Journal of Paleolimnology (2006) 35:571–592
DOI 10.1007/s10933-005-5264-1

Assessing recent eutrophication in coastal waters of the Gulf of Finland (Baltic Sea) using subfossil diatoms

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Received 19 April 2005; accepted in revised form 3 October 2005

Key words: Baltic Sea, Diatoms, European Water Framework Directive, Eutrophication, Species richness, Transfer function

Abstract

Marine eutrophication of estuaries and coastal waters is considered to be a significant problem worldwide. In the semi-enclosed Baltic Sea, where the nutrient load has strongly increased from its natural level, this has led to marked changes in the coastal ecosystems. A key to successful management of coastal waters is reliable scientific evidence of their past state. The palaeolimnological record of subfossil diatoms was used to study the rate and magnitude of eutrophication over the last ca. 200 years in two urban and three rural sites. The urban sites showed marked increases in the percentage abundance of planktonic diatoms (from <50 to ca. 90% and from <5 to ca. 70%) and diatom-inferred total dissolved nitrogen (from <800 to ca. 3000 $\mu g\ l^{-1}$ and from <400 to ca. 800 $\mu g\ l^{-1}$), and a decrease in species richness starting in the 19th – early 20th century with increased urbanisation. At both sites a clear recovery was observed after the cessation of waste water loading by the mid 1980s. The present planktonic diatom assemblages of these embayments, however, show no change back to the pre-disturbance diverse benthic communities. In contrast, the changes observed in the rural sites were only moderate and occurred later starting in the 1940s. No marked increases in diatom-inferred total dissolved nitrogen were seen, however, all sites showed an increase in small planktonic taxa (from ca. 1–6% to 8–36%) indicating increased nutrient enrichment and turbidity. These small floristic changes could be seen as an early warning signal despite little change in the inferred nutrient concentrations. The results have implications for the European Water Framework Directive, which requires European surface waters to be of good ecological status, defined both by biological and chemical quality elements.

Introduction

Eutrophication of the world's coastal areas has become a significant problem in recent times (e.g., Nixon 1995; Billen and Garnier 1997; Jickells 1998). The same trend can be observed in the Baltic Sea, where the nutrient load has strongly increased from its natural level (Larsson et al. 1985; Cederwall and Elmgren 1990; HELCOM 2005). This has resulted in increased plankton biomass and increased amounts of filamentous algae, decreased transparency of the water body, changes in community structure and abundance of zoobenthic communities, and anoxia/hypoxia of deeper basins (Jørgensen et al. 1990; Bonsdorff et al. 1997; Grall and Chauvaud 2002; Gray et al. 2002).

In contrast to the pelagic areas of the Baltic Sea, which are predominantly influenced by nutrient loading from large rivers and the atmosphere, the trophic status in estuaries and sheltered embayments is mainly determined by local factors, particularly the nutrient load from coastal catchment areas (e.g., Pitkänen, 1994; Weckström et al. 2002). The trophic conditions of these systems are strongly affected by mixing of the water column and water exchange with the open sea, which are controlled by climate and weather conditions and the geomorphology of the coast (Pitkänen et al. 1990). As the nutrient load has a much greater effect on these more closed coastal ecosystems compared to the open sea, they are a focus for management efforts.

The setting of functional management targets requires knowledge about present biogeochemical nutrient cycles in coastal waters. Equally important, however, is the need to know how nutrient concentrations have varied through time and what the chemical and ecological background conditions have been. Without knowledge of baseline conditions, it is difficult to assess the magnitude of eutrophication, which could lead to inappropriate management practices. The recent European Water Framework Directive (WFD) (Anonymous 2000) requires all surface waters in Europe to fulfill the criterion of 'good ecological status', where the biological and chemical status departs only slightly from undisturbed baseline conditions, during the realisation period 2015–2027. The WFD requires the determination of these baseline conditions against which the extent of anthropogenic eutrophication and the present ecological status of a system can be assessed.

Attempts to determine long-term nutrient enrichment using existing monitoring data are hampered by the limited time span of contemporary monitoring programs; for example, the Finnish coastal monitoring program only started in 1966 (Kohonen 1974). In the absence of historical water chemistry data, paleolimnologists have been using the information preserved in lake sediment profiles to reconstruct missing data sets both in Finland (e.g., Hynynen et al. 2004) and elsewhere (e.g., Tibby 2004; Reid 2005; Wolin and Stoermer 2005). However, fewer studies have attempted to use similar approaches to address longer term changes in the trophic status of coastal waters. By using qualitative analysis of indicators preserved in sediments, it is possible to determine changes in community structure and diversity (e.g., Andrén 1999, 2000; Cooper et al. 2004), while the relatively recent development of quantitative approaches, which are based on large modern reference data sets, make it possible to obtain reconstructions of actual nutrient concentrations (e.g., Clarke et al. 2003; Weckström et al. 2004). For the purpose of the present study, diatoms were chosen as an indicator group, as they (1) are present in diverse, numerically abundant assemblages; (2) have a siliceous cell wall that preserves well in freshwater and marine sediments; (3) can be identified to species level due to their taxonomically distinct frustules; and (4) are widely studied and hence known to respond quickly to physical and chemical changes in their environment (see Battarbee et al. 2001).

In this study, five sediment cores were collected from both urban and agriculturally impacted embayments from the Gulf of Finland (Figure 1), which is one of the most affected parts of the Baltic Sea (Pitkänen et al. 1990; Kauppila and Bäck 2001). The objectives were (1) to determine post-industrial eutrophication trends in coastal waters of southern Finland with particular emphasis on the effect of different land-use practices on their trophic development; and (2) to provide knowledge of background or reference conditions in the Gulf of Finland for the purposes of the European Water Framework Directive.

Study sites

Urban sites

Töölönlahti is a small, shallow and very turbid (transparency <1 m) embayment in the centre of Helsinki, the capital of Finland (Tables 1 and 2). It is the only embayment in the centre, which has not been dredged or filled in (Tikkanen et al. 1997). As a result of post-glacial isostatic land uplift and construction work since the 19th century, the site is presently connected to the open sea through a narrow system of straits, which has increased the residence time and restricted the water exchange of the embayment. The salinity (4.8‰), however, is close to that of the outside archipelago (Table 2). The original catchment area of Töölönlahti was ca. 4.7 km^2, but due to the diversion of sewage

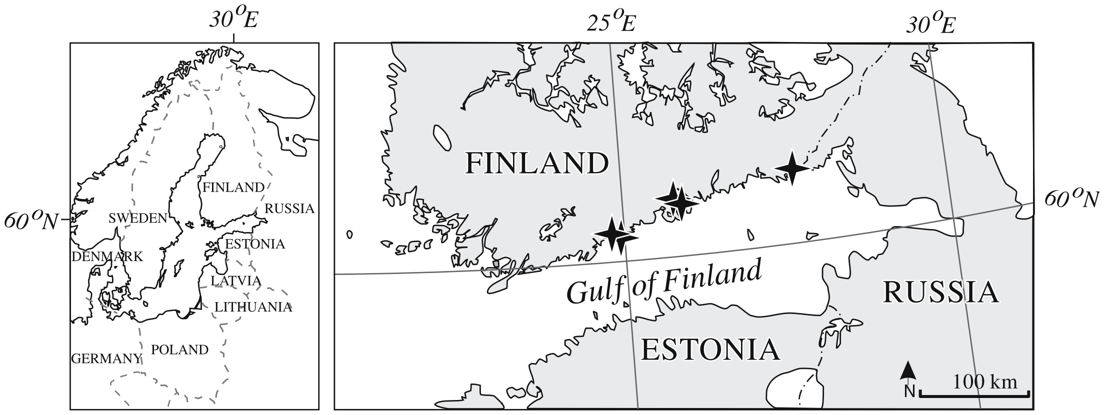

Figure 1. The index map shows the Baltic Sea and its bordering countries. In the detailed map the study sites are indicated as stars. From west to east: Laajalahti, Töölönlahti, Pieni Pernajanlahti, Fasarbyviken, and Hellänlahti.

water elsewhere its effective catchment is presently only ca. 0.4 km^2 (Korhola and Blom 1996). The embayment is surrounded by a densely built residential area, a railway yard, and a park. Extensive urbanisation of the catchment took place in the late 19th and early 20th century, which led to increased amounts of waste water being directed into the embayment. Töölönlahti received non-treated and later partially treated waste water until the 1960s. A sugar mill was located on the northern shore between 1823 and 1965 adding to the waste water load of the embayment. Töölönlahti is still eutrophic with an annual mean total phosphorus (TP) concentration of ca. 70 μg l^{-1} and a total dissolved nitrogen (TDN) concentration of ca. 600 μg l^{-1}.

