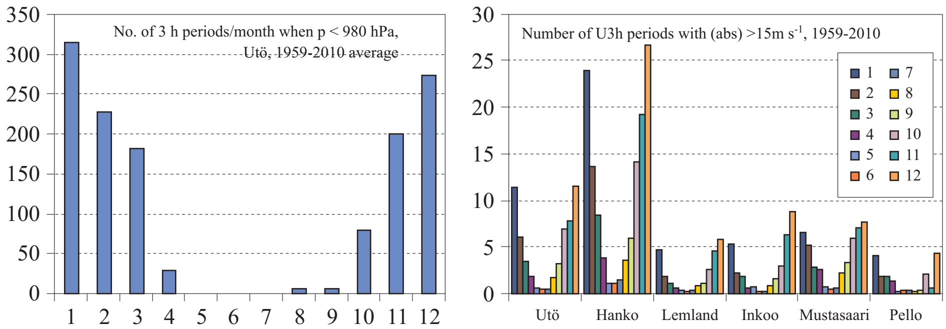

Figure 13. Monthly variation in the average number of 3 h periods/month when surface pressure p0 < 980 hPa and the monthly variation in the number of 3 h periods when 10 m wind velocity U_{abs} is $U_{abs} > 15$ m s^{-1}, averaged over the time period 1959–2010

> 15 m s−1 averaged over the whole period) we can see that high wind speed events do also occur in summer. Surface pressure maxima at these marine stations occurred on average in May.

From Figure 14, showing the number of 3 h periods/year when the wind speed was higher than 15 m/s, one can conclude that high wind speeds were more frequent before 1975 and again between 1991–1995. The frequency was highest in the winter months, and Figure 13 can be used to partly explain the higher deposition values in the winter of 1993–1994.

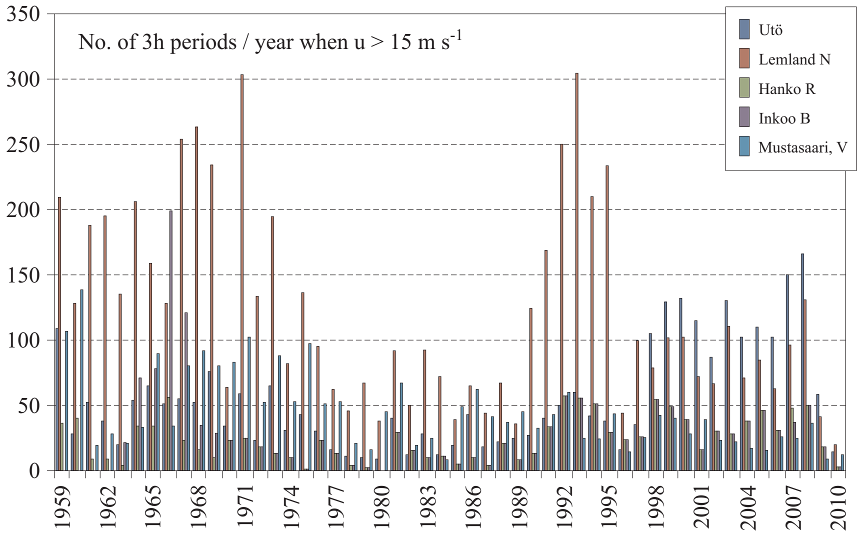

Figure 14. Number of 3 h periods/year with u > 15 m s^{-1} at Finnish marine meteorological stations

During the period 1993–2009 the correlation coefficient R of the winter (JFM) NO_y deposition with the length of the ice season varied between R = −0.52 over B2 and R = −0.19 over B4. The minimum probability (P-value) was 0.028 and the explanation factor $R^2 = 26.7\%$ over B2. The anti-correlation is stronger when December is included in the winter period. When winter is defined either as DJF or DJFM, the correlation is extremely significant ($P < 0.0006$, $R^2 > 54\%$) over B2, the Gulf of Finland, and significant ($P < 0.01$, $R^2 > 35\%$) over B3. For B2 the variation in the length of the ice season is important, because over this sea area the share of the annual airborne load due to winter and autumn deposition is 55–70%. However, ice conditions depend on the frequency of northerly or easterly continental airstreams, and all other MBL parameters vary with the cold air as well.

