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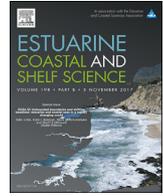
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Reconstructing the history of eutrophication and quantifying total nitrogen reference conditions in Bothnian Sea coastal waters



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ABSTRACT

Reference total nitrogen (TN) concentrations for the Gårdsfjärden estuary in the central Bothnian Sea, which receives discharge from an industrial point-source, have been estimated from diatom assemblages using a transfer function. Sedimentological and diatom evidence imply a good ecological status before 1920 with an assemblage dominated by benthic taxa indicating excellent water transparency, high diatom species richness and less organic sedimentation resulting in homogeneous well oxygenated sediments. A change in the diatom assemblage starts between 1920 and 1935 when the species richness declines and the proportion of planktic taxa increases. Increased organic carbon sedimentation after 1920 led to hypoxic bottom waters, and the preservation of laminae in the sediments. The trend in the reconstructed TN-values agrees with the history of the discharge from the mill, reaching maximum impact during the high discharge between 1945 and 1990. The background condition for TN in Gårdsfjärden is 260–300 $\mu\text{g L}^{-1}$, reconstructed until 1920.

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1. Introduction

The Baltic Sea is one of the most polluted seas in the world (Fonselius, 1972), an effect of 85 million inhabitants in the catchment of a basin with limited exchange with the open ocean. Humans have influenced the environment in the Baltic area for thousands of years, both locally via land-use changes (Bradshaw et al., 2005) and regionally as an effect of e.g., metal industry (Brännvall et al., 1999). Despite the duration of human occupation, it is mainly after the industrialization and introduction of artificial fertilizers following World War II that major changes in nutrient loads occurred (Gustafsson et al., 2012) and effects of eutrophication are recorded in Baltic Sea coastal waters (e.g., Elmgren, 1989; Bonsdorff et al., 1997). Eutrophication, defined as an increase of the rate of supply of organic matter to an ecosystem (Nixon, 1995), increases primary production and causes more frequent hypoxia at the sea bottom both in open water (Jonsson et al., 1990; Carstensen et al., 2014) and in the coastal zone (Persson and Jonsson, 2000; Jonsson et al., 2003; Conley et al., 2011). The limiting nutrient for

primary production in the open Baltic proper is nitrogen (Granéli et al., 1990), whereas it is phosphorus in the Bothnian Bay (Andersson et al., 1996). Model simulations show that the entire Baltic Sea was more limited by phosphorus 100 years ago than today (Savchuk et al., 2008). The Bothnian Sea, situated between the Baltic proper and the Bothnian Bay, has previously been considered as a transition zone with varying nutrient limitation, but as a result of eutrophication of the Baltic Proper a shift towards increased nitrogen limitation has occurred the last 20 years (Rolff and Elfving, 2015). Estuarine systems are, however, more complex than the open sea and display seasonal switches in nutrient limitation with in general phosphorus limitation in spring and nitrogen in summer (Conley, 2000).

Systematic monitoring of water quality in the Baltic Sea began, at the best, in the late 1960s to early 1970s (Cederwall and Elmgren, 1990) and background nutrient levels of pristine ecosystems are not known (Larsson et al., 1985). The establishment of reference conditions is needed to identify how present day conditions depart from them and to set targets for achieving good ecological status of all European waters within the European Union Water Framework Directive, EU WFD (European Union, 2000; Andersen et al., 2004). Further, a historical perspective is important for managing impacted marine ecosystems as it can indicate trajectories of

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change that more traditional local ecological studies cannot (Hughes et al., 2005). A palaeoecological approach extends beyond the time covered by instrumental records and allows present day observations and ecosystem function to be viewed in a long-term perspective (Bennion and Battarbee, 2007).

Abiotic factors, both physical and chemical, can be quantified and inferred from sub-fossil biotic assemblages, using calibration models (transfer functions) based on the modern relationship between species distribution and environmental parameters in an aquatic ecosystem (Birks, 1995). The transfer function concept was developed within palaeoceanography to reconstruct sea surface temperature (Imbrie and Kipp, 1971) and is today widely used also in palaeolimnology to reconstruct past pH and phosphorus concentration in lakes (Birks et al., 1990; Bennion, 1994) and climate change (Seppä and Birks, 2001). Diatom-based transfer functions have been used to reconstruct nutrient records of Danish (Clarke et al., 2003, 2006; Ellegaard et al., 2006) and Finnish coastal waters (Weckström et al., 2004; Weckström, 2006).

The aim of this study is to use palaeoecology to enhance the knowledge of historical nutrient concentrations in Bothnian Sea coastal waters which are affected by discharges of industrial point sources to establish reference conditions for annual mean total nitrogen. Reference conditions are needed for the EU WFD and our results are discussed in a wide context important to achieve sustainable environmental governance of the Baltic Sea.

2. Material and methods

2.1. Study site

Gårdsfjärden (61°37'N, 17°09'E) is a small, sheltered estuary of 4 km², situated on the east coast of central Sweden (Fig. 1). The estuary is connected to the Bothnian Sea via the narrow Dukarsundet with an 8.7 m deep threshold, which limits the exchange of water between the inner estuary and the open Bothnian Sea. The deepest part of Gårdsfjärden, 18 m, is situated close to the threshold and the mean water depth is around 6 m. The estuary was dredged in 1991, 1992 and 1994 to allow large ships to enter a harbour in the inner part. The most extensive dredging was carried out in 1994 when 240,000 m³ of sediment were removed from the threshold area. Gårdsfjärden is salinity stratified, with an annual mean

salinity, measured on the Practical Salinity Scale, of 2.4 at the surface and 4.7 at 10 m depth (measured between 1990 and 2001), which is similar to the surface water salinity of the open Bothnian Sea value of about 5.

