

## Long-term trends in eutrophication and nutrients in the coastal zone

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### *Acknowledgments*

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### Abstract

We used high-resolution paleoecological records of environmental change to study the rate and magnitude of eutrophication over the last century in two contrasting coastal ecosystems. A multiproxy approach using geochemical and biological indicators and diatom-based transfer functions provides a long-term perspective on changes in nutrient concentrations and the corresponding biological and sedimentary responses. In Roskilde Fjord, Denmark, total nitrogen (TN) increased 85% during the last century, with the most rapid increase occurring after the 1950s, corresponding to the postwar increase in N fertilizer use. In Laajalahti Bay, an urban embayment near Helsinki, Finland, total dissolved nitrogen (TDN) increased with growing wastewater inputs and decreased with the remedial actions taken to reduce these discharges. These changes are small relative to the order of magnitude increases in nutrient loading that have occurred in northwestern Europe, where the dissolved inorganic nitrogen (DIN) load has increased more than threefold in certain areas.

Coastal waters and estuaries are naturally fertile ecosystems that receive nutrient inputs from terrestrial, riverine, groundwater, marine, and atmospheric sources. The increase in anthropogenic effects on the loading and transport rates of the limiting plant nutrients nitrogen (N), phosphorus (P), and silica (Si) during the last century (Conley 2000; de Jonge et al. 2002) has led susceptible areas to display undesirable eutrophication effects. The traditional view of the coastal eutrophication process (Nixon 1995) is that increased inputs of nitrogen stimulate aquatic primary production, leading to an increase in the amount of autochthonously produced organic carbon. The study of eutrophication is inherently linked with the temporal and spatial distribution of the biologically available inorganic forms of N, P, and Si in the aquatic environment. Nutrients have always been transported from terrestrial through freshwater to oceanic ecosystems; what has altered over time (and with a quickening pace over the last 100–150 yr) is the quantity of nutrients being mobilized and moved (de Jong and de Oude 1988).

The recently implemented water framework directive (WFD) (Anonymous 2000) requires European Union member states to manage water resources to obtain “good ecological status,” where biological and chemical elements deviate only slightly from those expected under undisturbed conditions, during a realization period between 2015 and 2027. This requires the determination of reference, or baseline, conditions against which the effects of anthropogenic activities can be measured in terms of concentrations and ecosystem changes, and efforts to achieve good ecological status can be assessed.

The setting of appropriate restoration targets requires more than an understanding of nutrient use, partitioning, loss, and recycling through the aquatic continuum. There is also a need for information on baseline concentrations of nutrients in coastal waters, or at least concentrations when anthropogenic perturbations had minimal effects on the structure and function of coastal ecosystems. Without knowledge of these baselines and natural variation, assessing the magnitude of eutrophication in susceptible areas is difficult and could lead to inappropriate restoration targets and management practices. The science behind management is critical; it needs to demonstrate environmental damage while providing realistic errors and uncertainties in both measurement and prediction (Gray 1999).

A number of techniques can be used to estimate baseline conditions (Anderson 1995). Mathematical models integrate data from a range of sources and have the potential to sep-

arate natural and anthropogenic effects. For example, the Phison River model (Billen and Garnier 1997), a coupled model of a riverine and associated coastal ecosystem, allowed the testing of several nutrient reduction scenarios, including major changes in watershed use. Their results indicated that under pristine conditions phosphorus limitation may have been widespread in coastal ecosystems and that it is only since the onset of industrialization that silica and/or nitrogen limitation has occurred. Numeric models are effective tools for assessing various components of ecosystems, but it is not always possible to assess directly their results, particularly when they refer to hindcast scenarios.

Data mining from early studies, monitoring programs, and environmental databanks is also a possibility. However, the use of archived data, especially from before the 1970s, can be compromised by the heterogeneous nature and uncertain quality of older biological and chemical records; certainly archives should not be used blindly for decision making processes unless strict protocols for data validation and quality control and assurance are applied. Poorly designed monitoring programs, unable to answer the questions they were designed to answer because inadequate sampling strategies led to poor detection capability (Gray 1999), should also be avoided when choosing sources for data mining.

Paleoecology is another option. Numerous methodologies can be used to provide a chronology of environmental change, including sediment profiles of major elements (e.g., N, P, and biogenic silica (BSi)), as in Cornwell et al. [1996] or interpretations based on biological indicators. A wide variety of biological proxies exist, e.g., diatoms, dinoflagellate cysts, foraminifer, ostracods, and seeds of higher plants and biogeochemical markers such as plant pigments and lipids.

Diatoms are powerful paleoecological indicators, since their taxonomically distinct frustules allow identification to species level, and they are usually present in diverse, numerically abundant assemblages (Charles et al. 1994) that preserve well under a variety of sedimentary environments (Anderson and Vos 1992). There is a long history of the use of diatoms as indicators of environmental change in marine systems (*see* reviews in Stoermer and Smol 1999), and diatoms have been used as indicators of eutrophication in coastal waters (e.g., Cooper 1995; Andr n 1999). However, the current lack of precise autecological knowledge for many coastal taxa makes interpretation of biostratigraphic records difficult. More importantly from a management perspective, while a qualitative analysis of floristic changes can be interpreted in terms of changes in trophic status, it cannot recon-

struct actual nutrient concentrations. For this a more quantitative approach is needed.

Statistically robust methods are available to quantify modern diatom distribution in terms of optima and tolerances with respect to particular hydrochemical gradients. These, in turn, can be used to infer historical changes in water chemistry from fossil assemblages. This so called “transfer function” methodology was pioneered in paleoceanography, where it provided the first quantitative estimates of past sea-surface temperature from foraminiferal sedimentary assemblages (Imbrie and Kipp 1971). Modified, with the statistical basis changed to weighted averaging, diatom-based transfer functions are now common in freshwaters where they have been used to help quantify the effects of acidification (Birks et al. 1990), climate and salinity interactions (Fritz et al. 1991), and eutrophication (Bennion et al. 1996).

This method is not yet common in coastal marine systems. Juggins (1992) developed a diatom-based salinity transfer function for the tidal stretch of the river Thames; Jiang et al. (2002) developed a sea-surface temperature transfer function from diatom assemblages around the shelf seas of Iceland; and Ryves et al. (2004) developed a salinity transfer function from diatom assemblages in fjords and brackish lakes in Denmark. The first weighted averaging transfer functions for nutrient reconstructions in coastal systems have been developed using modern surface sediment training, or calibration, data sets from the Baltic Sea area: a total nitrogen (TN) transfer function developed in Denmark (Clarke et al. 2003) and a total dissolved nitrogen (TDN) transfer function developed in Finland (Weckström et al. 2004). Each calibration site has diatom assemblage data and associated environmental variables related to water quality. All sites are harmonized regarding diatom taxonomy, and quality assessment of the environmental data ensures that only sites with robust estimates of annual mean chemistry are used in transfer function development.

Here we compare two multiproxy paleoecological investigations from contrasting coastal ecosystems affected by eutrophication: Roskilde Fjord in Denmark, a mesohaline estuary with a predominantly agricultural catchment, and Laajalahti Bay in Finland, an oligohaline embayment that was, for a period, subject to municipal sewage discharges. Combining quantitative reconstructions of nutrient concentrations from diatom-based transfer functions with a multiproxy approach to long-core analysis allows us to provide environmental reconstructions based on both empirical modeling and the ecological information contained in geochemical and biological analyses. Sediment, organic carbon, and biogenic silica accumulation rates are calculated, and relative abundances of diatom life forms and a measure of diatom species richness are provided. Reconstructions of nitrogen concentrations that are independent of the sediment accumulation rate are determined. These proxies are used to reconstruct ecosystem changes in two coastal areas of the Baltic Sea over the past 100–150 yr. Ecosystem changes observed in Roskilde Fjord and Laajalahti Bay that are relevant to modern management concerns are discussed.

