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The Effect of Riverine Nitrate Loads on Winter Concentrations in the Great Belt, Denmark

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This paper presents a method that models the fraction of the NO_X concentrations (nitrate and nitrite) in a marine recipient resulting from riverine runoff from land. The analysis is performed on the surface waters in the Great Belt, Denmark, during winter seasons. The method is based on simulation of two independent NO_X fractions: First, the "background" fraction, which results from the oceanographic mixing in the Belt Sea between Kattegat water and Baltic Sea water, and second, the nutrient load from land. The calculation of the background fraction is based on the finding that the Kattegat water has high salinity and high NO_X contents whereas the Baltic Sea water has low salinity and low NO_X contents. By means of an empirical relation between salinity and NO_X a specific salinity measurement in the mixing zone can be related to a "background" NO_X concentration. Measurements in the Great Belt show to be higher than the calculated "background" concentrations, which indicates the presence of a second NO_X fraction. The second fraction is defined as the difference between the calculated background concentration and the measured NO_X concentration. The hypothesis of the present paper is that this second fraction is dominated by the river runoff. Cross correlation analysis between the time series of the river NO_X load and the time series of surplus concentration in the Great Belt reveals a time lag between maximum river load and maximum surplus concentration during winter seasons of approx. 1 month. An empirical transfer function has been developed in order to connect the daily NO_X river loads with the surplus concentration in the Great Belt. The development of the NO_X-concentration in the Great Belt due to river run off is modelled for 8 specific winter periods of 3 months (December – February) between 1988/1989 and 1995/1996.

The model simulates concentrations on a weekly basis and quantifies the NO_X-increase during each winter based on the river load data. The analysis indicates that the NO_X concentration can be doubled during winters with high runoff, giving rise to an increase of approx. 60 µgN/l at the end of February. In winters with minimal runoff no significant increase above the background level is found.

Introduction

Eutrophication

During the last 20-30 years severe eutrophication effects have been observed in the Danish marine areas. In HELCOM (1996) it is concluded that the long term development of the inorganic nitrogen concentration follows the development of the nitrogen load which approximately doubled between the mid 1950s and the 1980s. In order to reduce the eutrophication effects in the marine environment the Danish government passed The Action Plan for the Aquatic Environment in 1987. This law includes two key assumptions: 1) The nitrogen concentration in the sea has increased, and 2) a substantial contribution of nitrogen comes from (farm-) land. The law states that loads of nitrogen from the open land as well as from cities should be reduced by 50% compared to the level during 1980-1985. In the years after launching the action plan the nitrogen loads of the rivers varied due to the varying freshwater discharge, only. When the load was corrected for varying discharge, the reduction aim of the plan was not reached (Fyns Amt 1997).

Hydrographic Processes

The general hydrographic processes in the Kattegat and the Belt Sea are of specific importance, because they are responsible for the transport and mixing processes of nutrients. In the inner Danish waters these processes are highly complex, resulting in changes of the state variables within few metres in depth, few kilometers in space and within hours to few days. The hydrography of the Belt Sea and the Kattegat has been studied for approx. 100 years (Knudsen 1900). A brief introduction of the most important transport and mixing processes in the Belt Sea and Kattegat will be given in the following.

The three narrow sea straits through Denmark connect the North Sea with the Baltic Sea, see Fig. 1. The position between two seas makes the hydrography for this region different from oceans and estuaries. The water from the southwest Baltic Sea is characterized by low salinity (~8 PSU) as well as low NO_X concentration (~60 µgN/l) during winter, whereas the water from the Skagerrak is characterized by high salinity (~33,5 PSU) and high NO_X concentration (~150 µgN/l), (HELCOM 1996). In the transition zone the brackish and therefore less dense Baltic water floats above the more saline and hence more dense Kattegat water and forms a vertical density