Gårdsfjärden receives discharge from Delångersån, a regulated river with an annual mean flow of 10 m³ s⁻¹. The estuary also receives a discharge of about 1 m³ s⁻¹ from the pulp and paper mill at Iggesund. This site has an industrial history from 1685 CE, initially as an ironworks, expanding at the end of the 19th century to include a saw mill. After World War II, it became solely a timber concern. The discharge from the mill supplies the estuary with three times the natural annual input of phosphorus and nitrogen which amount to about 17 tons y⁻¹ TP and 170 tons y⁻¹ TN between 1992 and 1993 (Nilsson and Jansson, 2002). Technical improvements have drastically reduced the discharge below the maximum discharge in the 1960s (Nilsson et al., 2003). Water discharge data from the paper mill are available from 1940 and monitoring of the water quality in Gårdsfjärden was initiated in 1969. At first, monitoring concentrated on oxygen content, turbidity, colour and chemical oxygen demand (COD). Time series of nutrient concentrations start in 1980 with 6 measurements yearly. The annual mean nutrient concentrations between 1990 and 2001 of the upper 10 m of the water column was 23.2 µg L⁻¹ for total phosphorus (varies between years with 17.3–34.3 µg L⁻¹) and 337 µg L⁻¹ for total nitrogen (varies between years with 253–428 µg L⁻¹).

2.2. Core collection, geochemistry and dating

Core sites were selected after thorough investigation of the sediment accumulation in the estuary with a low frequency echo sounder (14 kHz) and side scan sonar (500 kHz) from R/V Sunbeam in September 2002. An east-west transect of five sites situated in the deeper part of the estuary, showing bottoms with soft layered sediment accumulation, were cored with an 8 cm diameter Gemini gravity corer which takes two parallel cores. One of the parallel cores at each coring site was sub-sampled in the field and refrigerated while the other was transported to the laboratory for lithological description.

The sediment was analyzed for total organic carbon (TOC) and nitrogen using a LECO CHNS-932. Water content was measured by weighing the sediment before and after freeze drying. Peak concentrations of ¹³⁷Cs, measured on ground, dry sediment using an Intertechnique 2000 gamma counter, were used as a marker horizon indicating the release of radioactivity from the 1986 Chernobyl accident. The investigated area is situated in the direct plume of the radioactivity fallout and there was no delay in the signal recorded in the sediment as seen in other parts of the Baltic Sea (Meili et al., 2000). Laminae in the cores were visually inspected and counted, and used together with the ¹³⁷Cs measurements to construct age models. The sediment core at site IGG 2 (Fig. 1) was found to have highest resolution and was accordingly selected for further analysis.

2.3. Diatoms

The sediment was cleaned for diatom analyses following Battarbee (1986) and mounted for permanent slides with Naphrax™. Diatoms were identified and counted under an Olympus BX51 light microscope using Nomarski differential interference contrast with magnification of 1000× and oil immersion. At least 500 valves, excluding *Skeletonema* spp. and *Chaetoceros* spp. were enumerated in each sample following the protocol of Schrader and Gersonde (1978). The vegetative cells of the excluded taxa were very rarely recorded, and were excluded from the count because of their weakly silicified frustules easily dissolve and therefore could

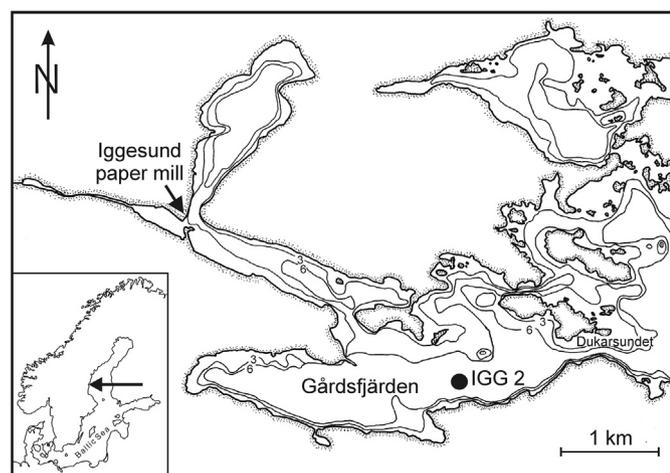


Fig. 1. Map of the Gårdsfjärden area on the Swedish coast of the central Bothnian Sea showing the selected coring site IGG 2 (61°37.25'N; 17°09.39'E). The bathymetric isolines of 3 and 6 m and the discharge point from the Iggesund paper mill are indicated. Water exchange between Gårdsfjärden and the open Bothnian Sea is limited by the narrow Dukarsundet.

give erroneous distribution records in sediments. *Chaetoceros* spp. resting spores were frequently recorded, but since there is a lack of knowledge about the formation of resting spores and they could not be differentiated to species level they are excluded from the basic sum and presented separately.

Diatom species identification and criteria to assign marine affinity and life-form followed Krammer and Lange-Bertalot (1986–1991), Snoeijs (1993), Snoeijs and Balashova (1998), Snoeijs and Potapova (1995), Snoeijs and Kasperoviciene (1996), Snoeijs and Vilbaste (1994) and Witkowski et al. (2000), together with papers listed in Andrén et al. (2007). Taxonomic intercalibration for the training set was accomplished through workshops, cross-counting exercises, and the adoption of species aggregates (Andrén et al., 2007). *Cyclotella* sensu lato taxa has been transferred to *Discostella* and *Lindavia* following Houk and Klee (2004) and Nakov et al. (2015).

