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Tracing pollution and recovery using sediments in an urban estuary, northern Baltic Sea: are we far from ecological reference conditions?

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ABSTRACT: One of the primary challenges of the Water Framework Directive (WFD) of the European Union is to provide a guide for the recovery of surface waters from pollution. However, few studies deal with reference conditions according to the WFD in coastal waters. Using the urbanised Laajalahti Bay (Helsinki, Finland) as an example, reference conditions and pollution history were defined using the stratigraphy of diatoms, sediment geochemistry, stable isotopes, sedimentary pigments, long-term monitoring results of water quality, and nutrient-loading. Principal components analysis was used to generate a multidimensional index of water quality on the basis of the sediment data. We distinguished 5 phases: (1) a pre-industrial phase (before ca. 1815); (2) a phase of slight human disturbance (ca. 1815 to 1900); (3) an onset of anthropogenic impact (ca. 1900 to 1955); (4) a severe pollution phase (ca. 1955 to 1975); (5) basin recovery and a phase of internal loading (from ca. 1975 onwards). Phase 2 was used to define reference conditions. Phase 1 was not used as it occurred before the formation of the semi-enclosed bay by post-glacial isostatic land-uplift. There was good agreement between the sedimentary record and the water-quality data during Phase 4. Despite an improvement in water quality after the local municipal treatment plant closed in 1986, Laajalahti Bay is still far from reference conditions due to internal loading.

KEY WORDS: Pollution · Recovery · Reference conditions · Palaeolimnological methods · Urban estuary · Baltic Sea

INTRODUCTION

Cultural eutrophication and other anthropogenic sources of pollution are major concerns in the Baltic Sea (Cederwall & Elmgren 1990, HELCOM 2002) and other marine areas (Vollenweider et al. 1992, Nixon 1995). Pollution of coastal waters results in changes in community structure and abundance of biota (Cederwall & Elmgren 1990, Bondsdorff et al. 1997, Grall & Chauvaud 2002), anoxia and hypoxia of the sea bottom (Jørgensen et al. 1990, Anderson et al. 2002, Gray et al. 2002), and the accumulation of harmful substances in the sediment, water column and biota (Burton & Statham 1990, Fowler 1990, Luoma 1990, Olsson et al. 2002, Poutanen et al. 2002, Schneider et al. 2002). Information on cultural eutrophication and pollution is, in many locations, preserved in the sediment record. Management decisions to counter eutrophication and pollution could be better informed with the use of baseline reference conditions for these areas. These can be obtained from the sediment record using well-established palaeolimnological techniques.

The European Union Directive 6000/60/EC, applying the status-assessment approach of the US Environmental Protection Agency (EPA), established a framework for community action in the field of water policy

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in 2000. The overall objective of the directive (WFD) is to maintain and improve the ecological quality of surface waters and, ultimately, to achieve good environmental quality by controlling the pollution sources that impact them. The implementation of the WFD requires the establishment of reference conditions, or baselines, against which the changes can be measured. This is hampered by the lack of monitoring data in many areas. Thus, more appropriate palaeolimnological methods, which provide such historical insights, are needed.

Historical data and models have been used to define reference conditions for the ecological indicators of the WFD in coastal settings, but they have their limitations (see e.g. Nielsen et al. 2003). Palaeolimnological techniques offer yet another potent tool. Sediment archives have been successfully used in assessing past anthropogenic impacts and cultural eutrophication in freshwater ecosystems (Bennion et al. 1996, Rippey & Anderson 1996), but their application to coastal systems is still limited (see, however, Cooper 1993, 1999, Sakson & Miller 1993, Vos & De Wolf 1993, Tikkanen et al. 1997, Nees et al. 1999, Sullivan 1999). In particular, palaeolimnological techniques allowing nutrient concentrations to be inferred quantitatively from the remains of organisms preserved in the sediment wait to be tested in coastal environments. Diatoms and algal pigments are known to be especially sensitive indicators of trophic conditions (Battarbee 1991, Korhola & Blom 1996, Andren et al. 1999, Leavitt & Hodgson 2001). Additionally, effects of sewage, atmospheric deposition and land-use can be seen in the sediment chemistry of nutrients (Rippey & Anderson 1996) and heavy metals (Blomqvist et al.1992). The chronology of these changes is usually derived from the record of ^{210}Pb, a natural radioactive isotope of lead (Appleby et al. 1986, Appleby 2001).

Water-quality data indicates that the semi-enclosed Laajalahti Bay on the west coast of Helsinki, Baltic Sea, is a good example of a coastal site recovering from pollution after the cessation of sewage-loading in the mid-1980s (Varmo et al. 1989, Pesonen et al. 1995, Lappalainen & Pesonen 2000). At present, the estuary receives only little external loading, but is still affected by release of phosphorus from the sediment (internal loading), both in anaerobic and aerobic conditions (Rekolainen 1982). According to the classification scheme by the Finnish Environment Institute for Finnish coastal waters, the water-quality status of the estuary is still only 'moderate' (Antikainen et al. 2000, Autio et al. 2003) due to internal loading. Because of the WFD, it is important to determine how far the estuary is from natural reference conditions, and how the estuary has been affected by human disturbance.

This study aimed at establishing and quantifying long-term historical trends in biological and chemical parameters of the water column using the sediment record and to confirm the recovery of the estuary from pollution. In addition, we have established ecological reference conditions for the estuary according to requirements of the WFD. We apply a multi-proxy approach to trace recovery of Laajalahti Bay from pollution by connecting the stratigraphy of diatoms, sediment geochemistry, stable isotopes and sedimentary pigments with long-term monitoring results of water quality and loading. Some of the diatom and sediment geochemistry data has been previously published or is being published separately (Vaalgamaa 2004, Weckström et al. 2004). Here we present a synthesis of the overall results including re-analyses of published data and a substantial amount of new data.

MATERIALS AND METHODS

Study area. Laajalahti Bay (surface area 5.3 km^2, mean depth 2.4 m) is a semi-enclosed estuary west of the City of Helsinki (Fig. 1). It receives freshwater from 2 brooks, and is connected to the Gulf of Finland first by 2 narrow straits and then by 2 sounds, which restrict horizontal water-exchange. The theoretical residence

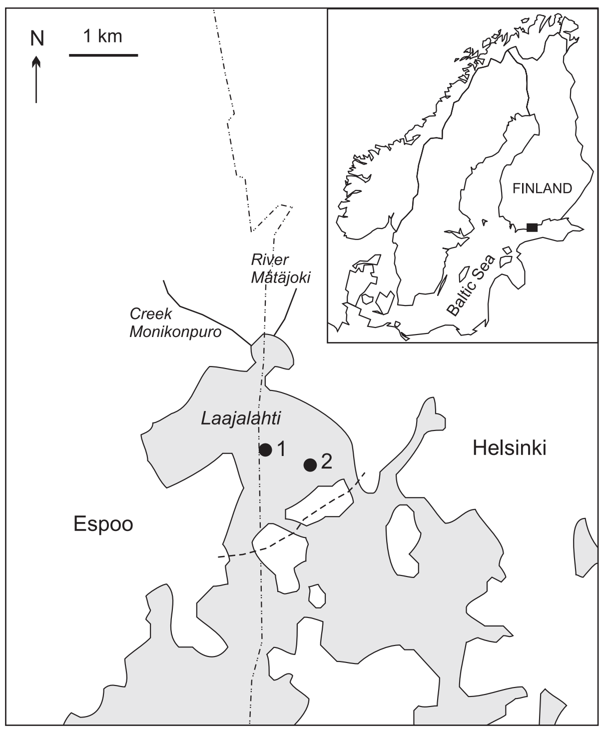


Fig. 1. Location of study area and sampling sites for water quality (1) and coring (2)

time is 0.11 yr and the average salinity (4.6 psu) of the estuary is close to that of the open archipelago. The water is turbid with Secchi depth varying between 0.5 and 1 m. There is no clear stratification in the estuary, therefore oxygen conditions near the bottom are usually good during summer and the period of ice cover between December and April. The bottom topography of Laajalahti is flat and the sediment quality is quite homogenous (P. Munne unpubl. data). The bottom consists mainly of soft sediments, and the shoreline is dominated by the reed *Phragmites australis* (Lappalainen & Pesonen 2000). According to the Finnish typology, Laajalahti Bay is included in the inner coastal type of the western Gulf of Finland (Kangas et al. 2003).

In the 1960s, Laajalahti Bay was one of the most polluted coastal areas of Helsinki, receiving annually ca. 60 *t* yr^{-1} of total phosphorus (TP), 300 t yr^{-1} of total nitrogen (TN) and 1000 t yr^{-1} of organic matter (measured as BOD_7) from a treatment plant built in 1957. The nutrient and organic matter loads started to decrease in the 1960s (Fig. 2). The decline of TP was more pronounced in the mid-1970s after the introduction of chemical removal of phosphorus and the redirection of overload to other treatment plants in Helsinki. The trend for the organic load was similar to that of phosphorus, but the load of TN remained at 150 t yr^{-1} from the early 1970s to the closing of the plant in 1986, when purified wastewaters of the capital area were redirected to the outer archipelago. During the operational time of the plant, the loads of TP and organic matter decreased by more than 90% and the load of TN by nearly 60%. The loads of phosphorus and organic matter reached levels below 5 and 200 t yr^{-1}, respectively, in the late 1970s. At present, Laajalahti Bay receives only diffuse loading from 2 brooks discharging 11.6 t yr^{-1} of TN and 0.9 t yr^{-1} of TP. Scattered dwellings are the main anthropogenic source of nutrients (58 and 22% of the total loading of TP and TN, respectively) and agriculture is the second largest source (22% of the total loading of TP and TN). On average, the input from the brooks (1293 kg TN km^{-2} yr^{-1} and 97 kg TP km^{-2} yr^{-1}) relative to the surface area of the estuary is 1 order of magnitude smaller than in other Finnish estuaries (P. Kauppila unpubl. data).