Laajalahti is a shallow and turbid (mean depth ca. 2.4 m, transparency ca. 1 m) urban embayment west of Helsinki (Tables 1 and 2). It has a surface area of ca. 5.3 km^2 and is connected to the open archipelago by two narrow straits restricting the water exchange. Laajalahti receives fresh water from two small brooks, which, however, do not affect the salinity of the site (4.6‰), as it is close to that of the outside archipelago. In the catchment area, much of the agricultural land, which presently constitutes 12% of the catchment, has been converted to urban area (54%) and the amount of forests has markedly decreased since the 1950s. In the 1960s, Laajalahti was heavily impacted by waste water loading from a sewage treatment plant built in 1957 close to the shore. The plant was closed in 1986 (Kauppila et al. 2005). At present, the embayment receives only diffuse loading from the catchment area. Despite the reduction in the nutrient load Laajalahti is still eutrophic (ca. 400 μg l^{-1} TDN and ca. 65 μg l^{-1} TP, Table 2).

Rural sites

Pieni Pernajanlahti is a long and narrow estuary, which was formed along a tectonic fault-line. Its surface area is ca. 9 km^2 and the approximate mean depth ca. 5 m, while maximum depth is 15 m (Table 1). The water is turbid with a mean transparency of 1.2 m (Table 2). Pieni Pernajanlahti is connected to the open sea through a maze of islands restricting the water exchange; the mean salinity is, however, ca. 4.5‰. The catchment area of the estuary measures 356 km^2, of which agricultural land comprises ca. 26%, while the rest is almost entirely covered by forests (64%). River Ilolanjoki, which has a mean water flow of ca. 3 m^3 s^{-1} (L. Villa, unpublished data), empties into the site. Pieni Pernajanlahti is presently eutrophic with an annual mean TDN concentration of ca. 400 μg l^{-1} and a TP concentration of ca. 45 μg l^{-1} (Table 2).

Fasarbyviken is a small and turbid (water transparency ca. 1 m) embayment with a surface area of ca. 1.3 km^2, an approximate mean depth of ca. 2 m and a salinity of ca. 4.8‰ (Tables 1 and 2). The catchment area of Fasarbyviken is dominated by forests (ca. 69 %), while agricultural land, which is mainly located in the immediate vicinity

Table 1. Description of the five study sites and core details.

	Urban sites		Rural sites		
	Töölönlahti	Laajalahti	Pieni Pernajanlahti	Fasarbyviken	Hellänlahti
Location	60.18° N, 24.93° E	60.18° N, 24.87° E	60.38° N, 25.90° E	60.37° N, 25.98° E	60.58° N, 27.77° E
Catchment area (km^2)	4.7/0.4	52	356	18	386
Surface area (km^2)	0.2	5.3	9.0	1.3	2.0
Appr. mean depth (m)	2.0	2.4	5.0	2.0	1.6
Date of coring	October 2003	September 1998	September 1998	September 1998	September 1998
Depth of coring site (m)	2.2	3.7	9.6	4.0	2.6
Core length (cm)	84	90	83	89	89

Values for catchment and surface area are rounded up. Approximate mean depth was estimated from charts. For Töölönlahti, both past and present size of the catchment area is given.

of the site, covers ca. 24%. Urban and industrial land comprises only ca. 7% of the small catchment area (ca. 18 km^2). A relatively large pig farm (appr. 700 heads), which is located in the catchment, may affect the water quality of the embayment, as the manure is spread on nearby fields. Fasarbyviken is moderately eutrophied with an annual mean TDN concentration of ca. 370 $\mu g\ l^{-1}$ and a TP concentration of ca. 40 $\mu g\ l^{-1}$ (Table 2).

Hellänlahti is a small (ca. 2 km^2) and shallow (mean depth ca. 1.6 m) estuary at the bottom of a much larger estuarine complex called Virolahti (Table 1). It is presently turbid (transparency < 1 m) and eutrophied (ca. 390 $\mu g\ l^{-1}$ TDN, ca. 50 $\mu g\ l^{-1}$ TP) (Table 2). River Virojoki, which drains into the site, constitutes the bulk of the nutrient load received by the embayment (Kokko and Turunen 1986). Three fish farms are located in the outer part of the larger estuarine complex, which, depending on the currents, may also affect the water quality of the site. The salinity is around 3.7‰. The catchment area of Hellänlahti covers 386 km^2, and is dominated by forest (77%). Agricultural land comprises 15% and urban and industrial area only about 4%.

Material and methods

Core collection and dating

The sites were sampled with a mini-Mackereth corer (Mackereth 1969) in September 1998, except for Töölönlahti, which was sampled in October 2003. The cores were taken from the deepest area at all sites, and sectioned into 1 cm intervals (Table 1). The main characteristics of the obtained sediments were described and sub-samples were stored at 4 °C in small plastic bags.

All five cores have been analysed for their organic matter content (percentage loss-on-ignition, % LOI) (Boyle 2004) and a range of geochemical variables. Detailed descriptions of these analyses are given in Vaalgamaa (2004) and Vaalgamaa and Conley (unpublished data). Analyses conducted on the Töölönlahti core have not yet been reported.

In order to date the sediment, the cores from Hellänlahti, Fasarbyviken, Pieni Pernajanlahti and Laajalahti were analysed for ^{210}Pb, ^{226}Ra and ^{137}Cs by direct gamma assay in the Liverpool

Table 2. Summary characteristics of the study sites.

	Urban sites		Rural sites		
	Töölönlahti	Laajalahti	Pieni Pernajanlahti	Fasarbyviken	Hellänlahti
Transparency (m)	0.7	1.0	1.2	1.0	0.8
Salinity (‰)	4.8	5.4	4.4	4.8	3.7
TDN (μg l^{-1})	600	393	398	365	385
TP (μg l^{-1})	68	63	45	39	48
Chl *a* (μg l^{-1})	46	14	17	13	17

Water chemistry, chlorophyll *a* and water transparency data were collected for a diatom-nutrient calibration data set between August 1996 and February 1998. TDN, total dissolved nitrogen; TP, total phosphorus; chl *a*, chlorophyll *a*. Mean values of selected variables are given. Water transparency was measured using the white lid of a Limnos-water sampler (diameter 11 cm).

University Environmental Radioactivity Laboratory, using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors. The technical procedures are described in Appleby et al. (1986, 1992) and Appleby (2001). Sediment chronologies were calculated using the constant rate of supply (CRS) and the constant initial concentration (CIC) ^{210}Pb dating models together with chrono-stratigraphic dates determined from the ^{137}Cs record. To obtain a chronology for the Töölönlahti core, it was correlated using % LOI with another core from a previous study, which was dated using ^{210}Pb, ^{137}Cs and spheroidal carbonaceous particles (SCP) (Tikkanen et al. 1997). This was possible, as there were distinctive changes in % LOI. Chronologies for the cores collected in 1998 are given in Table 3.

Diatom analysis

A total of 20–40 sub-samples from selected levels of each core were prepared by oxidation using H_2O_2 (Renberg 1990). The resulting suspensions were centrifuged and washed with distilled water. A few drops of the cleaned slurry were air-dried on cover slips and subsequently mounted on glass slides with Naphrax®. Diatoms were identified using an Olympus BX40 microscope with phase-contrast at 1000× and valve counts per sample were 400–500. The main taxonomic sources used for identification were Krammer and Lange-Bertalot (1986–1991), Snoeijs et al. (1993–1998), Witkowski (1994) and Witkowski et al. (2000). The use of taxa aggregates followed taxonomic conventions of the MOLTEN project (http//:craticula.ncl.ac.uk/MOLTEN/jsp). Fragile and weakly silicified taxa that are often not preserved in coastal sediments were found in some of the cores. These taxa were also enumerated, but were excluded from the main count and not used in subsequent numerical analyses, as preservation factors could have altered their original abundance. The relative percentage abundance of the excluded taxa was expressed as a percentage of the total number counted, while for all other taxa it was expressed as a percentage of the 400–500 included valves.

Data analysis

The ratio of planktonic to benthic diatom taxa was calculated for each fossil sample. Detrended correspondence analysis (DCA), which is a unimodal indirect ordination method (Hill and Gauch 1980), was used to summarise the compositional changes in diatom assemblages over time. Species richness of the fossil assemblages was assessed using rarefaction analysis, which gives realistic estimates of richness without any bias associated with the variability of count size between samples. Rarefaction analysis was implemented with the program RAREPOLL (Birks and Line 1992). The diatom stratigraphies were zoned with constrained optimal sum of squares partitioning using the software package Zone v. 1.2 (Juggins 1991). The number of statistically significant zones was calculated using the broken-stick model described in Bennett (1996).

Diatom-inferred total dissolved nitrogen (DI-TDN) reconstructions were calculated for each core using a transfer function, which had previously been generated from a calibration data set consisting of 49 relatively small, shallow and sheltered embayments along the southern coast of

Table 3. Radionuclide chronologies of the four cores collected in 1998.