2.4. Numerical methods

The training set used for the transfer function development comprises diatom samples from coastal Baltic Sea surface sediment samples and associated environmental variables collected within the projects MOLTEN (Clarke et al., 2006; Weckström and Juggins, 2006) and DEFINE (Andrén et al., 2007) between 2001 and 2005 (<http://craticula.ncl.ac.uk/Molten>). The water chemistry data used for the training set were provided by local, regional and national monitoring programs with long time series of high quality environmental data. The mean for the upper 10 m of the water column over the five years prior to sediment sampling was used. Stations were selected to cover a long nutrient gradient. Surface sediment samples were sampled using a gravity corer or grab and the top 1 cm, representing the most recent sediment deposition, was collected. A thorough description of the methodology can be found in Andrén et al. (2007).

A weighted-average transfer function model, using square root transformed assemblage data and inverse deshrinking, was used to reconstruct TN concentrations. Reconstruction significance was assessed with the randomTF method from Telford and Birks (2011) using the palaeoSig package version 1.1–1 for R (Telford, 2012).

The dissimilarity between each fossil assemblage and its closest modern analogue was estimated using the squared chord distance (Overpeck et al., 1985). A good analogue was defined as a fossil sample having a squared chord distance less than the 5th percentile of the distribution of all distances among modern samples in the training set (Juggins and Birks, 2012). A squared chord distance less than the 10th percentile was considered fairly good.

Diatom stratigraphical data were divided into diatom assemblage zones using cluster analysis (CONISS), on taxa occurring with at least 2% in one level, available in the software Tilia v. 2.0.38. Detrended correspondence analysis (DCA), a unimodal indirect ordination method (Hill and Gauch, 1980), was used to summarise compositional changes in the whole diatom assemblage over time and performed using the Vegan package in R (Oksanen et al., 2007). Species richness of the fossil diatom assemblages was estimated using rarefaction analysis (Birks and Line, 1992).

3. Results

3.1. Training set and transfer function

The training set consists of 341 sites from the Baltic Sea coastal areas together with sites from the Swedish west coast, Danish waters, Oslo Fjord and the Netherlands (Andrén et al., 2007). The sites span a wide environmental range with water depth between 0.5 and 101 m, salinities between <0.1 and 31.7, total phosphorus

between 5.7 and 476 $\mu\text{g L}^{-1}$ and total nitrogen between 150 and 3890 $\mu\text{g L}^{-1}$. To reduce the environmental heterogeneity in the data set, only sheltered sites are used in the analyses below: 229 sites remain after excluding the exposed sites.

The significance of the environmental variables on the biotic data was tested using a constrained correspondence analysis (CCA), which confirms that all four variables used (salinity, water depth, TP, TN) have a significant independent effect with $p < 0.01$. Variance partitioning (Borcard et al., 1992) shows that the environmental variables explain 14.4% of the variance, of which total nitrogen has a small but significant and independent proportion.

The weighted average, with inverse deshrinking, transfer function model has a leave-one-out RMSEP of 0.38 log TN $\mu\text{g L}^{-1}$, about 10% of the log TN range in the dataset (Fig. 2).

3.2. Lithology, age model and geochemistry

Sediment is accumulating on approximately 56% of the bottom of Gårdsfjärden. About 28% of the sediment was gas charged. The bathymetry surrounding the core site is relatively smooth and flat, becoming steeper close to the shore (Fig. 1). Coring, at 12 m water depth in the centre of the embayment, recovered two parallel 55 cm-long sediment cores labelled IGG 2 ($61^{\circ}37.25'N$; $17^{\circ}09.39'E$). The lithology of IGG 2 is described in Table 1. Laminations in the top 35.5 cm of the sediment core were counted. Their thickness varied between 10 and 23 mm in the uppermost part and between 2 and 3 mm in the lowermost. The rapid increase in ^{137}Cs activity between 18 and 17 cm is interpreted as representing year 1986 (Fig. 3). The ^{137}Cs dating indicated that the laminations were annual and could be used to construct an age model. Assuming that the sediment accumulation rate in the lowermost laminated part, where laminae were 3 mm thick, was similar to that in the non-laminated section at the bottom of the core, the age model could be extended back to approximately 1870 at 55 cm. Dredging in the threshold area in 1991, 1992 and 1994 resulted in light clayey layers in the sediment core, which could be used as marker horizons to validate the age model.

Total organic carbon (TOC) varies between 2.5 and 3% in the lowermost 15 cm of the homogeneous sediments and starts to increase around 40 cm to a maximum value of 9.2% at 20 cm (Fig. 3). Around 15 cm there is an abrupt decrease in TOC which stabilizes at

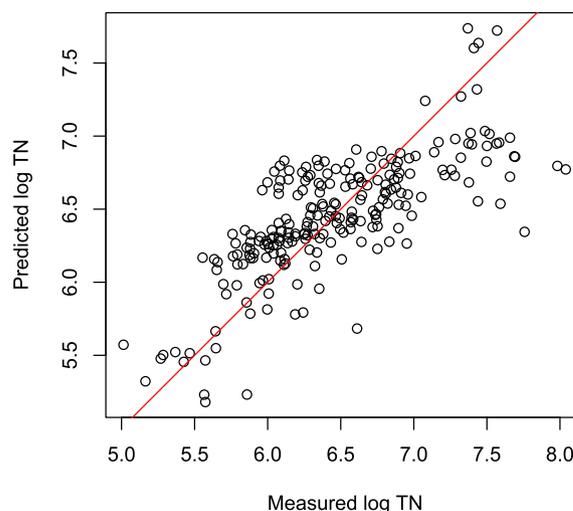


Fig. 2. Predicted log TN calculated with a weighted average transfer function against measured log TN for the DEFINE diatom training set (Andrén et al., 2007; <http://craticula.ncl.ac.uk/Molten>).