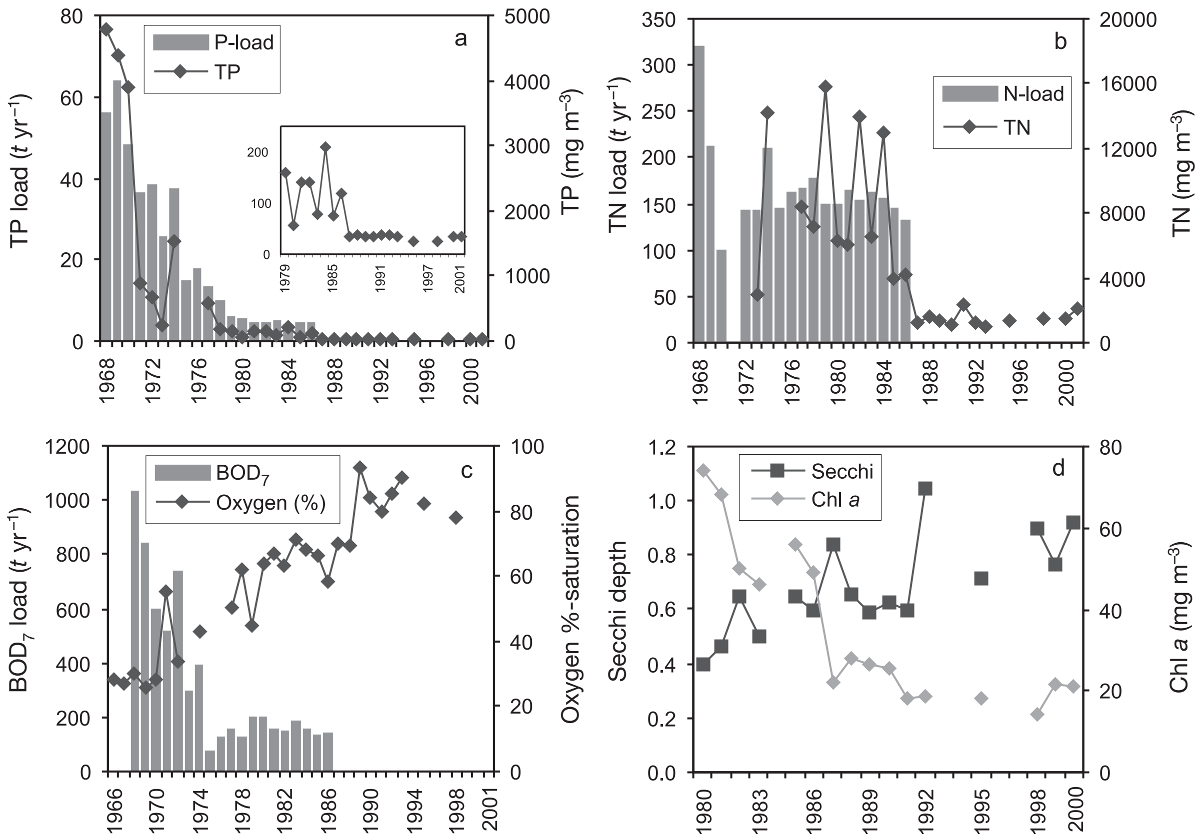
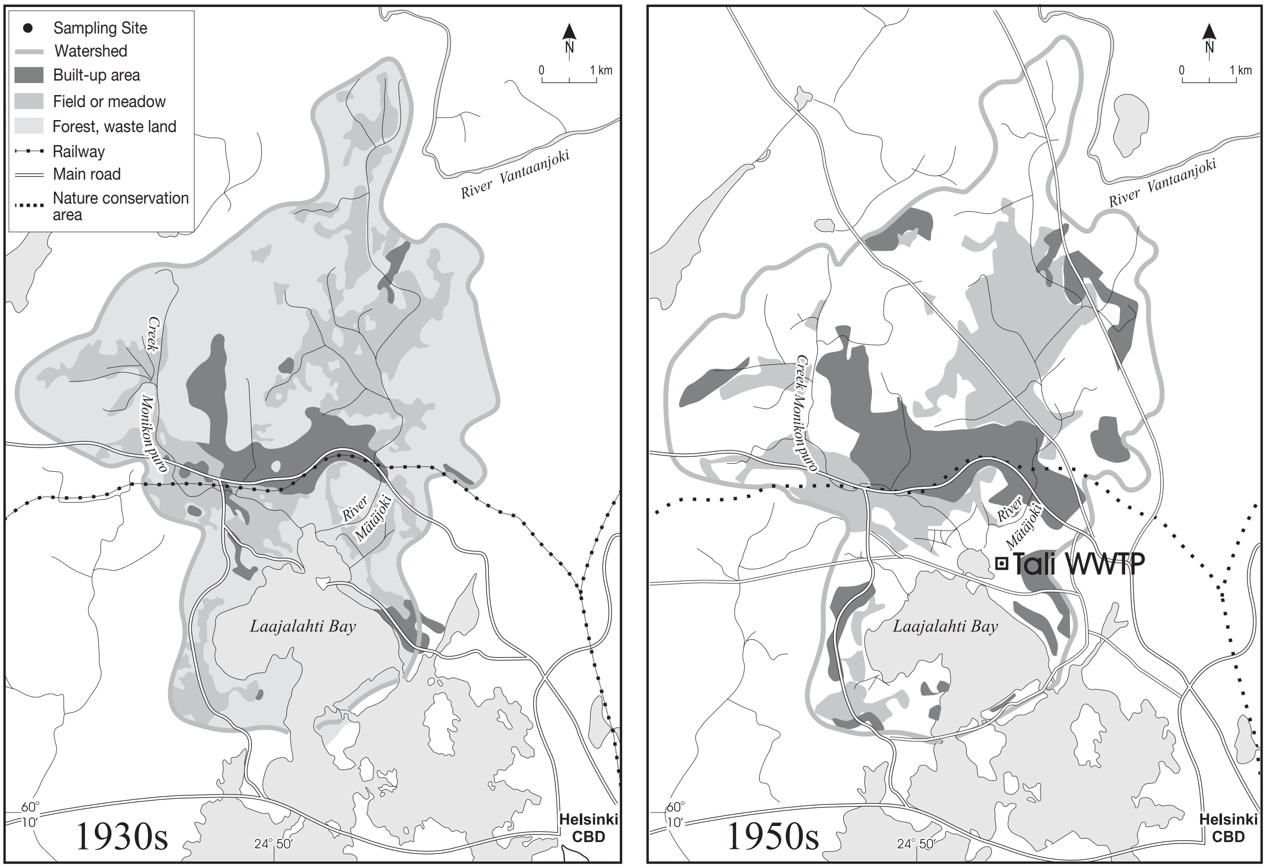


Fig. 2. Trends in water-quality variables and annual municipal loading of nutrients and organic matter into Laajalahti Bay during 1966 to 2001. (a) Surface wintertime concentrations and municipal load of total phosphorus (TP); (b) surface wintertime concentrations and municipal load of total nitrogen (TN); (c) near-bottom summertime oxygen %-saturation and municipal load of organic matter (as BOD_7); (d) summertime concentrations of phytoplankton chl *a* and Secchi depth. Loading data during 1968 to 1971 based on calculations by Rekolainen (1982)



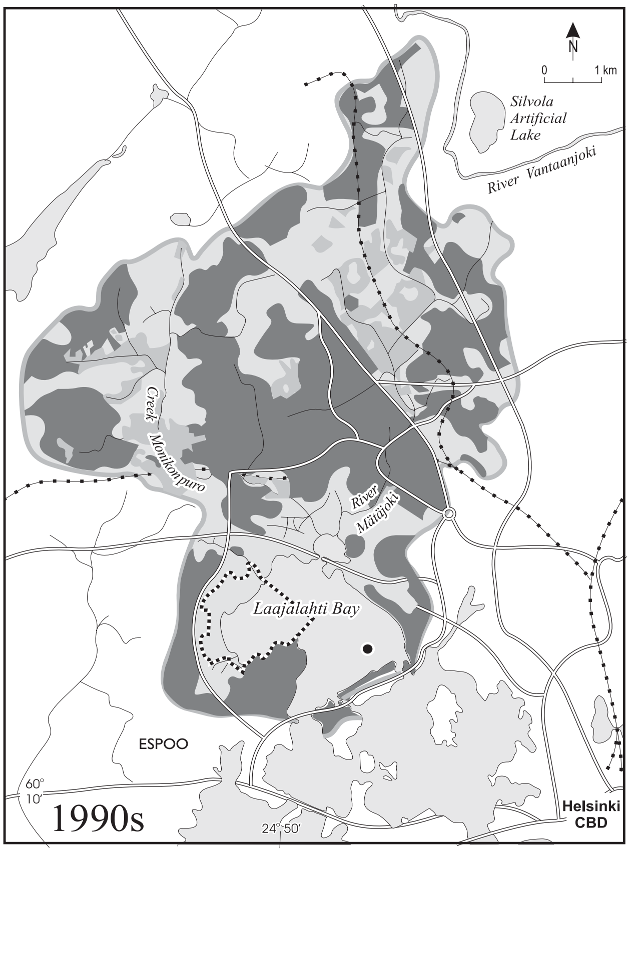


Fig. 3. Land-use in Laajalahti Bay catchment area in 1930s, 1950s and 1990s (data modified according to Vaalgamaa 2004)

Land-use in the catchment of the estuary (25.4 km^2) changed significantly during urban development. In the 1930s and 1950s, most of the area was covered with forests and swamps (ca. 61 and 59%, respectively), and fields and meadows comprised 24 to 20% of the total catchment area (Fig. 3). The change between these decades was slight, the urbanised area growing by only 6%. The major changes in land-use have occurred since the 1950s with the urbanised area, including an industrial district in the middle of the catchment, growing from 21 to 54% of the total area. Therefore, much of the agricultural land has been converted to urban area and the forested area in the catchment has markedly decreased. Today, the main land-use types of the catchment are urbanised area (54%), fields and meadows (12%) and forests and wasteland (34%, including swamps and wetlands), with a nature conservation area of 70 ha located in the NW corner of the estuary. The human population of the catchment is approximately 120 000.