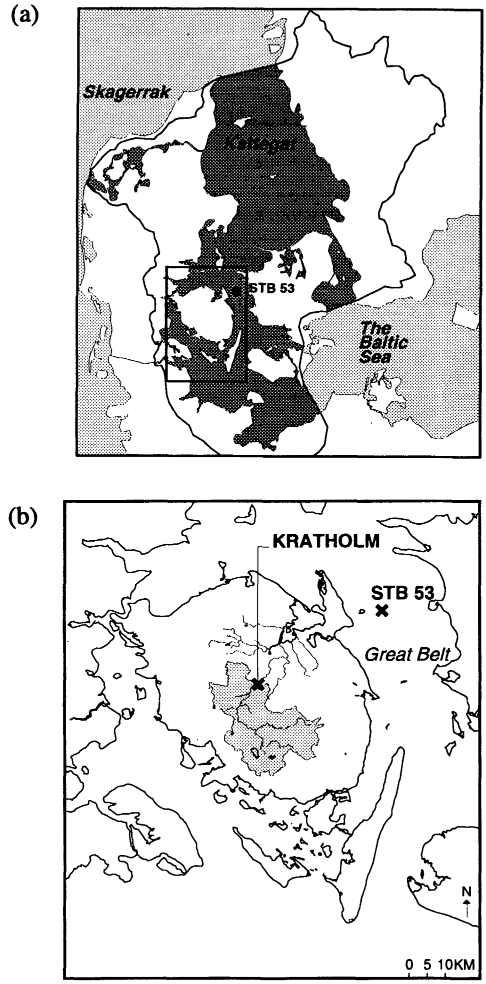

Fig. 1. a) Map of the Kattegat and the Belt Sea with surrounding catchment area. b) The island of Funen with the position of the marine monitoring station STB53 and of the station "Kratholm" in river Odense.

stratification. The continuous mixing due to current and wind results in horizontal gradients of both salinity and NO. Due to meteorological forcing of wind and air pressure differences between the North Sea and the Baltic Sea these salinity- and NO_X-gradients are moved southward through the Belt Sea for westerly winds resulting in increasing salinity and NO_X values in the Great Belt. For easterly winds the salinity and NO_X values in the Great Belt decrease correspondingly. The above addressed processes are analysed in detail in (Jacobsen 1980; Lass 1988; Stigebrandt 1983; Stigebrandt 1992). In recent work (Holtegaard 1998) the baroclinic and geostrophic effects within the Kattegat/Skagerrak region are investigated in detail, explaining frontal behavior and detailed circulation patterns, including the adjustment to rotation. The work, however, verifies the general flow patterns as described above.

The Modelling Approach

The scope of the present model is to describe the fractions of background and surplus concentration and the sum of these two fractions based on measured river load of NO_X and the momentary salinity in the Great Belt.

Qualitative descriptions of the general eutrophication problems in the specific area have been performed by numerous authors (Andersson and Rydberg 1988; Miljøstyrelsen (Danish Environmental Agency) 1988; Richardson and Jakobsen 1988). The system of relevant processes are outlined by a few (Miljøstyrelsen 1979; Schrøder and Møller 1986; Schrøder and Malmgren-Hansen 1988; Ærtebjerg 1986). Modelling approaches including the hydrographic processes in terms of deterministic models have been made by very few authors (Hansen *et al.* 1990, Hansen *et al.*1994; Jürgensen *et al.* 1996). Although a relation between the runoff of NO_X from land and the NO_X concentration in the Danish waters during winter season is intuitively evident and statistically illustrated in several publications on averaged data (HELCOM 1996), it has not been possible to model the development of the NO_X concentration on a marine location with an accuracy that allows for comparison with individual field measurements. Therefore, it is assumed that the achieved accuracy of the present model must allow the comparison of model results with (weekly) field measurements of NO_X on the specific location, Fig. 1.

This way the present work differs in principle from the existing (descriptive, integrating) relations that are based on winter mean values of river runoff and winter mean values (typically 1-3 winter measurements per station) averaged for selected marine stations in the inner Danish waters (Ærtebjerg 1990).