Table 1
Lithological description of sediment core IGG 2.

Depth (cm)	Lithology
0–14.1	Laminated olive-green/dark-grey clay gyttja
14.1–19.1	Diffuse laminated clay-gyttja
19.1–23.9	Dark laminated clay-gyttja
23.9–26.8	Black homogeneous or possibly laminated clay-gyttja
26.8–30.7	Laminated clay-gyttja
30.7–35.5	Diffuse laminated clay-gyttja
35.5–39	Homogeneous clay-gyttja
39–54.5	Light homogeneous gyttja-clay

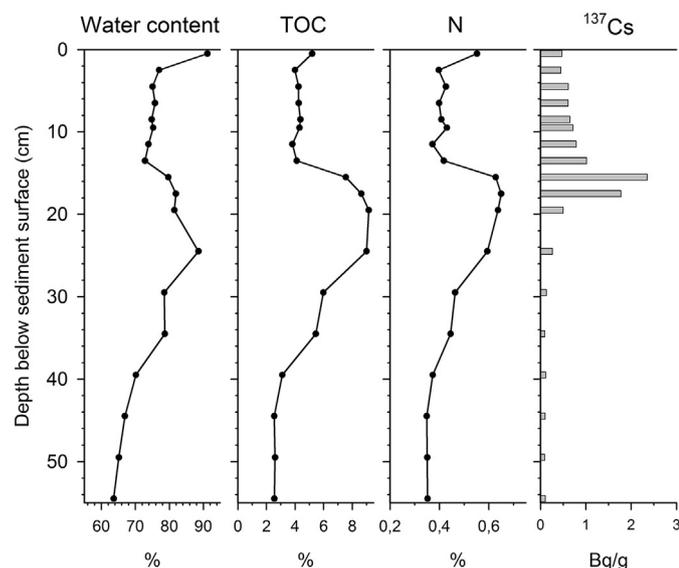


Fig. 3. Water content, total organic content (TOC), nitrogen (N) and measurements of ^{137}Cs in the sediment core from Gårdsfjärden. The peak between 18 and 17 cm indicates the radioactive fallout from the Chernobyl power plant in 1986.

4–5% in the uppermost part of the core corresponding with the extensive dredging to deepen the channel inlet via Dukarsundet into Gårdsfjärden. This resulted in a substantially increased bulk sediment deposition rate in the estuary. Nitrogen content varies between 0.35 and 0.65% with maximum values between c. 25–15 cm (Fig. 3).

3.3. Diatoms

Diatom assemblages were enumerated from twelve levels and a total of 206 taxa were identified. The diatom assemblages are a mixture of brackish, brackish-freshwater and freshwater taxa with both planktic and benthic life-forms throughout the core. A complete record of the diatom core data together with diatom species authorities can be found on <http://craticula.ncl.ac.uk/Molten/jsp/>. Diatoms with an abundance of more than 2% in at least one stratigraphic level are shown in Fig. 4. There are some clear trends in the diatom stratigraphy identifiable as shifts in life-form (plankton living in open water, benthos living attached to or associated with different substrata) and salinity requirements. The stratigraphy was divided into 3 local diatom assemblage zones (DAZ 1–3) described as follows:

DAZ 1 (54.5–32 cm) is dominated by brackish water taxa e.g., *Rhoicosphenia curvata*, *Martyana atomus*, *M. schultzi* and *Fragilaria elliptica* agg. as well as some brackish-freshwater taxa e.g., *Mastogloia smithii*, *Amphora pediculus* and *Epithemia sorex*. All these taxa are typical of Baltic Sea coastal waters (Snoeijs, 1993). Freshwater

taxa e.g., *Fragilaria exigua*, *Staurosira construens* var. *binodis* and *Aulacoseira subarctica* are also abundant in the zone. The zone has a high proportion, more than 80%, of benthic taxa, of which around 20% are epiphytic (growing on plants or other algae). The zone has the highest species richness with 84 taxa per sample (Fig. 6).

In DAZ 2 (32–12.5 cm), the proportion of planktic taxa increases to a maximum value of 54% and it is dominated by 60–70% of freshwater taxa e.g., *Aulacoseira subarctica*, *Aulacoseira ambigua*, *A. granulata*, *Lindavia radiosa*, *Discostella stelligera* and *Tabellaria flocculosa*. There is a low abundance of benthic taxa in the zone, especially epiphytic taxa, which occur at less than 10% in some levels. Species richness is lower than in the previous zone with 72–76 taxa per sample.

In DAZ 3 (12.5–2 cm), the uppermost zone, the abundance of brackish water taxa e.g., *Thalassiosira levanderi*, *Pauliella taeniata*, *Diatoma moniliformis* and brackish-freshwater taxa e.g., *Thalassiosira baltica*, *Cyclotella meneghiniana* increases. The abundance of freshwater taxa in general decreases, but some taxa reach about 10% each e.g., *F. exigua*, *A. subarctica* and *A. ambigua*.

