Biostratigraphical and geochemical evidence for late Holocene water quality and limnoecological changes in a small humic lake in southern Finland

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Multiple anthropogenic stressors on lake ecosystems demand effective measures towards improved protection and management of water bodies. The Water Framework Directive defines a common goal for sound water management and obliges EU member countries to monitor and protect the ecological status and water quality of all relevant surface waters. However, major problems hindering the attempts for effective water management are the lack of long-term observational data on reference status and an inadequate understanding of the responses of lake ecosystems to environmental pressures. With this regard, paleolimnological techniques are invaluable as they provide means to seek for past analogies of lake-environment interactions.

Long-term development in the water quality and ecological status of Lake Storträsk, located in southern Finland, was assessed using a variety of paleolimnological proxies. The aim was to determine the reference status of this dystrophic lake, and to attain holistic understanding of late Holocene environmental changes and their influences on the lake's status. The principal hypothesis of the study was that late Holocene climate changes, catchment development and contemporary human activities have affected the status of the lake leaving records in the abiotic and biotic features of the sediment deposits.

A 43-cm sediment core was obtained from the lake basin and studied for its physicochemical and biological attributes. The core was dated with radiometric methods, and a time frame of ca. 4500 years was established for the sediment sequence. Loss-on-ignition and magnetic susceptibility were measured and the elemental content of the sediment was assessed by ICP-MS- and CNS-analyses. Fossil diatom assemblages were studied to reconstruct long-term development in lake-water pH. Ordination techniques and diversity indices were applied to identify temporal patterns and relationships in the bioassemblages, and Spearman's correlation coefficients were calculated to assess statistical relationships between the studied abiotic and biotic parameters.

The results reveal that, whereas long-term climate changes have had the most profound impact on the water quality and ecology of Lake Storträsk, the status of the lake has also been altered by early catchment disturbances and historical human activities as well as more intensive anthropogenic disturbances after the establishment of intensive agriculture in the area. The base of the sediment core reflects the transition from the warm and dry Holocene Thermal Maximum to the cooling and increasingly humid late Holocene. After the initial phase, a more stable development took place, disrupted by possible signs of forest fires and early clearance periods in the area. From the late Middle Ages onwards, human activities within the vicinity of the lake became more intensive leaving highly distinct marks in the sediment, but were receded towards the present. Regardless of the current location of Lake Storträsk in a conservation area, and the apparent inability of the barren catchment to support any intensive agricultural practices, it is clear that human influences on the lake have been significant.

The results of this study highlight the importance of long-term perspective in the assessment of lake reference conditions since lake ontogeny is often far from linear. Furthermore, this study emphasizes the importance of combining information from multiple paleolimnological proxies as they provide a more robust and holistic basis for understanding lake-environment dynamics particularly in humic boreal lakes which often respond to environmental change in distinct ways.

Avainsanat – Nyckelord – Keywords
Paleolimnology, reference status, late Holocene, dystrophy, Östersundom

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Tiivistelmä – Referat – Abstract

Ihmistoiminnasta aiheutuva järvien kuormituksen lisääntyminen edellyttää määrätietoista vesienhoidon suunnittelua sekä tehokkaita vesiensuojelun välineitä. Euroopan unionin vesipuitedirektiivi pyrkii osaltaan yhtenäistämään ja tehostamaan vesiensuojelun tavoitteita ja toimia edellyttäen vesistöjen tilan säännöllistä seurantaa sekä asettaen tavoitteita vesien hyvän ekologisen ja kemiallisen tilan saavuttamiseksi. Kestävän vesienhoidon esteenä on kuitenkin usein puutteellinen tieto taustatilasta sekä riittämätön ymmärrys järviekosysteemien ja ympäristön välisestä vuorovaikutuksesta. Paleolimnologiset menetelmät tarjoavat tärkeän välineen menneiden ympäristömuutosten tarkasteluun, sillä nämä muutokset heijastuvat usein järvisedimenttien fysikaalisissa, kemiallisissa ja biologisissa ominaisuuksissa.

Tutkimuksessa selvitettiin Etelä-Suomessa, Östersundomissa sijaitsevan Storträskin vedenlaadun sekä ekologisen tilan kehitystä myöhäis-holoseenin aikana. Pyrkimyksenä oli määrittää järven taustatila paleolimnologisin menetelmin, sekä tutkia myöhäis-holoseenin aikaisia ympäristömuutoksia ja niiden vaikutusta järven tilaan. Tutkimushypoteesina oli, että myöhäis-holoseenin ilmastonmuutokset, järven valuma-alueen kehitys sekä ihmistoiminta alueella ovat vaikuttaneet Storträskin tilaan heijastuen sedimentin piirteissä.

Järven syvännealueelta otettiin 43 cm pitkä sedimenttisarja, joka ajoitettiin radiometrisin menetelmin. Sedimentistä määritettiin hehkutushäviö sekä magneettinen suskeptibiliteetti, ja sedimentin alkuainekoostumusta tutkittiin ICP-MS -menetelmällä sekä hiilen, typen ja rikin pitoisuuksia CNS -analysaattorilla. Sedimenttisarjasta tehtiin lisäksi piileväanalyysi, jonka avulla rekonstruoitiin järviveden pH-muutoksia myöhäis-holoseenin aikana. Ordinaatioanalyysien, lajiston monimuotoisuutta kuvaavien diversiteetti-indeksien sekä korrelaatioanalyysin avulla pyrittiin hahmottamaan ajallisen muutoksen luonnetta sekä yhteyksiä tutkittujen abioottisten ja bioottisten muuttujien välillä.

Tutkimus osoittaa, että myöhäis-holoseenin ilmastokehitys on ollut pääasiallinen muutosta ajava tekijä Storträskin limnologisessa ja ekologisessa kehityksessä, mutta myös valuma-alueen luonnolliset sekä ihmistoimintaan liittyvät häiriöt ovat vaikuttaneet järven tilaan merkittävällä tavalla. Sedimenttisarjan alaosa edustaa siirtymävaihetta holoseenin lämpömaksimin ja myöhäis-holoseenin välillä ja poikkeaa ominaisuuksiltaan selkeästi järven myöhemmistä kehitysvaiheista. Myöhäis-holoseeniin siirtymisen jälkeen järven kehitys on ollut kohtalaisen tasaista, joskin myös merkkejä erilaisista valuma-alueen häiriöistä on tallentunut sedimenttiin. Erityisesti intensiivisemmän maatalouden leviäminen alueelle keskiajalla sekä teollistumiseen liittyvä raskasmetallilaskeuma ovat heijastuneet huomattavina muutoksina järven ekologisessa tilassa ja vedenlaadussa huolimatta järven soistuneesta ja kallioisesta valuma-alueesta, joka rajoittaa ihmistoimintaa järven välittömässä läheisyydessä. Tulokset osoittavat Storträskin reagoivan herkästi erilaisiin ympäristön paineisiin, minkä vuoksi järven tilan seuranta on ensiarvoisen tärkeää ihmistoiminnan lisääntyessä alueella.

Tutkimuksen tulokset korostavat pitkän ajan näkökulman merkitystä paleolimnologisessa tutkimuksessa ja vesistöjen taustatilan selvityksessä, sillä järviekosysteemien kehitys on usein kaukana lineaarisesta. Tulokset painottavat myös useiden paleoindikaattorien hyödyntämisen tärkeyttä, sillä ympäristömuutoksia ajaa tyypillisesti kirjo toisiinsa sidoksissa olevia muuttujia ja muutoksen ilmenemiseen vaikuttaa lisäksi eri järvityyppien sisäinen dynamiikka ja kyky puskuroida kuormitusta.

Avainsanat – Nyckelord – Keywords
Paleolimnologia, taustatila, myöhäis-holoseeni, dystrofia, Östersundom

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1. INTRODUCTION

1.1 Sedimentary research as a tool for effective lake management

Lake ecosystems have become subject to increased environmental pressure during the Anthropocene (i.e., the recent period of human interferences *sensu* Crutzen & Stoermer 2000) due to intensifying human activities since the beginning of the 19th century. While change is natural to all ecosystems, the rate of change has escalated in many areas to such extent that ecosystems are not able to adapt and may suffer significant ecological deterioration. The impacts of, for example, increased loading of pollutants and acidic compounds as well as enhanced nutrient input have been extensively studied (Smol 2002, Battarbee & Bennion 2011), yet more research is needed to gain better understanding of the complex dynamics controlling ecosystem responses to these stressors. Other processes, such as human-induced climate change, are still vastly unexplored and need to be further delved into. Climate change is suspected to have a growing impact on the structure and functioning of lake ecosystems, which highlights the urgency of achieving improved knowledge of lake-climate interactions (Wrona et al. 2006, Schindler 2009, Adrian et al. 2009, Battarbee & Bennion 2012). Understanding the variation in lake ecosystems, be it as a result of natural or anthropogenic causes, is of utmost importance if we are to fully comprehend how these ecosystems function and how they can be restored and protected (Smol 2002, Sayer et al. 2010). The EU Water Framework Directive requires European Union member countries to ensure good ecological and chemical status for all relevant water bodies by the years 2015-2027 (European Commission 2000, 2003), which further emphasizes the importance of assessing reference conditions to allow for sound restoration and management practices (Rosen et al. 2011, Bennion et al. 2011).

Lakes respond sensitively to environmental change and these changes are often reflected in the physical, geochemical and biological features of the sediments accumulating in lakes. Hence, sedimentary research plays a key role in deciphering the effects of increasing environmental pressure on aquatic ecosystems as sediments collect and unify environmental signals of both local and regional change (Battarbee et al. 2001a, Smol 2002, Cohen 2003, Battarbee & Bennion 2012). Furthermore, modern instrumental measurements and historical monitoring cover only a short time-span and do not solely provide adequate data for understanding the effects of human activity and environmental variability on lake ecosystems. In order to determine accurately the reference status, which provides the basis for setting goals within the Water Framework Directive, an understanding of long-term lake development is needed, for which reason paleolimnological techniques provide an invaluable tool for water management.

1.2 Multi-proxy approach in interpreting environmental signals

The responses of individual lakes to natural and human-induced environmental change are complex and distinguishing the effects of different sources of disturbances can be difficult (e.g., Korsman et al. 1994, Sayer et al. 2010, Battarbee & Bennion 2012). While climatic factors, for instance, can generally be regarded as fundamental drivers forcing changes on lake ecosystems, catchment characteristics and internal lake dynamics determine the ways in which different types of lakes respond to external stressors. Dystrophic lakes are abundant in the boreal region and hence understanding their particular dynamics is important. The high amount of refractory dissolved organic carbon (RDOC) and other humic, acidic compounds inherent to these lakes affect significantly the light, temperature and oxygen regimes in the water column (Wetzel 2001). Moreover, dystrophy poses controls on lake productivity, food web dynamics and predation regimes (Wissel & Boeing 2003, Korosi & Smol 2012, Zawiska et al. 2013), and hence affects the biotic signals recorded in the sediment. Furthermore, characteristics of the catchment, such as the extent and type of soil cover and vegetation as well as features of the bedrock and catchment topography, are significant with regard to lake response dynamics (Korsman et al. 1994, Virkanen et al. 1997). Employing multiple proxies in paleolimnological assessment provides tools for unraveling the complex and interdependent signals of different stressors on lake ecosystems, and allows for more comprehensive and precise interpretations of environmental change.

1.3 Objectives of the study

The overall objective of the study is to reconstruct long-term variation in the water quality and limnoecological status of Lake Storträsk with paleolimnological techniques, for the assessment of reference status of the lake. As a member country of the European Union Finland is obliged by the Water Framework Directive to monitor the ecological status and water quality of surface waters and to establish reference conditions for water bodies. Furthermore, the results should provide data for the monitoring of water quality in the area and contribute to the planning of protection of small water bodies in Finland. The detailed aims are 1) to explore the environmental gradients behind the observed changes, 2) to determine how, and to which extent, they are reflected in the physical, geochemical and biological features of the sediment, and 3) to disentangle the effect of different causative mechanism by employing a multi-proxy approach.

Particular interest is placed on large-scale climatic variations during the late Holocene and the effects of anthropogenic disturbances on the lake. With this regard, physicochemical features of the sediment are studied to identify lake-catchment interactions, for example, balance between allochthonous and autochthonous sources of organic material, weathering and erosion, and catchment vegetation. Biological assemblages are used to infer changes in water quality, particularly acid-base equilibrium and oxygen status, and also to explore variation in depth and stratification regimes, aquatic habitats and food web.

1.4 Principal hypotheses

The main hypothesis of the study is that late Holocene climatic variation and recent anthropogenic disturbances have affected the catchment characteristics, limnology and ecology of Lake Storträsk. It is hypothesized that these effects have left a record in the physical (organic content of the sediment, magnetic properties) and geochemical (trace element concentrations, nutrient components) features of the sediment. In addition, it is presumed that lake biota have been affected by these factors thus leaving a record in the biostratigraphy (community composition and abundance of aquatic organisms) of the sediment. The results are expected to improve the understanding of the relationship between late Holocene environmental variation and the development of lake ecosystems with special reference to small, dystrophic boreal lakes.

2. STUDY AREA

2.1 Study site

Lake Storträsk is located in Östersundom, a new subdistrict in eastern Helsinki on the southern coast of Finland (Figure 1a). The area, which formerly belonged to the adjacent municipalities of Sipoo and Vantaa, was consolidated to City of Helsinki in 2009 (Östersundom-toimikunta 2013). The district is fairly sparsely populated and the landscape is characterized by boreal forests, small population centers, agricultural land and peatlands. However, due to becoming a part of the City of Helsinki the area is currently under major development and is planned to become a dense detached house district. The development of Östersundom will likely have a significant effect on the streams and waters in the area.