Nutrient load and budget calculations. The point source loads of TN, TP and organic matter (BOD_7)

were taken from a database maintained by the Finnish Environment Administration. Annual nutrient fluxes into Laajalahti Bay from different sources were calculated using the database developed by the Finnish Environment Institute (SYKE) for the estimation and control of nutrient loads into watercourses (Rekolainen 1993, Kauppila et al. 2001).

The available concentration data and the calculated residence time was used to assess the role of sediments (Inkala et al. 2002) after the cessation of direct wastewater loading in 1986. There are no direct measurements available on nutrient-exchange rates after the study of Rekolainen (1982). Data on the classical salinity–concentration relationships (see Aston 1980) were divided into 3 periods: Period 1 (1971 to 1978) was characterised by extremely high TP concentrations, which declined steeply; Period 2 (1979 to 1986) exhibited high TP concentrations, which declined gently; the 3rd (1987 to 2002) period showed no significant trend of wintertime TP concentrations after the closing of the wastewater treatment plant in 1986 (Table 1 & Fig. 2).

Monitoring water quality. Water-quality data originated from 2 monitoring stations of the Helsinki Environmental Center: 1 station at the mouth of the Mätäoja Brook draining into the estuary, and 1 in Laajalahti Bay (Fig. 1). Samples were taken from the surface (1 m) and near the bottom (3 or 4 m) at least 6 times per year during the period 1966 to 2001. Total nitrogen (TN), nitrite-N (NO_2), nitrate-N (NO_3), ammonium-N (NH_4), total phosphorus (TP) and phosphate-P (DIP) were analysed from unfiltered samples according to Finnish standard methods (Koroleff 1976, 1979). Oxygen concentration and percentage saturation were determined by the Winkler method (Grasshoff et al. 1983). Phytoplankton chlorophyll *a* (chl *a*), measured from a composite sample (0 to 4 m), was analysed according to Lorenzen (1967). The chl *a* samples were extracted with acetone in 1980 to 1994 and with ethanol (ethyl alcohol) thereafter.

Sediment coring and sampling. Two 90 cm long sediment cores were collected with a mini-Mackereth corer (Mackereth 1969) in 1998 in 3.7 m of water. The master core, LaA, was used for dating, diatom analysis and sediment geochemistry. Core LaB was used for analysing grain size and stable isotopes. Core LaC (32 cm) was collected from the same location in 2002 with a HON-Kajak gravity corer (Renberg 1991) for pigment analysis. Coring a single well-chosen location has been shown to be sufficient for assessing the eutrophication history of a site (e.g. Anderson 1998). The cores were subsequently sectioned into 1 cm intervals and stored for further analysis in small plastic bags at 4°C in the dark. The upper 40 cm of LaA and LaB were used for the present study. The cores were correlated using loss-on-ignition (LOI) analysis.

Dating. The master core was dated in the Liverpool University Environmental Radioactivity Laboratory

Table 1. Annual trends in concentrations of nutrients and chl *a* in the surface-water layer and oxygen concentrations and percentage saturation in near-bottom water layer in Laajalahti Bay in winters (February to March) and summers (July to September) of 1968 to 1986, 1987 to 2002 and 1968 to 2002. Trends analysed by Kendall's tau *B*-test with significance level of 0.01. Units for medians and annual slopes in brackets. n: number of observations; –: no trend; TN: total nitrogen; TP: total phosphorus; DIN: dissolved inorganic nitrogen; DIP: dissolved inorganic phosphorus

Parameters	1968–1986				1987–2002				1968–2002			
	n	Trend	Median yr^{-1}	Slope	n	Trend	Median yr^{-1}	Slope	n	Trend	Median yr^{-1}	Slope
Summer												
TN (mg m^{-3})	13	Decrease	1200	155	14	–	658	–	27	Decrease	858	68
TP (mg m^{-3})	14	Decrease	170	24	14	Decrease	68	1.9	28	Decrease	95	11.6
PO_4-P (mg m^{-3})	14	Decrease	40	7.2	14	–	16	–	27	Decrease	17	3.1
DIN (mg m^{-3})	13	–	19	–	14	Decrease	15	1.0	27	Decrease	18	0.8
TN/TP	13	Increase	7.7	0.18	14	Increase	9.5	0.17	27	Increase	8.9	0.15
DIN/DIP	13	Increase	0.5	0.12	14		0.9	–	27	–	0.7	–
Chl (mg m^{-3})	6	–	53	–	12		22	–	18	Decrease	26	2.2
Secchi (m)	13	–	0.5	–	14		0.7	–	27	Increase	0.6	0.02
Winter												
TN (mg m^{-3})	12	–	6900	–	11	–	1300	–	23	Decrease	2920	486
TP (mg m^{-3})	17	Decrease	210	244	11	–	35	–	28	Decrease	99	108
PO_4-P (mg m^{-3})	14	–	148	–	11	–	19	–	25	Decrease	62	28
DIN (mg m^{-3})	14	Decrease	5752	412	11	–	947	–	25	Decrease	1938	455
TN/TP	12	–	47	–	11	–	41	–	13	–	43	–
DIN/DIP	12	–	43	–	11	–	66	–	23	Increase	50	2.6
Oxygen (mg l^{-1})	19	Increase	7.2	0.4	11	–	10.2	–	30	Increase	8.6	0.3
Oxygen (%)	19	Increase	51	2.8	11	–	72	–	28	Increase	61	2.3

(UK). It was analysed for ^{210}Pb, ^{226}Ra and ^{137}Cs by direct gamma assay, using Ortec HPGe GWL series, well-type, coaxial, low-background, intrinsic germanium detectors (Appleby et al. 1986, 1992). Radiometric dates were calculated using the constant rate of supply (CRS) and the constant initial concentration (CIC) ^{210}Pb-dating models (Appleby & Oldfield 1978). There was a significant discrepancy between the two ^{210}Pb chronologies in the upper zone. According to the ^{137}Cs date of the 1986 Chernobyl accident, the CIC model is more applicable to recent sediments. Hence a composite model chronology was constructed using the CIC model for the upper zone (0 to 13 cm) and the mean sedimentation rate from both models for the deeper sections (Fig. 4).

Sediment lithology and geochemistry. The water content and loss-on-ignition of the sediment were determined after SFS (1990, standard No. 3008).

Samples for grain-size analysis were homogenised by ultrasonification and subsequently measured on a Malvern Mastersizer. Then mean grain size was calculated from 6 subsamples. For chemical analyses the samples were dried in plastic bags at 80°C (Tanner & Leong 1995), and subsequently homogenised in an Ika A10 mill with a W-Co blade. A subsample was digested by autoclaving in 7 M nitric acid at 125°C for 30 min for analysis of Cu, Zn and TP (SFS 1980, standard No. 3044). Concentrations of metals were measured using a Varian SpectrAA 10+ atomic absorption spectrophotometer. The TP concentration was analyzed from the bulk sample using the ammonium molybdate method with ascorbic acid reduction (SFS-EN 1997, standard No. 1189). Inorganic phosphorus (IP) was measured by digesting in 1 M HCl for 18 h (Aspila et al. 1976) and then analysed according to SFS-EN (1997, standard No. 1189). Organic phosphorus (OP) was obtained by subtracting the inorganic fraction from TP. The nitrogen (TN) and carbon (TC) content of the sediment was measured using a Leco-analyser. The concentration of biogenic silica (BSi) was analysed using a modification of the method of DeMaster (1981).

The repeatability and precision of the procedures used for digestion and metal and TP determinations were verified using certified reference materials (NIST 1646 estuarine sediment, VKI CMR municipal sludge A) (see Vaalgamaa 2004). Quality control of BSi analyses was carried out using the same reference samples as those used in the interlaboratory comparison conducted by Conley (1998).

Stable isotope analysis. Sediment ^{15}N was determined using continuous-flow isotope mass spectrometry (CF-IRMS). Sediment samples (1 g) were oven-dried and split. Splits for ^{15}N were ground with an agate pestle and mortar and loaded into tin capsules (25 mg sample^{-1}).

Samples of ground material were combusted with a Carlo Erba/Fisons 1108 CHN analyser before being passed into a single-inlet dual collector mass spectrometer (automated nitrogen carbon analysis [ANCA] SL 30-30 system; PDZ Europa); 2 samples of an internal reference (flour) were analysed after every 5 samples. Isotope ratios were calculated using the formula $\delta^{15}N(‰) = [(^{15}N/^{14}N)_{sample}/(^{15}N/^{14}N)_{reference} - 1] \times 100$. Standard deviation of the reference was 0.2‰.

Pigments. Pigments were extracted from 0.5 g of homogenised freeze-dried material using 100% acetone, and were prepared for analysis as described by Chen et al. (2001). Quantitative analyses of all pigments were conducted using a Waters HPLC equipped with an on-line photodiode-array detector (Waters 996 PDA). The gradient programme used followed the methods of Wright et al. (1991) as modified by Chen et al. (2001). Identification of individual pigments was performed by comparing retention times and PDA spectra with authentic standards obtained from The International Agency for ^{14}C Determination, DHI, Denmark.