3.4. Nutrient reconstruction

Based on the significant test (Fig. 5), the TN reconstruction is statistically significant; reconstructions of salinity, depth and TP are not, implying that these variables did not drive the changes in the diatom stratigraphy. Diatom-inferred total nitrogen fluctuates between 306 and 264 $\mu\text{g L}^{-1}$ in the lowermost part of the core (Fig. 6). TN starts to increase between 1935 and 1950 and reaches 350–365 $\mu\text{g L}^{-1}$ between 1950 and 1980. The maximum value of ca 400 $\mu\text{g L}^{-1}$ is reached in 1990. After this, there is a decreasing trend, with concentrations falling 357 $\mu\text{g L}^{-1}$ in the uppermost sample representing 2001.

Fossil assemblages in DAZ 1 had good analogues (Fig. 6). Assemblages in DAZ 2 had, with one exception, fairly good analogues, as did assemblages in DAZ 3.

4. Interpretation and discussion

4.1. Environmental history of Gårdsfjärden

The industrial history of Gårdsfjärden estuary's catchment began in 1685 CE, almost 200 years before the start of the 135 year long sediment record from the estuary. The addition of a saw mill to the ironworks at the end of the 19th century seems to have had little effect on the environmental status of the estuary. At this time, the estuary had quite a high proportion of epiphytic diatoms indicating that light conditions at sea floor were sufficient for macrophyte growth. The accumulating sediment had low organic carbon content and the absence of laminae suggests that bottom-water oxygen was not depleted and benthic organisms were abundant. The diatom assemblage consisted of mainly brackish/brackish-freshwater species until 1920 (Fig. 6), indicating that the water exchange through the narrow Dukarsundet (Fig. 1) was high and the inflow of freshwater and sediment from River Delångersån were moderate.

The clear deterioration of the environmental status in Gårdsfjärden estuary is closely connected to the history of the Iggesund pulp and paper mill. Before 1920 the estuary could be considered to have good ecological status based on water transparency, primary production and oxygenation of the sea floor. In 1916–1917, the pulp mill initiated production of sulphate and sulphite pulp (Nilsson et al., 2003) and between 1920 and 1935 there is palaeoecological evidence of early environmental disturbance in Gårdsfjärden. The perturbation is evident as a change in diatom species composition with an increase in freshwater planktic taxa at

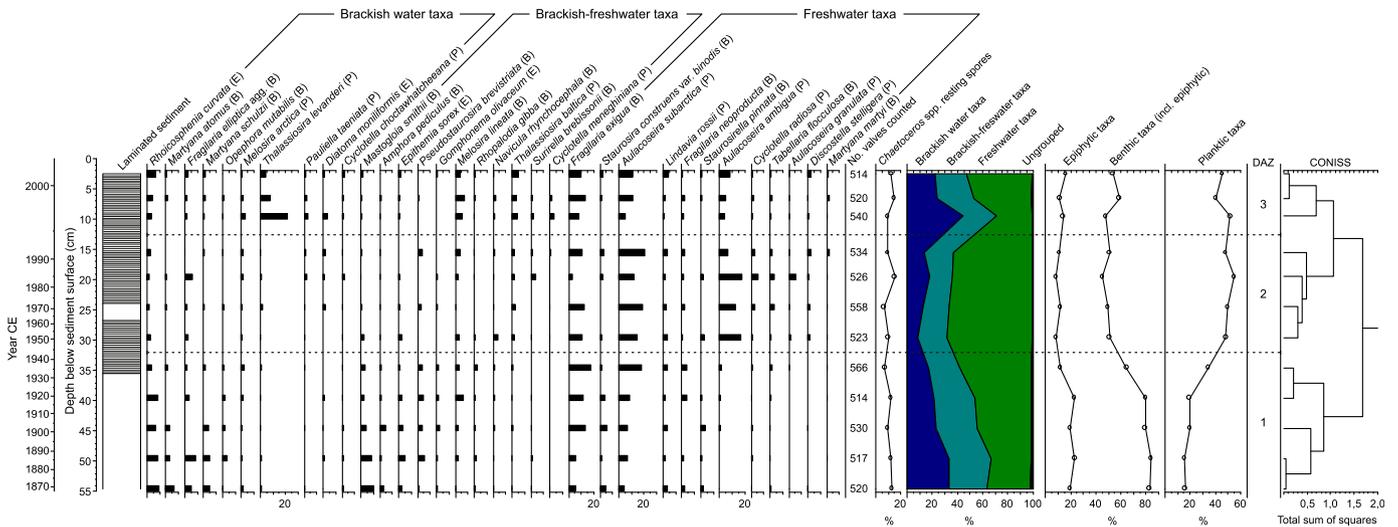


Fig. 4. Diatom stratigraphy of core IGG 2 plotted on linear depth scale from Gårdsfjärden, Bothnian Sea showing relative abundance of species occurring at 2% in at least one level. Taxa are grouped into salinity requirements and life-forms (P-planktic, B-benthic, E-epiphytic) and sorted in order of first appearance. *Chaetoceros* spp. resting spores are not included in the basic sum but presented as percent of the total diatom count. The age model shown to the left is constructed from laminae counts and ^{137}Cs measurements. The diatom stratigraphy has been divided into three local diatom assemblage zones (DAZ 1–3) using cluster analysis, CONISS.