The catchment area is relatively small (~60 ha) and comprises mostly exposed bedrock and a small *Sphagnum* bog around the north-western margin of the lake (Figures 1b, 2). The lake basin is shallow with a maximum depth of slightly over 5 meters and covers roughly 7 % of the catchment area. The threshold of the lake is 32.4 m above the present sea level. Storträsk has one small outlet channel; Gumbölenpuro (Kujala 2011), which originates from the southern end of the lake and drains southeast (Figures 1b, 2). The lake has no inlets and thus the hydrologic input is controlled by precipitation and water seeping through the surrounding peat deposits and mineral soil. The annual average flow of water in Gumbölenpuro is around 25 $l\ s^{-1}$ (Kujala 2011).

Storträsk is a naturally acidic lake; the pH and alkalinity measured from the epilimnion in May 2012 were 5.4 and <0.05 $mmol\ l^{-1}$, respectively. The buffer capacity of the lake is weak and there is little soil in the catchment area to replenish the base cation reserve of the lake. Storträsk has been treated with calcium carbonate ($CaCO_3$) during the past two decades (Esa Tiihonen, personal communication, 2013) to counteract the acidity, but due to the lake's natural tendency to acidification the effects do not last (Kujala 2011).

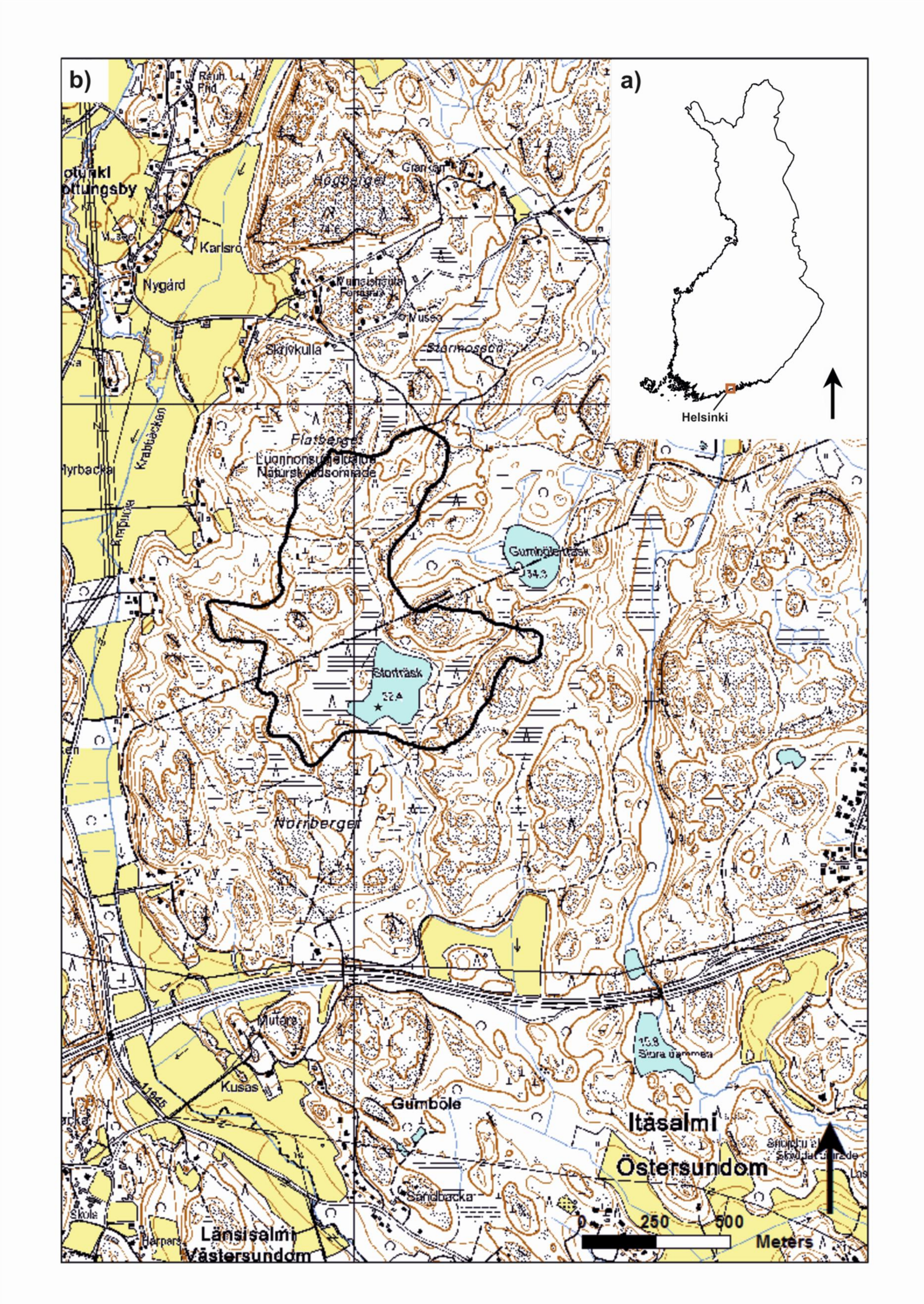

Figure 1. a) Location of Lake Storträsk in southern Finland and b) the catchment area (black line) adapted from Kujala (2011) and surroundings of the lake based on National Land Survey of Finland basic map 1:20 000 (PalTuli).

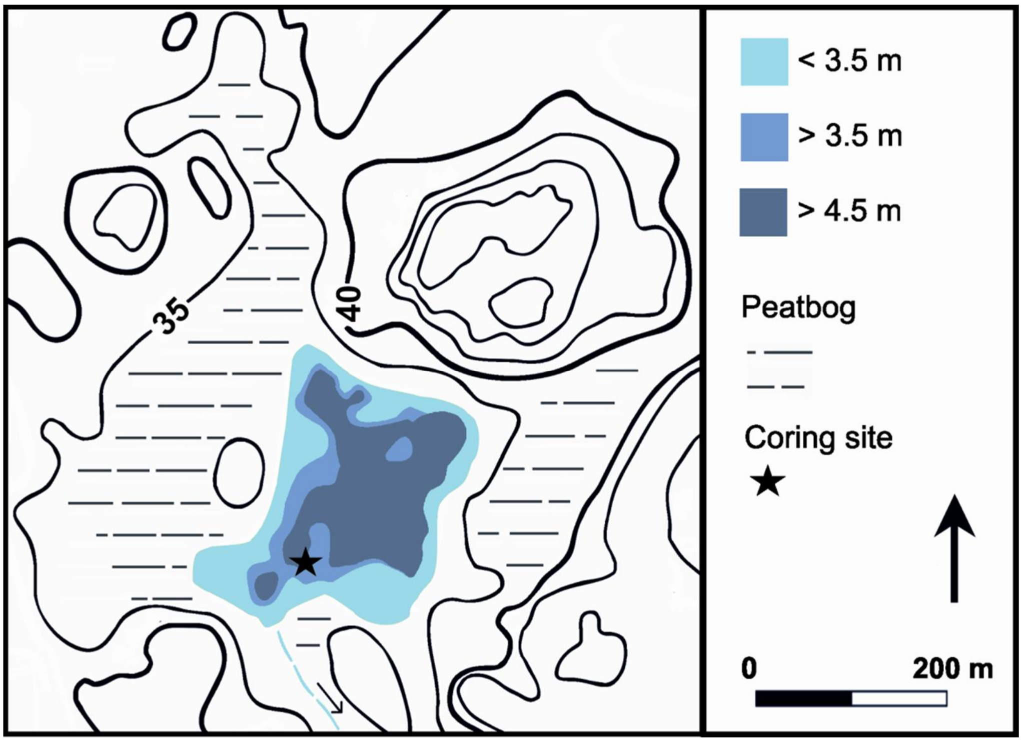

Figure 2. Rocky outcrops surrounding Lake Storträsk and basin morphology.

The first water quality measurements from the lake were performed in 1992 after which there has been significant variation in the observed pH (ranging between 4.5 and 6.9) and alkalinity (ranging between <0.05 and 1.44 mmol l^{-1}) (Finnish Environment Institute). The measurements do not, however, give an accurate picture of the natural pH and alkalinity variation due to the recent liming practices. The lake has a high content of humic acids originating mostly from decaying organic material in the surrounding peat deposits. The measured total organic carbon (TOC) of the lake was 19 mg l^{-1} in 2005 (Finnish Environment Institute) which is typical for polyhumic lakes. Hypolimnetic oxygen measured from the depth of ~4 m was 5.4 mg l^{-1} in May 2012. Total phosphorus concentrations in the lake have shifted between 10 and 19 µg l^{-1}, whereas total nitrogen concentrations show more variation, the values ranging from 530 to 1200 µg l^{-1} since 1992. The catchment area is not well suited for cultivation and hence the effects of contemporary agriculture on Storträsk remain low, although more extensive agriculture is practiced in the adjacent Krapuoja catchment (Kujala 2011). The catchment features and limnological variables in Lake Storträsk are presented in Table 1.

Table 1. Catchment features and limnology of Lake Storträsk. Limnological values are based on measurements from the epilimnion performed in May 2012.

Coordinates (ETRS-TM35FIN)		**pH**	5.4
N	6682978	**Alkalinity (mmol l^{-1})**	<0.05
E	398084	**Electrical conductivity (mS m^{-1})**	3.2
Catchment area (ha)	60.5	**Turbidity (FNU)**	1.3
Lake area (ha)	4.5	**Colour number (mg Pt l^{-1})**	160
Elevation (m als)	32.4	**Total phosphorus (µg l^{-1})**	16
Maximum depth (m)	5.1	**Total nitrogen (µg l^{-1})**	530

2.2 Geologic setting and catchment development

Lake Storträsk lies a few kilometers from the shore of the Baltic Sea and was formed as a result of land upheaval. Deglaciation occurred in the area about 13 000 years ago (Nenonen & Eriksson 2004), yet the area may have remained submerged until the end of the Anculys Lake phase. The lake basin was likely isolated from the Anculys Lake around 9000 years ago and was not affected by the Litorina transgression as the threshold limit of the lake is above the highest Litorina Sea limit in the region (Miettinen 2002).

The catchment comprises large areas of elevated bedrock cut through by narrow fault-line valleys, and the bedrock consists mainly of granite and quartz-feldspar gneiss (Kujala 2011). The depressions between the exposed bedrock are filled with thin deposits of till and peat. In addition, the stream Gumbölenpuro follows the hollows in the fractured rock. A larger fracture valley curves around the western margin of the catchment area and has been more intensively cultivated (Figure 1b).

After the emergence from the Baltic Sea basin, the vegetation in the area underwent a succession from seashore flora to a mixed conifer-deciduous forest with *Betula*, *Pinus* and *Alnus* as the dominant tree species (Sarmaja-Korjonen 1992). Broad-leaved trees (*Tilia*, *Quercus*, *Ulmus*, *Corylus*, *Fraxinus*) were also abundant. Norway spruce (*Picea abies*) spread to southern Finland and to the study region from the east about 3500 years ago and rapidly invaded the *Pinus-Betula-Alnus* forest (Sarmaja-Korjonen 1992). The vegetation of the study area is currently dominated by *Picea*, *Pinus* and *Betula* with *Picea* as the dominant species in the wetter areas.

2.3 Human activities

According to Sarmaja-Korjonen (1992), the area near Lake Storträsk was first influenced by humans about 2800 years ago. These disturbances were not, however, reflected in the sediment of Storträsk in the mentioned study, but in the sediment record of an adjacent Lake Hältingträsk few kilometers to the east. In addition, there is evidence of a short period of slash-and-burn cultivation during the Pre-Roman Iron Age (ca. 2300 years ago), which left a faint record in the sediment of Lake Storträsk. This early clearance period was followed by a long undisturbed period spanning from the beginning of the Iron Age to the beginning of the Viking Age. Intensive agriculture was not established in the area until about 1200 years ago. After the shift to agrarian economy, slash-and-burn cultivation was practiced until the beginning of the 20th century after which agricultural activities in the area have receded. (Sarmaja-Korjonen 1992).

Nowadays, land use in the area surrounding the lake is mainly dominated by agricultural activities. Lake Storträsk, on the other hand, has remained relatively undisturbed, and there is no extensive human activity in its instant vicinity. Nonetheless, the lake and its surroundings have important recreational value and will continue to provide ecosystem services in the future (Österdundom-toimikunta 2013). The lake was formerly located in a natural park and became part of the Sipoonkorpi natural park established in 2011 (Laki Sipoonkorven kansallispuistosta 325/2011). Hence the intensive development of the area should not bring about marked increase in direct external disturbance. The main human activities influencing the lake’s ecological state have been fish stocking and the treatment of water with calcium carbonate to neutralize the acidity of the water within the past twenty years (Esa Tiihonen, personal communication, 2013). Whitefish, trout, brook trout and rainbow trout have been stocked in the lake, rainbow trout being currently the most common species planted (Kujala 2011). Due to its location in the densely populated southern coast of Finland, the lake is subject to relatively high atmospheric loading of chemical substances through dry deposition (Verta et al. 1989, 1990, Skjelkvåle et al. 2001). Furthermore, the highway Porvoonväylä (E18) cuts Gumbölenpuro roughly 1 km south of the lake and may cause slight increase in particulate emission loading.

3. MATERIALS

3.1 Sampling and sample preparation

A 43-cm sediment core was obtained from the lake basin (depth ~4.3 m) with a Limnos-type gravity corer (Kansanen et al. 1991). The core was sliced after the sampling at intervals of 1-cm and packed into Minigrip® plastic bags. The collected samples were kept in an icebox during transport and were stored in a cold room at 4 °C.