Methods

It is the objective of the present work to describe the fraction of the NO_X concentration in the Great Belt that is caused by the river NO_X supplied to the Kattegat and the Belt Sea by the numerous rivers and creeks of the catchment area. Furthermore, the model for the entire NO_X concentration must achieve an accuracy which makes it possible to compare model results with (weekly) field measurements of NO_X on the specific marine location.

Field Data

The present work is based on a set of field measurements, including daily transports of NO_X in river Odense at the Kratholm station and measurements of NO_X and salin-

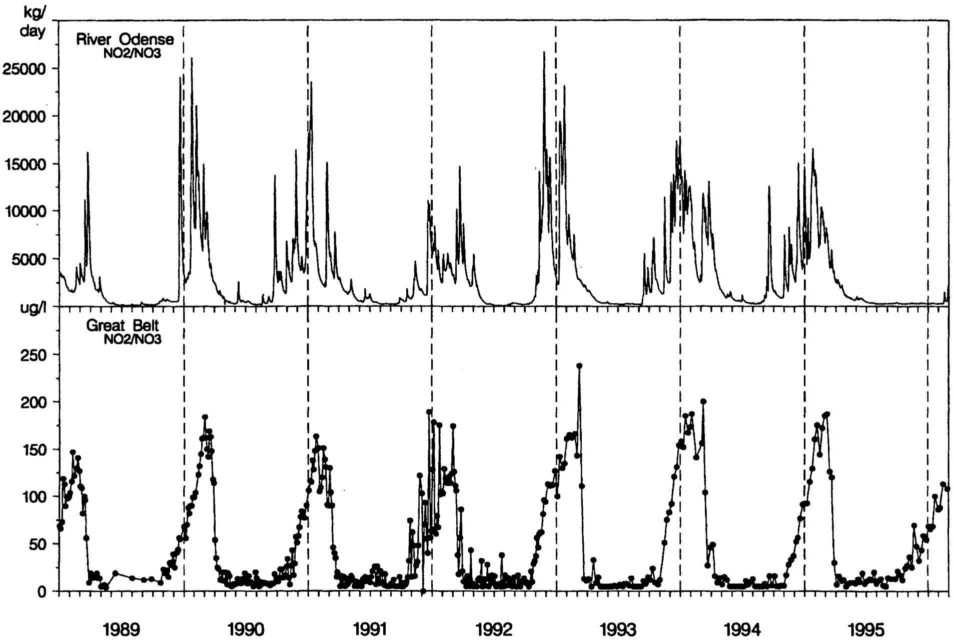

Fig. 2. Intensive measurements of daily river NO_x-transport and NO_x concentrations in the surface water of the Great Belt.

ity in the surface waters of the Great Belt at station STB53 with a relatively high sampling frequency of 2/week or 1/week, see Figs.1 and 2.

The river Odense is a representative for all rivers of the catchment area, because it represents the majority of tributaries of the region with respect to the temporal development of the runoff within each winter season (see Windolf 1998). The catchment area of the river Odense upstream from the monitoring station is 486 km^2, which represents approximately 4‰ of the total Swedish, German and Danish drainage area of 115,000 km^2.

In order to illustrate that the riverine nitrogen runoff is of consequence in the Kattegat and Belt Sea the following simple calculation can be performed: The total river runoff to the Kattegat and the Belt Sea of typical 130.000 tons total N (for 1990) (HELCOM 1996). The majority of this amount is discharged into the sea during the three winter months. For an order of magnitude consideration the entire transport is assumed to occur during this period. The validity of this assumption is illustrated by means of the daily riverine NO_x-transports in Fig. 2. The Kattegat and Belt Sea hence will receive 43.000 ton N per month during winter. Since the volume is approximately 683 km^3 (HELCOM 1996) the nitrogen increase per month would be of the order of 60 (gN/l, which is of the same order of magnitude as the natural background concentration. It shall be shown later that the typical time scale of residence is approximately 1 month. Therefore, there is good reason to anticipate that the riverine input is an important source for the increasing nitrogen concentration during winter.