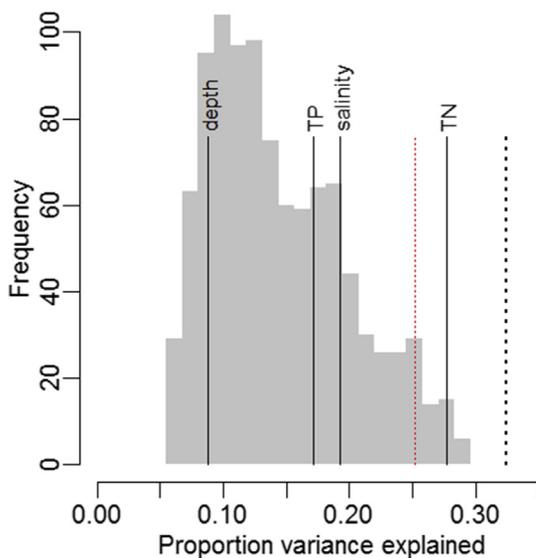


Fig. 5. Histogram of the proportion of variance in the IGG 2 diatom record explained by 999 reconstructions from transfer functions trained with random data. Red dotted line marks the 95th percentile of this null distribution. Solid black lines marks the proportion of variance explained by reconstructions of named environmental variables. The dotted line marks the proportion of variance explained by the first axis of a Principal component analysis (PCA) of the fossil data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the expense of brackish/brackish-freshwater epiphytes. The responding epiphytes are mainly *Rhoicosphenia curvata*, *Epithemia sorex* and *Gomphonema olivaceum* (Fig. 4) probably due to deteriorating water transparency causing loss of macrophytes. Similar changes in diatom life-forms have been observed in Mariager Fjord, Denmark (Ellegaard et al., 2006). An increase in the abundance of planktic diatom taxa has been ascribed to coastal marine eutrophication in the Chesapeake Bay (Cooper and Brush, 1991) and in the southern Baltic Sea (Andrén et al., 1999). Weckström et al. (2007) show that there is a link between elevated abundance of planktic taxa and high nitrogen concentrations. In Gårdsfjärden,

concurrent with the shift to a more plankton-dominated diatom assemblage, there is a decrease in diatom species richness (Fig. 6). Comparable declines in diatom species richness have been recorded in Finnish and Danish coastal embayments and estuaries and are attributed to increased discharge of nutrients following an increased human impact (Clarke et al., 2006; Ellegaard et al., 2006; Weckström, 2006). In coastal sites from the Gulf of Finland, structural changes in diatom assemblages occur after exceeding a total dissolved nitrogen (TDN) threshold of 400–600 $\mu\text{g}\text{L}^{-1}$ (Weckström et al., 2007). In Gårdsfjärden, even under the most deteriorated environmental conditions, diatom species richness remains quite high, although the shifts in life-form and species composition are considerable. The DCA axis 1 sample scores (Fig. 6) which reflect the major compositional changes in the diatom assemblage over time show a smooth succession without sharp transitions, except for the sample corresponding to 1996, interpreted as the result of dredging the estuary inlet.

The brackish water planktic taxon *Cyclotella choctawhatcheana* has in many studies been found to indicate eutrophic conditions (Cooper, 1995; Andrén et al., 2000; Weckström, 2006) and in Gårdsfjärden it is present but in very low abundance. Diatom taxa in Gårdsfjärden responding to increased nutrient availability are in particular the freshwater taxa *Aulacoseira ambigua*, *A. subarctica* and *A. granulata* (Fig. 4, DAZ 2). *Aulacoseira ambigua* and *A. granulata* are considered eutrophic freshwater taxa and have also been found in the Oder Estuary, southern Baltic Sea, during periods of increased eutrophic conditions in the last century (Andrén, 1999). *Aulacoseira* species have in previous studies been used as indirect palaeoenvironmental indicators of the persistence of strong seasonal wind stress and resultant turbulent water column mixing and nutrient upwelling conditions (Wang et al., 2008). Their frustules are heavily silicified, forming colonies that require turbulence-induced resuspension to remain in the photic zone (Rühland et al., 2008; Lotter et al., 2010).

In early 1930's, the homogeneous gyttja-clay was replaced by a laminated clay-gyttja (Fig. 6). The laminae suggest the absence/low abundance of benthic organisms caused by bottom water hypoxia. The hypoxia coincides with an increased accumulation of organic carbon and nitrogen in the sediment. The elevated C/N-ratio from 1935 (Fig. 6) reflects a large input of terrestrial organic carbon