Laajalahti					Pieni Pernajanlahti					Fasarbyviken					Hellänlahti				
Depth (cm)	Chronology		Sed. rate		Depth (cm)	Chronology		Sed. rate		Depth (cm)	Chronology		Sed. rate		Depth (cm)	Chronology		Sed. rate	
	Date AD	±	cm $year^{-1}$	±		Date AD	±	cm $year^{-1}$	±		Date AD	±	cm $year^{-1}$	±		Date AD	±	cm $year^{-1}$	±
0.0	1998	0			0	1998	0			0	1998	0			0	1998	0		
0.5	1998	1	0.83	15	0.5	1998	1	2.84	61	0.5	1998	1	2.52	16	0.5	1998	1	1.25	19
2.5	1995	2	0.57	11	4.5	1996	1	2.24	21	4.5	1996	1	2.32	15	2.5	1996	2	1.00	17
4.5	1991	2	0.51	11	6.5	1995	1	1.42	17	8.5	1994	2	2.04	17	4.5	1994	2	0.80	14
6.5	1987	2	0.43	14	8.5	1994	2	1.12	17	12.5	1992	2	1.42	18	6.5	1991	2	0.80	16
8.5	1982	3	0.31	19	10.5	1992	2	0.93	19	16.5	1989	2	1.15	16	8.5	1989	2	0.80	15
10.0	1974	4	0.21	24	12.5	1989	2	0.84	15	21.5	1984	3	1.35	17	10.5	1986	3	0.67	20
12.5	1963	6	0.16	28	16.5	1985	2	0.90	16	25.5	1982	3	1.60	26	12.5	1983	4	0.58	18
14.5	1949	8	0.14	28	18.5	1982	2	0.90	19	29.5	1979	3	1.43	19	14.5	1979	5	0.37	17
16.5	1935	10	0.13	28	21.5	1979	3	0.89	24	33.5	1977	4	1.24	23	16.5	1972	7	0.19	33
18.5	1919	18	0.12	28	25.5	1974	3	0.62	23	37.5	1973	5	1.04	21	18.5	1958	10	0.12	37
20.5	1903	23	0.12	28	29.5	1966	4	0.53	15	43.5	1967	7	0.92	27	19.5	1948	15	0.11	40
					33.5	1959	5	0.62	25	47.5	1962	9	0.77	35	20.5	1939	19	0.10	43
					37.5	1953	6	0.50	36	53.5	1954	11	0.61	31	21.5	1929	23	0.10	46
					43.5	1940	8	0.38	33	57.5	1946	15	0.44	38	22.5	1920	26	0.10	
					47.5	1927	11	0.32	39	59.5	1940	18	0.39	52					
					53.5	1908	16	0.31	63	61.5	1935	21	0.40	52					
					57.5	1895	23	0.29	58	63.5	1930	24	0.41	52					
					59.5	1888	26	0.30	62	65.5	1926	26	0.41	54					
										67.5	1921	28	0.42	58					
										69.5	1916	30	0.42	61					

Finland (Weckström et al. 2004). In Weckström et al. (2004), the responses of diatom taxa with at least 1% abundance in two or more samples were modelled against the log_{10}- transformed nutrient data using weighted-averaging partial least squares (WA-PLS) regression (ter Braak and Juggins 1993). WA-PLS calibration was then used to estimate the diatom-inferred modern nutrient concentrations for each of the 49 sites in the calibration data set. The model for TDN performed better than the TP-model in terms of the squared correlation between the observed and diatom-inferred values ($r^2 = 0.73$ and 0.57, respectively), and the root mean square error of prediction (RMSEP $= 0.09_{log}$ and 0.10_{log}, respectively), as assessed by leave-one-out cross-validation. As nitrogen is considered to be the limiting nutrient to phytoplankton growth in most parts of the Baltic Sea with the exception of the low-saline Bothnian Bay (e.g., Granéli et al. 1990; Pitkänen 1994), TDN was adopted for further analyses. The programme C2 (Juggins 2004) was used for model generation and down-core reconstructions.

The dissimilarity between each fossil sample and its closest modern analogue was estimated, as down-core reconstructions are potentially more reliable if the fossil diatom assemblages have close modern analogues in the calibration data set. The dissimilarities of all calibration set sites were first calculated using four dissimilarity coefficients: squared chord distance, squared Euclidean distance, squared chi-squared distance and Bray Curtis percentage similarity. These were subsequently checked for deviations from the normal distribution, as the cut-off values, which are determined by the percentiles of the distribution of the dissimilarity values across the calibration set, are strongly influenced by skewed distributions. Chord distance (d^2) (Overpeck et al. 1985) showed the least skewed distribution of the calibration set dissimilarity values and was therefore chosen. 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 calibration set. The modern analogue technique (MAT) was implemented using the programme C2 (Juggins 2004).

Squared residual distance of the modern samples to the TDN axis in a canonical correspondence analysis (CCA) was used as a criterion of lack of fit to TDN. Samples with a high residual distance from the TDN axis have a poor fit to this variable. Fossil samples can be passively positioned on the TDN axis through transition formulae. A fossil sample with a residual distance equal to or larger than the residual distance of the extreme 10% of the calibration data set was considered to have a poor fit to TDN. Both DCA and CCA were performed with CANOCO for Windows, version 4.0 (Ter Braak and Šmilauer 1998).

Results and discussion

Core chronologies

At Laajalahti and Hellänlahti, the total ^{210}Pb activity reached equilibrium with the supporting ^{226}Ra at a depth of ca. 20–25 cm. At Pieni Pernajanlahti and Fasarbyviken, equilibrium was reached at a greater depth of 60 and 75 cm, respectively. Since the maximum unsupported ^{210}Pb activity was between 102 Bq kg^{-1} (Laajalahti) and 166 Bq kg^{-1} (Fasarbyviken), it is likely that these represent a period of not more than about 100 years (Table 3).

All four cores taken in 1998 had a well-resolved subsurface peak of ^{137}Cs activity. Generally, the ^{137}Cs inventories of these peaks were large indicating that they record the fallout from the 1986 Chernobyl accident. The total inventory of Laajalahti (10, 233 Bq m^{-2}) was lower than at the other sites, but the $^{137}Cs/^{210}Pb$ inventory ratio was comparable, and well in excess of the expected weapons $^{137}Cs/^{210}Pb$ ratio. At Laajalahti and Hellänlahti, it was not possible to distinguish a peak recording the 1963 nuclear weapons fallout maximum, which presumably has been masked by downwards diffusion of Chernobyl ^{137}Cs, whereas at Pieni Pernajanlahti and Fasarbyviken the 1963 nuclear weapons fallout maximum is seen as a small shoulder on the ^{137}Cs profile at depths of 31.5 and 48.5 cm, respectively.

Using the 1986 ^{137}Cs date as a reference point, composite model ^{210}Pb chronologies (using both CIC and CRS) were constructed for Laajalahti and Hellänlahti following methods outlined in Appleby (1998). At Laajalahti, the sedimentation rate appears to have been relatively uniform (0.13 cm $year^{-1}$) during the first half of the 20th century. After a rapid increase since ca. 1980, the sedimentation rates during the past decade have had an average value of ca. 0.6 cm $year^{-1}$. At Hellänlahti, sedimentation rates have been relatively steady until the mid 1970s, with a mean value of ca. 0.12 cm $year^{-1}$. During the early 1980s accumulation rates increased dramatically and contemporary values are calculated to be ca. 0.83 cm $year^{-1}$. ^{210}Pb dates for Pieni Pernajanlahti and Fasarbyviken were calculated using the CRS model, together with the 1986 and 1963 depths indicated by the ^{137}Cs stratigraphy. Use of the CIC model was precluded by the non-monotonic variations in ^{210}Pb activity. At Pieni Pernajanlahti, the mean sedimentation rate until 1940 was ca. 0.31 cm $year^{-1}$. Since then sedimentation rates have increased generally, with peaks in the early 1950s, ca. 1980, and during the past few years. The mean post-1963 sedimentation rate has been 0.96 cm $year^{-1}$. At Fasarbyviken, the mean sedimentation rate until the mid 1940s was ca. 0.41 cm $year^{-1}$. Between 1950–1980 sedimentation rates increased sharply reaching peak values of more than 1.5–2 cm $year^{-1}$ in the 1980s and 1990s.

The core taken from Töölönlahti was correlated with a previously dated core using % LOI, as easily discernible changes occurred in the % LOI profiles. The original core was dated using ^{210}Pb, ^{137}Cs and spheroidal carbonaceous particles (SCP). However, the ^{210}Pb dating proved to be problematic; hence more weight was given to the ^{137}Cs and SCP dating, which were in good agreement, supported by the pollen record of park trees and known land use changes in the catchment area. The mean sedimentation rate of the core during the 20th century was ca. 0.6 cm $year^{-1}$. A more detailed discussion is given in Korhola and Blom (1996) and Tikkanen et al. (1997).

Diatom assemblage structure & diversity

Töölönlahti

A comparison with the diatom stratigraphies presented in Korhola and Blom (1996) and Tikkanen et al. (1997) was not attempted, as the method of preparing diatom slides was different. Many of the very common, small and light taxa were lost in the diatom counts of these previous studies as the samples were decanted only for some hours after H_2O_2 treatment. These taxa, however, account for 22–85% (mean 56%) of percentage abundances in the samples studied here. The amount of total diatom taxa found was also considerably lower in these former studies amounting only to 89 taxa compared to 180 taxa in the present study. Although the general trends in diatom assemblage structure and diversity are similar despite the difference in slide preparation, the percentage abundance data for single taxa are very different. More importantly, in Korhola and Blom (1996) and Tikkanen et al. (1997) the diatom assemblages show a dominance of benthic taxa at the bottom of the core and again at the top, which differs markedly from the results of the present study (discussed in detail later in this paper).