Sampling and analysis techniques for the river samples are described in Fyns Amt (1997). The concentrations of NO_x in the Great Belt are measured during the winter season (December, January and February) where primary production is negligible, and the NO_x can hence be considered conservative. Sampling and analysis techniques for the marine samples are described by Fyns Amt (1996).

Simulation Approach

The general approach of the NO_x-modelling is based on the ideas presented by (Andersson and Rydberg 1988). The measured concentration of NO_x in the Belt Sea during winter can be divided into two fractions:

a) Background concentration from the adjacent seas
b) Surplus concentration from river loads

Background Concentration – The background concentration is determined based on mixing of the two water masses from the North Sea and the Baltic Sea, respectively. North Sea water is characterized by high salinity and high NO_x concentration and the Baltic Sea water by low salinity and low NO_x concentration. In a Salinity – NO_x – plot the regression line between the North Sea and the Baltic Sea represents all possible mixing ratios of the two water masses. If no additional NO_x is supplied to the surface waters in the Belt Sea, a sample from the Great Belt (the mixing region) will give a value on the regression line between the two water masses. The momentary salinity in the sample from the Great Belt expresses the final result of all mixing processes prior to the time of sampling. In combination with the "salinity – NO_x – relation" the measured salinity hence gives the background concentration of NO_x for any specific mixing situation. A detailed modelling of all (highly complex) hydrographic processes within the Kattegat and the Belt Sea is not necessary because the salinity of the specific sample is measured and included in the further calculations.

In order to define the water masses and to elucidate the uncertainty two different methods are applied.

The first method is based on winter average values of surface measurements in the North Sea and the Baltic Sea, see Table 1.

Table 1 – Long term average values of salinity and NOx during winter in the surface of the North Sea and the Southwest Baltic Sea (HELCOM 96).

	Salinity (PSU)	NOx (μg N l^{-1})
North Sea water	33.5	150
Southwest Baltic Sea	8	60

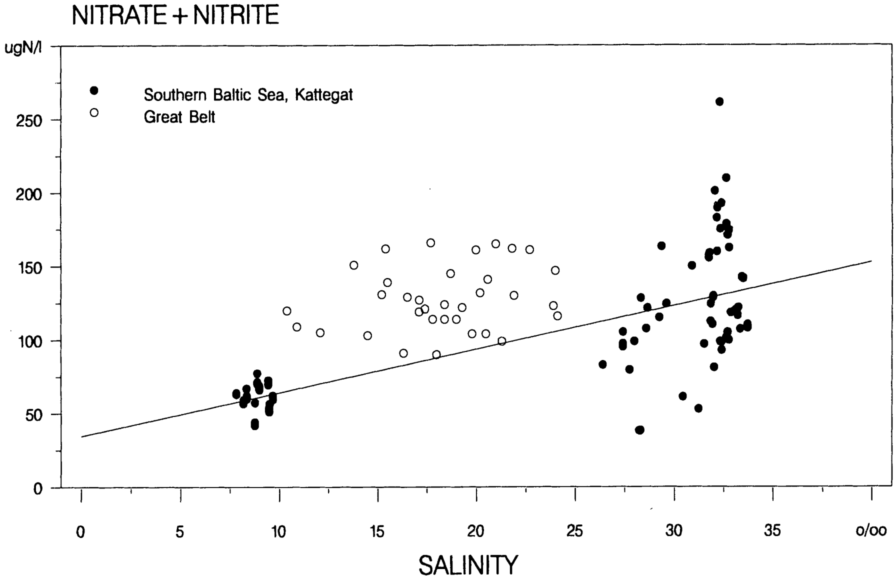

Fig. 3. Diagram of salinity and NO_X in the Kattegat bottom water (right) and the surface water of the southwesterly Baltic Sea (left) for all three winter months (Dec., Jan., Feb.). The line indicates mixing between the two water masses. The open circles in the middle are from the Great Belt surface. Open circles above the line indicate input of NO_X from land.

The advantage of this method is its simplicity. The disadvantage is that the impact of nitrogen loads on the surface waters within the same winter period has not been taken into account.