A total of 32 samples were analyzed for sediment geochemistry, physical features and diatom assemblages. In addition, the topmost 21 samples were analyzed for radiogenic lead (^{210}Pb) and cesium (^{137}Cs) isotopes, and the lower half of the core was dated with AMS radiocarbon (^{14}C) method. Four macrofossils used in the ^{14}C dating were isolated from the sediment core during sub-sampling and were stored in distilled water and kept cool at 4°C. The subsamples for physical analyses, ^{210}Pb and ^{137}Cs analyses and diatom analysis were separated from each 1 cm segment of fresh sediment, after which the rest of the sediment was refrigerated and freeze-dried for ICP-MS and CNS -analyses. All analyses were performed contiguously at intervals of 1 cm to a depth of 20 cm. Of the lower part of the core (21−43 cm), every other sample was analyzed as more recent changes were considered to be of most interest. In addition to diatom compositional data, results from fossil Cladocera analysis (Nevalainen, L., unpublished) and chironomid analysis (Luoto, T.P., unpublished) performed on the same sediment samples were used to gain complementary information on the changes in lake biology and a wider perspective on the environmental change within the lake and its catchment.

Lake depth was measured with a portable depth sounder (Speedtech Depthmate Portable Sounder, Speedtech Instruments trademark by Campbell Scientific Inc., USA) in order to locate a suitable sampling site. In addition, more extensive measurements were made to gain a picture of the bathymetry of the lake basin (Figure 2). Water quality data from the Environmental Information System HERTTA (Finnish Environment Institute) were used to acquire information about the current status of the lake.

4. METHODS

4.1 Physical methods

4.1.1 Water content and loss-on-ignition

The water content of the sediment was determined by heating the weighted wet sediment samples (approximately 5 g) in an oven at 105 °C for 12 hours after which the samples were weighted. Loss-on-ignition (LOI), reflecting the organic content of the sediment, was measured by igniting the dried samples at 550 °C for two hours, and by comparing the weight with the previous value following Dean (1974). LOI values can be used to describe the quality of the sediment (Salonen et al. 2002) and to infer lake productivity and input of material from outside the lake (Nesje & Dahl 2001, Cohen 2003, Willemse & Törnqvist 2013).

4.1.2 Magnetic susceptibility

The magnetic susceptibility of the sediment was measured with a Bartington MS2B magnetic susceptibility meter (Bartington Instruments LTD, England). The samples were homogenized in the Minigrip® bags in which the samples were originally collected and the measurements were performed through the plastic. Magnetic susceptibility reflects the degree to which material is magnetized when subjected to an external magnetic field and may indicate a variety of environmental disturbances (Thompson et al. 1975, Oldfield et al. 1983).

4.2 Radiogenic dating methods

In order to determine a temporal context for the sediment profile, the core was dated using radiogenic techniques. The uppermost 21 cm (at 1-cm intervals) of the sediment core were dated at the Institute of Geological Sciences (Polish Academy of Sciences) measuring ^{210}Pb and ^{137}Cs activities in the sediment by gamma spectrometry (Ritchie & McHenry 1990, Zaborska et al. 2007, Pittaeurová et al. 2011). ^{210}Pb was used to establish a chronology for samples with ages less than ~100 years (Pittaeurová et al. 2011) and radiocesium to trace the signals of nuclear weapons testing in the 1960's and the Chernobyl accident in 1986 which are often recorded as distinct horizons in lake sediments (Klaminder et al. 2012).

From the lower parts of the sediment core, four terrestrial macrofossils were analyzed using accelerator mass spectrometer (AMS) radiocarbon dating. The samples at depths of 17 cm and 42 cm were analyzed at the Poznan Radiocarbon Laboratory, Poland, and samples found at the depths of 28 cm and 36 cm were analyzed at the Beta Analytic Inc. Laboratory, Miami. The measured radiocarbon ages were calibrated to calendar years with the Calib Radiocarbon Calibration program (Stuiver & Reimer 1993) based on Libby half-life (5568 years) and measured radiocarbon ages without $\delta^{13}C$ correction. In addition, Calib was used to interpolate estimated age ranges for the samples between 42 and 17 cm based on the acquired calibrated ^{14}C ages. Radiocarbon measurements based on terrestrial macrofossils are an effective and accurate method for determining chronologies for lake sediments compared to bulk sediment measurements which are more prone to errors caused by mixing of carbon from different reservoirs (Oldfield et al. 1997, Barnekow et al. 1998).

Sedimentation rates were estimated for the sediment profile based on the obtained radiogenic dates and sediment accumulation. The results of physical and geochemical analyses are, nevertheless, expressed as weight percentages and concentrations in dry sediment material as calculating reliable estimations for accumulation rates would have required more detailed sediment chronology.

4.3 Geochemical methods

4.3.1 ICP-MS analysis

Inductively coupled plasma mass spectrometry (ICP-MS) was employed to determine the concentrations of iron (Fe), manganese (Mn), arsenic (As), cobalt (Co), chromium (Cr), copper (Cu), nickel (Ni), zinc (Zn), vanadium (V), antimony (Sb), cadmium (Cd), mercury (Hg), lead (Pb) and phosphorus (P). The samples were digested in acid using microwave assisted acid extraction method EPA 3051 (EPA 1994) to release the analytes from the inorganic and organic matrices in the samples. To prepare the samples approximately 0.25 g of the freeze-dried sample material was measured into a Teflon digestion tube and 10 ml of concentrated nitric acid (HNO_3) was added. In addition, six duplicate samples (every fifth sample), six replicates of reference material and six blank samples were prepared in a similar manner. The reference material used in the analysis was Canadian Certified Reference Material LKSD-4. A total of 50 samples were subjected to microwave treatment after which the clear solutions were diluted 1:20 in sterile Milli-Q® water.

Both external and internal standardization methods were used to assess and to ensure the accuracy of the analysis (Virkanen 2007). Calibration range was determined based on typical concentrations of the measured elements in stream sediments in the area (Lahermo et al. 1996) as well as the concentrations of each analyte in the reference material (Table 2). A total of seven standards with descending order of ion concentrations and a blank standard were prepared from certified standard solutions with known analyte concentrations. Same reagents were added to the standard solutions in same proportions as in the samples. Internal standards were used to normalize the data and clear the effects of arbitrary errors and instrument drift. In addition, interferences related to the technique were minimized or corrected by adjusting instrument operating conditions. The squared correlation coefficients (r^2) of the obtained calibration curves were revised and the absolute concentrations in the dry sediment were calculated using blank-correction.

Table 2. Calibration range (blank standard excluded), certified reference material concentrations, and estimated background concentrations based on Lahermo et al. (1996).

	Calibration minimum ppm	**Calibration maximum** ppm	**Background minimum** ppm	**Background maximum** ppm	**Certified value** ppm
P	0.0001	5	0.625	2	1.635
Fe	0.002	100	12.5	75	33.75
Mn	0.0001	5	0.25	2	0.5375
Zn	0.0001	5	0.025	0.19	0.23625
Pb	0.0001	5	0.00625	0.038	0.11625
Cr	0.0001	5	0.01875	0.125	0.02625
V	0.0001	5	0.025	0.125	0.04
Ni	0.0001	5	0.00625	0.05	0.04
Cu	0.0001	5	0.00625	0.05	0.0375
Co	0.0001	5	0.00625	0.05	0.01375
As	0.0001	5	0.00125	0.05	0.015
Cd	0.0001	5	0.000375	0.0013	0.002375
Sb	0.0001	5	0.0000125	0.0025	0.001875
Hg	0.0001	5	0.000025	0.00025	0.0002375

4.3.2 CNS analysis

The weight percents of carbon (C), nitrogen (N) and sulphur (S) in the freeze-dried sediment were determined with Elementar vario micro cube CNS analyzer. Canadian Certified Reference Materials Project’s LKSD-4 was used as reference material for the analysis. In addition, the relation of carbon and nitrogen contents in the sediment, i.e., the C/N ratio was calculated as it provides a useful tool for estimating the origin of the organic material in the sediment (Wetzel 2001, Håkansson & Jansson 2002). Accordingly, the relative significance of autochthonous organic material (generated within the lake) and allochthonous organic material (derived from outside the lake) was assessed (Wetzel 2001).

4.4 Diatom analysis

The sediment core was analyzed for fossil diatoms in order to depict changes in the community composition and associated past environmental conditions. The analysis was performed following standard procedures as described in Battarbee et al. (2001a). For the preparation of diatom slides approximately 0.5 cm^3 of sediment was oxidized with water/hydrogen peroxide (H_2O_2) solution (25:50 ml) at 55 °C in order to remove organic matter from the sediment. Mineral debris was removed by decanting and washing with tap water after which small amounts of the cleaned diatom-water suspensions were dried and mounted with Naphrax® on the microscope slides. A minimum of 300 diatom valves were counted from each sample, and the identification was performed with a light microscope at 1000x magnification. Identification was based mainly on the diatom flora of Krammer and Lange-Bertalot (1986-1991), whom also the nomenclature follows.

4.5 Data analyses

4.5.1 Diatom-based pH reconstruction

The diatom relative abundance data was used to reconstruct past variation in the acidity (pH) of the lake since lake-water pH is regarded as one of the most significant factors affecting the distribution of diatom taxa (e.g., Birks et al. 1990, Weckström et al. 1997, Battarbee et al. 2001a). The calibration dataset used for the reconstruction was The Surface Waters Acidification Programme (SWAP) dataset comprising 169 lakes from Sweden, Norway, Scotland, Wales and English Lake District (Stevenson et al. 1991). The training set places emphasis on acidic waters, the pH values varying from 4.3 to 7.3 (mean 5.6 pH units), and was thus chosen for the pH reconstruction. The calibration dataset and associated diatom-pH inference model were available at The European Diatom Database (Battarbee et al. 2001b). Due to the large number of rare taxa, only 90 species of the initial 133 identified taxa were used in the reconstruction. The pH reconstruction was performed applying weighted averaging (WA) by inverse deshrinking regression (Birks et al. 1990, Birks & Simpson 2013). The inference model had jackknifed coefficient of determination (r^2) of 0.82 and root mean squared error of prediction (RMSEP) of 0.33 pH units, with average and maximum biases of 0.01 and 0.35 units, respectively. The reconstruction was performed with the Ernie software (Environmental Reconstruction using the EDDI diatom database) v. 1.2 (Juggins 2001). The reliability of the reconstruction was assessed by comparing the presence of taxa in the fossil assemblages to that of taxa in the modern training set. In addition, modern analogue technique (MAT) was used to illustrate the dissimilarity between the calibration data set and fossil samples based on squared chi-squared distance measure and 5 nearest analogs using the Ernie software (Juggins 2001). Furthermore, correlation between the diatom-inferred pH and primary axis values derived from a principal component analysis (PCA) on diatom assemblages was calculated to evaluate the significance of pH in controlling the composition of the diatom assemblages.

4.5.2 Ordination analyses

In addition to data on diatom assemblages, supplementary data on fossil chironomid assemblages (Luoto, T.P., unpublished) as well as data on cladoceran assemblages (Nevalainen, L., unpublished) in the sediment of Lake Storträsk were analyzed with statistical methods in order to gain a holistic understanding of changes occurring in the ecology of the lake. The chironomid and cladoceran stratigraphies are shown in Appendices 1–2. Detrended correspondence analysis (DCA) and principal component analysis (PCA) were used as indirect ordination methods to identify patterns and describe relationships between the biological communities and environmental variation. DCA summarizes variation in the assemblages along DCA axes and was used in this study with the interest of assessing the lengths of the compositional gradients which indicate the nature of variation in the taxonomic distribution (linear or unimodal). Accordingly, linear indirect ordination method, PCA, was chosen for further evaluation of relationships between bioassemblages and environmental gradients. Both analyses were carried out separately on all three biological communities, and PCA was also conducted on diatom species data combined with independent environmental variables. The analyses were run using the program CANOCO 4.52 (ter Braak & Šmilauer 2002). Both ordination analyses were performed on square-root transformed relative species abundance in order to attenuate the influence of species with high variance, and DCA was run with down-weighting of rare taxa to diminish their high influence on the ordination.

4.5.3 Diversity indices

Taxonomic diversity across the diatom, cladoceran and chironomid stratigraphies was estimated with a set of statistical measures describing different aspects of the assemblages' composition. Species richness was expressed as the number of taxa in each sample. In addition, Hill's N2 index was used to describe species evenness (Hill 1973, Tuomisto 2010) and Shannon's H index to depict entropy in the assemblages (Jost 2006). Species richness and Shannon index were calculated with the Paleontological Statistics (PAST) program (Hammer et al. 2001) and Hill's N2 values with C2 data analysis program (Juggins 2003).

4.5.4 Correlation coefficients

Spearman's rank correlation coefficients (R_S) and associated levels of statistical significance (p) were calculated to assess the relationships between physicochemical parameters (LOI, relative proportions of C, N and S, C/N ratio, magnetic susceptibility, trace metals and sedimentary P obtained from ICP-MS analysis, inferred hypolimnetic oxygen and anoxia, and diatom-inferred pH) and biological (PCA axis 1 and 2 scores for diatom, chironomid and cladoceran assemblages) data. Calculations were made using the PAST program (Hammer et al. 2001).

4.6 Sources of error and uncertainty

Sampling and sample preparation are always critical steps that expose samples to degradation. Utmost caution was therefore used to avoid contamination and deterioration of the samples. The used containers, instruments and reagents were of required purity level and suitable for each analysis. Furthermore, sampling was done with the Limnos gravity corer to ensure minimum disturbance to the sediment sequence.

Changes in sedimentation rate affect the results significantly and may cause distortion to the gradients of environmental variables expressed as concentrations or weight percents. Hence, data interpretation was made acknowledging the effects of changes in the sedimentation rate since reliable estimations of accumulation rates could not be calculated for reference due to the imprecision of the obtained chronology.