Diatoms. A modern calibration data set of surface-sediment samples for diatom analysis and associated surface-water-chemistry data was collected between August 1996 and February 1998 along the southern coast of Finland, Baltic Sea (22°93′E to 27°77′E). The data set consisted of 49 relatively small (median area 117 ha), shallow (median depth 3.2 m) and sheltered coastal embayments with low water

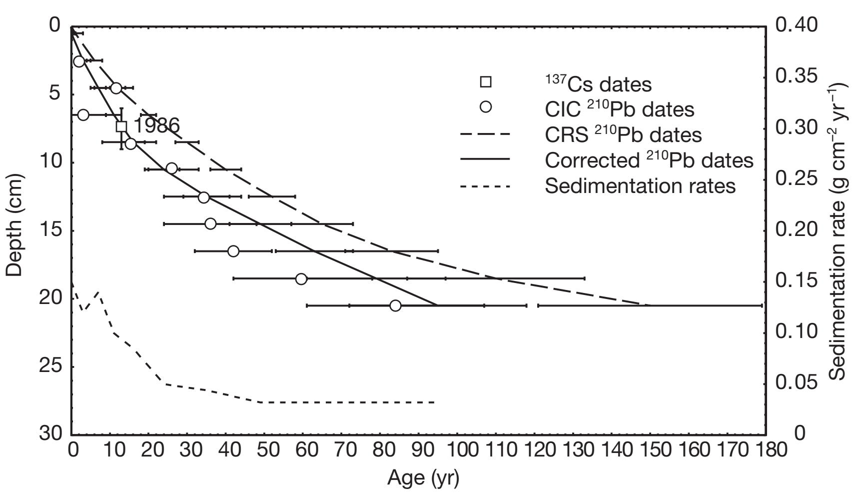


Fig. 4. Radiometric chronology of Laajalahti core LaA constant rate of supply showing (CRS) and constant initial concentration (CIC) model ^{210}Pb dates together with 1986 depth determined from ^{137}Cs stratigraphy. Also shown are corrected ^{210}Pb dates and sedimentation rate (Vaalgamaa 2004)

transparency (median 1.4 m). The sites were visited altogether 6 times during the field sampling; 4 additional sites were included in the data set and sampled twice in February and in August 1998 to increase the number of sites at the high end of the nutrient gradients. A detailed description of the modern calibration data set in terms of water sampling, analysing techniques and water-quality parameters is given in Weckström et al. (2002).

The surface-sediment samples were collected at the same location as the water samples with a small gravity corer (Glew 1989). The top 1 cm was extracted for the analysis of modern diatom assemblages. The samples were stored in small plastic bags at 4°C. Diatoms were prepared from the sediment samples 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®. Diatom counts for all samples were 500 to 600 valves.

Data analysis. In regard to the monitoring data, trends in measured TN, TP, inorganic N and P, oxygen, chl *a* and Secchi depth were analysed separately for the winter period (January to March) and the summer period (July to September). Based on the general overview of the plotted data, we used the non-parametric Kendall's tau *B*-test to examine the significance of the monotonic trends. This test is suitable for water-quality data, because it is not particularly sensitive to missing data and outliers, and requires no assumption of normality (Helsel & Hirsh 1992). Linear regression analyses were used to estimate the magnitude of significant trends. Additionally, linear regression analyses were used to model relationships between chl *a* and both TN and TP, which were normalised by log-transformation prior to analysis (Zar 1999).

A diatom-transfer function for total dissolved nitrogen (TDN), which had been previously generated from the 49-site calibration data set, was used to infer past TDN concentrations from the fossil diatom assemblages in the Laajalahti core (Weckström et al. 2004). Only taxa with at least 1% abundance in 2 or more samples were included in this model. Multivariate canonical ordinations were used to examine the relationships between diatom species and the measured environmental parameters. Detrended correspondence analysis (DCA) was used to measure the species turnover (the gradient length) in order to determine whether to use linear or unimodal ordination techniques. Canonical correspondence analysis (CCA) with forward selection and associated Monte Carlo permutation tests (999 unrestricted permutations; $p \leq 0.001$) was then used to identify a minimum subset of environmental variables that accounted for statistically significant variation in the diatom data (ter Braak 1986). The independent explanatory power of these variables was explored by using variance decomposition in partial CCAs (ter Braak 1988). Finally, weighted-averaging partial least squares (WA-PLS) regression and calibration (ter Braak & Juggins 1993) was used to estimate the diatom-inferred modern nutrient concentrations for each reference site.

Diatom diversity in the Laajalahti core was quantified by estimating the species richness of the fossil assemblages using rarefaction analysis, implemented with the programme RAREPOLL (Birks & Line 1992) with a base count of 502. Rarefaction analysis is well suited for stratigraphical species data and produces realistic estimates of richness without any bias associated with the variability of individual sample-count size (Birks & Line 1992). The major trends in the diatom assemblages of the Laajalahti core were summarised using correspondence analysis (CA). Correspondence analysis is a widely used technique for unimodal indirect gradient analysis (ter Braak 1995), which is useful for reducing the number of dimensions in a large and complex data set to detect the underlying patterns of variation in the data. The diatom biostratigraphy was also zoned with the constrained optimal sum-of-squares partitioning with untransformed species-percentage data using the software package ZONE Version 1.2 (S. Juggins, Department of Geography, University of Newcastle, UK, unpubl. computer programme). The number of statistically significant zones was calculated using the broken-stick model described in Bennett (1996).

Principal component analysis (PCA), an indirect linear ordination method (ter Braak 1995), was used to summarise the trends in both biological and chemical variables of the Laajalahti core and to provide easy visualisation of these patterns. The variables used to generate this multidimensional index for water quality comprised (1) diatom species richness, (2) CA axis 1 scores, (3) diatom-inferred total dissolved nitrogen (DI-TDN), and (4) the geochemical variables BSi, TN, TC, TP, OP and the Cu:Zn ratio. Additionally, these variables were zoned similarly to the diatom data with optimal sum-of-squares partitioning including the calculation of statistically significant zones. Pigments, stable isotopes and grain size were excluded from the PCA, as their record did not cover the whole sediment sequence.

RESULTS

Changes in water quality since late 1960s

The wintertime water quality of Laajalahti Bay in the late 1960s was characterised by extremely high phosphorus concentrations (4400 µg l^{-1} in surface water) due to heavy loading of municipal wastewaters (Fig. 2). The decrease in TP of 80% by the early 1970s was due to the implementation of wastewater purification. The concentrations of TP continued to decrease during the late 1970s and 1980s, but the decline was less pronounced. TN also decreased during these decades, mainly due to the reduction of wastewater-loading, but the concentrations showed great annual variation. After the closure of the treatment plant in 1986, the concentrations of nutrients rapidly decreased to half of their pre-closure level. In the 1990s, the wintertime concentrations of TP (32 µg l^{-1}) were close to those in the open Gulf of Finland (Pitkänen et al. 2001). TN concentrations (1200 µg l^{-1}) were still more than twice those of the open sea.

As a result of wastewater-loading until the mid-1980s, surface concentrations of nutrients were 3 to 10 times greater than near the bottom. Furthermore, anoxic or hypoxic conditions in the whole water column prevailed below the ice until the early 1970s (Fig. 2). After closing the treatment plant, the difference between the surface and bottom values of TP disappeared, but surface TN has still remained twice the values of the bottom. In addition, the cessation of wastewater discharge in 1986 resulted in changes in the abundance of inorganic nutrients: ammonium-N forming the main fraction of inorganic N until 1986, and nitrate-N thereafter. On the basis of the inorganic N:P ratio (ca. 60) before the spring bloom, primary production has been clearly P-limited during the past few decades.

In the summer, concentrations of nutrients in Laajalahti Bay showed similar decreasing trends to those in the winter before 1986. However, the levels of nutrients were lower and the slopes of trends gentler in the summer than in the winter (Table 1). In the 1990s, the summertime concentrations of TP (68 µg l^{-1}) and TN (660 µg l^{-1}) were twice those of the open Gulf of Finland (Pitkänen et al. 2001). TP and dissolved inorganic nitrogen (DIN) have continued to decrease since the closing of the treatment plant, whereas no significant trends could be found in wintertime concentrations of nutrients after the closure.

Summertime oxygen conditions have been good in the near-bottom water layer due to effective vertical mixing. In 1972, however, bottom oxygen percentage-saturation decreased below 70%, probably due to increased oxygen consumption caused by voluminous sedimentation of organic matter. Over-saturation values of oxygen in the late 1960s and the 1970s reflected high primary production not only in the surface but also in the bottom water layer.

Concentrations of phytoplankton chl *a* decreased significantly in the summers of the 1980s with the decreased nutrient-loading (Fig. 2). However, the high values (20 µg l^{-1}) of chl *a* in the 1990s suggest that Laajalahti is still strongly eutrophied. Phytoplankton primary production has been clearly N-limited during the summer periods, as indicated by the low inorganic N:P ratio (<1) and free phosphate present in the water during the growing season (Table 1). On average, 50 µg of extra phosphate-P l^{-1} were available for phytoplankton production during the period of wastewater-loading between 1957 and 1986, falling to approximately 13 µg l^{-1} after the treatment plant closed. Regression analyses showed that chl *a* was strongly associated with both TN and TP, which accounted for 91 and 74% of the variation in chl *a*, respectively (n =18, $p < 0.0001$).

Internal loading

Based on the classical salinity–concentration relationship, the nutrient distributions in Laajalahti Bay in the 1970s were strongly dependent on mixing conditions between sea water and the water from the Mätäjoki Brook, which was mixed with wastewaters from the purification plant (Fig. 5). The strong effect of wastewater-loading was still clear in the 1980s, although phosphorus concentrations were much lower due to the decreased loading from the purification plants.