Fig. 3 illustrates the second method which is based on measurements from a bottom station in the Kattegat in order to describe water of North Sea origin which is not influenced by the runoff of the respective winter season. Surface measurements from the Arkona Basin during winter are used to describe Baltic Sea water. Linear regressions are performed on the measurements for each of the three winter months (December, January and February) in order to include the general increase during the winter period.

Despite the different approaches, the synthetic background fractions from the two methods only differ with approximately 7%, corresponding to ~5 µgN/l for salinities often found in the Great Belt, which in this context can be regarded as negligible. In the following analysis the background concentration is determined by means of the second method.

Surplus Concentration – The measured NO_X concentrations in the Great Belt typically lie above the regression line during winter periods (see Fig.3), indicating that additional NO_X is supplied to the Great Belt. This surplus concentration is assumed to be caused by the nutrient input from land in the winter season and is defined as the difference between the measured NO_X concentration and the corresponding background NO_X concentration outlined above.

Cross Correlation

In order to describe the net transport and dispersion processes of nitrogen within the inner Danish Waters a cross-correlation function is determined between the measured transport in the river Odense (representing all rivers) and the calculated surplus time series in the Great Belt. The analysis gives information about the extent of correlation between the two time series and about the time lag between maximum input (river) and maximum concentration (sea).

The Transfer Function

The transport processes from the river Odense to the Great Belt is described empirically in the present analysis in terms of a transfer function, which mathematically ties the daily NO_X-transport in the river Odense to the surplus concentration in the Great Belt.

The basic concept for transferring an impulse time series into a response time series by means of a transfer function and a subsequent convolution is a well known concept in statistics Jørsboe (1976) or Madsen (1989). The transfer function transfers a single impulse (NO_X-transport in river Odense of one day) into a response (the time series of the surplus NO_X concentration in the Great Belt due to the one day impulse). The responses for all individual impulses are then integrated (in terms of a convolution integral) in order to obtain a continuous response time series.

Since, to the knowledge of the authors, no transfer function exists that describes the present process theoretically, a standard transfer function is chosen empirically. For comparison reasons three standard transfer functions were applied and their parameters calibrated with respect to the time series of the surplus concentrations. The following correlation coefficients were found between the modelled and the measured NO_X concentrations, see Table 2. The data applied in this comparison are from the 5 winter seasons 1988/1989 to 1993/1994.

It can be seen that the transfer functions described by Gamma- and Normal distributions both give correlation coefficients higher than 0.95. The Gamma (3,14) trans-

Table 2 – Correlation coefficients R^2 between the modelled and the measured winter NO_X concentration in the surface layer of the Great Belt calculated for different transfer functions. Data are from winter 1988/1989 to 1993/1994.

Optimised transfer function	Correlation R^2
Gamma	0.96
Normal	0.95
Exponential	0.73

fer function, $G_{(3,14)}$, is chosen because this function has the highest correlation. Furthermore, this function has a "tailed" distribution (skewness), which is a well known phenomenon in river transports. No effort has been put into the physical explanation of the chosen parameters.

The formula for the applied Gamma transfer function *f* is given in Eq. (1)

$$f(k,\beta,t) = \frac{1}{\beta\Gamma(k)} \left(\frac{1}{\beta}\right)^{(k-1)} \exp\left(-\frac{t}{\beta}\right) \quad t > 0 \qquad (1)$$

where

k – shape parameter ('number of occurrences')
β – scalar parameter ('inverse intensity')
t – co ordinate of time

$$\Gamma(k) \equiv \int_0^{\infty} t^{(k-1)} \exp(-t)\,dt \qquad k > 0 \qquad (2)$$

The construction of a response time series *R(t)* based on a impulse time series *I(t)* is conducted as a convolution integral which is illustrated in Eq. (3)

$$R(t) \equiv \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} f(k,\beta,i)\, I(t)\, di\, dt \qquad (3)$$

where

i – independent co ordinate of time

The optimization procedure for the chosen transfer function is illustrated in Fig. 4, which shows the correlation coefficient as a function of different distribution parameters in a 3-dimensional illustration. The "peak" represents the optimal parameter set for the Gamma transfer function. Fig. 4 illustrates that an alternative choice of parameter sets along the "ridge" might only give minor loss of correlation.