Paleoreconstructions are always merely estimations of the past and several sources of uncertainty are inherent to the techniques. For one, inaccurate identification of taxa due to the subjectivity of identification, taxonomic diversity and lack of references may lead to erroneous interpretations. Furthermore, the representativeness of the training set with regard to the studied sediment sequence as well as environmental parameters confounding the significance of the inferred variable significantly impact the eventual reconstruction. To reduce uncertainties, the pH reconstruction based on diatom assemblages was evaluated with statistical methods, as described above, and was examined in reference to other studied environmental parameters.

The sensitivity of ICP-MS analysis is a benefit, but also makes the method susceptible to disturbances. The common interferences are related to the occurrence of isotopes of different elements with equal mass (isobaric interferences), the formation of doubly-charged ions, and the formation of molecular ions between sample ions and matrix components (polyatomic interferences) (Virkanen 2007). These interferences were minimized by adjusting the method as described above.

5. RESULTS

5.1 Physical properties of the sediment

5.1.1. Lithology, water content and loss-on-ignition

The sediment core consists mostly of homogenous gyttja with high organic content and was classified based on LOI values applying Salonen et al. (2002) (Figure 3). The water content of the sediment varies between 92 and 95 % and follows a trend similar to that of LOI apart from the two uppermost samples with clearly elevated water content (Figure 3). Loss-on-ignition values are lowest near the base of the core, the values ranging from 55 % to 59 % between 42 and 36 cm (~4500–2800 cal yr BP). After the minimum values, LOI increases markedly to a value of 74 % at 32 cm (ca. 2200 cal yr BP). The rise is followed by a more gradual increase, and the overall trend is bent to a decline after the middle of the profile (~20 cm, ca. 900 cal yr BP).

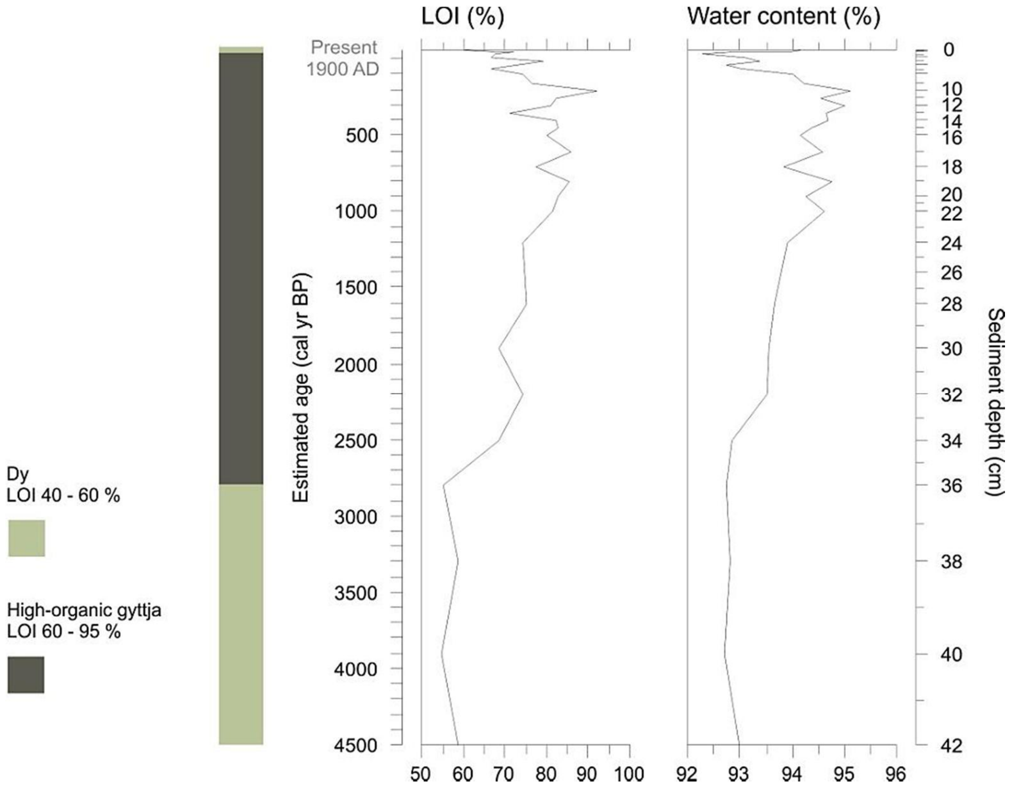

Figure 3. Lithology, loss-on-ignition (LOI) and water content in the sediment profile of Lake Storträsk.

In the upper half of the sediment core the most marked peak in LOI occurs between ~12 and 9 cm (~300–150 cal yr BP) with the core maximum of 92 % reached at the depth of 10 cm. The values vary markedly in the uppermost samples but the overall trend is towards values similar to those at the bottom of the core.

5.1.2 Magnetic susceptibility

The magnetic susceptibility of the sediment fluctuates substantially with no clearly distinguishable patterns (Figure 4). The minimum value of -0.73 SI occurs at the depth of 14 cm (~400 cal yr BP) and the maximum of 2.13 SI is reached in the topmost sample. The value in the surface sample is anomalous to some extent as most of the values remain below 1 SI (mean 0.3 SI). In addition, phases of clearly elevated values can be detected around the depths of 32 and 30 cm (~2200–1900 cal yr BP), and 22 and 19 cm (~1000–800 cal yr BP).

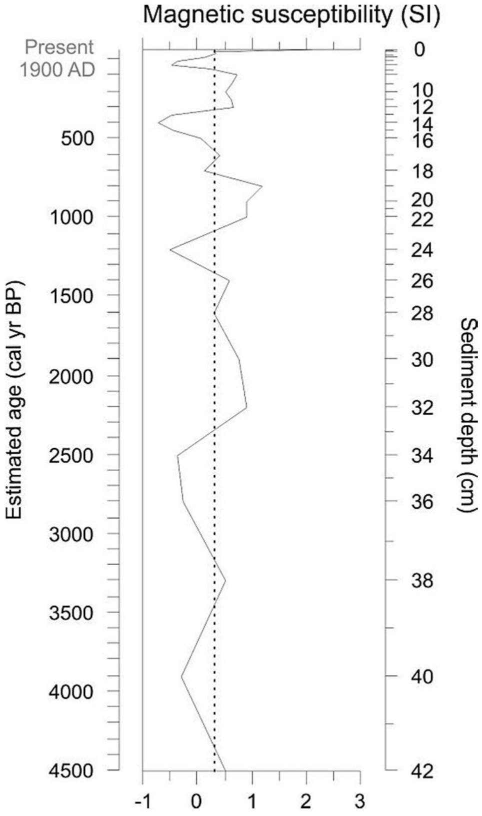

Figure 4. Stratigraphic variation in magnetic susceptibility in the sediment profile of Lake Storträsk. Mean value (0.3 SI) is indicated with a dotted line.

5.2 Temporal context

^{210}Pb-chronology could not be obtained for the upper half of the sediment core due to insufficient amount of sample material for the gamma spectrometric analysis. However, ^{137}Cs activities for the topmost 12 cm (samples 0–11 cm) were retrieved (Figure 5a). The results show a notable peak at the depth 3–4 cm probably associated with the Chernobyl nuclear accident horizon (1986) indicating that the uppermost samples represent recent accumulation. No clear signs of the nuclear weapons testing can be seen, which might be due to dilution of the signal by non-active material or the magnitude of the Chernobyl horizon obscuring the effects of nuclear weapons fallout (Klaminder et al. 2012).

Based on radiocarbon dating, the lowermost sample of the core (42–43 cm) represents an age of ca. 4500 cal yr BP, and the uppermost macrofossil from 17–18 cm yielded an age of ca. 600 cal yr BP (Table 3).

Table 3. Radiocarbon dates of terrestrial macrofossils from Lake Storträsk sediment core.

Depth (cm)	Dated material	Lab. No.	Age (^{14}C yr BP)	Median probability (cal yr BP)	2σ range (cal yr BP)
17–18	bark	Poz-49366	578 ± 30	600	532−646
28–29	bark	Beta-333963	1670 ± 30	1570	1505−1658
36–37	tree twig	Beta-333964	2660 ± 30	2778	2738−2820
42–43	pine needle	Poz-49364	4018 ± 30	4488	4407−4576

An age-depth model was constructed for the sediment profile based on the obtained ^{14}C ages and ages interpolated with Calib for samples between 42 and 17 cm, and based on the ^{14}C age at the depth of 17 cm and ^{137}Cs horizon at 3 cm for samples between 16 and 0 cm (Figure 5a). In addition, sedimentation rates based on the radiogenic age estimates and sediment accumulation are presented in Figure 5b.

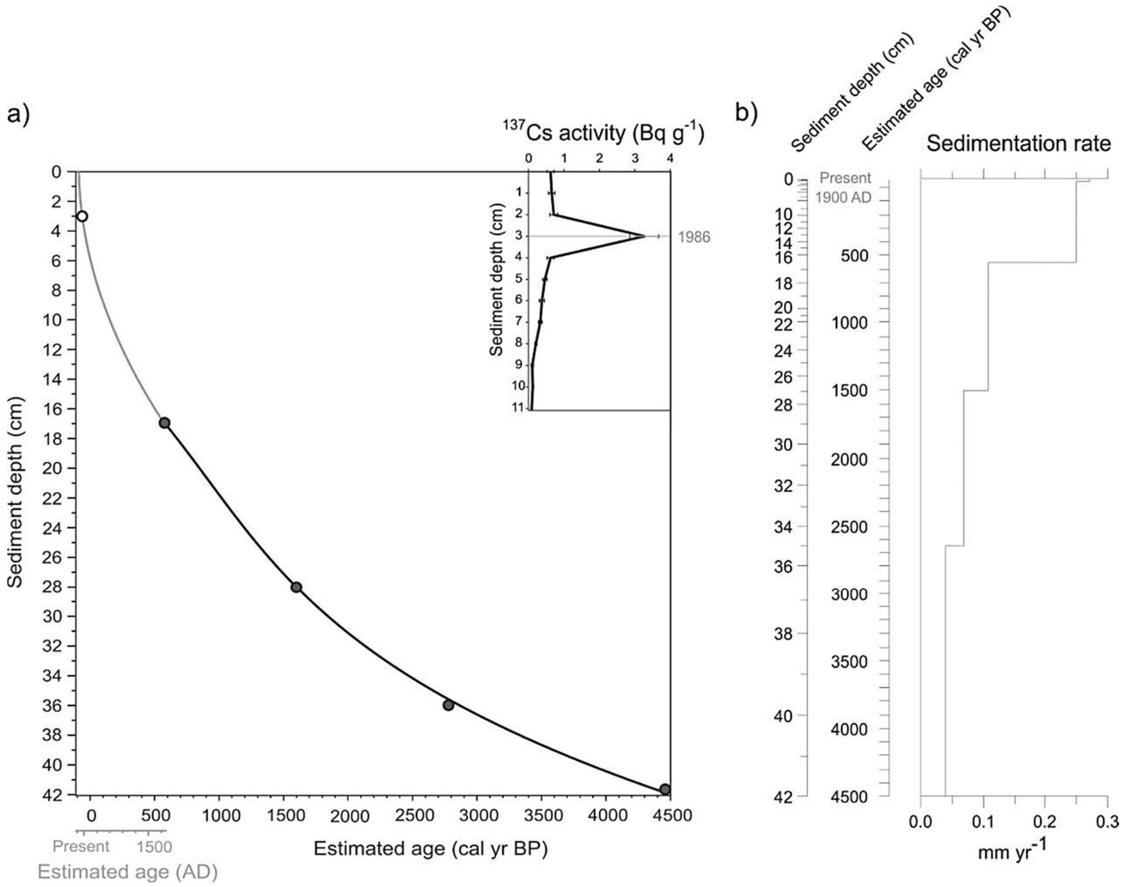

Figure 5. a) Age-depth model for Lake Storträsk sediment core. The filled circles represent ^{14}C dates for macrofossil plant remains at the depths of 42, 36, 28 and 17 cm and the blank circle represents the Chernobyl horizon suggested by ^{137}Cs chronology. Third order polynomial trend lines between 42 and 17 cm (black line) and between 17 and 0 cm (grey line) are shown. In addition, ^{137}Cs activity in the sediment core is shown. b) Rate of sedimentation in Lake Storträsk based on the radiogenic age estimates.

5.3 Geochemistry of the sediment

5.3.1 ICP-MS results

The studied elements display variable patterns along the sediment profile, although clear similarities can also be detected (Figure 6). The base of the core is characterized by elevated sedimentary P and slightly elevated values of Mn. Other elements have values close to the median value of each profile, apart from Pb and As which display minimum concentrations in the lowermost samples. Most of the trace metals close to their median concentrations show a declining trend between 42 and 40 cm (~4500–3900 cal yr BP) followed by slightly increased values between ~36 and 30 cm (~2800–1900 cal yr BP). Sedimentary P and Mn continue to decrease up to the surface with minor variation, whereas Pb begins to increase near the depth of 30 cm (~1900 cal yr BP) and more clearly after the middle of the core. Many of the trace metals show a more or less clear decrease in concentrations roughly between ~15 and 11 cm (~450–250 cal yr BP), which is most evident in the curves of Fe, V, Cu and Cr and barely detectable in Zn, Ni and As. After the decline, concentrations of all elements, apart from P and Mn, begin to increase rapidly, reach values preceding the drop between ~11 and 7 cm (~250 cal yr BP–early 20th century), and continue towards maximum values near the depth of 3 cm (late 20th century). The peak values are followed by a slight decline in most of the metals except for Fe which reaches its maximum value in the topmost sample.