After the cessation of wastewater-loading in 1986, no significant correlations could be found between TP concentration and salinity in the available data covering winter and summer periods (Fig. 5). However, the summertime average of TP (85 µg l^{-1}) is twice the corresponding wintertime average (35 µg l^{-1}). In similar coastal areas without any substantial current or previous wastewater-loading, the wintertime nutrient TP level is 30 to 40 µg l^{-1} (Pitkänen et al. 2001), while the summertime concentrations are lower, usually between 20 and 30 µg l^{-1}. This suggests that a considerable net input of phosphorus occurs from the bottom sediments, which evidently still contain substantial amounts of phosphorus from the period of high external loading.

The amount of the net sediment efflux for phosphorus could be roughly estimated for the summer season (June to September) by multiplying the average concentration difference between summer and winter (50 µg l^{-1}) by the surface area (5.3 km^2) and average depth (3 m) of the estuary, and then dividing the result by the average summer residence time (2 mo = 0.5 summer seasons, Inkala et al. 2002). Accordingly, the net sediment efflux of phosphorus in Laajalahti Bay was estimated to be 1.5 t per summer. This amounts to a purified wastewater load of about 50 000 inhabitants (93 % removal efficiency). Based on the active sediment accumulation area totalling 95 % of the entire area (P. Munne, unpubl. data), the estimated net sediment efflux of phosphorus equals 2.4 mg m^{-2} d^{-1}. This is much less than the average obtained experimentally for the reduced surface sediments of the coastal Gulf of Finland in late summer (13 mg m^{-2} d^{-1}, Pitkänen et al. 2001), but more than that obtained for oxidised bottoms (usually <2 mg m^{-2} d^{-1}, Lehtoranta & Heiskanen 2003).

Sediment

The upper part of the sediment core was dark brown with black streaks except for a lighter brownish top layer of 1 cm. From 35 cm downwards, the colour of the sediment was pale brown. The visual lithology seems to indicate oxic conditions in the older sediment, anoxic-sulfidic conditions in the recent sediments and oxic conditions at the sediment–water interface. For further details concerning the sediment lithology, see Vaalgamaa (2004).

Core chronology and sediment accumulation rates

Total ^{210}Pb activity in the Laajalahti core reached equilibrium with the supporting ^{226}Ra at a depth of ca. 20 to 25 cm, corresponding to ca. 150 yr accumulation. The ^{137}Cs activity has a relatively well-resolved subsurface peak at 7 to 9 cm. Although the total amount of ^{137}Cs was rather low, the ^{137}Cs:^{210}Pb ratio (3.8) is well in excess of the ^{137}Cs:^{210}Pb ratio expected from nuclear weapon fallout. The ^{137}Cs peak thus almost certainly records the fallout from the 1986 Chernobyl accident. It was not possible to distinguish a peak recording the 1963 fallout maximum from nuclear weapons, which has presumably been masked by downward diffusion of ^{137}Cs from the Chernobyl incident.

Fig. 4 shows ^{210}Pb dates calculated using both the CRS and CIC models, together with the 1986 depth indicated by the ^{137}Cs stratigraphy. The composite model chronology (see 'Materials and methods: Dating') suggests a relatively uniform sedimentation rate of ca. 0.032 g cm^{-2} yr^{-1} (0.13 cm yr^{-1}) during the first half of the 20th century. There appears to have been a rapid increase since ca. 1980, and sedimentation rates

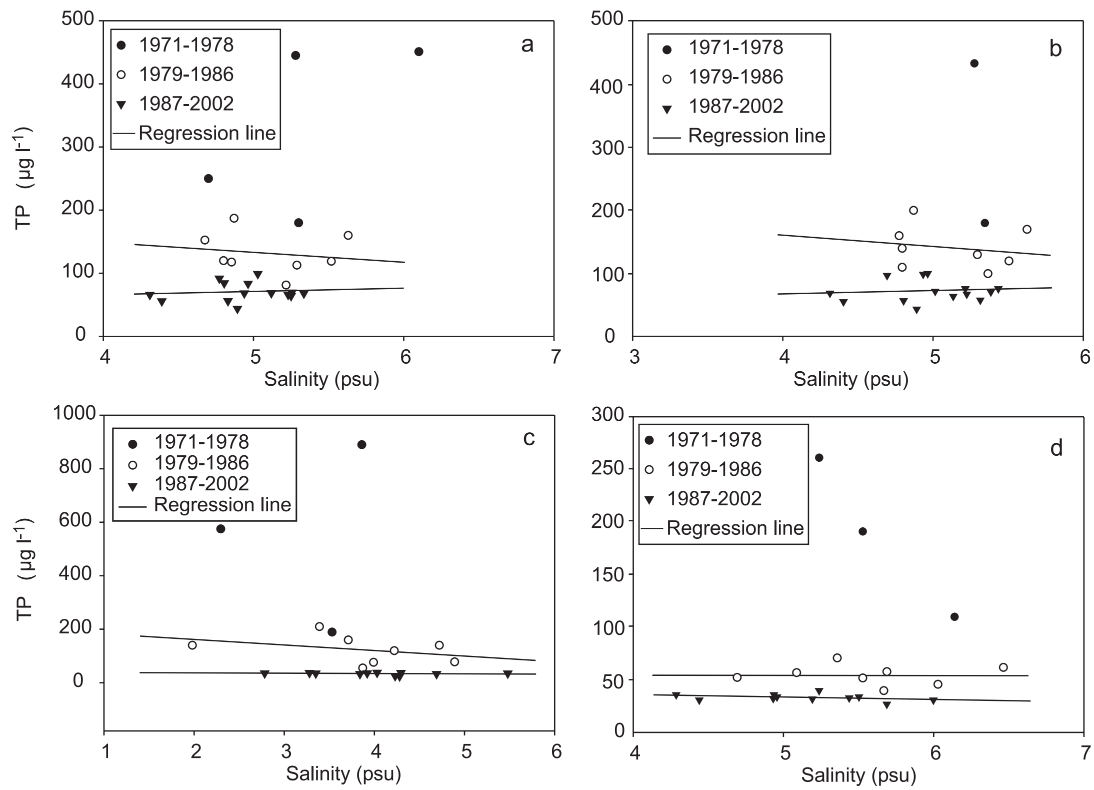


Fig. 5. Correlations between TP and salinity during 1971 to 2002. (a) Surface-water layer in summer; (b) near-bottom water layer in summer; (c) surface-water layer in winter; (d) near-bottom water layer in winter. Data divided into 3 periods: extremely high P concentrations and steep slope in the trend (1971 to 1978); high P concentrations and gentle slope in the trend (1979 to 1986); and no significant trend in P concentrations (1987 to 2002)

during the past decade have had an average value of ca. 0.13 g cm^{-2} yr^{-1} (0.6 cm yr^{-1}). The dates below 25 cm are extrapolated from an age–depth relationship based on 12 ^{210}Pb measurements and 1 ^{137}Cs date. The established chronology agrees well with the spheroidal carbonaceous particle (SCP) record from Laajalahti Bay, which showed a rapid increase in particle concentration starting in the late 1930s with a clear subsurface peak at 1975, consistent with the known energy consumption data from the Helsinki area (P. Leeson unpubl. data). The SCP method has been shown to give fairly reliable age determinations for lake and coastal sediments in Finland (e.g. Tolonen et al. 1992), and can thus be used as a control for ^{210}Pb-dating when determined from the same core.

Sediment lithology and geochemistry

There is a distinct decrease in sediment grain size at 36 cm (ca. 1680). Between 35 and 20 cm (ca. 1700 to 1900), the grain size reaches its highest values (6.4 to 7.2 µm); above this depth, the values decrease again and stabilise around 5.5 µm. TN, TC, and TP follow similar trends and correlate well with organic content (as determined by LOI) of the sediment. TN concentrations increase gradually upwards and reach their highest levels (<4 mg N g^{-1}) at the top of the core (Fig. 6). TP concentrations also increase gradually throughout the core, but more steeply in the top 3 cm. The trends in organic phosphorus (OP) and biogenic silica (BSi) profiles appear to be quite different. The OP concentrations increase slightly from 30 to 10 cm (ca. 1780 to 1972), then the concentrations decline notably, only to increase again in the top 3 cm. BSi concentrations increase gradually between 30 and 11 cm (ca. 1780 to 1967) and decrease slightly towards the top of the core.

Copper (Cu) and zinc (Zn) comprise a pair of elements that occur in reduced form in solution at different threshold redoxpotential (Eh) levels. Therefore, the Cu:Zn ratio has been used to describe sediment redox-conditions (Tolonen & Meriläinen 1983). However, the usefulness of this indicator may be limited in areas with irregular anthropogenic loading or massive organic sedimentation. Major changes in the Cu:Zn ratio can be seen at depths of 22 and 12 cm. The sharp rise at 22 cm corresponds to the beginning of the 20th century, and the fall at 12 cm dates to the late 1960s.