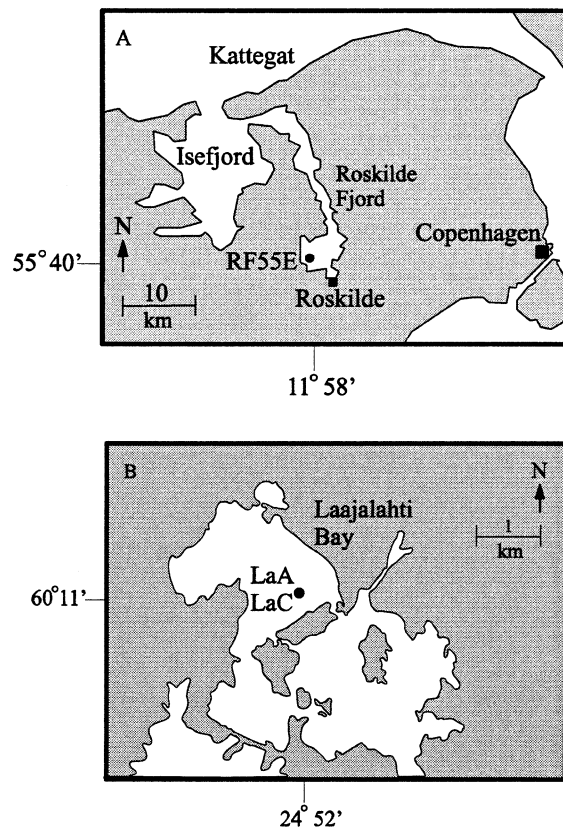


Fig. 1. (A) Map of Roskilde Fjord and surrounding area showing core collection site. (B) Map of Laajalahti Bay showing surrounding area and core collection site.

## Methods

**Site descriptions**—Roskilde Fjord is a 30 km long, shallow (mean depth 3 m, surface area 123 km<sup>2</sup>) estuary in north Zealand, Denmark (Fig. 1A). A sill restricts water transport between the northern part of the estuary, which connects to the Kattegat through the Isefjord, and the southern part, which is composed of three basins. The average water residence time is ca. 70 d. This results in strong north–south gradients of salinity (19–11) and nutrients (TN, from 37 to >100 μmol L<sup>-1</sup>; total phosphorus [TP], from 2 to >13 μmol L<sup>-1</sup>). Southern Roskilde Fjord has a catchment area of 450 km<sup>2</sup> and a surface area of 52 km<sup>2</sup>. Agricultural practices account for 67% of land use, urban areas 15%, and the remaining land is covered with forests, wetlands, and lakes. Roskilde, the only city in the catchment, lies on the shore of the southernmost basin.

Laajalahti Bay is a small, shallow (surface area 5.3 km<sup>2</sup>, mean depth 2.4 m) urban embayment west of Helsinki, Finland (Fig. 1B). It is connected to the open archipelago by two narrow straits, and water exchange of the embayment is restricted. Freshwater discharge comes from two small brooks; the average salinity of the embayment (4.6), however, is close to that of the outside archipelago. The theoretical residence time of the embayment is ca. 1 month. Much of the agricultural land in the catchment has been converted to urban areas, and the forested area has markedly

decreased since the 1950s. Contemporary land-use types of the catchment are urbanized areas (54%), fields and meadows (12%), and forests, swamps, wetlands, and wasteland (34%). In the 1960s, Laajalahti Bay, one of the most polluted coastal areas of southern Finland, received  $3 \times 10^5$  kg yr<sup>-1</sup> of total nitrogen,  $6 \times 10^4$  kg yr<sup>-1</sup> of total phosphorus, and  $1 \times 10^6$  kg yr<sup>-1</sup> of organic matter (measured as BOD<sub>7</sub>) from a sewage treatment plant built in 1957. Currently the embayment receives only diffuse loading from two brooks discharging  $11.6 \times 10^3$  kg yr<sup>-1</sup> of TN and  $9 \times 10^2$  kg yr<sup>-1</sup> of TP.

*Core collection, processing, and dating*—A 1-m-long sediment core (RF55E) from Roskilde Fjord was collected in 1995 with a Mackereth corer (Mackereth 1969) from a deep (15 m) location in the westernmost southern basin (55°40'N, 11°58'E), where pronounced accumulation of fine-grained sediments occurs and temporary anoxia is common each summer. The core was stored in a cold room at 5°C, sectioned at 1-cm intervals within 24 h of collection, and freeze-dried.

Core RF55E was analyzed for <sup>210</sup>Pb and <sup>137</sup>Cs in a Canberra germanium–lithium well-type gamma detector. Radiometric dates were calculated using the constant rate of supply (CRS) model (Appleby and Oldfield 1978).

A 90-cm long sediment core from Laajalahti Bay was collected in 1998 from the deepest area (3.7 m) of the embayment (60°11'N, 24°52'E) with a Mackereth corer (Mackereth 1969). This core (LaA) was used for dating, diatom analysis, and sediment geochemistry. Core LaC (32 cm) was collected from the same location in 2002 with a HON-Kajak gravity corer (Renberg 1991) for pigment analysis and was kept shaded until processed. The cores were sectioned at 1-cm intervals within 24 h of collection and stored in small plastic bags at 4°C. LaA and LaC were correlated using loss-on-ignition (LOI) analysis.

Core LaA was analyzed for <sup>210</sup>Pb, <sup>226</sup>Ra, and <sup>137</sup>Cs by direct gamma assay using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors, following the technical procedures of Appleby et al. (1986). Radiometric dates were calculated using the CRS and the constant initial concentration (CIC) models (Appleby and Oldfield 1978). A composite model chronology was constructed using the CIC model for the upper zone (0–13 cm) and the mean sedimentation rate from both models for the deeper section (*see* Vaalgamaa 2004 and Weckström et al. 2004 for further details).

*Geochemistry and pigments*—The total carbon (TC) and organic carbon (OC) content of the sediment cores were measured by direct determination on crushed, freeze-dried material. Subsamples were pretreated with 1 mol L<sup>-1</sup> hydrochloric acid (HCl) to remove the inorganic fraction, thus allowing determination of the organic fraction. Core RF55E was measured on a CHNS elemental analyzer (CE instruments EA1110). Core LaA was measured on a Leco analyzer. The concentration of biogenic silica (BSi) in both cores was measured using the Conley and Schelske (2001) modification of DeMaster (1981).

Pigments were extracted from 0.5 g of homogenized

freeze-dried material in 2 ml of 100% acetone, sonicated, and kept in the freezer (−20°) overnight. Extracts were separated from the sediment by centrifugation, filtered through a syringe filter (0.2 μm), and blown until dry under N<sub>2</sub> flow. Samples were redissolved in 100 μl acetone before analysis. Quantitative analyses of all pigments were conducted on a Waters high performance liquid chromatograph equipped with an online photodiode array detector (Waters 996 PDA). The run method was a modification of Wright et al. (1991) as described by Chen et al. (2001). Individual pigments were identified by comparison with retention time and absorption spectra of authentic standards and quantified at 438 nm, a wavelength that detects chlorophylls, their degradation products, and carotenoids.