Most of the taxa in the diatom stratigraphy of Töölönlahti were benthic, with only 26 planktonic taxa being observed. The assemblages of the core were divided into 3 statistically significant zones (Figure 2). Even Zone 1 (ca. 1800–1915), the oldest zone, had a marked planktonic component ranging between 42–79% (Figure 3). An increase in the abundance of planktonic taxa has often been observed in response to eutrophication (e.g., Cooper and Brush 1991; Andrén et al. 1999, 2000; Bennion et al. 2004; Cooper et al. 2004). These high abundances could be attributed to the location of the embayment, as urban development around Töölönlahti started in the early 1800s (Tikkanen et al. 1996). There was a clear increase in the relative abundance of planktonic taxa beginning in the 1830s, which is also reflected in the DCA axis 1 scores summarising the compositional change of diatom assemblages over time. Concurrent with these changes, species richness began to decline, which is consistent with several other studies, where severe cultural eutrophication has led to a decrease in species richness of algal communities (Wetzel 2001).

Species assemblages in Zone 1 were dominated by several *Cyclotella* (Kützing) Brébisson 1838 nom. cons. and *Thalassiosira* Cleve 1873 taxa associated with eutrophic waters (EDDI 2001; MOLTEN 2004), e.g., *Cyclotella atomus* Hustedt 1937, *C. meneghiniana* Kützing 1844 and *Thalassiosira guillardii* Hasle 1978 with increasing abundances of the latter two. The abundance of *C. meneghiniana* peaked markedly to 45% in a single sample dated to ca. 1910. *Skeletonema costatum* (Greville) Cleve 1878 and *Chaetoceros* spp. Ehrenberg 1844 started to increase towards the end of the zone, while benthic taxa such as

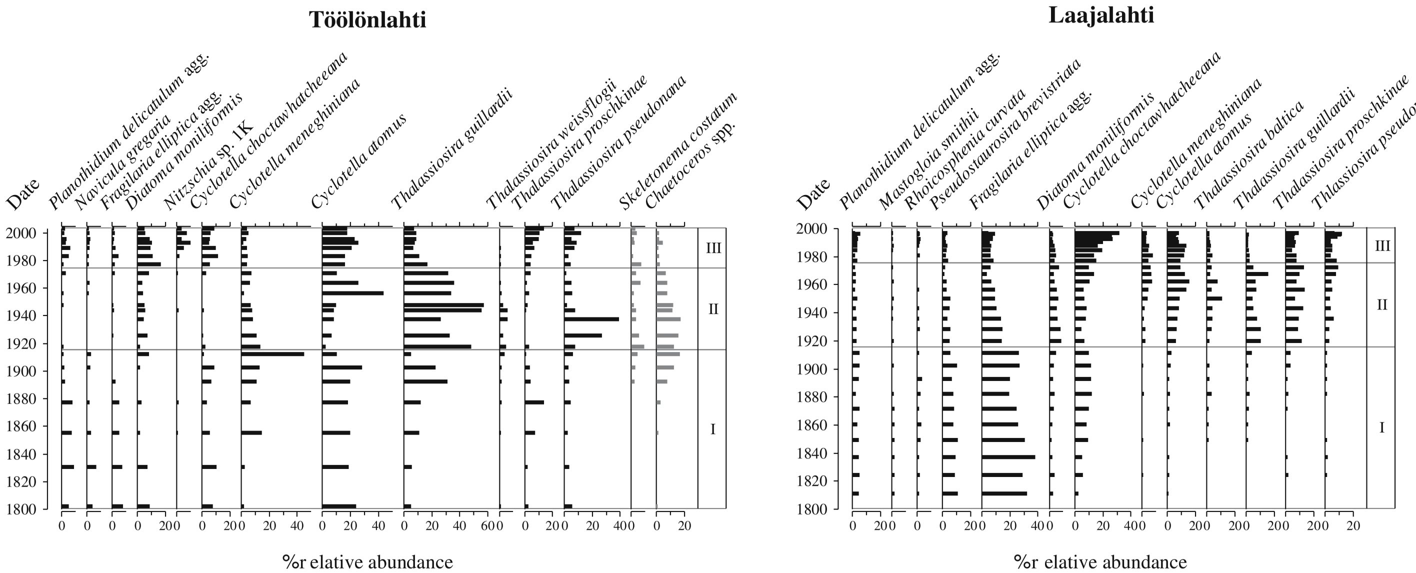

Figure 2. Stratigraphic diagrams of the urban sites showing the main diatom taxa (percentage abundance). Zonation of the species data was performed using constrained optimal sum of squares partitioning. All zones were statistically significant. *Planothidium delicatulum* agg. includes *P. delicatulum* (Kützing) Round & Bukhtiyarova nov. comb., *P. septentrionalis* (Östrup) Round & Bukhtiyarova comb. nov., *P. hauckianum* (Grunow) Round & Bukhtiyarova comb. nov. and *P. engelbrechtii* (Cholnoky) Round & Bukhtiyarova nov. comb. *Fragilaria elliptica* agg. includes *Staurosira elliptica* (Schumann) Williams & Round 1987, *Fragilaria elliptica* Schumann 1867 *sensu* Krammer & Lange-Bertalot 1991 and *Staurosira punctiformis* Witkowski, Metzeltin & Lange Bertalot spec. nov.

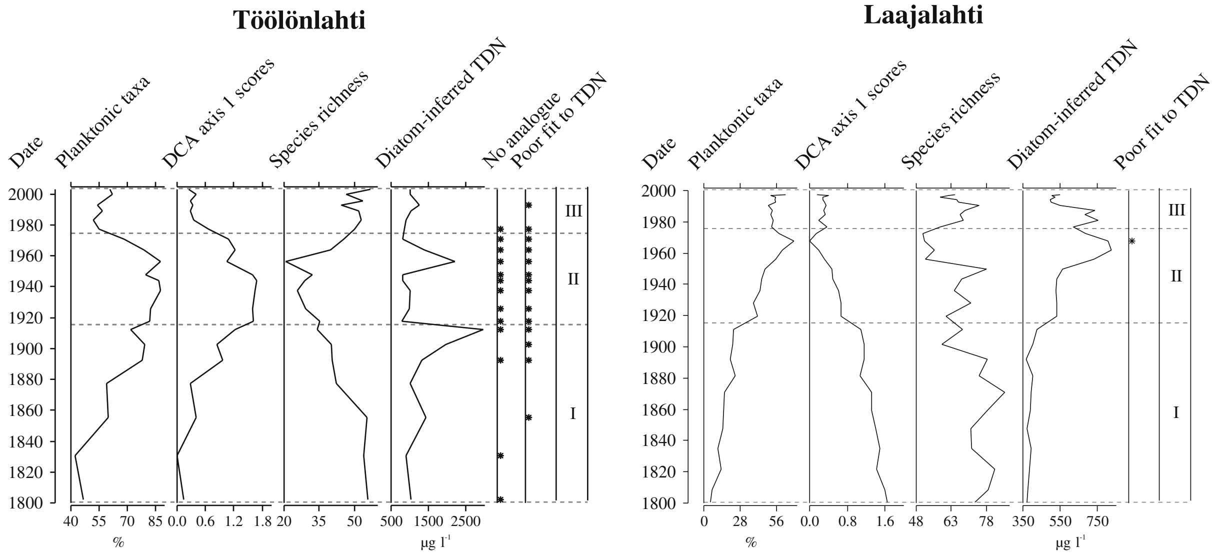

Figure 3. Summary diagram of the urban sites showing the main trends in (1) the abundance of planktonic diatoms, (2) diatom assemblage composition (DCA axis 1), (3) diatom species richness, and (4) diatom-inferred total dissolved nitrogen (TDN). Samples with poor fit to TDN and those lacking good modern analogues are indicated (*). Zonation based on the species data is shown as dashed lines.

Planothidium delicatulum agg., *Navicula gregaria* Donkin 1861 and *Fragilaria elliptica* agg. declined most likely in response to increased turbidity of the site caused by an increase in planktonic productivity due to nutrient enrichment (e.g., Cooper and Brush 1991; Andrén et al. 1999).

The changes observed in the diatom assemblages of Zone 1 can be attributed to the increasing urbanisation and population density of Helsinki and the start of point-source loading to the embayment by a sugar mill in 1823 and by the first enclosed sewage pipe in 1878 (Enkvist 1974).