Results

The results of the cross correlation analysis are illustrated in Fig. 5. This standard statistical analysis provides a continuous function that confirms the hypothesis of a correlation between the two processes. A clear correlation maximum is seen for a time lag between the impulse in the river Odense and the maximum response in the Great Belt of approx. 25 days, or 3-4 weeks. Since the NO_x transport in river Odense is considered typical for the entire catchment area, this means that the concentration of the surface waters in the Great Belt culminates approx. 3-4 weeks after culmination of the river loads. The period can be interpreted as a reaction time scale of the Belt Sea to major winter input of NO_x from rivers during winter.

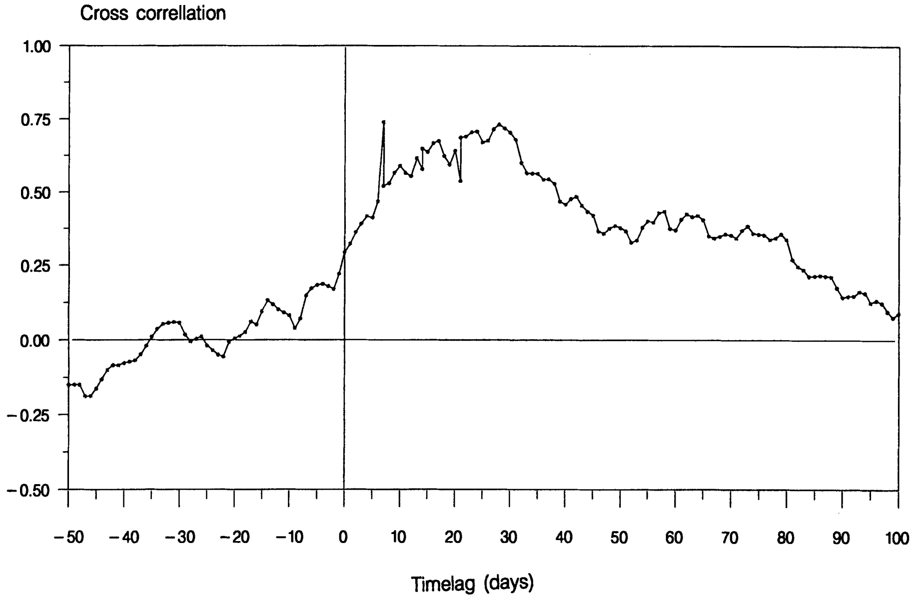

Fig. 4. Optimization of the parameter set in the gamma distribution. Maximum correlation between model and measurements is found for parameters on the "ridge" of the plot. The chosen point is for 3 on x-axis and 14 on y-axis.

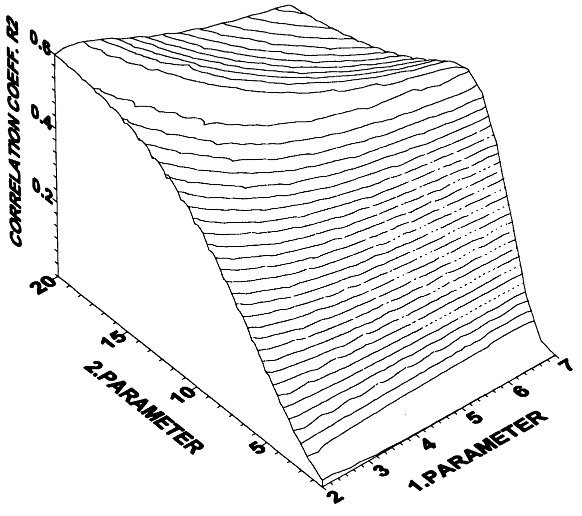

Fig. 5. Cross correlation function between the NO_X-load and the surplus NO_X-concentration in the Great Belt.