Stable isotopes

The $\delta^{15}N$ profile of nearshore sediments has been shown to be a specific tracer of eutrophication in nearshore environments (Voss & Struck 1997, Struck et al. 2000). The Laajalahti core demonstrates a distinct and continuous increase in $\delta^{15}N$ from around 3‰ at 19 cm depth (Fig. 6) which corresponds to the early

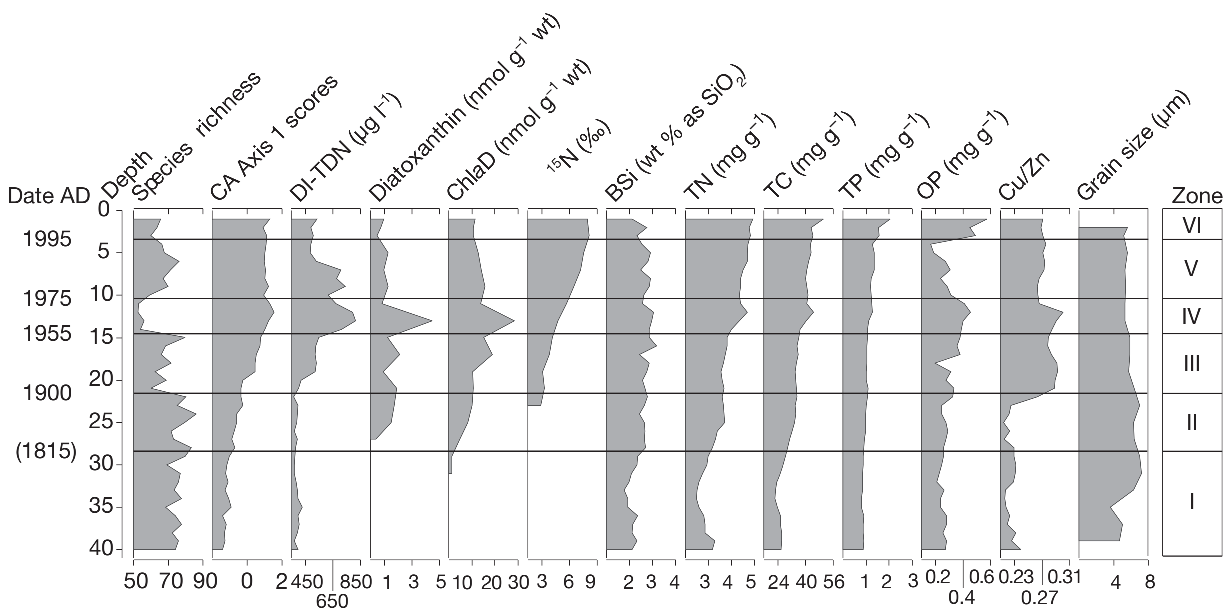


Fig. 6. Stratigraphic diagram of index of species richness, correspondence analysis (CA) Axis 1 scores summarising overall changes in diatom assemblages, diatom-inferred total dissolved nitrogen (DI-TDN), pigments, $\delta^{15}N$ and sediment geochemistry. Data divided into 6 zones: Zone I (ca. pre-1815), Zone II (ca. 1815 to 1900), Zone III (ca. 1900 to 1955), Zone IV (ca. 1955 to 1975), Zone V (ca. 1975 to 1995) and Zone VI (post-1995). Only boundary between Zones II and III is statistically significant, other zones are included to aid interpretation. Extrapolated date is given in parentheses. Chl*a*D: chl *a* plus its degradation products

1920s to around 8‰ at the surface. While there is a good correlation between $\delta^{15}N$ and other eutrophication indicators (DI-TDN, OP, chl*a*D [chlorophyll *a* degradation products]) from ca. 1900 to 1962, $\delta^{15}N$ is in direct contrast to these indicators and the water-chemistry data, which show a recent partial recovery from year 1975 onwards. However, it shows a similar pattern to TOC and TN. While sedimentary $\delta^{15}N$ is not a direct measure of TN input or production, the data from Laajalahti suggest that the N cycle at this site remains perturbed, probably by elevated levels of denitrification.

Sedimentary pigments

The sediment pigment profiles show a characteristic peak around 15 to 16 cm corresponding to the high nutrient load in the mid 1960s (Fig. 6). The concentration of chl *a* including its degradation products (chl*a*D), indicating total algal abundance, ranged from <2 to 28.4 nmol g^{-1} dry wt. It gradually increased from 30 cm (ca.1870) to a minor peak at 19 to 20 cm (ca.1946) and decreased after the major peak at 15 to 16 cm. Diatoxanthin, a relatively stable indicator of diatoms (Leavitt & Hodgson 2001), ranged from 0.40 to 4.45 nmol g^{-1} dry wt. Diatoxanthin showed the same overall trend as chl*a*D, however, the concentrations were higher below the major peak at 15 to 16 cm compared to the top part of the core. This could have been caused by the shift in diatom assemblages from benthic to planktonic dominance, which can affect the amount of pigment reaching the sediment without major exposure to degradation in the water column. The ratio between chl *a* and its degradation products was quite low, indicating that much of the relatively labile chl *a* has been degraded, as expected for a shallow, well-mixed estuary such as Laajalahti Bay.

Diatom assemblages

Altogether 226 diatom taxa were found in the Laajalahti core. The majority of these were benthic; only 14 taxa were planktonic. The diatom assemblages of the core were divided into 4 statistically significant zones (Fig. 7): In Diatom Zone 1, the diatom assemblages were dominated by benthic species such as small *Fragilaria* spp. Lyngbye, *Mastogloia smithii* Thwaites, *Rhoicosphaenia curvata* (Kützing) Grunow and *Planothidium delicatulum* aggregates. There was a marked increase in *F. elliptica* aggregates, and to a lesser extent *Pseudostaurosira brevistriata* (Grunow) Williams & Round in Diatom Zone 2, dating to the late

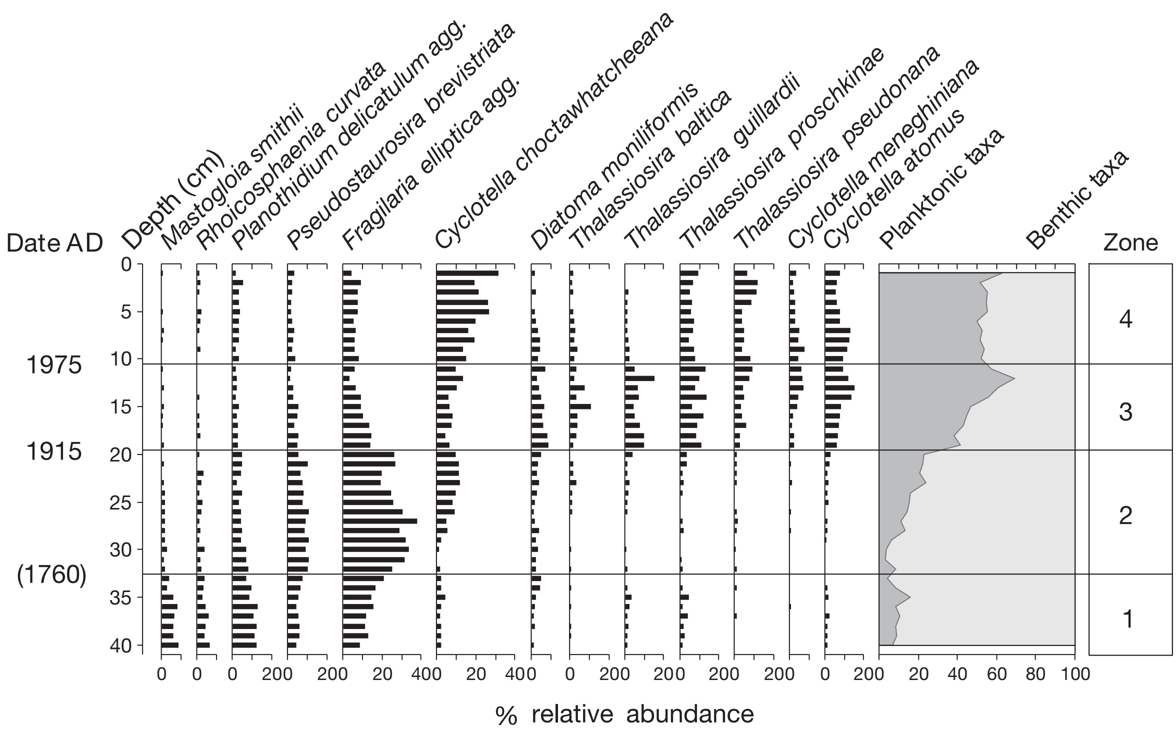


Fig. 7. Summary diatom diagram for Laajalahti Bay. Data are divided into 4 statistically significant zones: Zone 1 (ca. pre-1760), Zone 2 (ca. 1760 to 1915), Zone 3 (ca. 1915 to 1975) and Zone 4 (post-1975). Extrapolated date is given in parentheses

1800s, with a decrease in other benthic taxa. The predominance of *Fragilaria* spp. is a typical feature of shallow coastal embayments as well as recently isolated lakes (Stabell 1985). The water depth of the estuary has decreased over time due to post-glacial isostatic land-uplift and may have reached a threshold in Diatom Zone 2, after which conditions became favourable for benthic *Fragilaria* taxa. *Cyclotella choctawhatcheeana* Prasad, a taxon that seems to be favoured by anthropogenic disturbance (e.g. Cooper 1995), also increased halfway through this zone. The biggest change in the core occurs at the beginning of Diatom Zone 3, corresponding to the early 1920s, when the proportion of planktonic diatoms increased notably. At the start of this phase there was a clear decline in *Fragilaria* spp. and a reciprocal rise in many small planktonic diatoms, such as the genus *Thalassiosira* Cleve, *C. atomus* Hustedt, *C. meneghiniana* Kützing and also *Diatoma moniliformis* Kützing. The decline in benthic *Fragilaria* spp. was most probably due to increased turbidity of the water column, which in turn was caused by higher planktonic productivity with increasing nutrient enrichment of the estuary (e.g. Cooper & Brush 1991, Andren et al. 1999). There was an increase in the common eutrophic taxa *C. atomus* and *C. meneghiniana* (European Diatom Data Base: http://craticula.ncl.ac.uk:8000/Eddi/jsp/index.jsp) towards the end of Diatom Zone 3 corresponding to the late 1950s. Both *T. baltica* Ostenfeld and *T. quillardii* Hasle declined at the beginning of Diatom Zone 4. The last significant species shift occurred halfway through Diatom Zone 4 in the late 1980s, when both *C. meneghiniana* and *C. atomus* declined with a simultaneous rise in *C. choctawhatcheeana*.