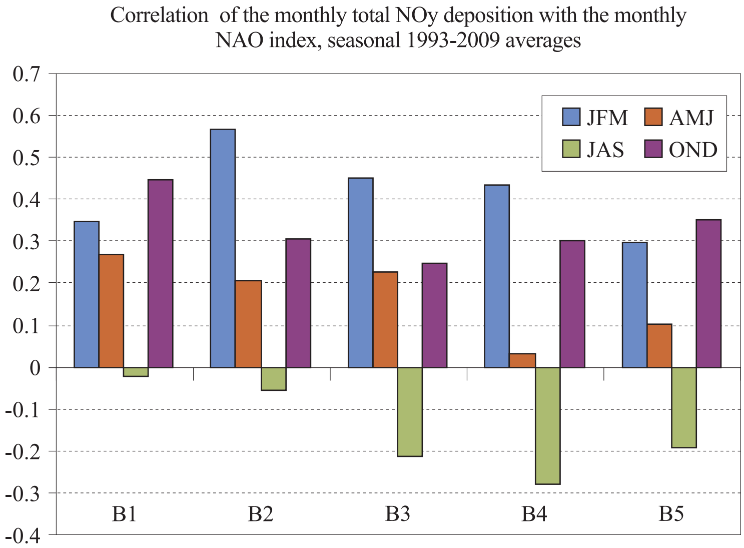

Figure 15. Correlation of the monthly NAO index with the monthly total NOy deposition in 1993–2009 presented as seasonal averages over the Baltic Sea sub-basins B1–B5 shown in Figure 1

Figure 15 presents the seasonally averaged correlation of the monthly NAO index with the oxidized nitrogen deposition to the Baltic Sea sub-basins in the years 1993–2009. The correlation was extremely significant over B2, the Gulf of Finland, in winter (JFM), and significant ($P < 0.01$) over B3 and B4 in winter and over B1 in autumn (OND).

5. Discussion

The reasons for and the origin of the episodically-received external load to the northern Baltic Sea sub-basins B1–B3 cannot be explained fully by instantaneous local meteorological factors (wind speed or direction, turbulence, state of other weather elements or the passing of a cyclone), because nitrogen compounds are transported long distances to the areas of deposition and they remain in the air for several days before being deposited. Each episode is the result of a chain of events connected to cyclonal and frontal activity. Precipitation and weather extremes are not concurrent with the cyclone centre crossing a given sea area, but tend to occur with a time lead, as the wind field connected with cyclones and fronts is complicated.

However, deposition does seem to depend on the frequency of extreme weather events and cyclone activity, which in turn depend on the variation of large-scale weather patterns, such as the NAO, prevailing over the Baltic Sea. The results of the analysis of wind velocity and pressure minimum extremes presented in the previous section can be compared with estimates of storm frequency along the Swedish coast in the southern and northern BS (Eek 2000) or along the western BS5 (Olsson 2002); these show that at the Vingas station there was a distinct minimum in storm frequency between around 1935 and 1968, a maximum in 1920–1930 and in the 1980s. A similar variation, with some differences in the details of years and periods, can be seen from the data of the other stations studied by Olsson and Eek. The total number of severe storms was highest in 1919–1929 and 1940–1949 (Eek 2000). In Finland the strongest storm on record occurred on 28.8.1880, 120 years ago.

The frequency of storms was studied because the number of extreme weather events is generally expected to increase with climate change. In this case nutrient deposition may increase if emissions do not decline. But, over the Baltic Sea, this analysis did not show any increase in storm frequency. Although the HIRLAM data period covered too few years for any conclusion to be drawn, no trend could be detected also in the measurement station data. The hypothesis of increasing extreme weather event frequency may not be valid either: according to Zahn & Storch (2010), in warmer climate conditions the frequency of North Atlantic polar lows will decrease and their latitude will be shifted further north because stability over the Atlantic Ocean will increase.

The latitude of a cyclone track does not necessarily determine the amount of deposition. Even if the cyclone crosses the central BS Proper, it still depends on the stability of the atmospheric boundary layer over the pollutant emission areas whether contaminants are accumulated there into the air or not. On the other hand, if the cyclone were to follow a more northerly route along the Norwegian coast, there might still be a wet episode over the BS connected with fronts, or a dry episode event caused by turbulence over the water, if a simultaneous favourable flow from intensive emission areas occurred. Areas of rain associated with cyclonic activities can be located quite far from the cyclone centre. The influence of weather has to be analysed by studying each episode case-by-case, using backward simulations and by checking weather conditions along the whole transport path: local instantaneous conditions over water bodies do not explain a great deal.

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