*Transfer functions and diatom analysis*—The diatom-based transfer functions were developed from two independent training sets (Fig. 2) composed of diatom relative abundance data and associated environmental variables (Clarke et al. 2003; Weckström et al. 2004). The Danish training set is composed of 70 sites sampled in 1996 and 1997 at stations of the Danish National Aquatic Monitoring and Assessment Program (Conley et al. 2002) that range from shallow, brackish fjords to the deeper, saline open waters of the Belt Seas. The Finnish training set is composed of 49 sites collected between August 1996 and February 1998 from small, shallow, sheltered embayments in the Gulf of Finland (Weckström et al. 2002).

Surface sediments for both training sets were collected with Renberg-type corers, and samples from the top 1 cm were prepared for diatom analysis using standard procedures (Renberg 1990). A minimum of 500 valves was counted for each sample, and diatom assemblages reported as percentage abundance data. Only taxa at a minimum of 1% abundance at two or more sites were included in statistical analyses (180 and 89 taxa in the Danish and Finnish training sets, respectively).

The environmental data for the Danish training set were provided by the National Environmental Research Institute of Denmark. Secchi depth (m), salinity, oxygen (ml L<sup>-1</sup>), chlorophyll *a* (Chl *a*) (μg L<sup>-1</sup>), TN, NH<sub>4</sub>, NO<sub>n</sub> (combined nitrate and nitrite), TP, PO<sub>4</sub> (μmol L<sup>-1</sup>), and NO<sub>n</sub>:PO<sub>4</sub> were calculated as averages (using monthly measurements from 5 yr of monitoring data integrated through the water column). Water depth (m) was recorded at the coring site. All environmental variables except salinity and water depth were log<sub>10</sub> transformed prior to statistical analysis.

The environmental data for the Finnish sites were collected through six sampling visits during the period August 1996 to February 1998, and sampling occurred at least once in each season. Secchi depth (m), temperature (°C), salinity, conductivity (mS cm<sup>-1</sup>), pH, alkalinity (mmol L<sup>-1</sup>), TDN, NO<sub>n</sub>, NH<sub>4</sub>, TP, total dissolved phosphorus, Chl *a* (μg L<sup>-1</sup>), DSi, K, Ca, Mg, SO<sub>4</sub> (mg L<sup>-1</sup>), Na, Cl, (g L<sup>-1</sup>), Fe, and Mn (μg L<sup>-1</sup>) were averaged over the whole sampling period, while water depth (m) was that recorded at the sediment sampling site. All environmental variables were tested for skewness and, if necessary, log<sub>10</sub> transformed prior to statistical analyses.

For both data sets, detrended correspondence analysis

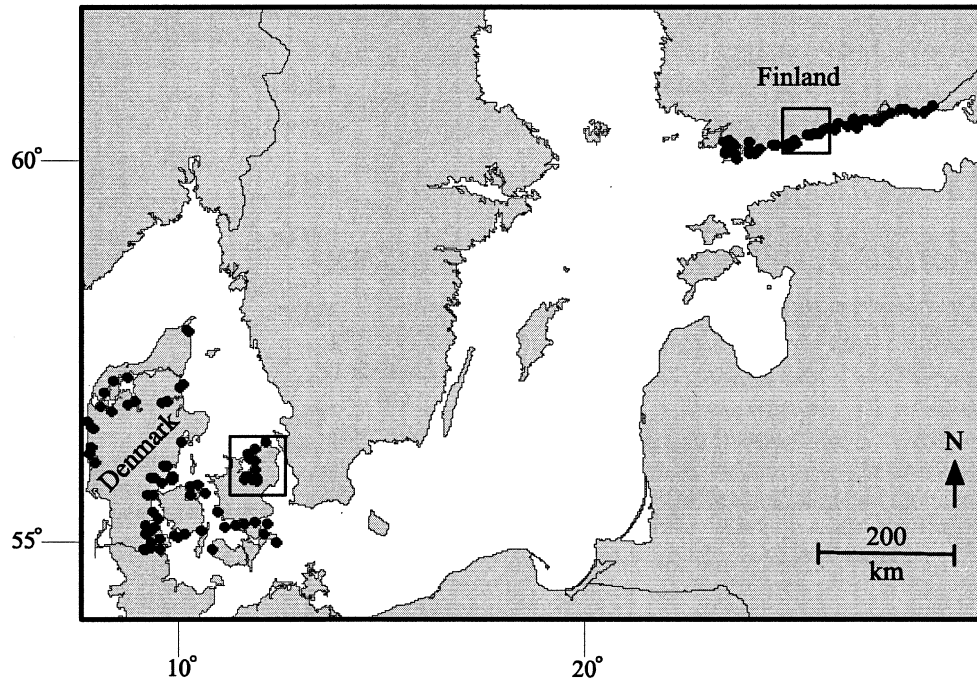


Fig. 2. Map of the Baltic Sea area indicating training set sites for Danish and Finnish transfer functions. The box in each country indicates the location of the core sites illustrated in Fig. 1.

(DCA) using detrending by segments and nonlinear rescaling was used to estimate gradient length in terms of standard deviation (SD) units of biological turnover. Both axes in both training sets had gradient lengths greater than 2 SD indicating unimodal methods of data analysis were appropriate (Birks 1995). Variance partitioning (Økland and Eilertsen 1994) of the diatom and environmental data using canonical correspondence analysis (CCA) with forward selection and associated Monte Carlo permutation tests (999 random permutations:  $p \leq 0.001$ ) showed that salinity, depth, and TN in the Danish training set, and salinity, depth, TDN, and TP in the Finnish training set accounted for significant and independent fractions of variation in the diatom data.

On the basis of the strong and statistically significant relationships between diatom distribution and TN in the Danish training set, and TDN in the Finnish training set, weighted averaging partial least squares (WAPLS) regression and calibration (ter Braak and Juggins 1993) was used to develop transfer functions to infer TN or TDN from the respective diatom relative abundances. Both of the resulting two-component WAPLS models show good agreement between observed and predicted TN or TDN under cross-validation via bootstrapping or jackknifing (Danish training set,  $r_{\text{boot}}^2 = 0.71$ , root mean squared error of prediction [RMSEP] =  $0.15 \log_{10}$  TN, 1,000 bootstrap cycles; Finnish training set,  $r_{\text{jack}}^2 = 0.73$ , RMSEP =  $0.09 \log_{10}$  TDN).

Diatom samples for long-core analysis were prepared in the same way as those for transfer function development, and a minimum of 500 valves enumerated in each sample. Since down-core reconstructed values are potentially more reliable if the fossil diatom assemblages have close modern analogues in the training set, the dissimilarity between each fossil sample and its closest modern analogue was calculated

using the squared chord distance ( $d^2$ ) (Overpeck et al. 1985). A good analogue was defined as a fossil sample having a  $d^2$  less than the value of the 10th percentile of the distribution of all distances among modern samples in the training set. Rarefaction analysis (Birks and Line 1992) was used to estimate diatom species richness in both cores, since this technique accounts for the bias that an unequal sample count size can have on estimates of species richness.

## Results

*Roskilde Fjord*—Visual inspection of the core indicated black sediment with a light brown oxidized layer restricted to the uppermost 0.5 cm. No laminations were visible, and no evidence (e.g., burrows or fragments of animals) could be seen of any macrofauna capable of bioturbating the sediments.