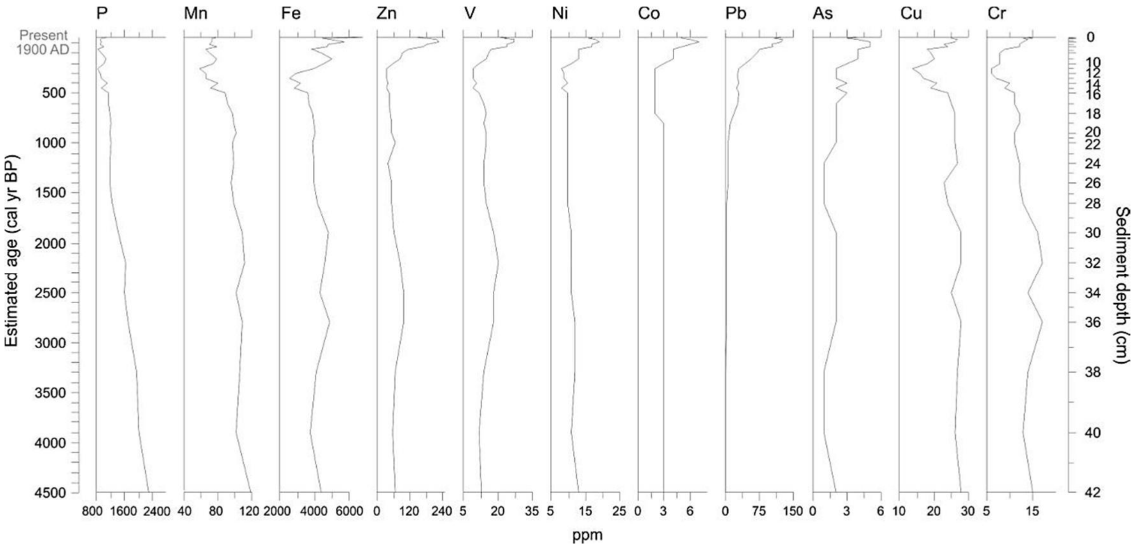

Figure 6. Stratigraphic variation in elemental concentrations in the sediment profile of Lake Storträsk.

The correlation coefficients (r^2) of the calibration curves obtained from the ICP-MS analysis had a good linear fit. The conversion of the initial results into absolute concentrations in dry sediment was done based on the measured dry weights of each sample and the median value of the six replicate blank samples used in blank correction. All measurements exceeded the assigned minimum detection limits (DL*3) which were based on the instrumental detection limits (DL). However, the concentrations of Cd, Sb and Hg were such diminutive in contrast to background levels in the analysis that the patterns were clearly interfered, and these elements were thus disregarded from the results.

The reliability of the analysis was assessed by examining the measured values of the six replicate reference samples and by comparing the obtained concentrations with the provisional values (Table 4). The deviation between the measured and provisional concentrations of the reference samples remained within the assigned limit of 90−110 % for Mn, Pb, Cr, V and Ni, whereas the yield for As slightly exceeded this limit. The measured value of Co was considerably lower than the provisional value (75 %) and also yields for P, Fe, Zn and Cu were below 90 %. Accordingly, the concentrations for elements for which the yield exceeded 100 % are probably slightly exaggerated, whereas the concentrations of elements with a yield below 100 % are probably slightly underestimated.

Table 4. Measured concentrations of the reference samples (LKSD-4 a…f) and reference samples average concentrations, relative standard deviations (RSD), provisional concentrations and respective yields.

	LKSD-4 a	**LKSD-4 b**	**LKSD-4 c**	**LKSD-4 d**	**LKSD-4 e**	**LKSD-4 f**	**Average**	**RSD**	**Certified value**	**Yield**
	ppm	ppm	ppm	ppm	ppm	ppm	ppm	%	ppm	%
P	1039.4	851.2	1215.7	1155.1	1131.5	1130.3	1087.2	11.8	1308	83
Fe	22750.2	18664.1	25692.5	24710.0	24655.5	24878.0	23558.4	11.0	27000	87
Mn	405.0	328.2	461.4	441.9	443.9	448.4	421.5	11.7	430	98
Zn	165.1	135.9	183.9	175.0	172.3	173.2	167.6	9.9	189	89
Pb	82.8	69.7	93.8	87.8	89.2	89.5	85.5	9.9	93	92
Cr	19.5	15.6	22.1	21.3	20.9	20.8	20.0	11.6	21	95
V	32.5	26.4	37.1	35.8	35.5	35.5	33.8	11.7	32	106
Ni	29.2	23.8	31.8	30.5	30.1	30.3	29.3	9.5	32	92
Cu	25.6	21.2	27.7	26.2	25.6	26.1	25.4	8.6	30	85
Co	8.2	6.7	9.0	8.6	8.5	8.6	8.3	9.8	11	75
As	13.2	10.6	14.7	14.1	13.8	14.0	13.4	10.9	12	112

The variation in the measured concentrations between the six replicate reference samples and between the duplicate samples from the sediment core was moderately low suggesting a relatively good precision for the analysis. The relative standard deviations (RSD) for reference samples are shown in Table 4 and for the duplicate samples in Table 5.

Table 5. Average, minimum and maximum relative standard deviations (RSD) for the duplicate samples of Lake Storträsk sediment core.

	P	**Fe**	**Mn**	**Zn**	**Pb**	**Cr**	**V**	**Ni**	**Cu**	**Co**	As
Average (%)	3.4	3.3	3.9	7.0	5.3	4.6	3.5	3.7	3.8	3.9	4.3
Min (%)	0.3	0.8	1.1	1.1	0.5	0.3	0.7	1.0	0.8	0.4	0.0
Max (%)	8.6	8.1	10.0	14.1	9.3	9.7	8.4	8.9	9.2	9.4	12.1

5.3.2 CNS results

The variation in the relative carbon and nitrogen content of the samples follow a very similar pattern (Figure 7). The base of the core is characterized by minimum values of both elements followed by a prolonged steady increase to maximum values of 41.3 % (C) and 2.5 % (N) near the middle of the core between ~22 and 20 cm (~1000–900 cal yr BP). After the peak values, C and N contents begin to decline and the decrease is leveled near 5 cm (~mid-20th century). Relative sulphur content in the sediment shows a declining trend between 42 and 40 cm (~4500–3900 cal yr BP) but begins to increase after the minimum value (0.41 %) at 40 cm. The increase remains somewhat constant until the depth of 11 cm (ca. 250 cal yr BP) where the relative S content increases sharply. The values remain high throughout the rest of the core, albeit a minor decline can be distinguished near the surface. The maximum value (1.02 %) is reached at 4 cm (~mid-20th century).

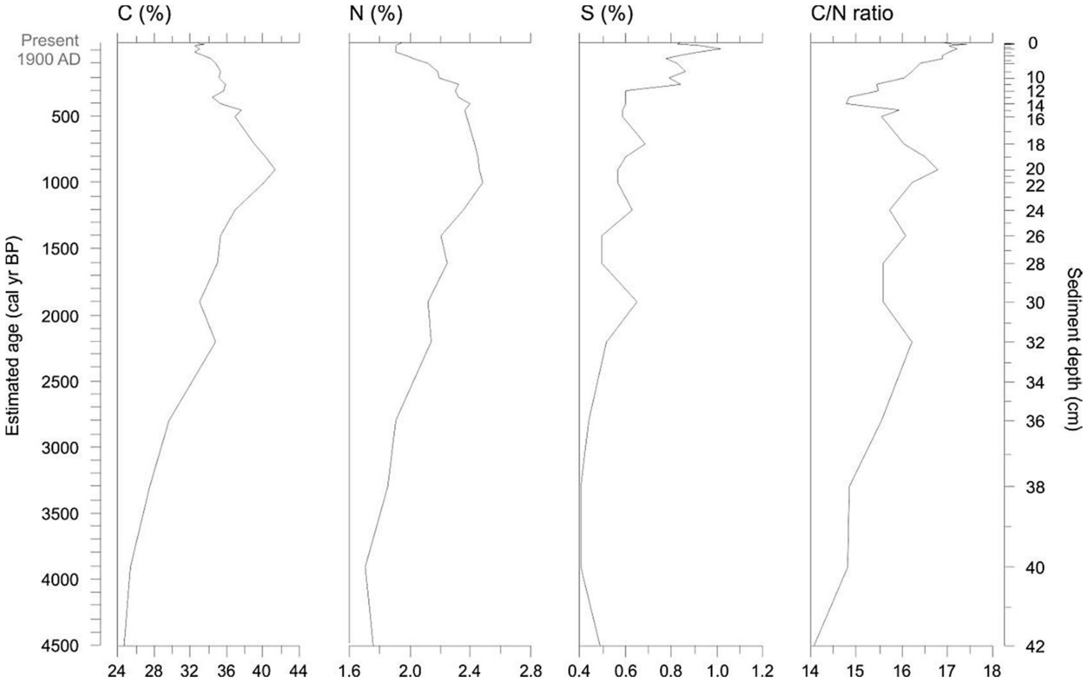

Figure 7. Stratigraphy of the relative amounts of carbon, nitrogen and sulphur and C/N ratio in the sediment samples of Lake Storträsk.

The C/N ratio in the sediment (Figure 7) varies between 14.0 and 17.4, the lowest values characterizing the base of the core and the highest values inherent to the topmost samples. The values increase gradually between ~42 and 32 cm (~4500–2200 cal yr BP) after which fluctuation increases. The increasing trend is disrupted by a decline between ~18 and 9 cm (~700–150 cal yr BP). The lowest values are reached around 14 and 13 cm (~400 cal yr BP) but are shortly followed by a rapid increase which continues up to the surface.

5.4 Diatom stratigraphy

A total of 133 taxa were identified from the sediment core and most abundant taxa were *Tabellaria flocculosa* (average abundance 18 %, number of occurrences 32), *Aulacoseira ambigua* (8 %, 31) and *A. distans* (7 %, 32). In addition, *A. lirata* (5 %, 27), *Frustulia rhomboides* (4 %, 32), *Pinnularia interrupta* (3 %, 31), *P.gibba* (2 %, 32), *P.viridis* (3 %, 32) and *Eunotia* spp. were present throughout the sediment profile. The relative abundances of most common diatom taxa are presented in Figure 8.

The base of the core (42–38 cm, ~4500–3300 cal yr BP) is characterized by the high abundance of *Aulacoseira distans* (~10–30 %), *A. ambigua* (~3–10 %), *P. interrupta* (~7–15 %), *Neidium ampliatum* (~4–7 %) and *N. affine* (~4 %). In addition, *Stauroneis anceps*, *Pinnularia legumen*, *Aulacoseira perglabra* and *Cymbella elginensis* are relatively well represented, their proportional abundances varying between ~1–4 %. The abundance of the *Aulacoseira* and *Neidium* species begin to decline markedly after the maximum abundances in the lowermost samples, and the latter genus nearly disappears from the profile after 38 cm.

Between 36 and 30 cm (~2800–1900 cal yr BP), the taxa dominant in the lowermost samples decline markedly whereas many taxa which were absent or scarce at the bottom of the core begin to increase. *Pinnularia gibba* (~3–5 %), *S. anceps* (~3–9 %), *Eunotia incisa* (~3–6 %) and *Anomoeoneis* spp. (~3 %) reach their maximum abundances in the stratigraphy but begin to decline after this phase. On the other hand, many taxa, such as *Aulacoseira lirata*, *Tabellaria flocculosa*, *Pinnularia viridis* and several taxa from the genus *Eunotia*, begin to increase after ~36 cm and remain relatively well represented throughout the rest of the profile. In addition, *Cymbella ehrenbergii* and *C. gracilis* become more abundant.

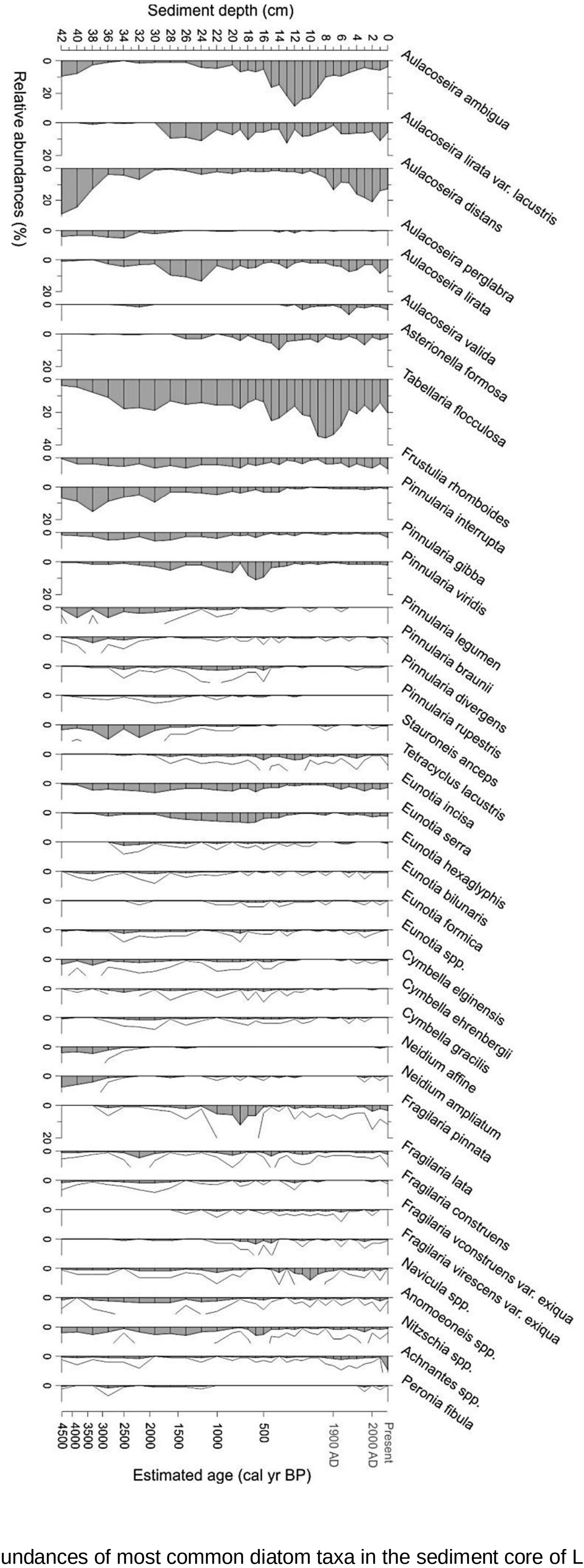

Figure 8. Relative abundances of most common diatom taxa in the sediment core of Lake Storträsk.

