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ORIGIN AND POSSIBLE EFFECTS OF EPISODIC NUTRIENT DEPOSITION EVENTS OVER THE BALTIC SEA

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Abstract: The focus of the paper is to introduce a tool for identification the main geographical areas from where the pollutants deposited to the Baltic Sea sub-basins during main episodic events are emitted to the air. This is important for estimating where emission control activities are required to be made in order to reduce harmful effects of eutrophication of the Baltic Sea. Additionally the possible influence of airborne nutrient load on the spring phytoplankton blooms is studied by looking causal relationships of deposition events and Chlorophyll-a concentrations (bloom intensity and length) over the Gulf of Finland and the Baltic Proper.

Key words: *Baltic Sea, airborne oxidized nitrogen load, episodes.*

INTRODUCTION

The Helsinki Commission (HELCOM) has kept a list of the major polluters (hot spots) of the Baltic Sea (BS) originally compiled within the Baltic Sea Joint Comprehensive Environmental Action Programme in 1992. The list contained mainly waste water sources and was used to reduce fluvial nitrogen (N) and phosphorus (P) loads to the BS. As a result several municipal wastewater treatment plants have been constructed around the sea. Because the airborne load will also cause significant adverse effects in the marine environment of the Baltic Sea area, the strategies for improving the environmental state of the BS should also include air emission control activities. Thus the location of individual sources influencing the airborne nutrient load to the Baltic Sea should be known.

Nitrogen deposition to the Baltic Sea is episodic: According to Hilatar model (Hongisto, 2003) simulations, between 2000-2009 over the Gulf of Bothnia 10 % of the annual wet load of oxidized nitrogen (NO_y) was received on average during 66 hours. In 2006, over all sub basins 50% of the total NO_y-load accumulated in 116-159 h, 80% in 333-434 h. The frequency of episodes had distinct minima in 1995-1997 and 2001-2005, and there was again a decrease in 2009.

The variation of the episodically received NO_y-load cannot be explained by instant local meteorological factors (wind, pressure, turbulence, state of other ABL-elements or cyclone overpass) because the dependency is not linear even with the precipitation; nitrogen compounds stay in air several days and each episode is a result of a chain of events connected to cyclone and frontal activity. Precipitation is connected to fronts and polluted air is mixed into varying vertical depths, circulated to the deposition area along complex paths and needs to be chemically converted to scavengable forms during the transport before deposited. The connection of episodes to meteorological events in 1993-2009 was studied in Hongisto, 2011.

To study the origin of the nitrogen deposition, either source receptor (SR)-matrixes or backward simulations can be used. SR-matrixes, used as by the EMEP-model, cannot be used to study episodes; the results have to be integrated over at least one month to avoid negative fields. Here we estimate with backward simulations from where the NOy of the ten highest deposition events for each BS sub-basin originate, in what kind of conditions the emissions were accumulated in the air, which kind of meteorological events typically persist over the emission area and how long the compounds stay in air.

The primary factors that limit the marine phytoplankton growth are light, temperature and nutrient availability (N, P, Si, Fe). Water stratification, storms and upwelling determine the mixing of the bottom nutrients to surface and in the BS the frequency of salt water inflow is important (Hagström et al., 2001). All algae species need both N and P for growth. Empirical data over the BS shows that for the vernal bloom species the N/P threshold limit is 7.2 by mass, (16:1 by mole M), while for the growth of cyanobacteria who can fix dissolved N, the N/P limit is 15 by mass (Håkansson and Bryhn, 2001).

In the Bothnian Bay (BB) where iron hydroxides transported to the water via rivers precipitate PO4, the inorganic N:P ratio is > 70 (M) and the limiting nutrient is P. Due to weak stratification and short light conditions no spring bloom occur there. During spring, in the Bothnian Sea the N:P ratio is around 10 (M), in the Northern Baltic Proper (NBP) and Gulf of Finland (GoF) N:P is around 3 (M; Hagström, 2001). Håkansson and Bryhn (2008) report very few monthly average mass-TN/TP-values lower than 7.2 concluding that the primary production (PP) is generally limited by phosphorus. But, nutrient limitation patterns have changed in time and switch during seasons and depend on the distance of the sampling site to the coast; the open waters of the GoF and the Baltic Proper were reported to be N-limited in springtime 2000-2005 by HELCOM (2009).