Unsupported  $^{210}\text{Pb}$  (Fig. 3) concentrations are variable ( $125\text{--}300 \text{ Bq kg}^{-1}$ ) over the surface 20 cm but decline in a near log-linear fashion from the core surface to ca. 55 cm, where background concentrations are approached. Supported  $^{210}\text{Pb}$  activity is nearly constant throughout the core (mean  $16 \text{ Bq kg}^{-1}$ , range  $8\text{--}25 \text{ Bq kg}^{-1}$ ). The  $^{210}\text{Pb}$  concentrations above 20 cm depth are inversely correlated with dry density ( $r = -0.68$ ; data not shown), indicating that the variability is due to variable sedimentation rates at the core site, probably through sediment resuspension and focusing. The overall decline in  $^{210}\text{Pb}$  activity with depth is relatively shallow, indicating high sedimentation rates at this site (see Fig. 4). The variability in  $^{210}\text{Pb}$  activity, particularly over the surface 20 cm, supports the application of the CRS model (Appleby and Oldfield 1978) to derive a core chronology, with 55 cm

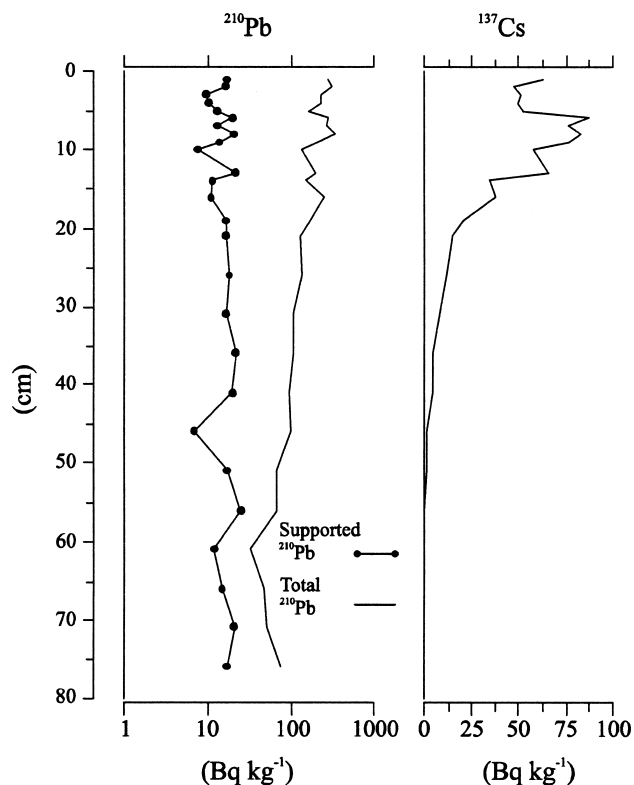


Fig. 3. Depth profiles of total and supported  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  in Roskilde Fjord core RF55E.

dating to ca. 1870. A well-defined peak in  $^{137}\text{Cs}$  activity between 5 and 7 cm (Fig. 3), attributed to Chernobyl fallout from 1986, is in reasonably good agreement with the  $^{210}\text{Pb}$  date for these levels (1986–1989). Both the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  profiles (Fig. 3) indicate that bioturbation is comparatively minor at this site.

Profiles of organic carbon (OC), biogenic silica (BSi), and sediment accumulation all show an increase in accumulation toward the top of the core, with the most marked increase beginning from ca. 1930 (Fig. 4). All three profiles show increased variability in the top section of the core, from ca. 1930 for BSi and from ca. 1970 for OC and sediment accumulation. The profiles of OC and BSi are driven by the changes in sedimentation rate since down-core changes in concentration of these two indicators are small (data not shown). There is a ninefold increase in sediment accumulation rate between the base of the analyzed section (ca. 1885) and the peak rate at the top of the core (ca. 1990).

Epipelagic, epipsammic, and epilithic diatoms dominate the sedimentary record, accounting for ca. 60% of the assemblages throughout the core (Fig. 4). Planktonic diatoms account for less than 10% of the assemblage before 1980, after this they increase to ca. 20%. Diatom species richness declined irregularly from ca. 1955.

Diatom-inferred total nitrogen (DITN) concentrations of around  $50 \mu\text{mol L}^{-1}$  are predicted for the period between ca. 1900 and 1955 (Fig. 4). After this time, and concomitant with a decline in species richness, DITN increases to a maximum of  $91 \mu\text{mol L}^{-1}$ , observed at the top of the core. This

value agrees well with the measured TN concentration of  $84 \mu\text{mol L}^{-1}$  from the monitoring data in 1995 and represents an 85% increase in TN concentration between 1950 and 1995. Confidence in the reconstruction is provided by the fact that at least 96% of the diatoms in each core sample were represented in the training set. Similarly, floristic matching using the squared chord distance ( $d^2$ ) indicated that only two samples (20–21 cm and 50–51 cm) lacked good analogues in the Danish training set. The sample-specific prediction errors associated with the reconstruction are shown in Fig. 4.

The rapid increase in TN concentration is associated with changes in the sedimentary diatom flora, although these changes are not dramatic. Several planktonic taxa, including *Coscinodiscus asteromphalus* Ehrenberg and *Cyclotella meneghiniana* Kützing, appear for the first time around 1955, while *Cyclotella choctawhatcheeana* Prasad maintains a presence throughout the core. Benthic and epiphytic taxa showed varying responses during the time period represented by the core. Of the benthic taxa, *Opephora mutabilis* (Grunow) Sabbe and Vyverman increased, while *Staurosira construens* Ehrenberg nearly disappeared from the record. Among the epiphytic taxa the *Tabularia fasciculata* aggregate decreased in relative abundance from ca. 1980, while *Cocconeis placentula* Ehrenberg showed virtually no change.

*Laajalahti Bay*—Visual inspection of core LaA indicated a 1-cm oxidized surface layer below which, to 35-cm depth, the sediment was dark brown with intermittent black bands that were particularly frequent between 17 and 24 cm. The remainder of the core was light brown in color. No evidence for the presence of large macrofauna was found at Laajalahti Bay, where the benthic fauna consists mostly of chironomids.

The sediment accumulation rate of Laajalahti Bay was relatively uniform at  $300 \text{ g m}^{-2} \text{ yr}^{-1}$  until the mid-20th century, after which it gradually increased to ca. five times this value at the present day. Both OC and BSi accumulation profiles appear to be strongly shaped by the sediment accumulation rate (Fig. 5). BSi accumulation rates began to increase slowly in the 1930s, while OC accumulation begins in the 1950s with markedly increased accumulation from the 1970s onward (see also Vaalgamaa 2004). The sediment pigment profile of the general biomarker Chl *a* and its degradation products shows an increasing trend, especially after 1940, with concentrations peaking in the mid-1960s and then declining toward the present. The high concentrations between 1940 and 1970, relative to the top of the core, were probably caused by increased production due to eutrophication of the embayment.