Near the depth of 28 cm (~1600 cal yr BP) *Aulacoseira lirata* var. *lacustris* (varying between ~4–13 % in the rest of the core), *A. lirata* (~2–10 %), *Eunotia serra* (~1–6 %) and *Asterionella formosa* (~0–6 %) increase clearly. On the contrary, taxa such as *P. legumen, S. anceps* and *A. distans* decline, the former two remaining scarce up to the top of the core. Between ~15 and 9 cm (~450–150 cal yr BP) a significant increase is seen in the abundance of *A. ambigua* (~15–30 %) and a slight increase also in *A. formosa* (~4–10 %). *T. flocculosa* is also present in high numbers and continues to dominate throughout the rest of the profile with an abundance of ~15–35 %. A decline in the abundance of several taxa occurs simultaneously with the sudden increase in *A. ambigua* and *T. flocculosa*. However, around the depth of 7 cm (~early 20th century) several species, such as *A. distans*, *A. lirata, E. incisa* and *E. serra*, recover distinctly. No particularly striking changes take place at the surface apart from a peak in *Achnantes* (namely, *A. minutissima*) in the topmost sample (~8 %).

5.5 Data analyses

5.5.1 Diatom-based pH reconstruction

The diatom-inferred pH variation in the sediment sequence (Figure 9) exhibits relatively large-scale fluctuation, the values ranging from 5.4 to 6.2 pH units. At the base of the core pH values decline towards increased acidity and remain low (pH ~5.5) for nearly a millennium between 36 and 30 cm (~2800–1900 cal yr BP). The minimum value is reached at the depth of 34 cm (ca. 2500 cal yr BP) from which point the values increase gradually from 5.4 at 34 cm to 5.7 at 16 cm (ca. 500 cal yr BP). A brief peak can be detected from the otherwise relatively even trend at the depth of 19 cm (ca. 800 cal yr BP) after which the long-term gradual increase is disrupted by a more pronounced rise in lake-water pH beginning near the depth of 15 cm (ca. 450 cal yr BP) and reaching the maximum pH value of 6.2 at 12 cm (ca. 300 cal yr BP). After the peak values pH decreases rapidly and becomes stabilized near ~8 cm (~late 19th century). The uppermost samples show no marked changes in lake-water pH and the values represent the median of the profile (~5.7).

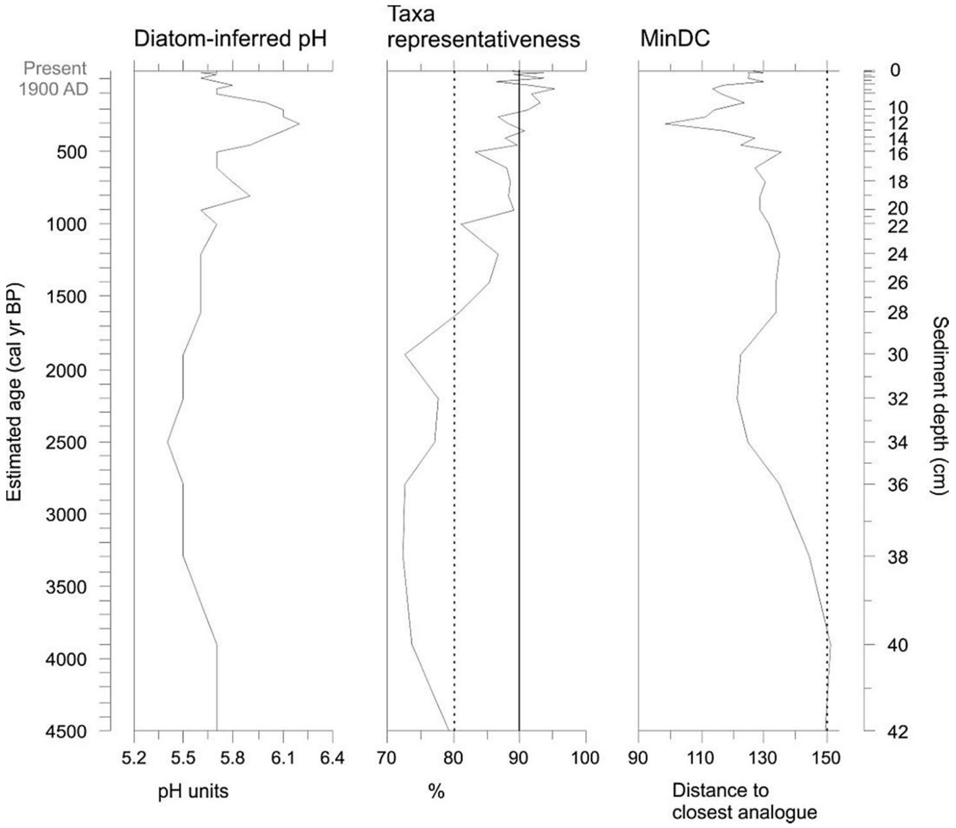

Figure 9. Diatom-based reconstruction of past lake-water pH changes in Lake Storträsk sediment profile. In addition, the proportion of fossil taxa in the training set (cut-off values at 80 % [dotted line] and 90 % [solid line]) and minimum dissimilarity values (minDC) in Lake Storträsk are shown. Samples with minDC values over 150 have poor analogues in the training set.

In most of the samples the taxa in the training set accounted for >80 % of the taxa present in the core samples, and nine samples out of 32 reached representativeness of >90 % (Figure 9). The seven lowermost samples failed to reach the representativeness of >80 % with values ranging between 72 % and 79 % due to the relatively high abundance of species, such as *Pinnularia interrupta*, *P. gibba* and *Neidium ampliatum* which are not present in the SWAP calibration dataset. Minimum dissimilarity values (MinDC) obtained from modern analogue technique (MAT) analysis (Figure 9) indicate that the chosen reconstruction model provides relatively credible predictions of the variation in the pH of the lake (Jones & Juggins 1995).

5.5.2 Ordination analyses

The DCA yielded relatively short gradient lengths for both axes 1 and 2 for all of the bioassemblages (Table 5). All values fall below 2 SD units supporting the use of a linear method in further data analysis (ter Braak 2003). There is relatively little resemblance between the PCA on diatom, cladoceran and chironomid assemblages apart from the distinction of the lowermost three samples as a separate cluster in all of the ordination diagrams. The respective eigenvalues and cumulative percentage variances are shown in Table 5.

Table 5. Gradients lengths for diatom, chironomid and cladoceran stratigraphies in Lake Storträsk based on DCAs, and eigenvalues and cumulative percentage variance of species data in the PCAs.

	Diatoms		**Chironomidae**		**Cladocera**	
	Axis 1	Axis 2	Axis 1	Axis 2	Axis 1	Axis 2
DCA						
Gradient length	1.59	0.86	1.98	1.69	1.12	0.80
PCA						
Eigenvalue	0.314	0.181	0.211	0.158	0.466	0.213
Cumulative percentage variance of species data	31.4	49.5	21.1	36.9	46.6	67.8

A two-dimensional PCA biplot depicting temporal variation in the diatom species composition is presented in Figure 10. In addition, a PCA diagram based on the diatom compositional data was created to arrange samples and the studied environmental variables along PCA axes (Figure 11). The PCAs on diatom assemblages show a distinct separation of samples into four separate clusters along the two primary axes (Figures 10–11). The two gradients explain 49.5 % of the total variation in the diatom assemblages. Axis 2 separates the lowermost samples, from 42 to 38 cm (~4500–3300 cal yr BP), and the uppermost 15 cm (~450 cal yr BP–present) from the samples in between (36–16 cm, 2800–500 cal yr BP), whereas axis 1 describes a more unidirectional succession. The lowermost three samples have the highest scores for both axes and are associated with high sedimentary P and high levels of hypolimnetic oxygen. Diatom species *Aulacoseira distans*, *Neidium affine* and *N. ampliatum* are most strongly affiliated with this set of samples. The four samples in the lower right in the biplot (36–30 cm, ~2800–1900 cal yr BP) have lower scores with regard to axis 2 and are linked with elevated concentrations of the trace metals Cu, Cr, and Mn, and various taxa from the genera *Pinnularia*.

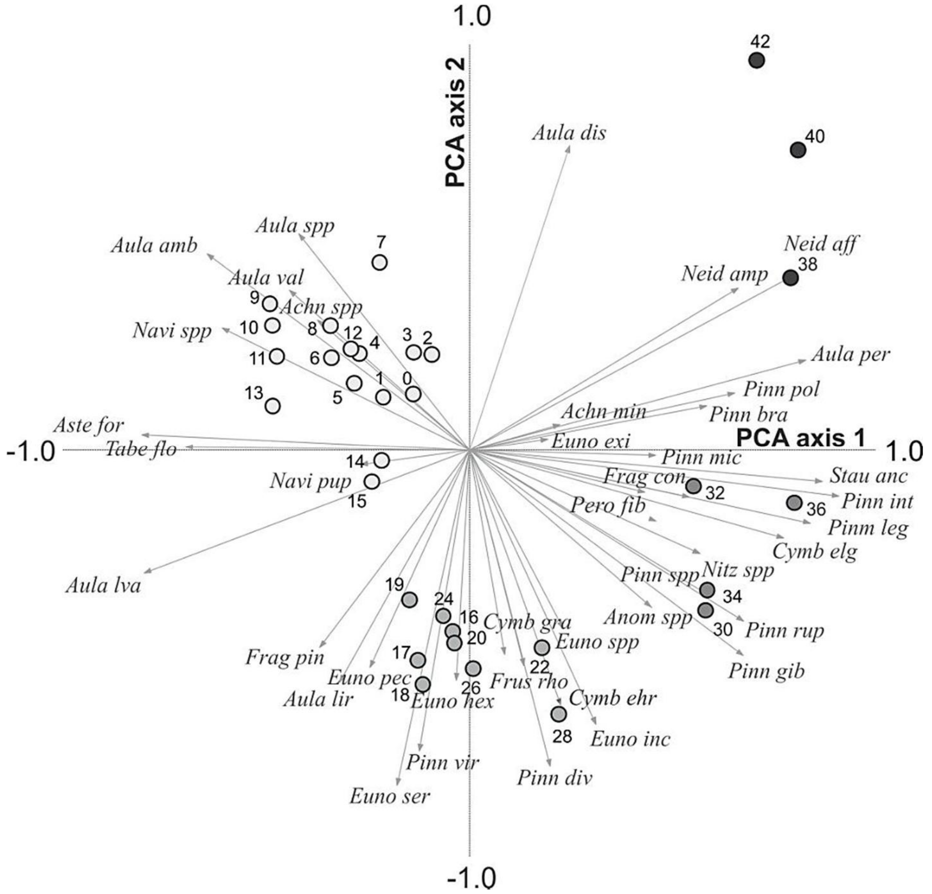

Figure 10. PCA biplot of diatom species (arrows) and samples (dots) in the sediment core of Lake Storträsk. The four separate sample clusters suggested by the analysis (samples 42–38, 36–30, 28–16 and 15–0 cm) are distinguished with different shades of grey. The abbreviations are based on the first four letters of the genus and the first three letters of the species name with the exception of *Aulacoseira lirata* var. *lacustris* (Aula lva).

The next cluster of samples (28–16 cm), describing a time period between ca. 1600 and 500 cal yr BP, shows a declining shift along the primary gradient and is characterized by high values of LOI, and high relative proportions of C and N. A variety of taxa from the genus *Eunotia* and *Cymbella* are inherent to this cluster of samples. The uppermost samples from 15 to 0 cm (450 cal yr BP–present) decline further along axis 1, but increase in axis 2 scores, though not to a level as high as in the lowermost samples. Elevated concentrations of S and various trace metals are characteristic to this phase, as well as diatom species such as *Aulacoseira ambigua* and *Asterionella formosa*.