Most of the sedimentation to the BS bottoms occur during the vernal bloom. The cycling of nutrients to keep up primary production is different during spring and summer blooms. 10-50 % of the total primary production is transformed to dissolved organic matter (DOM), which only bacteria can use and convert to dissolved nutrients to fuel the PP loop (Hagström et al., 2001). During spring, zooplankton and bacteria populations are low, in summer high, thus in summer the nutrients are recycled efficiently, and the PP is not so much dependent on external load.

The timing, intensity and length of the springtime phytoplankton blooms over the BS sub-basins vary considerably. It is traditionally explained by the large inter-annual variability on surface water conditions in spring due to varying ice winter strength, storm frequency and mixing conditions. According to Hagström et al., 2001 in early spring after vertical mixing the water is rich in nutrients, and sufficient light cause exponential growth of algae until the limiting nutrient is finished. In low wind conditions algae accumulate in the upper layer. However: spring blooms start over open water areas where the vertical mixing does not necessarily reach the bottom. Additionally the start time of the vernal bloom over the GoF varied with even by two moths (from 27 Feb to 26 Apr in GoF, from 3 March to 25 April in NBP and from 11 March – to 9. May over the Arkona Basin AB). The bloom intensity varied by a factor of five (241 - 1966 GoF, 140 - 522 NBP, 0-307 AB) and the bloom length from 28 to 78 days (GoF), 23 to 56 days (NBP), 0-32 days (AB) during 1992-2008 (Fleming-Lehtinen and Kaitala 2008). Additionally the biological species constituting the blooms can change from year to year.

The hypothesis is, that the airborne nutrient deposition episodes will influence the start time and intensity of the phytoplankton blooms. To test the hypothesis, the causal relationships of deposition events and the springtime Chlorophyll-a concentrations over the Gulf of Finland and the Baltic proper was studied using the Algaline data published online in HELCOM fact sheets. The satellite pictures contain time series data mainly of the summertime blooms, and the measurement data (ICES biological plankton data) proved to be rather fragmented in time and space and was difficult to interpret.

MATERIALS AND METHODS

The origin of the nitrogen deposition over the BS was studied using the results of the chemistry-transport model Hilatar (Hongisto, 2003), using the forecasts of the FMI operational hydrostatic weather prediction model HIRLAM (HIgh Resolution Limited Area Model). Only results of the year 2009 are presented here. The BS sub-basins: the Gulf of Bothnia (B1), the Gulf of Finland (B2), the Northern Baltic Proper (B3), the Southern Baltic Proper (B4) and the Kattegatt and the Belt Sea (B5), are presented in Hongisto, 2011 Fig 1. The location and time of ten highest accumulated 6h deposition events over these five BS sub-basins was picked up from the gridded time series of the model results data base. The model was modified to perform backward simulations by fixing to the grid point of the maximum deposition location a chemically inert delta function at all 21 vertical model heights and turning the wind direction and time to opposite directions. Vertical mixing and deposition processes were ignored in the backward simulations. The advection of the maximum concentration during 52 hours before the deposition was followed at each vertical layer. Time series of selected meteorological parameters and emission of grids along each transport path were picked out. Because most of the deposition was in form of NO_3 or HNO_3, in estimation of the possible emission areas 15 hours minimum conversion time was assumed. Actually the chemical transformation time is longer in winter, but a more specific analysis on the influence of the chemistry will be performed later.

For estimating the connection between the algae blooms and deposition, spring bloom intensity index, length, start date and mean and peak concentration data were picked from HELCOM web site for the period 1992-2008 (Fleming-Lehtinen and Kaitala, 2008). The same parameters for the years 2009 and 2010 were estimated from the graphical presentations found at the HELCOM fact sheets. Accumulated NO_y deposition over the respective BS sub-basins during the bloom and 10, 20 and 30 days before it were picked from the simulated 6h deposition time series over the period 2000-2010.

RESULTS AND DISCUSSION

The ten highest 6h deposition events occurred mostly in winter months. The wet deposition share during the episode moments was higher than the annual average, varying from 79 % to 62 % and being higher in the northern model areas. When meteorological conditions were investigated closer for some events, the deposition maxima seem to be connected to movement of low pressure areas from Central Europe towards northwest. Majority of the deposited compound was either NO_3 or HNO_3. From the chemical composition of the deposition it was estimated simply, that the air parcel washed down has to be over 15 hours old; the correct age of the sample is longer, but it depends on the relative emission amounts along the transport path, latitude, local time and season.