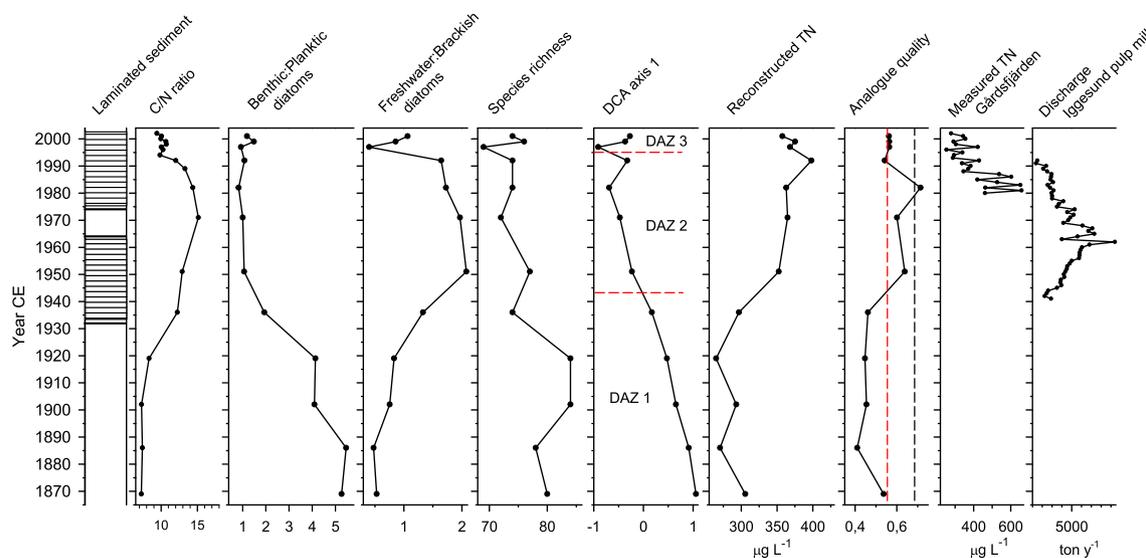


Fig. 6. Occurrence of laminated sediment, C/N ratio, diatom life-form ratio (benthic to planktic taxa), freshwater to brackish water diatom ratio, diatom species richness, diatom DCA axis 1 sample scores (including the DAZ 1–3), reconstructed diatom-inferred TN, analogue quality, measured TN in the Gårdsfjärden estuary (core IGG 2) plotted against date and discharge from the Iggesund pulp and paper mill (Nilsson et al., 2003). The red dashed line in the analogue quality graph shows the limit between good (less than the 5th percentile) and fairly good analogues and the black dashed line the limit between fairly good (less than the 10th percentile) and lack of analogues. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

derived from the increased discharge from the Iggesund pulp mill. The discharge from the mill has been measured since 1940 (Fig. 6; Nilsson et al., 2003) and there is a strong correlation between the deterioration of the environment in the estuary and the magnitude of the industrial discharge. The maximum C/N-ratio, around 1970, occurred after the discharge from the pulp mill peaked during the 1960s resulting in the deposition of black, possibly laminated, sediments. Since 1970, the C/N-ratio in the sediment core decreased slowly, likely reflecting the introduction of sedimentation basins for fibre recovery at the Iggesund pulp and paper mill in 1968 (Nilsson et al., 2003). Both TOC and N-values in the sediment were still very high and even increased until the early 1990s. This indicates that even if untreated effluents discharged from the pulp mill were reduced, nutrient availability within the estuary was still elevated as seen by measured TN (Fig. 6).

During years with maximum discharge from the pulp mill, a vast amount of wood fibres was lost into the estuary, requiring dredging every year to facilitate navigation (Nilsson et al., 2003). Gårdsfjärden is located in an area with about 7.5 mm year⁻¹ isostatic uplift (Ekman, 1996), reducing the threshold depth by nearly 1 m over the past 135 years. After introducing sedimentation basins for the waste water in 1968, dredging diminished until the early 1990s. Besides a substantial increase in bulk sediment accumulation rate in the bay the extensive dredging of the threshold area in 1994 seems to have had a direct impact on the diatom composition in the estuary. This is visible as a dip in the DCA axis 1 curve, an increase in brackish water taxa, especially the planktic *Thalassiosira levanderi*, minimum species richness (Figs. 4 and 6) and can be interpreted as an improved water exchange with the open Bothnian Sea.

4.2. Nutrient reconstruction

In a study of nitrogen and phosphorus turnover in the Gårdsfjärden estuary (Nilsson and Jansson, 2002) it is evident that the estuary was a source of dissolved phosphorus which accumulated in the sediment during the period with high nutrient discharge and is now being released from the hypoxic sediments. The estuary was at the same time a sink for dissolved nitrogen,

especially during spring, probably due to denitrification (Nilsson and Jansson, 2002). Which nutrient limited primary production prior to the increased industrial discharge is uncertain. In the open Bothnian Sea limiting nutrients have shifted from phosphorus 100 years ago (Savchuk et al., 2008) to nitrogen the last 20 years due to eutrophication of the Baltic Proper (Rolff and Elfving, 2015). The diatom-inference model shows that reconstruction of TN was the only significant environmental variable (Fig. 5). Our sediment archive allows us to reconstruct total nitrogen conditions in Gårdsfjärden since 1870. The reconstructed total nitrogen concentration of Gårdsfjärden fluctuates between about 260 and 300 µg L⁻¹ before 1920. Model simulations of pre-industrial trophic conditions show past concentration of TN in the Bothnian Sea of 16.5/17.0 µM L⁻¹ corresponding to about 231–238 µg L⁻¹ (Savchuk et al., 2008). These values are slightly lower compared to the result from the Gårdsfjärden estuary, but could be expected considering we are comparing the open sea with the coastal zone.

Data from a monitoring programme, dating back to 1980, provide an opportunity to validate the diatom inferred total nitrogen (DI-TN) reconstruction in Gårdsfjärden (Fig. 6). The reconstructed TN for 2001, the uppermost sample, is 357 µg L⁻¹, comparable with the monitored annual mean TN, 342 µg L⁻¹ (calculated as a mean of 5 years between 1997 and 2001). Our model performs well when the values are below 400 µg L⁻¹ and the diatom analogue quality is fairly good. Comparing the DI-TN values with the measured TN, it is clear that the model has difficulties reconstructing the high measured values in the early 1980s, which vary from 660 to 460 µg L⁻¹ (Fig. 6). In this part of the core, there is a decrease in the analogue quality, indicating that the modelled results may be less reliable. A diatom study from the Gulf of Finland similarly underestimated nitrogen concentrations during the most eutrophic phase due to a lack of modern analogues in the training set (Weckström et al., 2004; Weckström, 2006). The relationship between structural changes in the diatom communities and elevated nitrogen concentrations seems to end once total dissolved nitrogen concentrations are higher than 800 µg L⁻¹ and the diatoms seem to be unable to respond to very high nutrient concentrations (Weckström et al., 2007). The failure of the transfer function to

reconstruct the high nitrogen concentrations in the most polluted period does not affect our aim to quantify the low background nitrogen conditions. The four lowermost samples show palaeoecological evidence of a good ecological status and have good analogues in the modern diatom training set.