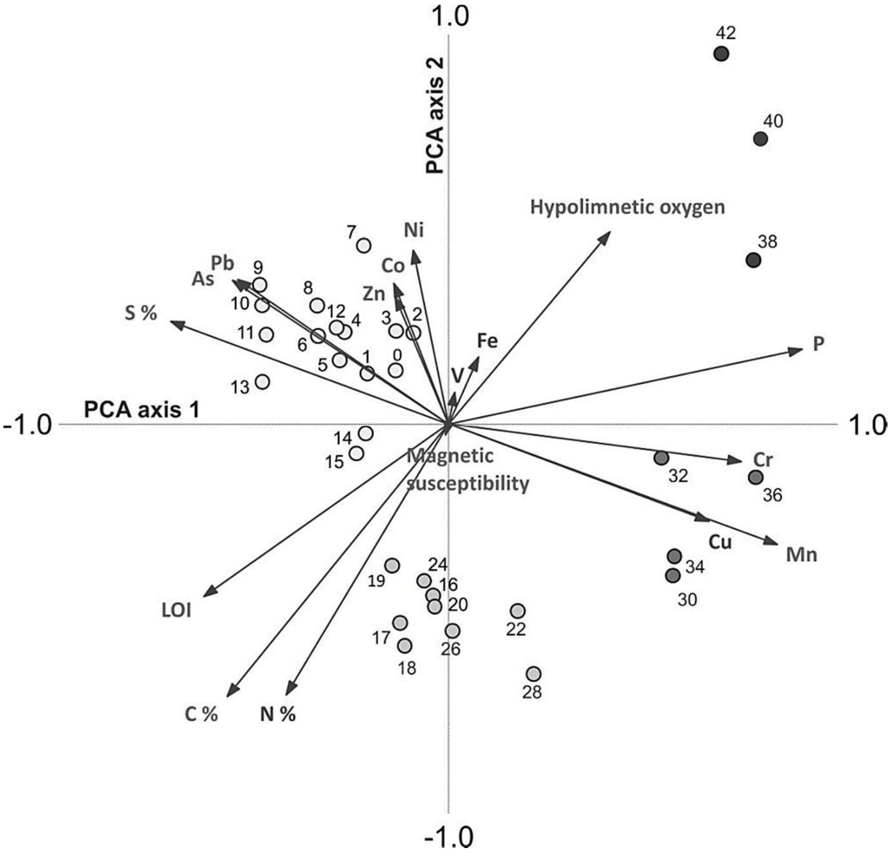

Figure 11. PCA biplot of samples (dots, based on diatom assemblages) and independent environmental parameters (arrows) in the sediment core of Lake Storträsk. The four separate sample clusters suggested by the analysis (samples 42–38, 36–30, 28–16 and 15–0 cm) are distinguished with different shades of grey.

The PCA diagram based on chironomid communities displays continuous variation along axis 1 and less extensive variation in axis 2 values apart from few distinctly separated samples (Figure 12). The two axes together explain 36.9 % of the total variance and the primary axis, along which most constant change occurs, accounts for 21.1 % of the variance. The lowermost samples between 42 and 36 cm (~4500–3300 cal yr BP) have highest axis 1 values and are characterized by high abundance of *Zalutschia zalutschicola*. The succession proceeds towards lower axis 1 scores, although the pattern is somewhat dispersed. Samples between 24 and 16 cm (~1200–500 cal yr BP) form a scattered cluster in the lower left corner of the biplot featuring high abundance of *Sergentia coracina*-type and *Tanytarsus lugens*-type. Samples between 13 and 8 cm (~350 cal yr BP–late 19th century) have clearly elevated axis 2 values, when compared with the rest of the profile, and are characterized by the abundance of a variety of taxa which are scarce elsewhere in the stratigraphy; for example, *Nanocladius rectinervis*-type and *Glyptotendipes pallens*-type (Appendix 1).

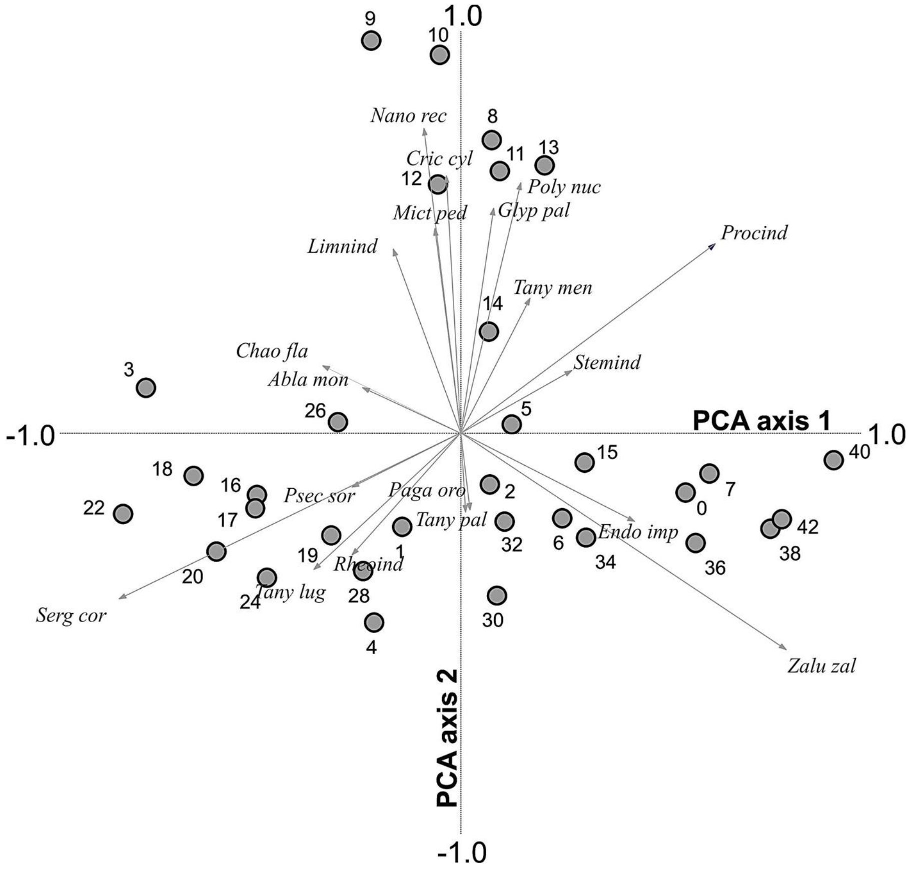

Figure 12. PCA biplot of samples (dots) and chironomid taxa (arrows) in the sediment core of Lake Storträsk. The abbreviations display the first four letters of the genus and first three letters of the species name with the exception of *Procladius* (Procind), *Stempellinella* (Stemind), *Rheotanytarsus* (Rheoind) and *Limnophyes* (Limnind).

In the PCA diagram on cladoceran assemblages, the distribution of samples along the primary gradient shows a succession from low axis 1 scores in the lowermost samples towards the surface with increasing axis 1 values (Figure 13). Axis 2 separates the lowermost samples between 42 and 38 cm (~4500–3300 cal yr BP) and uppermost six samples (6–0 cm, ~early 20th century–present) from the samples in between (36–7 cm, ~2800 cal yr BP–early 20th century), with the highest axis 1 scores reached in samples between 12 and 7 cm. Axes 1 and 2 explain 67.8 % of the total variation in the cladoceran communities. The lowermost three samples between 42 and 38 cm (~4500–3300 cal yr BP) have the lowest scores for both axes, and the taxa most distinctly associated with this cluster are *Eubosmina*, *Disparalona rostrata* and *Pleuroxus* *uncinatus*. The samples between 36 and 20 cm (~2800–900 cal yr BP) form a relatively uniform group with slightly higher axis 2 values and little change along axis 1. The most characteristic taxa in this set of samples are *Alonella exiqua* and *Leptodora kindti*, though both are generally scarce in the stratigraphy (Appendix 2). The following samples up to 7 cm (~early 20th century) are more sparsely distributed along both axis, although the strongest development occurs along axis 2. Samples between 13 and 7 cm (~350 cal yr BP–early 20th century), having the highest axis 2 scores, are characterized by *Alonella nana*, *Alonella excisa*, *Alona costata* and *Chydorus* cf. *sphaericus*.

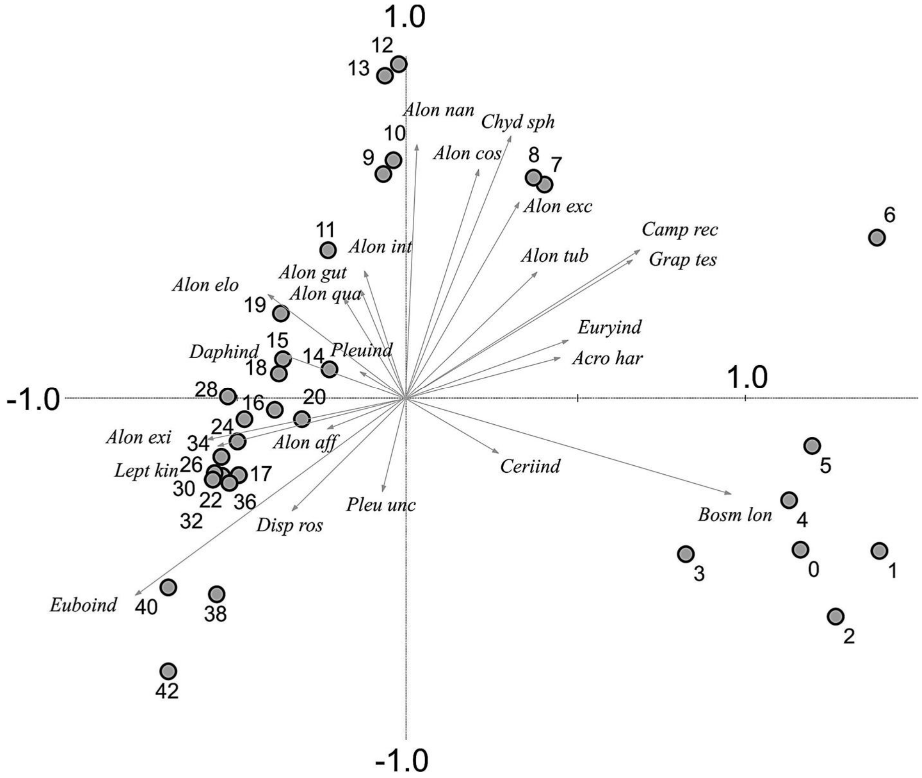

Figure 13. PCA biplot of samples (dots) and cladoceran taxa (arrows) in the sediment core of Lake Storträsk. The abbreviations display the first four letters of the genus and first three letters of the species name with the exception of *Eubosmina* (Euboind), *Daphnia longispina*-type (Daphind), *Pleuroxus* spp. (Pleuind), *Eurycercus* spp. (Euryind) and *Ceriodaphnia* spp. (Ceriind).

The sample at 6 cm (~early 20th century) with high axis 1 score is clearly separated from all other samples and associated with the cladocerans *Camptocercus rectirostris* and *Graptoleberis testudinaria*. The position of the sample is also linked to the notably low abundance of *Eubosmina* which is, accordingly, situated in the far opposite corner of the diagram. Samples between 5 and 0 cm (~mid-20th century–present) have also high axis 1 scores but lower scores for the secondary axis and are characterized by high abundance of *Bosmina longirostris*.

5.5.3 Diversity indices

Species richness in the diatom assemblage increases in the lowermost samples (~42–36, ca. 4500–2800 cal yr BP) after which the values begin a long-term gradual decline (Figure 14). Large-scale fluctuation is also inherent to the upper half of the core, though the overall declining trend is sustained. The number of taxa in the chironomid community increases more gradually and reaches maximum value at the depth of 11 cm (ca. 250 cal yr BP) from which point chironomid species richness declines (Figure 15). In the cladoceran community, the number of taxa present in the samples is slightly elevated at the base of the core and remains relatively constant throughout the lower half of the core (Figure 16). At the depth of 14 cm (ca. 400 cal yr BP) species richness increases notably, yet briefly, and is followed by a declining trend up to the surface.

Hill’s N2 and Shannon diversity indices follow a similar pattern in all biostratigraphies with few exceptions. Both species evenness and entropy are low at the bottom of the core in all of the assemblages (Figures 14–16) but increase rapidly in the diatom communities and more gradually in the chironomid stratigraphy. As with regard to species richness, the values in the diatom assemblages are most clearly elevated between 36 and 32 cm (ca. 2800–2200 cal yr BP) and remain high up to the middle of the profile, after which a clear decline can be detected (Figure 14). Both values, however, begin to increase again after ~7 cm (~early 20th century). In the chironomid community, highest evenness and entropy are reached between 15 and 9 cm (~450–150 cal yr BP) from which point a relatively stable trend continues up to the surface (Figure 15). The patterns in the evenness and entropy of the cladoceran stratigraphy differ clearly from those in the diatom and chironomid communities and also from that depicted by the cladoceran species richness. Both evenness and entropy increase slowly and with minor fluctuations until the depth of ~6 cm (~early 20th century) where particularly the evenness of the community is increased, though only temporarily (Figure 16).

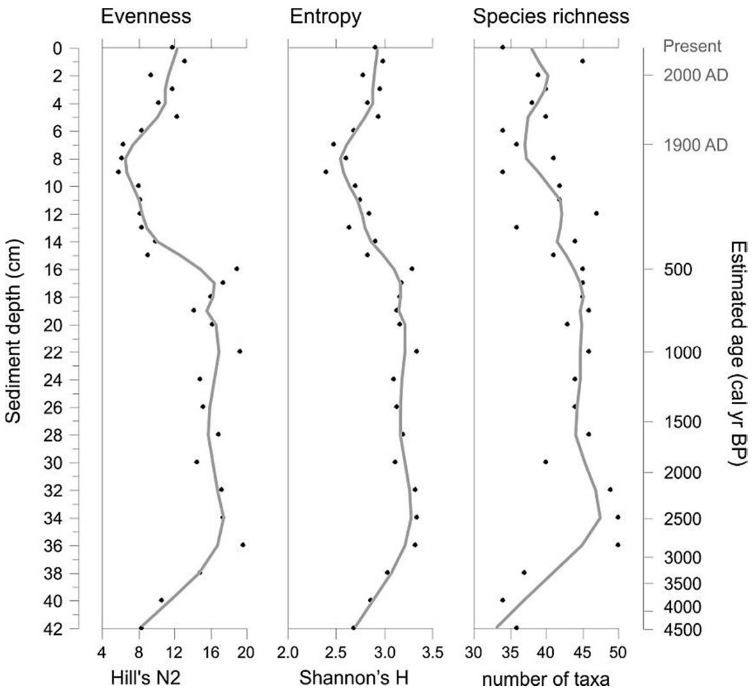

Figure 14. Evenness (Hill’s N2), entropy (Shannon’s H) and species richness (number of taxa, N) of diatom assemblages throughout the sediment stratigraphy of Lake Storträsk (dots). The grey line indicates a locally weighted scatter plot smooth (lowess, span 0.2).