In Zone II (ca. 1915–1975) planktonic diatoms dominated the diatom assemblages composing 80–90% of all taxa, which is mirrored in the DCA axis 1 scores (Figure 3). Species richness continued to decrease and reached the lowest value of 21 taxa in the mid 1950s. The number of planktonic species started to decrease towards the end of the zone concurrent with an increase in species richness. Two waste water treatment plants were installed in the catchment area of Töölönlahti in 1910 and 1915, respectively. These primitive treatment plants removed only organic matter from the sewage to reduce its biological oxygen demand, while serving a rapidly expanding population until their closure in 1935 and 1959, respectively. The sugar mill was closed in 1965, but its waste waters had been diverted elsewhere at an earlier date (Enkvist 1974).

Small, weakly silicified *Thalassiosira* taxa, especially *Thalassiosira guillardii* and *Thalassiosira pseudonana* Hasle & Heimdal 1970 dominated the diatom assemblages in Zone II (Figure 2). Prolonged nutrient enrichment of a water body causes dissolved silicate limitation (Conley et al. 1993). Under such conditions taxa with low silica requirements, such as the small *Thalassiosira* taxa in this study, will be strong competitors. *Cyclotella meneghiniana*, however, which is bigger and more heavily silicified, declined in abundance throughout Zone II. Also *Cyclotella atomus* declined during the first half of the zone, but increased again towards the end. *Skeletonema costatum* and *Chaetoceros* spp. exhibited their highest abundances in Zone II. These fragile taxa are often not well preserved in coastal sediments (MOLTEN 2004). Their increased abundance in Zone II may be a real response to higher nutrient concentrations or it may be a preservational artifact caused by increasing anoxia (and decreased bioturbation) of the sediment.

The abundance of planktonic diatoms clearly decreased to ca. 60% by Zone III (1975–2003). It did not, however, return to levels similar to those in the early 1800s, unlike species richness, which showed a recovery back to pre-1850s levels after the most pronounced eutrophication period of Töölönlahti in Zone II (Figure 3). The relative abundance of especially *Thalassiosira guillardii* decreased significantly from ca. 50% to less than 10% by Zone III. Also the abundances of *Cyclotella meneghiniana, Thalassiosira pseudonana, Skeletonema costatum* and *Chaetoceros* spp. decreased clearly by the mid 1970s (Figure 2). *Cyclotella choctawhatcheeana* Prasad 1990 and *Thalassiosira proschkinae* Makarova 1979, being almost absent in Zone II increased again in abundance during Zone III. Although both taxa have been associated with increasing nutrient enrichment (e.g., Cooper 1995; Andrén et al. 1999; Cooper et al. 2004), they do not seem to thrive well under hypertrophic conditions. Benthic *Achnanthes* Bory 1822 (incl. revised genera), *Navicula* Bory 1822 and *Fragilaria* Lyngbye 1819 (incl. revised genera) species, which had declined in abundance during Zone II, appeared again in Zone III.

Laajalahti

The diatom stratigraphy of Laajalahti consisted of 226 taxa. Most of these were benthic, only 14 planktonic taxa were observed. The assemblages of the core were divided into 3 statistically significant zones (Figure 2). Zone I (ca. 1800–1915) was dominated by benthic diatom assemblages. Species richness was greatest before the start of the 20th century being on average 76 taxa/sample (Figure 3). The diatom assemblages of Zone I were characterised by small *Fragilaria* spp., but also included many other benthic taxa such as *Mastogloia smithii* Thwaites 1856, *Rhoicospenia curvata* (Kützing) Grunow 1860 and *Planothidium delicatulum* agg. (Figure 2). There was an increase in *Cyclotella choctawhatcheeana*, a species that seems to indicate anthropogenic disturbance (e.g., Cooper 1995), in the mid 1800s. The population of Helsinki grew from 20,000 to 120,000 inhabitants during 1850–1910, but the wastewater treatment capacity of the city was minimal (Laakkonen and Lehtonen 1999). As a result the coastal area around Helsinki was already affected by municipal nutrient load (Finni et al. 2001).

The largest change in the core occurred at the beginning of Zone II (ca. 1915–1975) corresponding to the early 1920s, when the proportion of planktonic diatoms doubled to >40% (Figure 3) coinciding with a rapid expansion of the urban area (Laakkonen and Lehtonen 1999). At the start of this phase the dominant *Fragilaria* spp. declined and many small planktonic diatoms, such as *Thalassiosira* spp., *Cyclotella atomus* and *Cyclotella meneghiniana* increased clearly in abundance (Figure 2). The decline in benthic *Fragilaria* was most likely a response to increased nutrient enrichment (increased turbidity) of the site. There was a further increase in planktonic diatoms in the mid 1950s reciprocal with a marked decrease in species richness to 51 taxa/sample. This event was characterized by an increase in the common eutrophic taxa *Cyclotella atomus* and *C. meneghiniana* (EDDI 2001; MOLTEN 2004). Two waste water treatment plants were installed in the watershed of Laajalahti at the time; however, these treatment plants only removed organic matter from sewage (Weckström et al. 2004). A clear response to improved waste water purification in the early 1970s was seen as increased species richness and decreased abundance of planktonic taxa.

There was a further marked reduction in the nutrient load to the embayment in Zone III (ca. 1975–1998), when the last treatment plant was closed in 1986. In response, there was a decline in the eutrophic *Cyclotella meneghiniana* and *C. atomus* and a rise in the less eutrophic taxon *C. choctawhatcheeana* (Figure 2). Although a clear recovery of the system could be observed after the cessation of waste water loading, there is still a marked difference between the diatom assemblages of Zone I and the present. Even though there has been an increase in species richness to ca. 66 taxa/sample and a species shift within the planktonic taxa after the most pronounced eutrophication period (Figure 3), there still is no sign of a recovery back to the more diverse benthic assemblages characterised by *Fragilaria* species.

Pieni Pernajanlahti

The diatom stratigraphy of Pieni Pernajanlahti consisted of 252 taxa. The majority of these were benthic, only 28 planktonic taxa were observed. The diatom assemblages of Zone I (ca. 1880–1942) were dominated by benthic species such as *Planothidium delicatulum* agg., *Navicula gregaria*, many *Fragilaria* taxa and *Diatoma moniliformis* Kützing 1833 (Figure 4). The only notable change in Zone

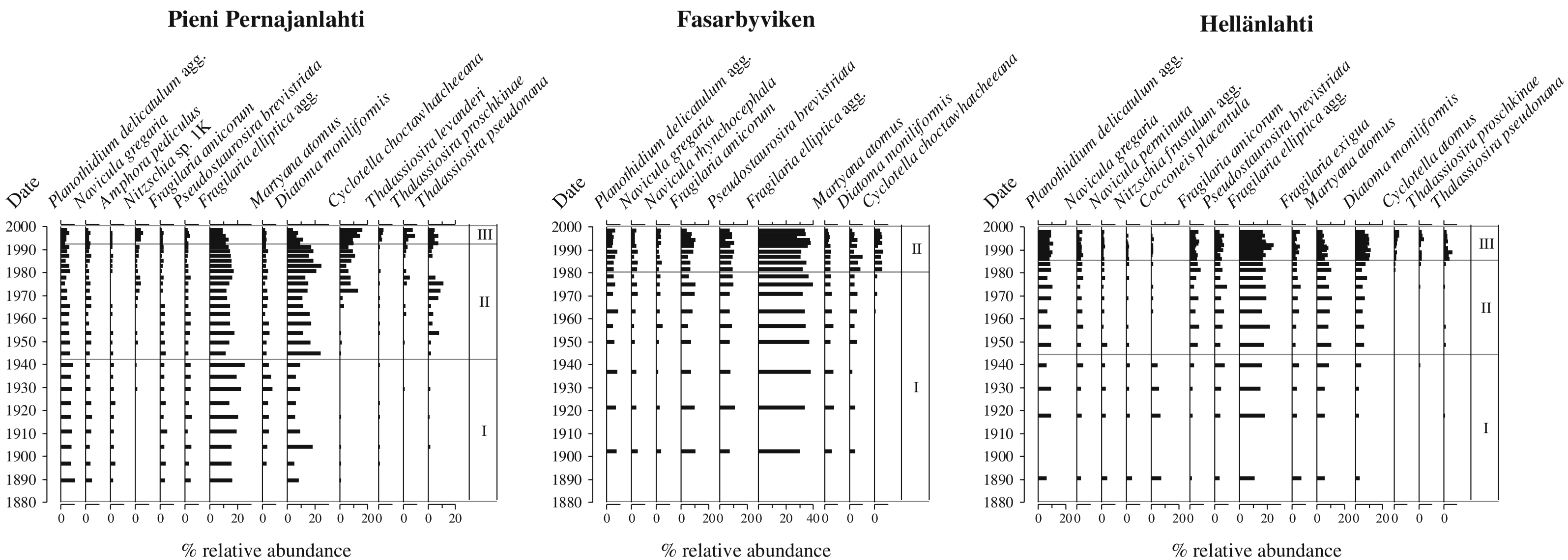

Figure 4. Stratigraphic diagrams of the rural sites showing the main diatom taxa (percentage abundance). Zonation of the species data was performed using constrained optimal sum of squares partitioning. All zones were statistically significant except at Hellänlahti, where the boundary between zones I and II is shown only to aid interpretation. *Planothidium delicatulum* agg. and *Fragilaria elliptica* agg. include taxa as in Figure 2. *Nitzschia frustulum* agg. includes *N. frustulum* (Kützing) Grunow in Cleve & Grunow 1880, *N. inconspicua* Grunow 1862 and *N. liebetruthii* Rabenhorst.