Both the CA primary axis scores summarising the overall changes in diatom assemblages and the diatom-inferred total dissolved nitrogen (DI-TDN) show very similar trends ($r = 0.84$, $p \leq 0.01$) (Fig. 6). DI-TDN concentrations were around 400 µg l^{-1} from the bottom of the core (pre-1815) until the early 1920s, corresponding to 19 cm, when DI-TDN increased to ~520–550 µg l^{-1}. The concentrations stayed roughly the same until the mid 1950s, when they started to increase and peaked to 826 µg l^{-1} at 13 cm dating to the mid-1960s. The DI-TDN then decreased steeply in the mid-1970s, maintained high levels of ~700 µg l^{-1} throughout the 1980s, and finally declined to present concentrations of ~500 µg l^{-1} at 6 cm corresponding to the late 1980s. The CA primary axis scores display the same phases, although more gradually than DI-TDN, except for the decline in the late 1980s. The similarity of the curves between the TDN reconstruction, the CA primary axis scores and the planktonic to benthic ratio in Fig. 7 suggests that increased nutrient concentrations most probably caused the observed change in the diatom assemblages. The discrepancy between the DI-TDN and the other 2 curves in the uppermost part of the core may be explained by the species shift within the planktonic community, whereby the eutrophic *Cyclotella atomus* and *C. meneghiniana* were displaced by *C. choctawhatcheeana*, which has a considerably lower TDN optima in the calibration data set than the other 2 *Cyclotella* taxa.

The species richness in Fig. 6 displays an almost inverse relation to DI-TDN and CA Axis 1 scores. It was greatest before the start of the 20th century, being on average 76 taxa sample^{-1}. There was a moderate decline to a mean of 68 taxa sample^{-1}, which coincides roughly with the slightly elevated DI-TDN concentrations in the 1920s to 1950s. Species richness decreased clearly to a mean of 58 taxa sample^{-1} during the most pronounced eutrophication period (late 1950s to mid-1970s) and recovered to approximately the level before the major disturbance at the beginning of the 1980s. The decreased species richness during the most pronounced eutrophication period of Laajalahti is consistent with data from several other studies, which indicated that severe cultural eutrophication has led to a decrease in diversity of algal communities (Wetzel 2001).

PCA

The results of the multidimensional PCA are illustrated as 2 ordination diagrams in Fig. 8. The first 2 PCA axes cumulatively explained 79% of the total variation in the sediment core data, with eigenvalues of $\lambda_1 = 0.68$ and $\lambda_2 = 0.11$, respectively. As the eigenvalues of Axes 3 and 4 were 0.09 and 0.05, respectively, together accounting for only 14% of the total variance, the discussion will be limited to the main Axes 1 and 2. PCA Axis 1, which alone accounted for almost 70% of the variation, was highly correlated with environmental variables that are essentially measures of the nutrient status of the estuary (DI-TDN, TC, TN, TP; Fig. 8a). High loadings on Axis 1 were attributable to all these variables, except species richness, which scored low on Axis 1. The variables OP and TP also achieved high scores on PCA Axis 2, while BSi received low scores at the other end of this axis (Fig. 8a). It appears, therefore, that PCA Axis 2 relates more closely to the degree of internal loading at Laajalahti.

Most of the core samples are distributed along the primary PCA axis with only a modest amount of variation along Axis 2. Samples from the basal section of the core scored lowest on PCA Axis 1, while samples from depths between 11 and 14 cm had the highest loadings (Fig. 8b). Only samples representing the top section of

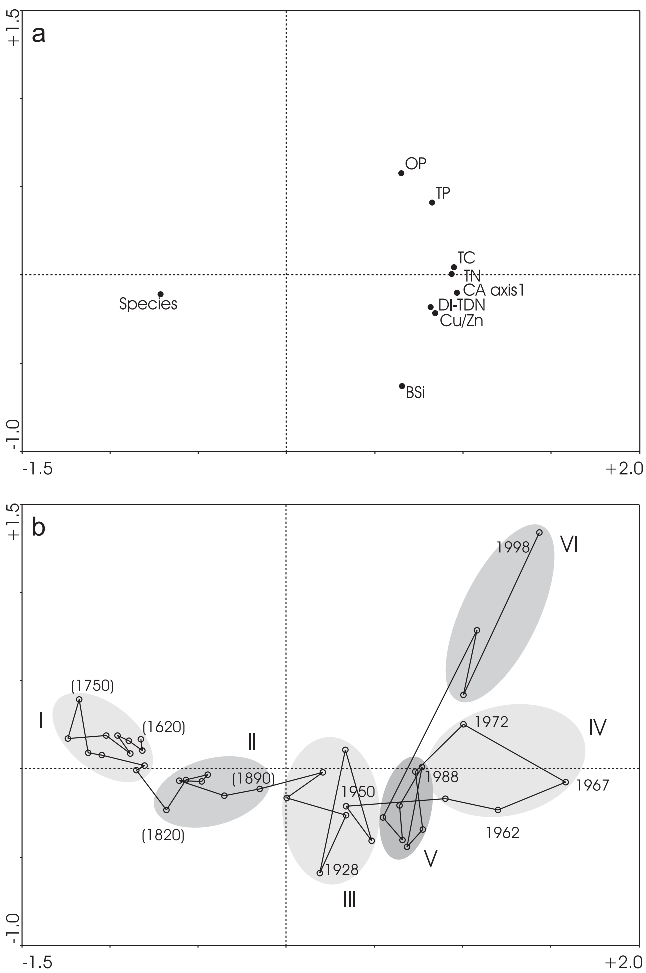


Fig. 8. Results of principal component analysis using species richness, correspondence analysis (CA) Axis 1 scores summarising overall changes in diatom assemblages, diatom-inferred total dissolved nitrogen (DI-TDN), and sediment geochemistry. (a) Variables; (b) samples of Laajalahti core, with adjacent samples joined by line. Zones are shown as grey circles labelled as in Fig. 6. Extrapolated dates are given in parentheses

the core show marked variation along PCA Axis 2, with high scores on this Axis. It was possible to distinguish 5 historical phases according to zones obtained by optimal sum-of-squares partitioning. These phases reflect the development of the nutrient status of Laajalahti on the basis of the created PCA index (Fig. 8b): (1) a pre-industrial phase (Zone I: before ca. 1815); (2) a phase of slight human disturbance (Zone II: ca. 1815 to 1900); (3) an onset of anthropogenic impact (Zone III: ca. 1900 to 1955), (4) a severe pollution phase (Zone IV: ca. 1955 to 1975); (5) basin recovery and the succeeding phase of internal loading (Zones V and VI: from ca. 1975 onwards). These interpretations facilitate the discussion of ecological reference conditions of Laajalahti Bay.

DISCUSSION

Natural conditions

On the basis of the PCA analysis and all the available data, Zones I and II represent natural conditions (Figs. 6 & 8). Depths of 29 to 40 cm (Zone I) dated back to the period before 1815, which represents the pre-industrial phase. Depths of 22 to 28 cm (Zone II) covered roughly the period 1815 to 1900, when human disturbance in Laajalahti Bay was minor. The load into Laajalahti Bay in the mid-1800s was estimated to be 1.5 and 17 t TN yr^{-1} based on the development of the average daily per capita nutrient load and the approximate population, which was assumed to have developed in parallel with the population in the catchment area of Helsinki (see Laakkonen & Peltonen 1999); there was no general sewage network at that time. However, pollution of coastal waters in Helsinki must have already started in the late 1880s on the basis of first-hand accounts at that time (Laakkonen 2001).

During the pre-industrial phase, sedimentary nutrient levels in Laajalahti Bay were quite stable, but showed a slow increase during Zone II, indicating general nutrient enrichment. This was most probably a consequence of post-glacial isostatic land-uplift, as embayments become more enriched by nutrients during their isolation process. This is also supported by changes in the grain size of the sediment and in the diatom community structure, where small *Fragilaria* spp. taxa dominate the assemblages. Annual average concentrations of TN were estimated to be ca. 600 µg l^{-1} based on the DI-TDN reconstruction and a subset of 30 calibration set sites, which was analysed for both TN and TDN in August 2001 and April 2002 to assess the squared correlation coefficient between these 2 variables. In order to obtain a rough estimate of past phytoplankton production levels, the original 49-site calibration data set was used to infer past chl *a* concentrations, which were estimated to be ca. 10 µg l^{-1} yr^{-1}. The predominance of benthic over planktonic taxa and the overall diverse diatom assemblages indicated by species richness suggest undisturbed conditions in the estuary during both periods, despite the fact that the abundance of planktonic diatom taxa started to increase slowly after the 1820s.

Onset of anthropogenic impact

Zone III (15 to 21 cm) roughly covered the years ~1900 to 1955. This period was characterised by a slow rise in the population and a lack of treatment of wastewaters (Laakkonen & Peltonen 1999). Most of the lavatory sewage in Helsinki at the time was discharged to coastal waters, because nearly 2 out of 3 apartments had flushing toilets (Laakkonen 2001). Wastewater-loading spread to a wider area, as the network of wastewater pipelines extended to the western part of Helsinki. On the basis of the average daily per capita load of TP and TN and the approximate population in the beginning of the 1900s, the loads into the estuary were estimated to be 9.9 and 110 t of TN yr^{-1} (Laakkonen & Peltonen 1999). These were similar to the loads in 1986, which was the last year in which the municipal treatment plant was operational (see Fig. 2).

The start of increasing pollution was reflected by clear changes in the sediment chemistry. An increase in the Cu:Zn ratio indicated a change towards more anaerobic conditions in the sediment as a result of increased organic production. The increase in nutrient enrichment, indicated by DI-TDN and $\delta^{15}N$, and the subsequent increase in organic production (measured as OP) indicated the start of eutrophication in the shallow estuary.