The backward trajectories at four model levels drawn over the emission map are presented in Figure 1. From the detailed maps one can roughly estimate where the most probable emission areas are located: mainly in Western and Central Europe. However, in order to identify areas more close, time series of meteorological and emission parameters at the grid points along the trajectories were processed to pick up those grids, where the age of the air parcel was over 15 hours, grid emission was higher than 100 t NO_2/month and simultaneously the mixing height was below 300 / 600 / 1000 / 1300 m (at model levels 1-4, 5-8, 9-13 and 14-17, respectively). Corresponding locations of the emission areas fulfilling the criteria are presented in Fig. 2. In Table 1 average meteorological conditions at those potential emission areas are presented. On average the mixing height was lower than expected and there was only weak rain if any. The wind velocity is the average wind velocity of the respective model height interval, thus at the lowest level (around 30 m) it must not necessarily be very slow even during an inversion. It can be estimated, that for the lowest trajectories the emission areas are most propable, while the inversion situations are clear. For the upper trajectories the inversion can prohibit mixing to the upper levels, although in the model weak vertical mixing across the hmix is allowed. This parameterization follows the results of one early US National Acid deposition study, where it was concluded that the atmospheric surface layer is never completely separated from the upper air.

When the timing of the episode events was compared with the start times of the algae blooms from the Algaline data over the GoF and NBP, no connection between the number of episodes per year or the time of a maximum episode with the start time of an algae bloom seems to occur. Also when the start of the algae bloom, bloom intensity index (BII), the length of the spring bloom (LSB) and peak chloropfyll *a* concentration over the GoF and NBP were compared with the maximum ice extent (MIE) of the BS by scatter plots and correlation analysis, no dependency was found. The strongest anticorrelation founded was -.35 between the start of the ice season and MIE over the NBP. Corrrelation was 0.35 between the LSB and BII and MIE without any statistical significance. The correlations of the cumulative NO_y deposition accumulated during 10, 20