The Gamma (3,14) transfer function is applied on the time series of the NO_X-transport in the river Odense. The transfer function (Eq. (1)) also gives a time lag between impulse and maximum response of approximately 1 month. Furthermore, the transfer function reveals a time lag of approximately 1 month between maximum and the time where the response is decreased to 37% ($= e^{-1}$). This can be interpreted as the time scale for how fast a nutrient peak in the Belt Sea will be reduced towards background concentration. This latter time scale can be interpreted as a time scale of adjustment to the boundary concentrations of the Belt Sea.

The transformed time series is interpreted as the concentration surplus .due to riverine runoff and is added to the computed time series of the background concentration. The sum represents a synthetic time series for the NO_X concentration in the Great Belt on a daily basis. This time series is plotted in Fig. 6 together with the measurements. Since the surplus fraction of the synthetic time series is a result of multiple integrations during the convolution process the output will be smoothed. The time scale of this smoothing is estimated to approximately 1 week, based on the visual appearance of the synthetic time series for NO_X. In order to match this average time scale the measurements have been slightly processed with a sliding average of one week. Compared with the measurement frequency of 1/week or 2/week this smoothing process does not have significant effect on the measurements. It should be noted that the model is calibrated only on the first 5 winter periods.

Discussion and Conclusions

The results indicate that the concentrations in the Belt Sea respond very clearly and predictable to the runoff from land.

The atmospheric NO_X deposition is not included in the input data of the model. The concept of surplus concentration implies that the effect of direct NO_X load by precipitation on the sea surface is included in the statistical analysis. A dominating atmospheric deposition would result in an immediate increase of the NO_X concentration, *i.e.* a reaction time of 0 days, which has not been found.

In general, mineralization is an important process for the NOx supply in open waters. The mineralisation process is a bio-chemical process, which is mainly governed by available organic matter, oxygen concentration and temperature in the bottom water. Vertical mixing processes bring mineralised NO_X from the bottom to the upper layer. Organic matter, oxygen and temperature can be considered quasi stationary during winter season and can therefore not be responsible for variations of NO_X concentrations that are as rapid as observed by measurements. A coincidental similarity between vertical entrainment and precipitation on the one side and mineralisation on the other hand is unlikely because the processes are governed by different mechanisms. The effect of mineralization in the adjacent waters is included in the model in terms of boundary values that increase from December to February. For the