I occurred in the early 1920s, when species richness of the diatom assemblages decreased from ca. 79 to 69 taxa/sample (Figure 5).

At the beginning of Zone II (ca. 1942–1992), the number of planktonic diatoms, especially the eutrophic taxon *Thalassiosira pseudonana* (MOLTEN 2004), started to increase (Figures 4 and 5). New fields were cleared in the catchment area of Pieni Pernajanlahti in the 1920s and 1940s (Linnasalo and Penttilä 2003), which could have increased erosion resulting in increased turbidity and higher nutrient concentrations of the embayment thus favouring planktonic diatoms over benthic forms. A further increase in planktonic taxa occurred in the late 1960s reciprocal with a decrease in species richness. *Thalassiosira proschkinae* and especially *Cyclotella choctawhatcheeana* increased in abundance at this time. River Ilolanjoki, which drains into Pieni Pernajanlahti, was dredged several times for purposes of flood protection in the 1950s and 1960s (Tamminen and Kukkamäki 1985; Villström 2002) with possible effects on turbidity of the site. The abundance of planktonic diatoms continued to increase in Zone III (ca. 1992–1998), which is mirrored in the decrease in DCA axis 1 scores. Species richness fluctuated substantially throughout Zone II, but a general decrease could be detected, which continued in Zone III (Figure 5).

Fasarbyviken

The diatom stratigraphy of Fasarbyviken consisted of 188 taxa; only 17 of these were planktonic. The assemblages of the core were divided into 2 statistically significant zones (Figure 4). Zone I (ca. 1880–1980) was dominated by benthic diatoms, especially *Fragilaria* spp., which constituted between 50–62% of all taxa. Species richness fluctuated somewhat during this zone, but was generally lower than in the other two rural sites due to the dominance of *Fragilaria* taxa. The only clear change in the species assemblages of Fasarbyviken occurred ca. 1980, when the abundance of planktonic taxa increased from an average of ca. 2% during Zone I to an average of ca. 7% during Zone II (ca. 1980–1998), which was mostly caused by an increase in *Cyclotella choctawhatcheeana* (Figures 4 and 5). This is mirrored in the DCA axis 1 scores, which summarise overall changes in the diatom assemblages. A small waste water treatment plant was installed in the catchment area in 1973, but its contribution to the total nutrient load has been estimated to be only ca. 10% (L. Villa, pers. commun.). It is more likely that this moderate increase in the relative abundance of planktonic taxa was due to e.g. intensified agricultural practices in the catchment area as diffuse loading is the main external nutrient source to the embayment.

Hellänlahti

The diatom stratigraphy of Hellänlahti consisted of 276 taxa. Most of these were benthic; only 23 planktonic taxa were observed. The assemblages of the core were divided into 3 zones, however, only the boundary between Zone I and Zone II was statistically significant (Figure 4). Zone I (ca. 1880–1945) was dominated by diverse benthic assemblages including e.g., *Planothidium delicatulum* agg., *Navicula gregaria, Cocconeis placentula* Ehrenberg 1838, *Diatoma moniliformis* and several *Fragilaria* spp. The percentage abundance of *Cocconeis placentula* declined towards the end of the zone, while abundances of *Fragilaria* spp. increased.

The first marked change in the Hellänlahti core occurred at the beginning of Zone II (ca. 1945–1985) when *Cocconeis placentula* almost disappeared from core assemblages, which is seen as a decrease in DCA axis 1 scores (Figure 4). As *C. placentula* is a strict epiphyte, this would suggest a decrease in the macrophyte abundance of the embayment. River Virojoki, which drains into Hellänlahti was repeatedly dredged during the 1930s–1960s (Seppälä and Huttunen 2002). This may have increased the turbidity of the embayment and led to a decline in benthic macrophytes such as stonewort and the water-milfoil and pondweed families due to light limitation. Species richness fluctuated notably during Zone II, but no major shifts in species assemblages could be observed after the beginning of this zone (Figure 5).

Another distinct change in the Hellänlahti core occurred at the beginning of Zone III (ca. 1985–1998) when the relative abundance of planktonic diatoms increased to $>10\%$ together with a moderate decrease in species richness (Figure 5). Planktonic taxa that increased most included *Cyclotella atomus, Thalassiosira proschkinae* and *Thalassiosira pseudonana*. According to monitoring data provided by the Finnish Environment Institute, the nutrient load of River Virojoki

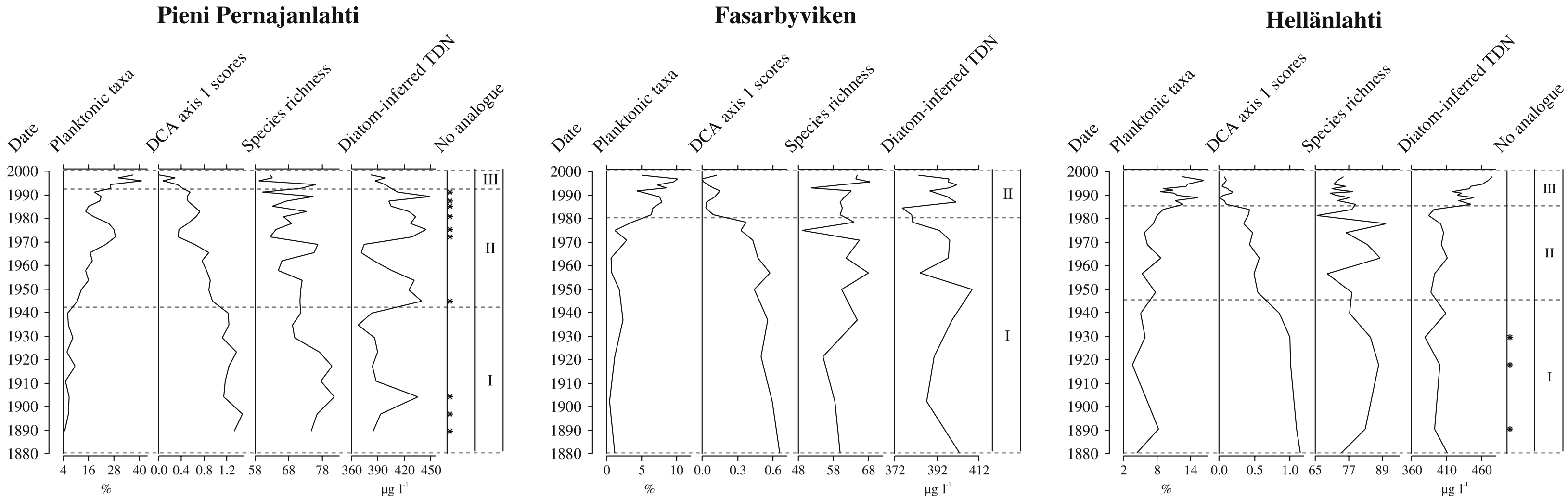

Figure 5. Summary diagram of the rural sites showing the main trends in (1) the abundance of planktonic diatoms, (2) diatom assemblage composition (DCA axis 1), (3) diatom species richness, and (4) diatom-inferred total dissolved nitrogen (TDN). Samples with poor fit to TDN and those lacking good modern analogues are indicated (*). Zonation based on the species data is shown as dashed lines.

increased in the late 1970s-early 1980s (data not shown), which together with the onset of fish farming in the larger estuarine complex of Virolahti may have caused this most recent change in the species assemblages of Hellänlahti.

Diatom-inferred TDN reconstructions

Urban sites

At Töölönlahti and Laajalahti, the diatom-inferred TDN concentrations showed a clear response to urbanization and waste water loading (Figure 3): There was an increase of DI-TDN in both cores coinciding with increasing urbanization of the catchment areas (Laakkonen and Lehtonen 1999). The inferred TDN concentration in the lower part of the core was substantially higher in Töölönlahti (on average ca. 1100 $\mu g\ l^{-1}$) compared to ca. 400 $\mu g\ l^{-1}$ in Laajalahti, and the increase occurred somewhat earlier, most likely due to the more central location of the embayment in the late 1800s. Highest DI-TDN concentrations peaking to 2948 $\mu g\ l^{-1}$ in Töölönlahti and 826 $\mu g\ l^{-1}$ in Laajalahti were observed during the period of waste water loading to these embayments. Also, a clear decrease in DI-TDN was seen in both cores after the closure of the waste water treatment plants. There was an unexpected decrease in DI-TDN concentrations in Töölönlahti between ca. 1920–1950, when diatom assemblage structure (high relative abundance of planktonic diatoms, low species richness) and known land use changes in the catchment area would have suggested the opposite. This decrease was mainly due to a marked decline in the relative abundance of *Cyclotella atomus*, which has the highest TDN optimum in the calibration data set and the dominance of *Thalassiosira guillardii*, which, although a eutrophic species, has a clearly lower TDN optimum (Weckström, unpublished data). This highlights the importance of community structure indices (e.g., abundance of planktonic diatoms, diversity) and changes in the abundance of individual species when interpreting diatom-inferred nutrient reconstructions. The observed shift between the two taxa may have been caused by silica limitation of the system with increasing nutrient concentrations, more intensive diatom blooms and resulting depletion of silica from the water column (Conley et al. 1993).