Concentrations of diatom-inferred total dissolved nitrogen (DITDN) are stable at around  $28 \mu\text{mol L}^{-1}$  until the early 20th century (Fig. 5). DITDN increased to  $37\text{--}39 \mu\text{mol L}^{-1}$  in the early 1920s. High DITDN concentrations from the mid-1950s to the late 1980s indicate an episode of severe eutrophication, with the highest concentrations (ca.  $59 \mu\text{mol L}^{-1}$ ) in the mid-1960s. This period was characterized by heavy wastewater loading from a sewage treatment plant and the start of the recovery of the embayment due to improved purification activities in the plant in the early 1970s. After

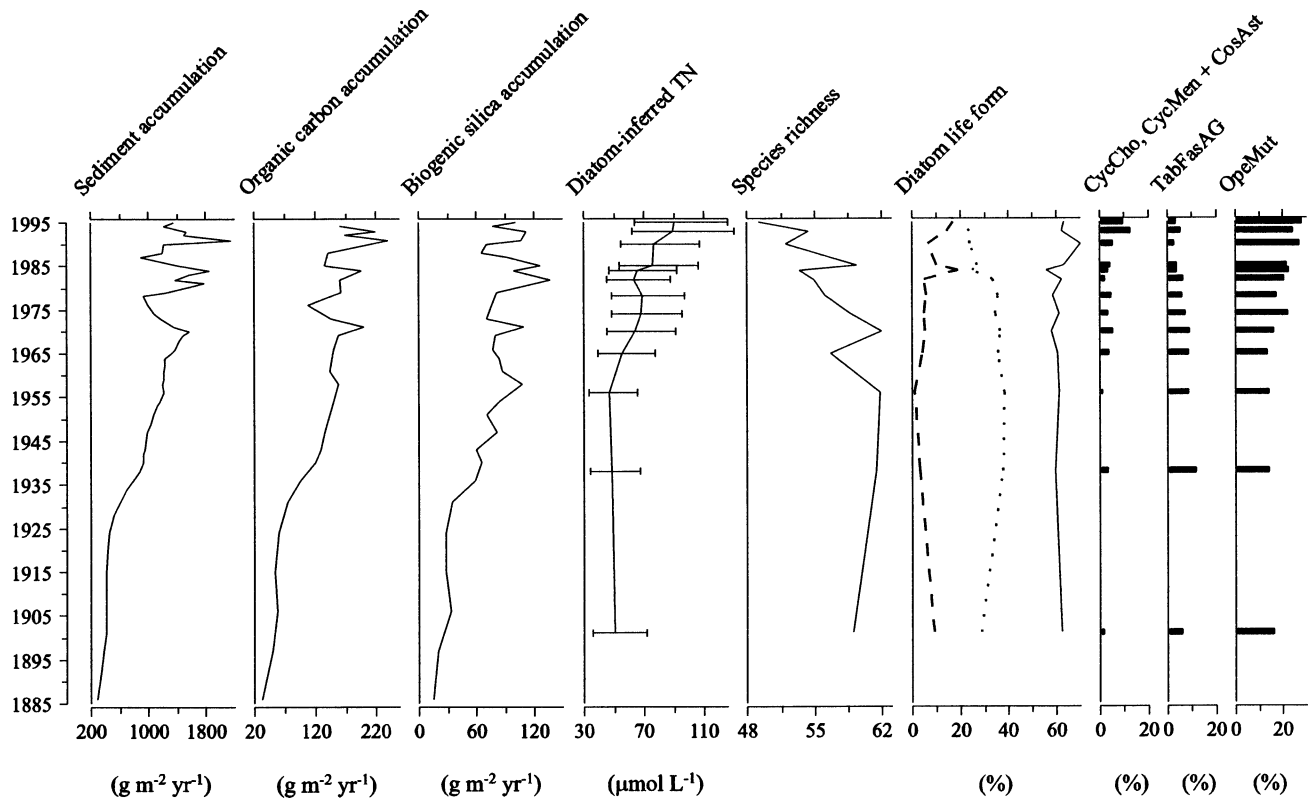


Fig. 4. Summary diagram, Roskilde Fjord. Stratigraphic plot of paleoecological proxies. Diatom-inferred TN panel shows sample-specific bootstrap standard errors (Birks 1995). Diatom life-form panel: dashed line = percentage of planktonic diatom taxa; dotted line = percentage of epiphytic taxa; solid line = percentage of epipelagic, epipsammic, and epilithic diatom taxa. Diatom taxa codes: CycCho, *Cyclotella choctawhatcheeana*; CycMen, *C. meneghiniana*; CosAst, *Coscinodiscus asteromphalus*; TabFasAG, *Tabularia fasciculata* aggregate; OpeMut, *Opephora mutabilis*.

closure of the treatment plant in 1986, DITDN concentrations dropped to ca.  $36 \mu\text{mol L}^{-1}$ , and similar concentrations persisted throughout the 1990s. Squared chord distance analysis ( $d^2$ ) reveals that Laajalahti Bay had good analogues in the modern Finnish training set throughout the core. The sample-specific prediction errors associated with the reconstruction are shown in Fig. 5.

The first notable change in the diatom assemblages occurred in the 1920s, when the proportion of planktonic diatoms, such as small *Cyclotella* (Kützing) Brébisson 1838 and *Thalassiosira* Cleve, increased markedly with a concurrent decrease in species richness (Fig. 5). The simultaneous decline in benthic *Fragilaria* Lyngbye could have been caused by increased turbidity, since the increase in DITDN suggests decreased benthic productivity. However, since the diatoms are expressed as percentage data, this decrease may also be an artifact caused by the increase in planktonic taxa. During the period of intense eutrophication (1950s to 1980s) there was a further increase in planktonic taxa and a pronounced fall in species richness. The common eutrophic taxa *Cyclotella atomus* Hustedt and *C. meneghiniana* Kützing (Korhola and Blom 1996) in particular became more abundant. Diatom species richness increased again in the late 1970s, most likely as a result of the improved wastewater treatment. After the closure of the treatment plant the two eutrophic *Cyclotella* taxa decreased in abundance.

## Discussion

Freshwater eutrophication has been a recognized environmental problem for several decades, but the concept of marine eutrophication was initially discounted due to the simple premise that, being large and dynamic, the marine ecosystem as a whole would be able to incorporate nutrient inputs without noticeable adverse effects. It was not until the mid-1980s that research into coastal eutrophication began to increase rapidly (Nixon 1995); by this time the most susceptible areas likely had been affected for a considerable time. The resulting sparsity of reliable long-term (greater than 20 yr) coastal monitoring data thus severely limits our ability to effectively manage estuarine and coastal waters. Using a multiproxy paleoecological approach, we have reconstructed eutrophication histories for two Baltic marine ecosystems with contrasting histories and demonstrated different responses to urban and agricultural nutrient sources.

*Eutrophication histories in Roskilde Fjord and Laajalahti Bay*—In Roskilde Fjord the sedimentation rate increases in three distinct phases: relatively slowly between the base of the core and ca. 1925; more rapidly until ca. 1970; after which the sediment accumulation rate oscillates sharply. The recent oscillation probably reflects increased resuspension of sediment within southern Roskilde Fjord after the almost