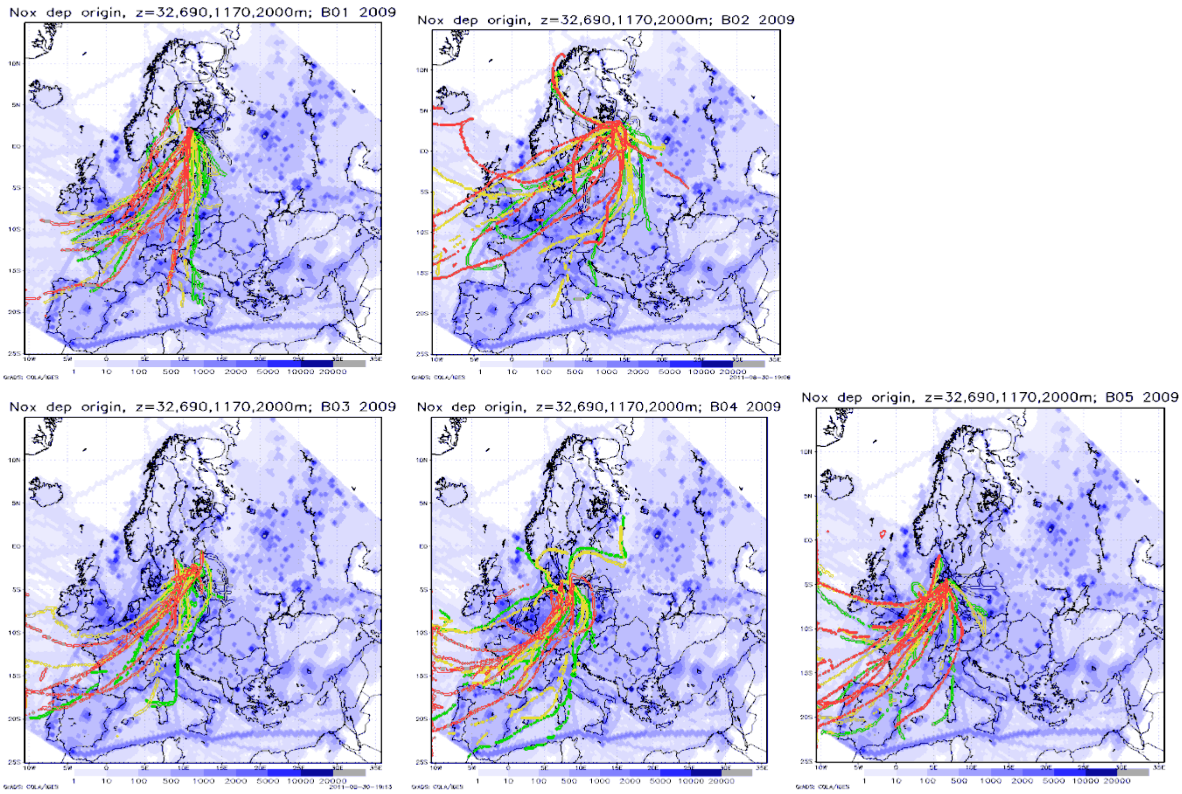

Figure 1. Backward trajectories from the 10 highest deposition episode locations. Black at z=around 30 m, green, yellow and read at heights of around 690m, 1170 and 2000 m, respectively.

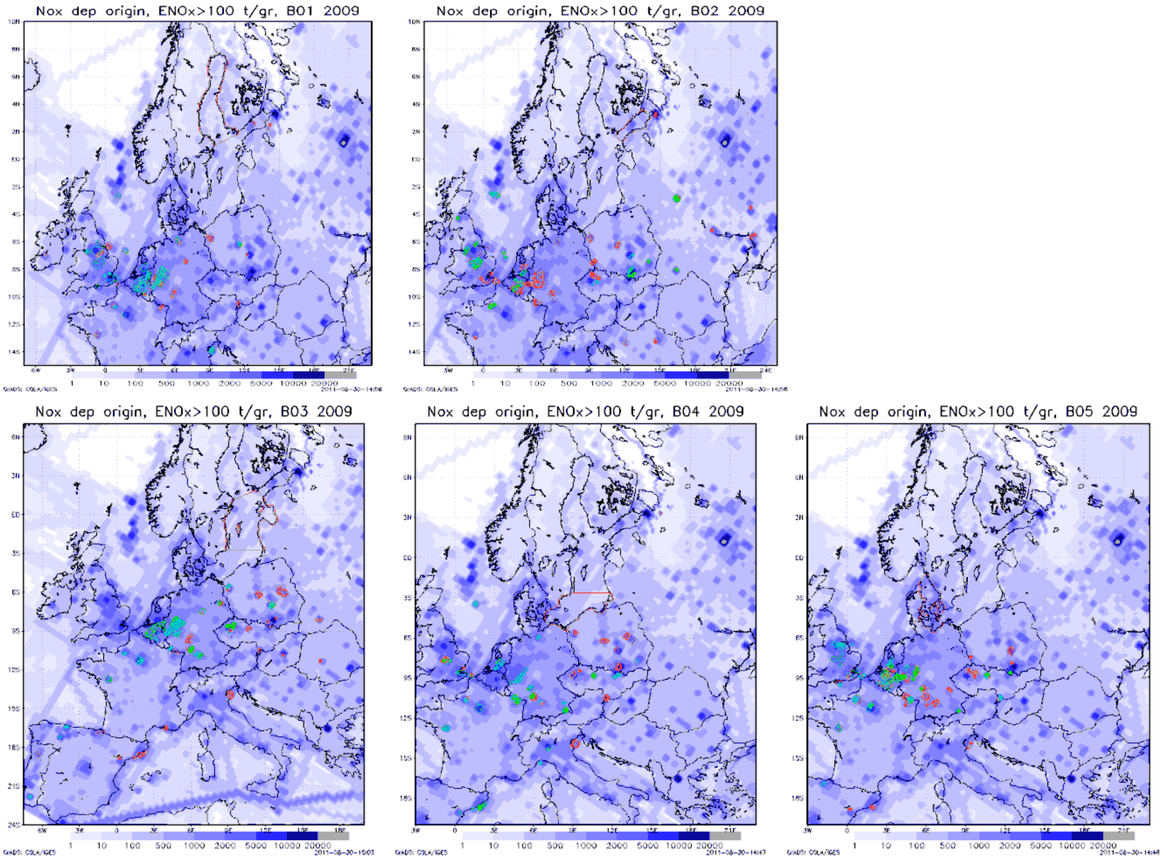

Figure 2. Possible locations of the emission areas of the peak deposition events. colours red, green and turquoise are at heights of z<650, 700-1200 and 1300-1400 m, respectively