Nutrient enrichment in the estuary also changed the biotic community structure. Benthic diatoms began to decrease in abundance simultaneously with a distinct increase in planktonic taxa, although benthic communities still dominated at this phase. However, as the diatoms are expressed as percentage data, this decrease may also be an artefact caused by the increase in planktonic species. Moreover, diatom species richness decreased relative to its natural level. The increase in chl*a*D indicated an increase in algal biomass. However, increases in sedimentary pigments arise through better preservation (Leavitt 1993). Blue-green algal blooms appear to have been a common phenomenon in the receiving waters near large cities such as Helsinki before the introduction of sewage treatment (Finni et al. 2001). The N_2-fixing cyanobacteria *Aphanizomenon* spp. and *Nodularia* spp. were reported from many coastal areas of the Baltic Sea before the Second World War (Finni et al. 2001).

Severe pollution phase

The fourth zone (11 to 14 cm) represented the time period of ~1955 to 1975. This period was characterised by heavy wastewater-loading and a clear inital recovery due to purification activities in the treatment plant during its operation (1957 to 1986) (Fig. 2). Rapid recovery from pollution after the reduction of external loading has also been recorded for some lakes (e.g. Schiendler 1974, Bradshaw & Anderson 2001).

The nutrient levels of the estuary during this phase were highly elevated despite the clear reduction in pollution and the subsequent decline of nutrient concentrations at the start of the 1970s. The sedimentary record generally supports the main trends of nutrients in the water column. The steep decline in DI-TDN in the mid 1970s could be validated by the data on N-loading and concentrations in the water (Figs. 2 & 6). Despite the lack of loading data before 1968, wastewater-loading most probably peaked at the end of the 1960s, as estimated on the basis of the length of the wastewater pipeline and the increase in the population in Helsinki (see Laakkonen & Peltonen 1999). The peak of the Cu:Zn ratio (Fig. 6) could indicate an anoxic period during the monitoring history of the estuary; this was verified by low oxygen concentrations in the near-bottom water layer in the early 1970s.

The concentrations of DI-TDN during the peak loading period were, however, underestimated by the transfer-function model. This problem has also been observed with diatom–TP transfer models created for lakes (e.g. Hall et al. 1997). There are 2 main reasons for this: (1) our model does not currently include sites with very high nutrient concentrations, which would provide modern analogues for the most eutrophic phase of Laajalahti, and (2) diatom communities may be unable to respond to higher nutrient concentrations by structural changes. Also, it is important to keep in mind the inherent errors of the water-chemistry data. The oldest monitoring data, especially, is less reliable because of less accurate analytical methods and instruments at that time (L. Pesonen pers. com). However, the transfer-function model seems to accurately reflect nutrient concentrations, which are in the range of the sites included in the model (248 to 2068 µg l^{-1}), as indicated by the close agreement between the measured TN and inferred TDN values in the 1990s. This would imply that the model would also closely reconstruct pre-disturbance reference conditions.

Sedimentary OP as a rough measure of organic production showed a decreasing trend from the 1970s towards the top layers (with the exception of the reactive top 3 cm sediment layer). Similar decreases can be also seen in the water-chemistry data. As OP is more indicative of algal production than TP, it may have more palaeolimnological value (Aminot et al. 1998). Moreover, OP might prove to be a better indicator of eutrophication, because it is less mobile and degrades more slowly than TP (Ruttenberg & Goñi 1997).

Along with OP, variations in BSi profiles have been used to infer changes in the productivity of overlying waters (Conley & Schelske 2001). Unlike OP, no significant increase in BSi concentrations was observed during the peak nutrient-loading at the end of the 1960s (Figs. 2 & 6). This may be a consequence of the silicate limitation of diatoms and competition with other phytoplankton groups such as green algae, flagellates and dinoflagellates, which may have responded more strongly to the elevated nutrient concentrations at that time.

Changes in the biotic community structure reflected changes in nutrient levels. The abundance of *Cyclotella atomus and C. meneghiniana*, both indicators of eutrophication, increased during the pollution stage. The increased dominance of planktonic taxa suggests significant nutrient enrichment. Maximum eutrophication, evidenced by water-chemistry data, was reflected as simultaneous peaks of nutrients (sedimentary TN and DI-TDN) and pigments (chl *a* and its degradations products and diatoxanthin). Additionally, a clear loss of species richness was associated with the most eutrophied period of Laajalahti Bay. A similar loss in diatom biodiversity has been observed in another urban embayment in the Helsinki area during a severe eutrophication phase (Korhola & Blom 1996).

Phase of internal loading

Zone V is found between 10 and 4 cm. The uppermost reactive surface was designated as Zone VI. This phase, covering the period from ca. 1975 onwards, represents conditions of substantial internal loading and the termination of external wastewater-loading in 1986.

The sediment record partly supported the monitoring results of water quality during this phase. The decline of DI-TDN and OP since the late 1980s reflected the closing of the wastewater treatment plant. However, nutrient levels in the water column have not decreased since the 1990s (Fig. 2). This may be explained by internal loading from the sediments. The clear increase in OP in the reactive sediment surface (top 3 cm) is mainly a consequence of remineralisation and decomposition of fresh organic material.

There was a clear discrepancy between the monitoring data and the sediment records of TC, TN and TP, as their concentrations did not decline towards the top of the sediment. The observed pattern was most probably a combination of 2 factors. Firstly, the surface sediments tend to have a greater percentage of organic material, TN and TP, resulting from a steady-state input–decomposition balance, because less of the labile portion has decomposed (Berner 1980). Secondly, increased inputs of organic material to the sediment may also alter the rate of sediment loss through oxidation associated with changing redox conditions (Cornwell et al. 1996). These facts are supported by the dating results, which suggest that rates of organic production were closely balanced by rates of oxidation loss during the earlier part of the studied period. Rapid burial during high-flow years appears to limit early sedimentary diagenesis by aerobic respiration (Callender 2000). Since the sedimentation rates have increased significantly since 1974, probably less of the labile portion has degraded from the recent sediments.

Having recovered from the severe pollution phase in the 1960s and 1970s, Laajalahti Bay is still clearly N-limited in the summer. This is supported by the fact that TN explained 92 % of the variation in chl *a*, while TP explained 74 %. N-limitation in the estuary is maintained by several processes. Firstly, substantial amounts of phosphate-P are released from the water–sediment interface despite the good oxygen conditions in near-bottom waters (Rekolainen 1982 and present Fig. 2). Secondly, the estuary exchanges water with N-limited open coastal areas. Finally, the role of denitrification is probably substantial in removing large amounts of N at the sediment–water interface on the basis of high organic production (measured as sediment OP and water chl *a*), and constantly high wintertime concentrations of nitrate-N in the water column. Moreover, the increase in the $\delta^{15}N$ profile indicated that increasing amounts of N in the site have been subjected to denitrification.

On the basis of nutrient concentrations, productivity and the community structure of the biota, Laajalahti Bay is still eutrophied. This is characterised by the decline of species richness since the late 1980s and the continuing dominance of planktonic taxa in the diatom assemblages. Also, during the 1990s phytoplankton was still dominated by cyanobacteria (Autio et al. 2003).

The present-day estuary is still far from reference conditions despite the clear recovery recorded in the 1970s and 1980s. The net release of phosphorus from the sediment is substantial and a much more important source than terrestrial load into the estuary. During the last 15 yr, no improvement in the condition of the estuary could be seen after the immediate response after the closure of the local municipal treatment plant (Fig. 2). Restricted water exchange with the open Gulf of Finland means that the bay will continue to be affected by internal loading causing high blue-green algal production. The delay of recovery due to internal loading has been reported for many lakes and coastal water areas, where P is released from a pool accumulated in the sediment during a period of high external loading (Arnesen 2001, Pitkänen et al. 2001, Wilander & Persson 2001, Soendergaard et al. 2002a,b).

Establishing reference conditions

Information on reference conditions is required for the ecological classification according to the EU Water Framework Directive. In the case of Laajalahti Bay, there is more than 1 option for establishing reference conditions. Palaeolimnological analyses on sediment geochemistry and diatom community structure suggested that natural conditions in Laajalahti Bay prevailed in the mid- to late 1800s. The time before the early 1800s does not represent realistic reference conditions due to the gradual land-uplift, which affected the biotic communities of the estuary. Natural changes of a given system driven by e.g. climate, hydrology or, as in this case, post-glacial land-uplift, pose a problem for the concept of reference conditions. However, these kinds of changes are included in the principles of the directive, which states that 'in controlling anthropogenic pressures it is unrealistic to base reference conditions upon historic landscapes that no longer exist in modern Europe'. Based on our study, potentially useful tools for defining reference conditions in similar environments to Laajalahti Bay include diatom community structure, quantitative diatom-based transfer functions, sedimentary pigments and organic phosphorus. More studies will be needed in the future to assess if these proxies can be used as valid indicators of human disturbance in more complex coastal environments both inside and outside the Baltic Sea area.

The ecological classification according to the WFD is based on the calculations of the ecological quality ratio (EQR), which is the relationship between currently measured and reference values. At present, the annual levels of chl *a* (ca. 20 µg l^{-1}) and TN (ca. 750 µg l^{-1}) in Laajalahti Bay are still higher than the references concentrations (10 µg chl *a* $l^{-1}$and 600 µg TN l^{-1}, respectively) in the late 1800s and the early 1900s. According to the WFD, Laajalahti Bay appears not to meet the environmental quality objectives because of internal loading. This means that management decisions will be required to restore the community structure present at the beginning of the last century. The expense and advantages of restoration activities must be considered in river basin management plans. If it proves unreasonably expensive to achieve a good status, the directive may set less stringent environmental objectives, especially if further deterioration has already been prevented, as is the case in Laajalahti Bay. However, internal loading delays recovery from pollution in lakes and coastal systems, and this must be taken into account in management plans. As a whole, more information on the effects of restoration activities on nutrient dynamics and eutrophication processes are needed for decision-makers who implement the WFD or other environmental protection programmes.

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