The two urban sites have a measured total nitrogen record of ca. 25 years (provided by the Finnish Environment Institute), which enabled the validation of the reconstructions for the upper part of the cores. The DI-TDN concentrations were compared with the annual, seasonally weighted mean TN concentrations. When comparing the records, it should be noted that in the study area TDN composes only ca. 60–70% of TN (Weckström et al. 2004). The main trends in the measured TN of Laajalahti were recorded well by the diatom-inferred TDN (Figures 3 and 6). In particular, the steep decline in the mid 1970s, the maintained high levels in the 1980s and the subsequent decline to present concentrations in the late 1980s were all tracked well by the model. There was also a good agreement between the measured and diatom-inferred values throughout the 1990s (mean TN and TDN, respectively: 830 $\mu g\ l^{-1}$ and 540 $\mu g\ l^{-1}$). However, the actual TN concentrations were systematically underestimated by the transfer model during the most pronounced eutrophication period. At present, the modern calibration data set lacks sites above ca. 2000 $\mu g\ l^{-1}$, which would parallel the most eutrophic phase of Laajalahti. In such conditions WA-PLS underestimates the true TDN. The inclusion of more sites at the high end of the TDN

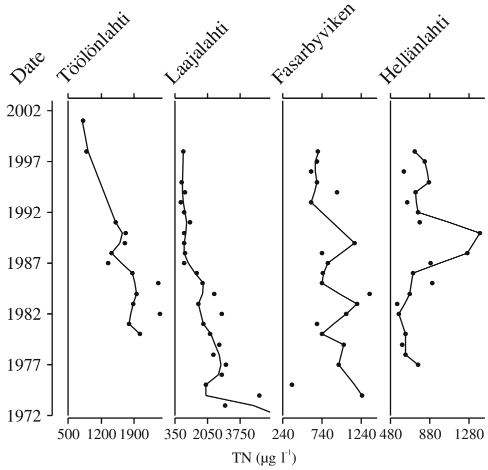

Figure 6. Monitoring data for total nitrogen (TN) at four of the study sites covering ca. 25 years. The measured TN concentrations are annual, seasonally weighted means (shown as circles). A LOWESS scatter plot smoother (span = 0.20, iterations = 4) was fitted to help detect major trends.

gradient should increase the accuracy of the model over this part of the gradient. This may, however, be difficult as the nutrient concentrations of similar coastal sites are now markedly lower than concentrations existing in these urban embayments during their past. In Töölönlahti, measured TN and diatom-inferred TDN concentrations were both relatively uniform throughout the 1980s (Figures 3 and 6). The peak in the mid 1990s in DI-TDN could not be confirmed by measured TN, as there was no monitoring data available between 1991 and 1998. The measured TN and DI-TDN concentrations (ca. 1700 and 1060 $\mu g\ l^{-1}$ on average) agreed reasonably well, except for the very top (1998–) part of the core, where concentrations were somewhat overestimated by the model. Generally, the measured record gives confidence to the DI-TDN reconstruction of Töölönlahti for samples pre-1980.

A number of samples in the Töölönlahti core appeared to have no good analogues in the modern calibration data set, especially during the most pronounced eutrophication period from ca. 1880 to ca. 1975 (Figure 3). This lack of modern analogues results, in part, from the high abundance of *Thalassiosira guillardii*, a taxon that only occurred at low abundances in the calibration data set, and from the rarity of analogous, extremely eutrophied sites in the calibration set (Weckström et al. 2004). Many of these samples also had a poor fit to TDN. In addition to the unexpected drop in DI-TDN during Zone II, these measures of assessing the reliability of diatom-inferred reconstructions suggest that the TDN reconstruction for Töölönlahti should be interpreted with some caution.

The other urban site Laajalahti, however, showed good analogues with the modern calibration data set throughout the core with only one sample having a poor fit to TDN (Figure 3). This is somewhat surprising, as the monitoring record from Laajalahti indicates concentrations as high as ca. 5000 $\mu g\ l^{-1}$, whereas the calibration data set only includes sites $< 2000\ \mu g\ l^{-1}$. This would suggest that the diatom assemblages of Laajalahti have not responded to these anomalously high nutrient concentrations occurring in the embayment in the 1960s and 1970s, which is supported by the clearly underestimated TDN-reconstruction.

Rural sites

In Pieni Pernajanlahti, diatom-inferred TDN fluctuated between ca. 370 $\mu g\ l^{-1}$ and ca. 450 $\mu g\ l^{-1}$ (Figure 5). There was a clear increase in DI-TDN at the beginning of the 1940s coinciding with the initial increase in the abundance of planktonic diatoms. Another marked increase in DI-TDN occurred at the beginning of the 1970s after which the concentrations declined towards the surface. Taking into account the dating uncertainties in the top part of the core (± 3 years on average), the DI-TDN appears to be to some extent correlated with the mean flow of the River Ilolanjoki ($r = 0.48$) (data, which is not shown here, was provided by the Uusimaa Regional Environment Centre from the 1970s onwards). As River Ilolanjoki is the main source of external loading to Pieni Pernajanlahti, changes in the mean flow would affect the nutrient concentrations of the embayment. The reconstructed TDN for Fasarbyviken showed little variation (380 $\mu g\ l^{-1}$ to 410 $\mu g\ l^{-1}$) throughout the core, whereas at Hellänlahti there was a distinct increase in the mid 1980s after relatively stable concentrations around 400 $\mu g\ l^{-1}$ (Figure 5). This increase at Hellänlahti coincided with an increase in the abundance of planktonic taxa. The DI-TDN concentrations at the rural sites were considerably lower than at the urban sites and no large increases comparable to the urban sites were observed.

All rural cores were dominated by benthic species, which could affect the performance of the DI-TDN model, as the distribution of benthic diatoms is likely to be related to factors such as water depth, light availability and variations in substrate rather than to open water nutrients directly (e.g., Bennion 1995; Michelutti et al. 2003). Benthic taxa may also have access to enhanced nutrient levels at the sediment-water interface while epiphytic taxa may derive nutrients from their host plants. The TDN optima of taxa in the model are, however, based on measured surface water TDN concentrations. Nevertheless, studies from the Baltic Sea show that nutrient enrichment of shallow coastal areas affected benthic diatom assemblages by increasing production and changing the assemblage composition (e.g., Sundbäck and Snoeijs 1991).

Both Fasarbyviken and Hellänlahti have monitoring records of TN spanning ca. 25 years

(Finnish Environment Institute), which provided the opportunity to validate the DI-TDN reconstructions. This was not possible for Pieni Pernajanlahti, which has not been monitored regularly. The TN values at Fasarbyviken were moderately underestimated by the model, which inferred relatively uniform TDN concentrations around ca. 400 $\mu g\ l^{-1}$ compared to the average measured TN values of 850 $\mu g\ l^{-1}$ (Figures 5 and 6). The monitoring data showed no clear trends, but displayed peaks of high ($>1000\ \mu g\ l^{-1}$) TN concentrations, which were not recorded in the DI-TDN. These peaks were, however, only short-lived (1–3 years), and may have been smoothed out in the diatom-reconstruction. The explanation for the underestimated N values inferred by the model may be the dominance of benthic *Fragilaria* taxa throughout the core. These species have wide ecological tolerances, which make them poor indicator taxa, and their optima for TDN lie close to the centre of the sampled TDN gradient (mean ca. 420 $\mu g\ l^{-1}$/ median ca. 350 $\mu g\ l^{-1}$) (Weckström and Juggins, unpublished data). A similar situation has been described in Bennion et al. (2001) using the northwest European calibration data set for lake TP reconstructions. At Hellänlahti, the measured and diatom-inferred values compare reasonably well both before and after the increased concentrations in the mid 1980s (ca. 630 $\mu g\ l^{-1}$ and ca. 400 $\mu g\ l^{-1}$ before; ca. 790 $\mu g\ l^{-1}$ and ca. 440 $\mu g\ l^{-1}$ after). Bearing in mind that TDN composes only approximately 60–70% of TN in the study area (Weckström et al. 2004), the values are relatively similar. There was a peak of high ($>1000\ \mu g\ l^{-1}$) TN concentrations in the late 1980s, which was not recorded in the DI-TDN. This peak, however, only lasted for 2 years, and may have been smoothed out in the diatom-inferred TDN reconstruction as in the case of Fasarbyviken. At Pieni Pernajanlahti the DI-TDN value for the surface sample (387 $\mu g\ l^{-1}$) was in good agreement with the measured annual mean TDN value (398 $\mu g\ l^{-1}$), which adds confidence in the reconstruction of the site.