Table 1. Average conditions at potential emission areas along trajectories. Sub-basin, model level, trajectory height, emission of the point, mixing height (hmix), surface temperature (T_s) , wind velocity (uabs), surface pressure (p_0), precipitation (prec), average cloudiness of the height interval (clf(z)), average total cloudiness of the vertical column (clf), Number of locations filling the criteria hmix < selected level and emission > 100 t NO2/month, (N), age of the air packet (age).

sub-basin	level	tr.height (m)	emi (t NO_2/m)	hmix (m)	Ts (°C)	uabs (ms^{-1})	p_0 (Pa)	prec. (mmh^{-1})	clf(z)	clf	N	age (h)
B01	1	32	227	188	1.2	7.8	100498	0.03	0.14	0.84	10	36
B02	1	32	142	219	-1.7	2.2	99820	0.05	0.59	0.48	41	41
B03	1	32	181	167	16.7	3.6	99057	0.01	0.03	0	15	25
B04	1	32	182	196	6.5	4.2	99895	0	0.11	0.12	43	40
B05	1	32	123	156	-0.6	5.6	98830	0	0.78	0.67	34	32
B01	2..6	..350	152	294	2.3	11.6	99431	0.05	0.24	0.56	60	38
B02	2..6	..350	162	197	4.1	7.2	100258	0	0.14	0.32	261	40
B03	2..6	..350	180	284	16.6	6.9	98907	0.11	0.02	0.04	99	34
B04	2..6	..350	186	286	14.4	8	99183	0.32	0.07	0.07	86	38
B05	2..6	..350	170	216	6.1	8.8	98060	0.02	0.3	0.42	158	27
B01	9..12	..815	153	371	7.7	15.3	99736	0	0.07	0.07	62	40
B02	9..12	..815	164	332	8.1	12.8	99988	0	0.01	0.41	87	31
B04	9..12	..815	152	412	11.2	9.8	100013	0.1	0.02	0.16	95	33
B04	9..12	..815	225	394	13	7.1	99166	0.26	0.09	0.12	163	38
B05	9..12	..815	138	235	8.4	8.9	99807	0.04	0.1	0.07	174	34
B01	13..18	..1420	146	503	3.5	14.3	99795	0.09	0.13	0.17	341	42
B02	13..18	..1420	220	543	4.2	18.1	98879	0.13	0.03	0.18	117	26
B05	13..18	..1420	176	500	9.7	15.7	98856	0.22	0.06	0.01	193	26
B04	13..18	..1420	126	438	7.7	13.1	98700	0.13	0.04	0.2	161	28
B05	13..18	..1420	146	461	7.6	12.7	99455	0.11	0.12	0.18	291	29