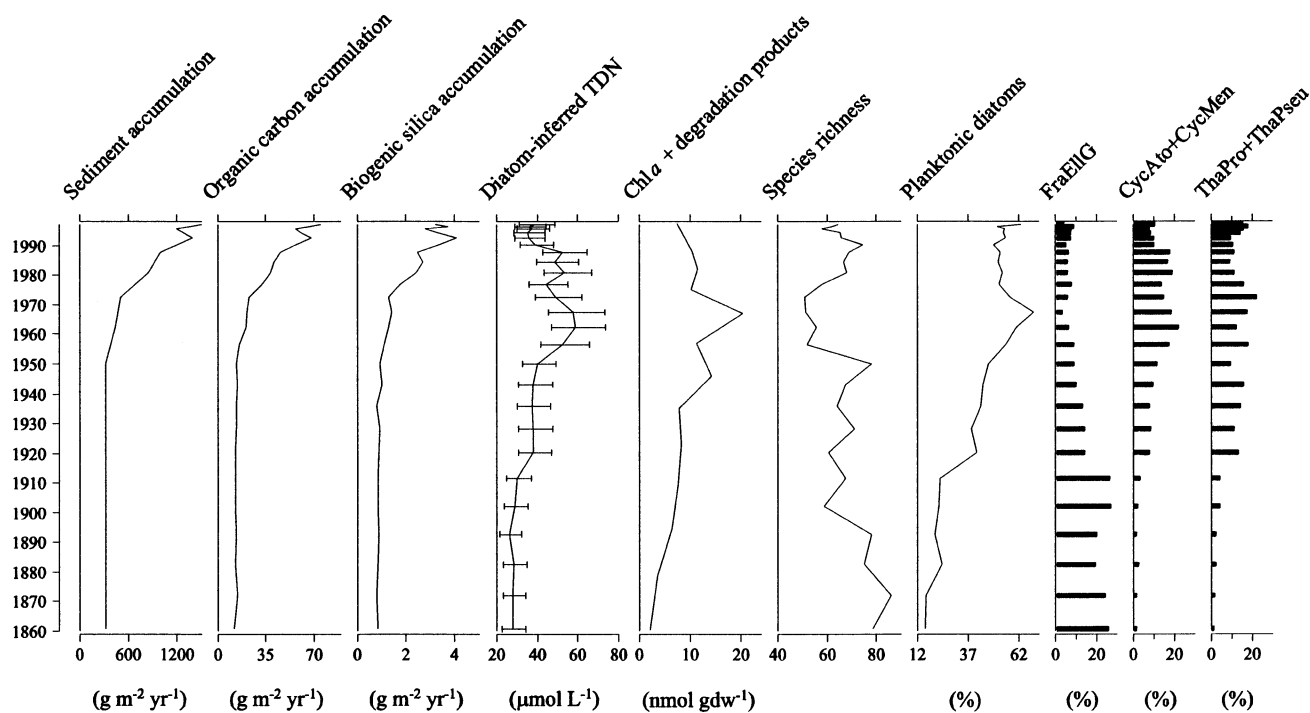


Fig. 5. Summary diagram, Laajalahti Bay. Stratigraphic plot of paleoecological proxies. The diatom-inferred TDN panel shows sample-specific jackknifed standard errors (Birks 1995). Diatom taxa codes: FraEllAG, *Fragilaria elliptica* aggregate; CycAto, *Cyclotella atomus*; CycMen, *C. meneghiniana*; ThaPro, *Thalassiosira proschkinae*; ThaPseu, *Thalassiosira pseudonana*.

complete loss of macrophyte cover during the 1970s destabilized the sediment surface, although macrophyte cover began to diminish from the 1930s (Adser unpubl. data). Concentrations of OC and BSi are relatively stable throughout the core, and their profiles are strongly driven by the sediment accumulation rate (Fig. 4); care should therefore be taken with the interpretation of these profiles.

The diatom-inferred TN (DITN) reconstruction is based on fossil diatom percentages and so is independent of the sediment accumulation rate. This profile shows a rapid increase in TN concentration after ca. 1950, which coincides with the increased use of artificial fertilizers in Denmark (Clarke et al. 2003). The DITN profile shows no indication of a decrease in TN concentrations in the fjord, despite management efforts to reduce nitrogen inputs to aquatic environments since the mid-1980s (Conley et al. 2002). Grimvall et al. (2000) in a study of the temporal aspect of nutrient transport from land to sea suggested that, because of decadal-scale processes governing the turnover of nitrogen, water quality response in highly fertilized systems to a decrease in N application “may be slower than the response to the post-war increase in the input.”

Diatom assemblage species richness declined irregularly during the period of rapid increase in DITN. Between the base of the core and the 1980s little change is seen in the relative proportion of diatom life forms (Fig. 4). Epiphytic taxa decline from 1980 onward: probably in response to loss of the macrophyte population. After ca. 1980 the percentage of planktonic diatoms increases but does not exceed more than 20% of the assemblages. Of these planktonic taxa, *C. meneghiniana* is associated with eutrophication in brackish

waters (Korhola and Blom 1996) and *C. choctawhatcheeana* (identified as *C. cf. caspia*) was associated with anthropogenic disturbance in Chesapeake Bay by Cooper (1995), who suggested that its tolerance for changes in environmental conditions gave it a competitive advantage. The presence of *C. choctawhatcheeana* throughout the profile might indicate that Roskilde Fjord has been perturbed by anthropogenic activities for the entire time period represented by this core, which, in the cultural landscape of Denmark, is not unexpected.

The percentage of epipelagic, epilithic, and epipsammic taxa also increases slightly after ca. 1980. This suggests that despite eutrophication, where an increase in planktonic taxa relative abundance would be expected at the expense of benthic taxa (Cooper 1995; Andr n 1999), the shallow fjord has remained suitable for benthic diatoms. The average Secchi depth from the four monitoring stations in the southern part of Roskilde Fjord is 3.4 m (Clarke unpubl. data). Since the average water depth is 3 m (Kamp-Nielsen 1992), the potential for production by the microphytobenthos is clear. The large population of blue mussels (*Mytilus edulis*) in the fjord (M hlenberg 1999) helps maintain high Secchi depth and may also reduce the percentage of planktonic diatoms that sediment out.

It appears that the sedimentary diatom record does not faithfully record the major species of the autochthonous planktonic community in Roskilde Fjord. Large blooms of *Skeletonema costatum* (Greville) Cleve occur in the fjord (Kamp-Nielsen 1992), but this lightly silicified diatom does not preserve well in the sediments, despite the temporal anoxia experienced in the basin. The poor preservation of this

taxon will increase the underrepresentation of planktonic diatoms in the core and partially explain why the expected increase in planktonic taxa is not observed at this site.

In Laajalahti Bay, the sediment accumulation rate increases slowly from ca. 1950 to ca. 1970, after which it shows more rapid accumulation rates, which continue to the top of the core. As in Roskilde Fjord, this pattern of sediment accumulation drives the accumulation of OC and BSi (Fig. 5), with the result that these variables should be interpreted with caution. The continued increase in sediment accumulation rate after reduction of the eutrophication pressure was not expected and is possibly a preservational artifact. Vaalgamaa (2004) suggested that modern conditions of periodic anoxia have slowed down degradation of organic material in the sediment, altering the steady-state input-decomposition balance observed prior to 1950. Dearing and Jones (2003) suggest that anthropogenic influence has been the principal influence on increasing sediment loads to aquatic systems through the Holocene and that small drainage basins (such as Roskilde Fjord and Laajalahti Bay) are more responsive to increases in sediment flux and therefore show the largest changes in recent accumulation rates.

The DITDN profile gives a different eutrophication history for Laajalahti Bay. The initial increase in DITDN in the 1920s coincides with a rapid expansion of the urban area in Helsinki (Laakkonen and Lehtonen 1999). The major effect on Laajalahti Bay was from the discharge of sewage wastewater between the mid-1950s and 1986, and this is clearly shown by the DITDN profile. The period of the greatest effect between the 1960s and the 1970s is confirmed by the sedimentary peak in Chl *a* and its degradation products. Assessment of the reconstruction using the squared chord distance shows that the fossil samples have good analogues in the modern Finnish training set, which adds confidence in the reconstruction. However, monitoring data from the early 1970s show TN concentrations as high as  $360 \mu\text{mol L}^{-1}$ , but the modern Finnish training set does not include sites above  $143 \mu\text{mol L}^{-1}$ . Since good analogues exist throughout the core, this would suggest that the diatom assemblages of Laajalahti Bay have not responded to the anomalous, high nutrient concentrations reported from the 1960s and 1970s, and this is supported by the clearly underestimated TDN concentrations in the reconstruction. Compared with the entire record of monitored TN (1972–1998), the DITDN reconstruction tracks the main trends reasonably well but systematically underestimates the actual concentrations during the most eutrophic period. There is, however, a good agreement between the measured and diatom-inferred concentrations throughout the 1990s (Weckström et al. 2004).