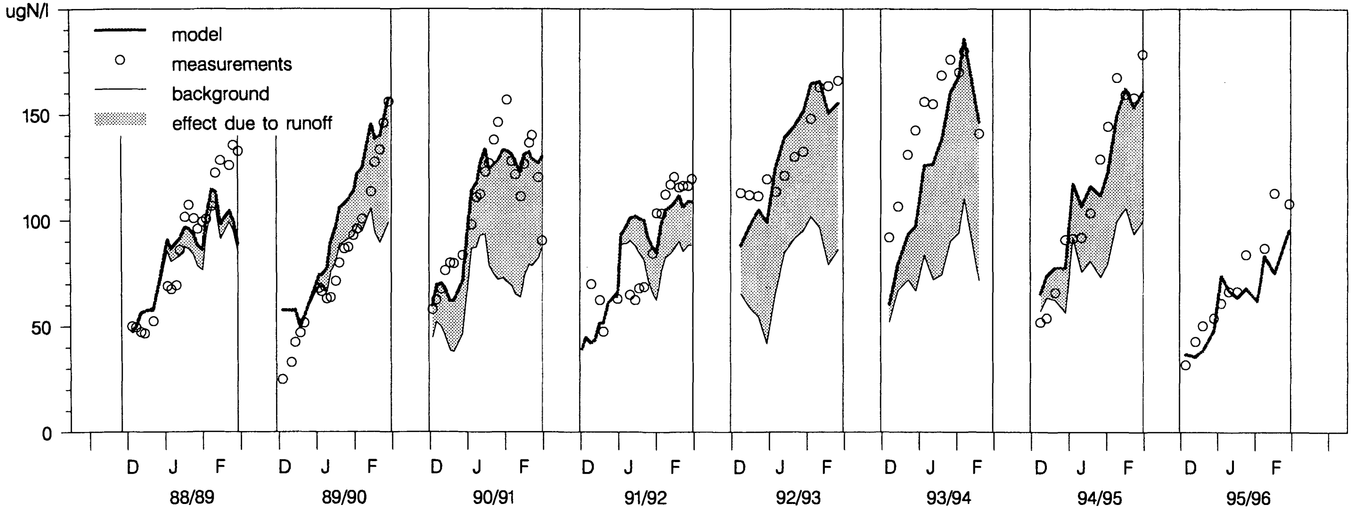

Fig. 6. NO_x-concentrations for the winter seasons (December through February) 1988/1989 – 1995/1996 in the Great Belt. The thin line indicates the background concentration, the thick solid line indicates the sum of background plus runoff induced concentration and the circles are the direct measurements. The shaded area illustrates the increase due to the run off from land. The winters from 1988/1989 to 1993/1994 are used for calibration. The remaining winters are used for verification.

present model the results seem to indicate that in this area the effect of local mineralisation is less significant than advection and river load.

Several features in the result plot in Fig. 6 should be stressed:

1) The time lag between culmination of the river loads and culmination of the surplus concentration in the surface waters in the Great Belt is approximately 1 month.
2) The time scale for concentration decrease after culmination in the surface waters of the Great Belt is also approximately 1 month.
3) Winter periods with relatively small run off from land (*e. g.* the winter 1995-96) show no or relatively small NO_X increase compared to the background concentration, whereas periods with relatively high run off (*e. g.* the winter 1994-1995) can double the natural background concentration in the Great Belt, corresponding to an increase of approximately 60 µgN/l.
4) The development within each specific winter season of the modelled concentration shows high qualitative similarity with the corresponding measured time series.
5) The general accuracy of the modelled concentrations is found to be satisfactory compared to the measurements in the calibration period 1988/1989 to 1992/1993. The accuracy for the following winter periods does not seem to differ significantly from the accuracy of the first winter periods, although the later winter periods were extreme compared to the winters in the calibration period.
6) It is seen from Fig. 6 that the accuracy of the model increases within each winter season, indicating a high degree of stability of the model. It is of high environmental relevance that the final concentration surplus, just before the start of the spring bloom, is modelled with the highest accuracy.
7) The model is able to reproduce individual features of the NO_X development in the Great Belt within the 3-month winter period. The features are governed by the specific run off events.
8) In operational mode the model is able to forecast the surplus NO_X concentration for a period of approximately one month during winter.

The presented result gives new insight into the nitrate concentration response in the central areas of the inner Danish waters due to input from land for each specific winter. A typical time scale for concentration reaction is approximately 1 month for increase as well for decrease and illustrates how rapid the mechanisms for transport and mixing are in the inner Danish waters (on average and in winter seasons).

Further, the NO_X concentration is described accurately at the end of each February, just before the start of the spring bloom. Instead of operating with winter mean concentrations this model shows the continuous development of the anthropogenic surplus-concentration. The present method represents a fast and economic way to model the key eutrophication parameter NO_X in the open parts of the Danish waters with high accuracy. The results demonstrate, that high frequent field measurements (as in the river Odense and in the Great Belt) provide data that make a quantitative analysis possible. The term "high frequent" is in this connection defined as 8-16 measurements per time scale. The time scale of the runoff events is approx.1-2 weeks, which demands daily measurements, the time scale of the dilution process in the Great Belt is found to be approx. 2 months, which demands weekly or half-weekly measurements.

The presented work indicates the need for key monitoring stations with sampling frequencies that match the time scales of the relevant processes. Key monitoring stations should measure all relevant parameters and processes (physical, chemical and biological) and monitoring should be combined with modelling of the same parameters and processes.

Finally, the present results confirm the basic hypothesis in the national "Aquatic Environment Plan" from 1987 that assumed a close relation between the nitrogen loading from land and the nitrogen concentration in the open sea.

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