4.3. Reference conditions and recovery

It is not likely that severe impact on the environmental status occurred in the Baltic Sea coastal zone before modern agriculture practice started around 1950, as evident in the time series of annual average total nutrient load (Gustafsson et al., 2012). Likewise, a reconstruction of nutrient concentrations in Baltic Sea surface water shows that only slight changes occurred before 1950–1960, which suggests that early measured data can be used to reflect reference conditions (Schernewski and Neumann, 2005). In the Gårdsfjärden estuary, however, considerable environmental changes; loss of epiphytes, decreased species richness and deteriorated bottom water conditions occurred before 1950, which would make this date a poor choice for reference conditions. The Gårdsfjärden core is too short to extend beyond the first human impact on the estuary and the conditions we reconstruct may not be considered pristine, but impact before 1870 was probably minor. The reconstructed nutrient conditions prior to 1920 could represent reference nutrient conditions and good ecological status as defined by the European Union Water Framework Directive, EU WFD and HELCOM Baltic Sea Action Plan, BSAP (European Union, 2000; HELCOM, 2007; Andersen et al., 2011). There is a need for a long-term perspective to determine natural variability when considering sustainable environmental governance (Willis and Birks, 2006) and to decide a well-founded point of departure from reference conditions (Bennion et al., 2011). This long-term perspective is at present even more important as models show that in a future warmer climate Baltic Sea water quality will be deteriorated due to strengthened internal feedbacks (Meier et al., 2012). Extensive human pressure on landscapes has occurred for thousands of years, with examples such as airborne pollution, which has continued for four millennia, interruption of natural acidification of lakes when clear cutting for agriculture started about 2000 years ago, and subsequent eutrophication of lakes as agriculture was initiated and expanded (Renberg et al., 2009). In the Baltic Sea there is evidence for changes in the diatom record as early as the turn of the twentieth century along the German coastal zone (Andrén, 1999; Andrén et al., 1999) but not until 1950 in the open Baltic proper (Andrén et al., 2000). Before 1920–1930 about a quarter of the diatoms in Gårdsfjärden were epiphytic, indicating that a large part of the sea floor in the estuary was covered with macrophytes. Diffuse laminations started to form already in the early 1930's with fully developed laminations in late 1940's, indicating deteriorated oxygen conditions and loss of bottom fauna. The palaeoecological evidence from Gårdsfjärden indicates that the worst environmental conditions occurred between the 1960s and 1980s, when the water discharge from the Iggesund pulp mill peaked, but before water quality was monitored. The deteriorated water transparency following increased effluent discharge from the mill efficiently changed available benthic habitats resulting in predominantly planktic diatom assemblages, decreased biodiversity and altered ecosystem functioning (Weckström et al., 2007). The palaeoecological evidence from Gårdsfjärden indicates that the worst environmental conditions occurred between the 1960s and 1980s, when the water discharge from the Iggesund pulp mill peaked, but before water quality was monitored.

Diatoms are evidently a sensitive and early warning indicator for changes in nutrient input. In the coastal zone the most obvious change occurs when the macrophytes are affected by deteriorated

water transparency and suitable habitats for epiphytes disappears. A reduction in nutrient discharge does not necessarily result in immediate recovery and re-establishment of macrophytes in the estuary and a switch back to background diatom assemblages, as observable in Gårdsfjärden estuary. The diatom assemblages indicate that planktic life forms were still dominating in the estuary at the time of core collection in 2002 and the epiphytes had not recovered. Recent studies in the area indicate however an improvement of environmental status as indicated by oxygenated bottoms, colonisation of bottom fauna and retention of phosphorus in sediment (Karlsson and Malmaeus, 2012). Apart from nutrient discharge, water exchange between the open Bothnian Sea and the coastal zone seems to be important for the environmental status, as evidenced by the short-lived impact on the diatoms due to dredging of the Gårdsfjärden threshold area. There is ongoing land uplift in the Bothnian Sea area of 5–8 mm/yr (Ekman, 1996), which makes the coastal zone very dynamic with previously settled and potentially toxic, nutrient rich sediment raised over the wave-base and recycled in the ecosystem (Karlsson et al., 2014). However, the increase in nutrients in the open Bothnian Sea (Rolff and Elfving, 2015) indicates that the trophic situation in the coastal zone is ameliorated compared to the open sea (Andersen et al., 2011). The open Bothnian Sea has been classified to have a poor trophic status based mainly on phytoplankton indicators, while in the Bothnian Sea coastal zone 9 out of 21 investigated sites were considered unaffected by eutrophication (Andersen et al., 2011).

5. Conclusion

The reconstructed total nitrogen values of 260–300 $\mu\text{g L}^{-1}$ before 1920 could be considered reference nutrient conditions for the Gårdsfjärden estuary, even though the industrial history of the paper mill at Iggesund dates back to 1685 CE. Until about the early 1930s, the estuary was mesotrophic, with well oxygenated bottom-water, macrophyte stands, and high diatom species richness dominated by benthic taxa.

Provided the requirements of quantitative reconstructions are met (Juggins, 2013), in addition to continuous sediment archives, good silica preservation, credible chronology and good diatom species analogues, diatom-inferred transfer functions can readily be used to establish reference nutrient conditions for the EU Water Framework Directive and HELCOM Baltic Sea Action Plan in other Baltic Sea coastal waters.

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