The diatom assemblages show a sharp decline in species richness during the period of maximum eutrophication (Fig. 5) and an increase following the cessation of wastewater discharge to Laajalahti Bay. As a consequence of the marked reduction in the nutrient load, the embayment has returned to the trophic status of the 1930s. Although the chemical recovery of Laajalahti Bay seems to be evident from the DITDN reconstruction, there still is no sign of a recovery back to a diatom assemblage dominated by benthic *Fragilaria* taxa. Laajalahti Bay shows the increase in planktonic diatoms that would be expected as a response to eutrophication.

The high planktonic production, indicated by the very low Secchi depth in Laajalahti Bay (0.5–1 m, Weckström et al. 2004), is maintained by internal loading (Kauppila et al. 2005).

*Long-term trends in eutrophication and paleoecological methods*—The changes in nitrogen concentration indicated from our reconstructions appear small compared with the order of magnitude changes in nutrient loading that have been suggested for northwestern Europe (e.g., Howarth et al. 1996; Conley 2000; van Raaphorst and de Jonge 2004), although links between increases in nutrient loading and resulting concentrations in open waters are not yet well known. Despite concern over recent eutrophication, the increase in nutrients between pristine conditions and the end of the 19th century may actually have been larger (Conley 2000). Billen et al. (1999) estimated the potential pollution load to the river Seine at the turn of the 19th century. Using historical technical records, they computed budgets for the major industries contributing to nutrient loads (such as the felt industry, clothes washing, and candle and soap production). Their results indicated that point discharges of nutrients may actually have been at their maximum at this time, and that rivers flowing through industrialized areas could have been carrying nutrient loads similar to those observed today. Billen et al. (1999) suggest this could have led to 19th and early 20th century coastal eutrophication. As we learn more about the effect of anthropogenic activities on nutrient mobilization over longer timescales, it is becoming apparent that changes have occurred throughout the history of human occupation. A multiproxy paleoecological reconstruction of anthropogenic effects on a Danish lake over a 6,000-yr period (Bradshaw 2001) indicated that while recent phosphorus-driven eutrophication of the lake is problematic, the largest changes in phosphorus concentration occurred during the Medieval period.

A multiproxy quantitative paleoecological approach offers a site-specific history of a system's response to recent environmental disturbances such as eutrophication. A range of methods can be used to hindcast nutrient concentrations, but their value is limited unless we can estimate their reliability (Birks 1995). Weighted averaging-based transfer functions are particularly valuable because they provide both quantitative reconstructions and statistically robust, sample-specific error estimates (e.g., Figs. 4, 5). In combination with analogue matching using a dissimilarity measure such as the squared chord distance, it is possible to assess the reliability of any reconstruction sample-by-sample down-core. Based on percentage data, transfer functions are also independent of the sediment accumulation rate, which may have a large influence on profiles of other proxies when presented as fluxes. This applies to estimates of diatom accumulation rates that have been used to show the increased productivity of both benthic and planktonic diatoms in lakes undergoing eutrophication (Anderson 1989). Such information is not shown by constant-sum percentage data when planktonic species increase disproportionately compared with benthic species (Battarbee et al. 2001). Obtaining accurate estimates of diatom accumulation rates is complex, since factors such as sediment focusing lead to variable rates of accumulation,

even in morphometrically simple (i.e., single basin) lakes (Anderson 1989), necessitating the use of a multiple-core approach. In our opinion it is likely that most marine basins will have complex sedimentary patterns and we query whether diatom accumulation rates from a single core could be considered representative of the area studied. For example, the recent oscillations in sediment accumulation rate in core RF55E (Fig. 4) probably reflect increased resuspension and hint at the likelihood of complex depositional patterns for sediment. Hall and Smol (1999) note that the costs associated with analyzing multiple cores will prevent the routine use of diatom accumulation rates and agree with Anderson (1995) that diatom-based transfer functions based on percentage data are less affected by core location and offer a cost-effective alternative.

Weighted averaging-based transfer functions for freshwaters appear to be robust when the variable being reconstructed (i.e., pH, TP, salinity) is the dominant variable controlling diatom communities for a given set of lakes. Problems do occur when transfer functions are developed for weaker, secondary gradients, such as lake water temperature (Anderson 2000). In the marine environment, the development of weighted averaging transfer functions is less straightforward, not only because of the correlation between N and P, but also because of the strong salinity gradients that occur in estuaries (e.g., Juggins 1992) and regionally as in Danish coastal waters (Conley et al. 2000) and the Baltic (Stigebrandt 2001). It is important, therefore, that training data sets are carefully designed to limit the salinity signal and to provide good coverage along nutrient gradients. Results presented here are encouraging, however, and indicate the potential of the method.

Another concern in applying paleoecological methods in coastal waters is bioturbation, which, added to the effects of resuspension and sediment mixing from tides, waves, and currents, can result in homogenized sediments that have a limited temporal resolution. Open marine systems generally have higher energy than freshwater systems (Nixon 1988), but sheltered coastal areas do exist. A coastline with fjords (e.g., Denmark, Finland, Sweden, British Columbia), or reduced tides (e.g., the Baltic Sea), may contain localities sheltered enough that erosion and transport are sufficiently reduced to permit the accumulation of fine-grained sediments. Bioturbation may still be of concern in such sheltered environments, and benthic macrofauna, through burrowing or feeding activities, can disturb temporal sequences by mixing sediments over periods of years or seasons depending on the accumulation rate. The  $^{210}\text{Pb}$  data from both Roskilde Fjord (Fig. 3) and Laajalahti Bay (Vaalgamaa 2004; Weckström et al. 2004) indicate that bioturbation is minimal at these sites. The  $^{210}\text{Pb}$  profile from Roskilde Fjord (Fig. 3) suggests that resuspension and increased bulk sediment rates are more important than bioturbation, justifying the application of the CRS-dating model.

Bioturbation can, however, affect the biogeochemistry of sediments. The burrows of species such as *Corophium volutator* (Amphipoda) and *Nereis diversicolor* (Polychaeta) can, by increasing irrigation of deeper sediment layers, cause an increase in the release of nutrients from the sediment (Mermillod-Blondin et al. 2004). Bioturbation can alter di-

atom assemblages by promoting dissolution of frustules through repeated exposure to seawater undersaturated in silica (Sancetta 1989) and through mechanical damage to fragile valves (McMinn 1995). In an analysis of factors affecting recent diatom thanatocoenoses of two deep (>250 m) British Columbian fjords, of which only one had permanent anoxia of the bottom waters, Sancetta (1989) concluded that in both locations bioturbation did not have a measurable effect while water column processes (zooplankton grazing and dissolution while settling) dominated the formation of the sedimentary assemblages.