In general, the fossil samples of the rural sites had good modern analogues in the calibration data set. However, at Hellänlahti, the three oldest samples had poor analogues due to a relatively high percentage of *Cocconeis placentula* (up to 13%) compared to the calibration set average of only 0.6%. Also at Pieni Pernajanlahti, there were some fossil samples at the bottom and again towards the top of the core which had poor analogues with the modern data set. All these samples had a high percentage of *Diatoma moniliformis* Kützing 1833 (up to 24%) compared to the calibration data set average of 3%.

Trends in eutrophication

The two urban sites exhibited the first clear signs of nutrient enrichment much earlier than the three rural sites. The first changes in Laajalahti were observed in the 1920s, while marked changes occurred as early as the 1800s in Töölönlahti owing to the more central location of this embayment. All observed changes (abundance of planktonic diatoms, species richness and DI-TDN) were also much greater in magnitude at the urban sites compared to the rural sites. Municipal waste water treatment techniques in Finland began to improve in the 1970s, which enhanced the water quality of coastal areas (see Pitkänen et al. 1987). This is evident at the urban study sites, where a clear improvement was indicated by decreased DI-TDN, a decline in the abundance of planktonic diatoms and increased diversity.

The first change in the rural sites was observed in the mid 1940s and a later one in the 1980s–early 1990s. A defining feature of these changes was an increase in small planktonic taxa, although this was not as marked as at the urban sites, where abundances of planktonic species reached ca. 70 and 90%, respectively. There was a general moderate decrease in species richness at Hellänlahti and Pieni Pernajanlahti, whereas no trend was seen at Fasarbyviken. No clear trends could be observed in the DI-TDN concentrations, except at Hellänlahti, where an increase of ca. 50 $\mu g\ l^{-1}$ occurred in the mid 1980s (Figure 5).

Taxa that increased in abundance with nutrient enrichment included *Cyclotella choctawhatcheeana, Thalassiosira proschkinae* and *T. pseudonana*. Only the hypertrophic urban sites showed high abundances of the eutrophic taxa *Cyclotella atomus, C. meneghiniana* and *Thalassiosira guillardii* (EDDI 2001; Bradshaw et al. 2002). It may be that at low salinities species with a freshwater affinity such as *C. atomus* and *C. meneghiniana* are stronger competitors at very high nutrient levels than e.g. *Cyclotella choctawhatcheeana*, which has a higher optimum for salinity (Weckström and Juggins, unpublished data).

Both urban sites showed trends of anthropogenic disturbance and a subsequent recovery after the cessation of waste water loading. The recovery of these sites is, however, not complete, despite the low external nutrient load at present. Although the chemical recovery of the sites is evident (as indicated by DI-TDN and the long-term monitoring record of TN), there is a marked difference at Laajalahti between the diatom assemblages of Zone 1 and the present. Current assemblages are still heavily dominated by planktonic taxa with lower species richness compared to the diverse benthic assemblages of Zone I. In Töölönlahti, the difference between diatom assemblages of Zone I and the present is not as distinct due to the fact that the site was already affected by urban development around the time corresponding to the base of the core. However, the abundance of planktonic diatoms is clearly higher now compared to that of the beginning of the 1800s. The recovery of biological communities is known to be complex and influenced by many internal and external factors (e.g., Niemi et al. 1990). At present, the planktonic assemblages at both sites are maintained by internal loading from the sediments (Tikkanen et al. 1997; Kauppila et al. 2005) as the embayments continue to be nutrient enriched despite decreased external loading, and by high turbidity, which has resulted in a loss of submerged macrophytes (Häyrén 1921) and affects benthic diatom communities adversely. This phytoplankton-dominated state may be preserved despite nutrient reductions due to the inability of aquatic plants to recolonise in highly turbid waters. These results have implications for the European Water Framework Directive, which requires European surface waters to be of good ecological status, defined both by biological and chemical quality elements (Anonymous 2000).

The small floristic changes observed in the rural sites of this study could signal that an ecologically important threshold has been crossed even if there is no change in the inferred nutrient concentrations (e.g., Jones et al. 1997; Bennion et al. 2004; Weckström and Juggins, unpublished data). Although diffuse nutrient loading from agriculture is the most important single anthropogenic source of nutrients to Finnish coastal waters (Pitkänen 1994), the effects are likely to be moderate in this study, as the catchment areas of the rural sites are mostly forested with agriculture comprising < ca. 30% of the catchment. More pronounced impacts on coastal waters due to agricultural loading could be expected in SW-Finland, which is the main agricultural area in Finland due to fertile clay-rich soils (Suomen kartasto 1990). Several sites in the area are currently analysed as part of a national research programme, BIREME, funded by the Finnish Academy. The relatively late timing of the first changes in the diatom assemblages of the rural sites coincided with the intensification of agriculture in Finland after the Second World War. New fields were cleared and the use of artificial fertilisers increased dramatically at this time (IFA 1996–2005).

At four of the sites, significant changes in diatom assemblages and associated DI-TDN occurred after the 1920s, hence background or reference conditions could be set at early 1900s. In the case of Töölönlahti, a longer sediment core would have to be taken in order to establish pre-urbanisation conditions at the site. However, at Töölönlahti this would mean recreating conditions that existed in the embayment over 200 years ago. At that time, however, the embayment did not exist in its present form due to post-glacial land uplift and urban development, but was a more open system affected by the open sea (Korhola and Blom 1996). Also Laajalahti was a more open system at this time, which was clearly reflected in the diatom assemblage structure pre-1800 (Kauppila et al. 2005). Facing challenges of this kind, the WFD acknowledges that it is unrealistic to base reference conditions upon historic landscapes that no longer exist in modern Europe.

The reference conditions for the four sites are defined by diverse benthic diatom assemblages ($>80\%$ benthic taxa) that are characterised by small *Fragilaria* spp. The diatom-inferred TDN, however, is surprisingly high (ca. 400 $\mu g\ l^{-1}$ TDN, which corresponds with ca. 600 $\mu g\ l^{-1}$ TN in the study area). These lowland catchment areas have been cultivated at least since the Bronze Age, although population densities were low and slash-and-burn cultivation was practised along with field cultivation until the 19th century on the southern coast (Soininen 1974). Hence these higher than expected TDN concentrations could be a consequence of longer term anthropogenic influence (e.g., Bradshaw 2001). Natural leaching is also higher in catchment areas with clay-rich soils as in this study (Suomen kartasto 1990) compared to catchments dominated by tills. The established reference conditions seem to, however, represent relatively stable systems before recent, post-industrial anthropogenic eutrophication. Another reason for the higher reference TDN concentrations could be the dominance of *Fragilaria* spp., as discussed earlier in this paper. High occurrences of *Fragilaria* spp. appear to be associated with environmental instability (e.g., Haworth 1976; Denys 1990). It is likely that these opportunistic, fast reproducing species respond to factors such as increased turbidity rather than directly to higher nutrient supply. Several embayments with small rocky catchments in the calibration data set showed background TDN concentrations between 250 and 300 $\mu g\ l^{-1}$. The benthic assemblages of these embayments were not dominated by *Fragilaria* spp., but included taxa such as *Amphora pediculus* (Kützing) Grunow in A. Schmidt et al. 1875, *Cocconeis neothumensis* Krammer 1990, *C. placentula, Epithemia sorex* Kützing 1844, *Mastogloia smithii, Navicula gregaria, Opephora mutabilis* (Grunow) Sabbe & Vyverman 1995, *Rhoicosphenia curvata* and *Tabularia fasciculata agg*. (incl. *T. fasciculata* (C.A. Agardh) Williams & Round 1986, *T. tabulata* (C.A. Agardh) Snoeijs 1992 and *T.* cf. *laevis* Kützing) at 1–10% abundances (top-bottom sampling; Weckström, unpublished data). Thus, the dominance of *Fragilaria* spp. in the late 1800s–early 1900s may be a first indication of anthropogenic disturbance.

Palaeolimnological techniques such as those employed in this study can help to assess ecological changes in coastal waters over centennial time scales and provide information on background conditions. They are also a valuable means for detecting early community changes, where water quality problems are not yet evident and hence provide a management tool for ecosystem protection in addition to ecosystem restoration. The used diatom indices (DI-TDN, species richness, and % planktonic diatoms) employed here proved to be good indicators of changes in the trophic status when validated against the existing monitoring data and known land use changes. This study suggest that these diatom indices could be used as biological quality elements in the WFD for defining trophic reference conditions as well as for current water quality monitoring in coastal waters of the Baltic Sea area.

Acknowledgements

The author would like to thank F. Adser and S. Vaalgamaa for assistance with collecting the sediment cores and P. Appleby for the dating. The manuscript has benefited from discussions with A. Korhola and J. Weckström. This research was supported by the Finnish Ministry of Education, the EC Energy, Environment and Sustainable Development Programme (contract EVK3-CT-2000–00031, MOLTEN) and the Academy of Finland (contract 202436, DETECT).

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