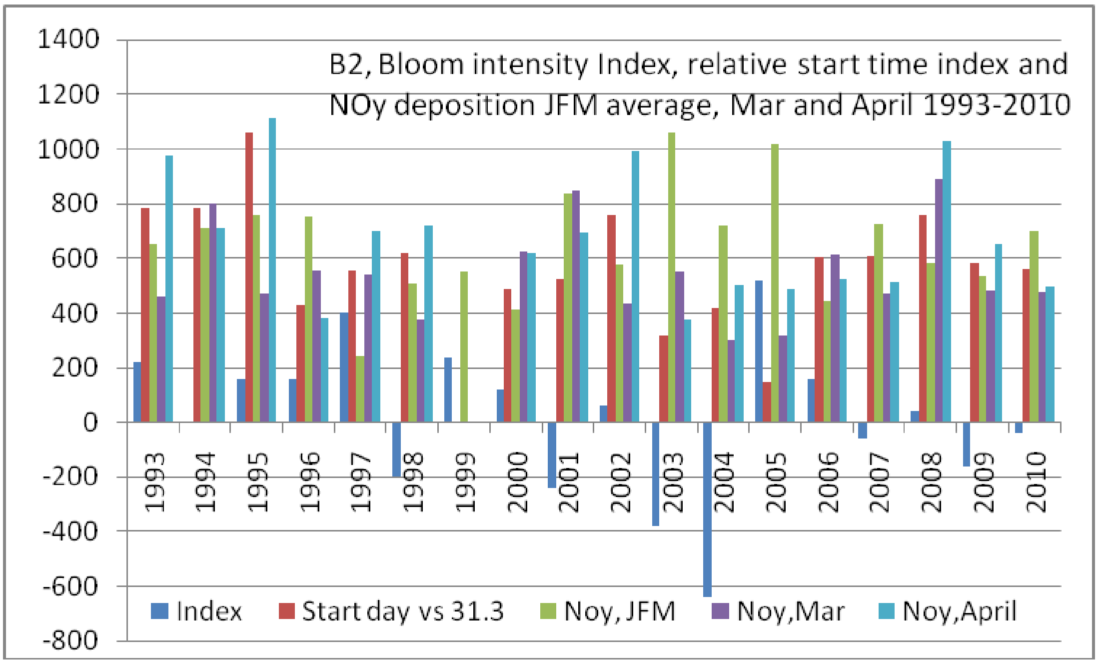

Fig. 3. The western GoF vernal bloom intensity index, the scaled start time index relative to 31 March and the average NO_y-deposition to the GoF in January-March, in March and in April, 1993-2010.

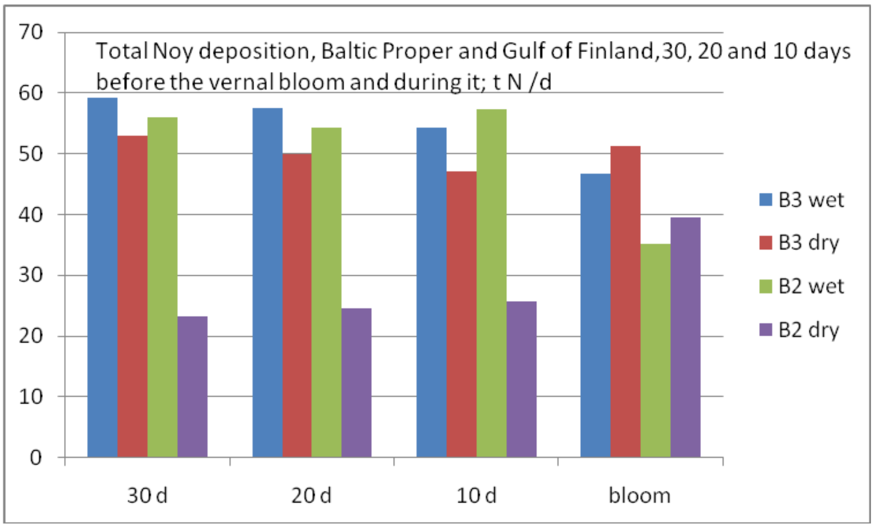

Fig. 4 Average NO_y deposition to the Baltic Proper and the Gulf of Finland, t/day 30, 20 and 10 days before the bloom and during it.

and 30 days before the vernal bloom with the bloom intensity index in 2000 – 2010 were negative, -0.35, -0.24 and -0.36 respectively. To illustrate the variation between the years, the spring bloom intensity index over the Western GoF, the start time index relative to 31 March and the average NO_y-deposition to the GoF in January-March, in March and in April over the years 1993-2010 are presented in Fig. 3. Correlation of the airborne NO_y deposition with the bloom intensity index was 0.58 over the GoF B2 and 0.64 over the NBP, B3, but this is obvious and comes from the definition of the bloom intensity index which is proportional to the bloom duration: the accumulated deposition is normally higher if the time period is longer. Actually the daily NO_y deposition during the spring bloom was below the average daily deposition before the bloom as indicated in Fig. 4. There was an increase of the dry deposition share during the bloom. This comes from the general shift of the stratification of the atmosphere towards later spring: the atmosphere becames more stable, precipitation and wind velocity decrease. Both wet and dry deposition have their minimum seasonal values in spring over the Baltic Sea. Work for studying the connection between the meteorological conditions and spring bloom intensity parameters is going on.

FURTHER RESEARCH NEEDS

The study to identify origin of deposition episodes will be extended to cover more years and more frequent episodes instead of 10 per sub-basin in order to better estimate where the main emission areas contributing to the deposition locate. The areas where episodes are originated in this study are much more in South-West and Central Europe than which can be concluded from the EMEP source-receptor matrixes.

The airborne NO_y deposition is important for the vernal blooms to fuel the nutrient cycle, although there was no clear connection between the start time and the episode frequency, and the hypothesis set in introduction, a causal relationship between the deposition and start of a bloom, could not be proven to be a clear fact. The biogeochemical marine cycle is very complex; at local scales changing nutrient concentration does not always elicit a response of changing phytoplankton biomass or production (Li et al.,2010). It is necessary to study longer time periods over different BS sub-basins in detail.

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