The potential consequences of bioturbation can be reduced by sampling for sediment cores in areas where anoxia either reduces or eliminates benthic fauna. McMinn (1995) found that diatom assemblages from an anoxic basin with no bioturbation, within Ellis Fjord in Antarctica, included a high abundance of fragile, weakly silicified taxa that were not present in an oxic basin of the same fjord. Dating models that can account for the mixing of radionuclides in the zone near the sediment-water interface exist (e.g., Christensen 1982) and can be used in situations where bioturbation is suggested by flattened profiles of  $^{210}\text{Pb}$  activity. More studies on the factors affecting the formation of paleoecological records in coastal areas are needed, since it is becoming apparent that their quality can vary significantly from site to site and that different factors can have the strongest influence over the information retained in the sediment record at various sites. Both the  $^{210}\text{Pb}$  data and the well-resolved  $^{137}\text{Cs}$  profile from Roskilde Fjord (Fig. 3) suggest that mixing is not extreme at this site. In this sense, these sheltered marine basins have more in common with culturally affected lakes than higher energy estuarine environments with greater faunal densities where  $^{210}\text{Pb}$  dating can be problematic.

*Applicability for management of coastal eutrophication*—The long-term perspective gained from a reliably dated sediment record can provide details of the timing and rate of ecosystem response to anthropogenic disturbance, and if the temporal resolution is sufficient, an indication of natural variability beyond that offered by most neoecological studies (e.g. Smol 1992; Anderson 1995; Hall and Smol 1999) or dynamic models. Anderson (1995) focuses on the possibility of using paleoecology to validate empirical and dynamic models used for hindcasting or forecasting water quality variables. Anderson (1995) notes that empirical models, being based on modern relationships at a regional scale, are generally extensions of the space-for-time approach. Independent validation through paleoecology can highlight situations where regional variability can lead to reference conditions being outside the parameters of such models. Coastal paleoecological studies could potentially be used in a similar manner as their sophistication increases.

Following the requirements of the WFD, Andersen et al. (2004) used the DITN reconstruction from Roskilde Fjord to provide reference concentrations in the first attempt to measure deviation of observed values (seasonally weighted measured TN) from reference conditions for coastal TN concentrations. They took a range of the lowest inferred TN concentrations (50, 54, and 58  $\mu\text{mol L}^{-1}$ ) as potential reference conditions and compared annual TN concentrations

from 1989 to 2002 with concentrations expected under 15% and 25% deviation from each reference concentration. Classification of the ecological quality of the fjord varied depending on the reference concentration chosen and the percentage deviation considered acceptable, but even under the most lenient model ( $58 \mu\text{mol L}^{-1}$  reference concentration and 25% deviation) the fjord could only be considered as having good ecological status during 4 yr out of the 13 for which records existed.

During selection of the DITN reference conditions for Roskilde Fjord, Andersen et al. (2004) compared the reference concentrations from the paleoecological approach with results from modeling historical TN concentrations for various Danish fjords. Modeled concentrations for Roskilde Fjord were between  $60$  and  $65 \mu\text{mol L}^{-1}$ , and, for inner fjord areas in two other comparable systems, concentrations between  $48$  and  $57 \mu\text{mol L}^{-1}$  were obtained. These independent approaches show good agreement and add reliability to the estimates of reference TN concentrations in Danish estuaries from both approaches. A dynamic mass balance model for the Baltic Sea (Jansson and Dahlberg 1999) indicated a doubling of nitrogen concentration in coastal waters associated with a threefold increase in N loading. The 85% increase in TN concentration observed in Roskilde Fjord is also in close agreement with this model.

The ability of paleoecological methods to track ecosystem response to a reduction in nutrient load is shown by the example from Laajalahti Bay (Weckström et al. 2004). This embayment does not represent the conditions typical of many other inner coastal waters of southern Finland. The high internal P loading from the sediments due to the historical sewage inputs means Laajalahti Bay is N limited, while a number of other embayments in the low-salinity Gulf of Finland are typically P limited (Kauppila et al. 2005). The internal loading has resulted in Laajalahti Bay still not approaching reference conditions despite closure of the municipal sewage treatment plant 18 yr ago. The DITDN reconstruction indicates a TDN concentration in 1997 (the top of the core) that at ca.  $40 \mu\text{mol L}^{-1}$  is similar to concentrations inferred for the early 1950s but is still higher than the ca.  $27 \mu\text{mol L}^{-1}$  inferred from the lower levels of the core that represent reference conditions. The paleoecological record also indicates that diatom assemblages are currently dominated by planktonic taxa, while reference conditions clearly show assemblages dominated by benthic taxa (Weckström et al. 2004). Benthic diatoms are not part of the biological elements of the WFD in coastal and transitional waters, possibly reflecting the lack of information about benthic systems and eutrophication in a management context. In the case of Laajalahti Bay it may be prohibitively expensive to achieve good ecological status, and the directive may set less stringent environmental objectives (Kauppila et al. 2005). This indicates the need for historical information at a local scale to allow appropriate management.

By providing a long-term perspective, paleoecological techniques make it possible to define and justify a suitable time period to use as reference conditions. This approach can provide better definition at a local scale than many mathematical models, which are frequently spatially restricted and do not always cover the innermost coastal areas—as, for

example, Baltic-wide models. Estimates of the degree of environmental change will be more robust if they are based on two or more independent methods.

Lake sediments have been used to provide long-term records of cultural eutrophication and acidification at many sites (e.g., Christie and Smol 1993; Charles et al. 1994; and reviews in Stoermer and Smol 1999). In some locations applied paleolimnology has been used in conjunction with other approaches to elucidate site-specific responses to anthropogenic disturbance (*see* examples in Hall and Smol 1999). The potential for paleoecology to be used in a similar manner in coastal waters is becoming apparent.

Both Laajalahti Bay and Roskilde Fjord are relatively sheltered sites and probably have more in common with lake sediment records than many marine basins. This is indicated by the relatively undisturbed  $^{210}\text{Pb}$  profiles at these sites, which contrast substantially to the near uniform  $^{210}\text{Pb}$  profiles or surface mixed layers that can occur in marine coastal sediments. In many marine basins sediment reworking means that high-resolution sediment records with a high information content, such as we have identified at Laajalahti Bay and Roskilde Fjord, are unlikely to be present. It is important, therefore, that the benefits of working with marine sediment core studies are balanced against the potential problems. A detailed examination of these problems is not possible here, but some of the more obvious problems that need to be addressed are dating, diatom dissolution and its affect on transfer function development, pigment degradation processes in both the water column and sediments, re-suspension and winnowing of organic sediments, and spatial and temporal heterogeneity of sediment deposition. All of these topics have been addressed within paleolimnology over the last 20–30 yr. In comparison, paleoecological studies of marine sediments are in their infancy, but, given the clear need in management for long-term records from coastal embayments, these issues need to be addressed.

The multiproxy paleoecological approach is a powerful tool for providing evidence of historical biological and chemical reference conditions. By observing changes in a variety of independent indicators, a more complete ecological history can be provided, with different indicators providing complementary information that reduces uncertainty in interpretation. In spatially and temporally complex coastal systems this is likely to be of prime importance. However, to ensure reliability of results care must be taken in sites chosen to apply this approach. Only areas of constant sediment accumulation are suitable, and dependable assessments of trophic conditions through time will require a rigorous, multiproxy approach. This method will not be applicable everywhere, but in suitable environments, combined with other integrated physical, chemical, and biological indicators and information such as that proposed by Elliott et al. (1999), it can be applied to management needs. The WFD will have a large influence on the management of European aquatic resources, as reference conditions are defined and management techniques to achieve them are implemented. Clearly, knowledge of long-term change is needed to understand natural variability, both within and between sites, and to allow effective implementation of the WFD. To a certain extent this will drive the development of applied science.

Science-based monitoring and assessment that contains a strong quality-control element, such as the paleoecological approach, is therefore essential.

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