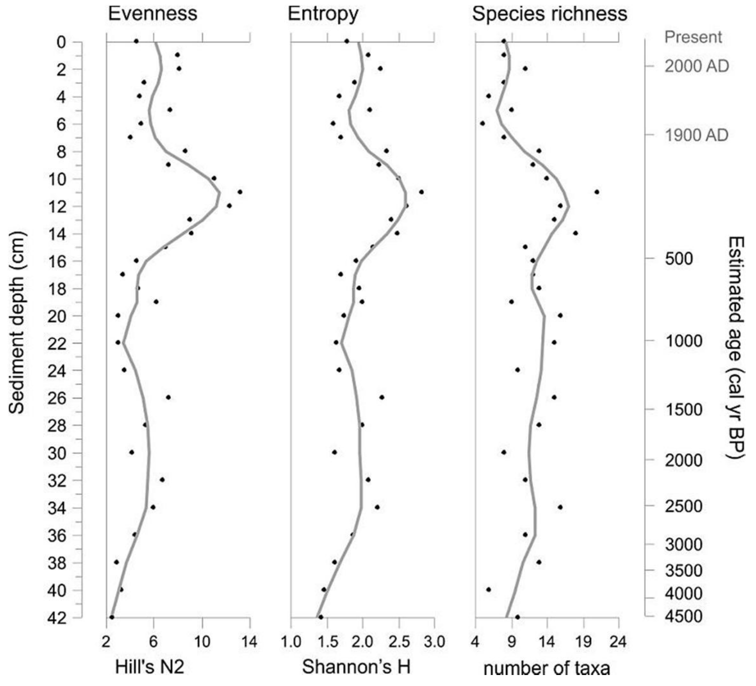

Figure 15. Evenness (Hill’s N2), entropy (Shannon’s H) and species richness (number of taxa, N) of the chironomid assemblages throughout the sediment stratigraphy of Lake Storträsk (dots). The grey line indicates a locally weighted scatter plot smooth (lowess, span 0.2).

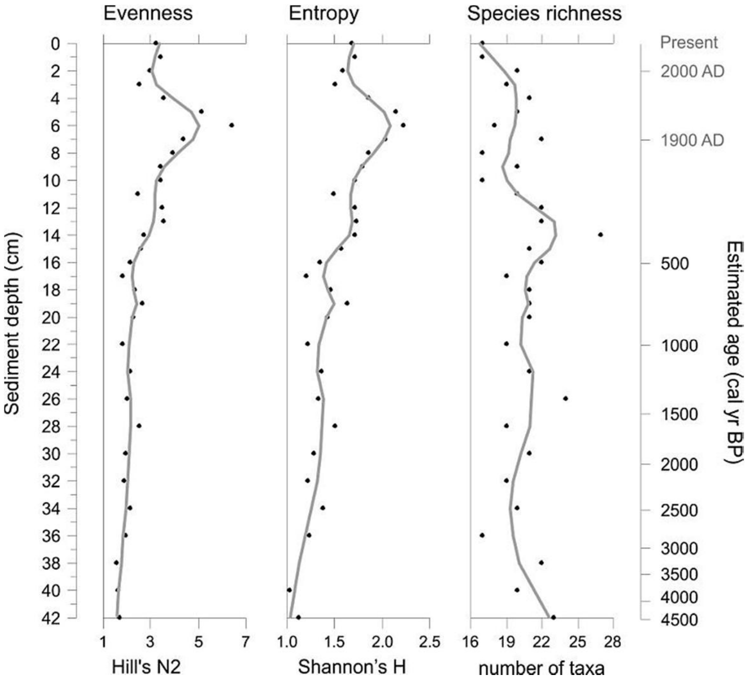

Figure 16. Evenness (Hill's N2), entropy (Shannon's H) and species richness (number of taxa, N) of the cladoceran assemblages throughout the sediment stratigraphy of Lake Storträsk (dots). The grey line indicates a locally weighted scatter plot smooth (lowess, span 0.2).

5.5.4. Correlation analyses

Spearman's correlation coefficients indicate that there is a significant negative correlation between diatom-inferred pH, which was excluded from the ordination analysis, and the primary axis in the PCA based on diatom assemblages ($p < 0.001$, $R_S = -0.75$). In addition, sedimentary P shows a strong positive correlation with axis 1 ($p < 0.001$, $R_S = 0.83$). Axis 2 in the diatom PCA is most strongly correlated with LOI ($p < 0.05$, $R_S = -0.40$) and relative contents of C ($p < 0.001$, $R_S = -0.56$) and N ($p < 0.001$, $R_S = -0.64$).

The primary ordination axis in the chironomid PCA has a moderately high correlation with chironomid-inferred hypolimnetic oxygen ($p = 0.001$, $R_S = 0.55$) and anoxia ($p < 0.01$, $R_S = -0.47$). In addition, axis 1 is negatively correlated with LOI ($p < 0.001$, $R_S = -0.58$) and relative C ($p < 0.001$, $R_S = -0.70$) and N ($p < 0.001$, $R_S = -0.64$) contents. Axis 2 in the chironomid PCA has a strong correlation with the diatom-inferred pH ($p < 0.001$, $R_S = 0.72$).

The primary PCA axis based on cladoceran assemblage has a strong negative correlation with sedimentary P ($p < 0.001$, $R_S = -0.84$), and also C/N ratio shows somewhat high negative correlation with axis 1 ($p < 0.001$, $R_S = -0.67$). Axis 2 has highest correlations with LOI ($p < 0.001$, $R_S = -0.56$), relative proportion of N ($p < 0.001$, $R_S = 0.56$) and diatom-inferred pH ($p < 0.001$, $R_S = 0.64$).

With regard to the elemental concentrations provided by the ICP-MS analysis, the most visible connection is distinguished between Mn and sedimentary P; the correlation between the two is $R_S = 0.96$ at a significance level $p < 0.01$. The trends in the concentrations of Fe, Zn, Ni, Co and V resemble each other ($p < 0.001$, $R_S \approx 0.80–0.90$), whereas Cr and Cu are most strongly correlated with each other ($p < 0.001$, $R_S = 0.80$). Furthermore, the trend of As correlates strongly with that of Pb ($p < 0.001$, $R_S = 0.90$).

6. DISCUSSION

6.1 Transition from the Holocene Thermal Maximum to the late Holocene

Based on the radiogenic ^{14}C age estimates (Table 3, Figure 5a), the lowermost sample in the sediment core of Lake Storträsk represents an age of ca. 4500 cal yr BP which coincides with the final stages of the Holocene Thermal Maximum (HTM) preceding the transition to the late Holocene around 4200 cal yr BP (Walker et al. 2012). The HTM occurred in southern Finland between ca. 8000 and 4200 cal yr BP and was characterized by elevated summer temperatures (Korhola et al. 2000, Heikkilä & Seppä 2003, Sarmaja-Korjonen & Seppä 2007) and relatively low effective precipitation (Väliranta et al. 2007, Luoto et al. 2010). The HTM was followed by a long-term decline in temperatures and an increase in effective precipitation (Bigler et al. 2002, Heikkilä & Seppä 2003, Luoto et al. 2010).

The low values of both LOI, which describes the overall organic content of the sediment (Nesje & Dahl 2001), and C/N ratio, which indicates the origin of the organic material, i.e., autochthonous or allochthonous (Wetzel 2001), imply that the relative significance of organic material originating within the lake was highest at the base of the core, but the overall amount of organic matter lowest (Figure 17). With regard to the observed pattern in LOI values, it should be noted that changes in sedimentation rate (Figure 5b) may distort the results to some extent. The low sedimentation rate in the lowermost samples (Figure 5b) suggests that the annual accumulation of organic material may have been notably lower in the lower part of the stratigraphy in relation to the upper half of the core due to the progressive increase in sedimentation rate throughout the profile. The apparent changes in sedimentation rate in the studied sediment profile (Figure 5b) may be related to actual changes in the accumulation of sediment into the lake, but due to the high organic and water content of the studied sediment sequence it is likely that the sedimentation rate gradient is at least partly related to sediment compaction and possibly also to decomposition of organic material.

The overall variation in C/N ratio (between ~14 and 17) suggests that both terrestrial sources and in-lake production were significant sources of organic matter in Lake Storträsk (Meyers & Teranes 2001). The increase in C/N ratio at the bottom of the core, however, suggests that the relative proportion of allochthonous organic material grew more significant in time. The low organic content of the sediment could be an indication of comparatively low primary productivity (Nesje & Dahl 2001, Shuman 2003) possibly driven by low influx of nutrients from the catchment due to low precipitation during the HTM as suggested by, for example, Luoto et al. (2012). The values, however, remain relatively low also during the early phases of the late Holocene. Even though the measured LOI values represent the minimum in the sediment sequence in the lower part of the core, they are quite high (>50 %) indicating that the sediment of Storträsk has been organic-rich through much of the lake’s history. LOI values generally increase along with lake depth (Shuman 2003, Nevalainen et al. 2013a), and it possible that the low values at the base of the core were partly related to lower water level caused by the warm and dry climatic conditions during the HTM. However, the lake has a small outlet channel that makes significant fluctuations in water level unlikely. Lake level bears significant impacts on, for example, light and mixing regimes and aquatic habitats (Laird et al. 2010), and hence such conditions should probably be reflected in other features of the sediment as well.

The observed patterns in LOI and C/N ratio could also be related to the dominance of minerogenic fraction in the material washed into the lake, replaced gradually by increased accumulation of organic matter. Such a trend has been observed in the development of many lakes created by retreating ice related to the post-glacial vegetation and soil development (Norton et al. 2011). Undeveloped vegetation cannot bind mineral soil as effectively, and the amount of organic matter in the soil generally increases as soils and vegetation mature. Accordingly, the trends observed in LOI and C/N ratio in the sediment profile of Lake Storträsk could be related to post-glacial catchment development processes.

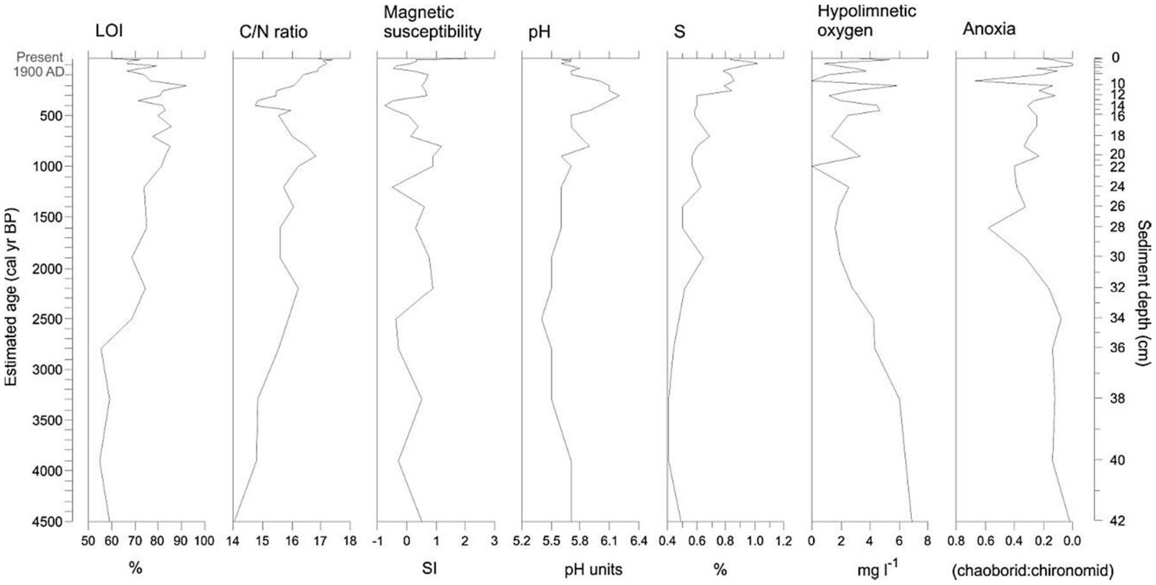

Figure 17. Summary of significant paleoenvironmental parameters (LOI, C/N ratio, magnetic susceptibility, diatom-inferred lake-water pH and relative sulphur content) from Lake Storträsk sediment core. In addition, quantitative estimation of late-winter hypolimnetic oxygen based on fossil midge assemblages from the sediment core of Lake Storträsk and estimation of hypolimnetic anoxia based on chaoborid:chironomid -ratio by Luoto, T.P. (unpublished) are shown. Larger proportions of Chaoborus represent increased anoxia.

The diatom-inferred pH reconstruction from Storträsk indicates a gradual decline in pH at the onset of the late Holocene (Figure 17). A variety of environmental controls may be associated with the observed trend. For one, declining pH might be an indication of natural long-term acidification caused by vegetation and soil development in the catchment area (Renberg & Hellberg 1982, Battarbee et al. 1990, Renberg et al. 1993a, Korsman 1999, Bigler et al. 2002), which would fit well with the previously discussedcatchment processes. The development of podsol soils and peat is generally accompanied with increased input of acidic compounds and organic acids, whereas vegetation growth and gradual leaching decreases the amount of leachable base cations in the soil (Renberg et al. 1993a, Korsman 1999, Norton et al. 2011). In addition, weathering of, e.g., apatite consumes protons (H^+) thus supporting higher pH and, accordingly, gradual leaching of such minerals would have led to declining pH (Norton et al. 2011). The small catchment area of Storträsk and scarcity of soil, bearing leachable base cations, increases further the vulnerability of the lake to acidification (Virkanen et al. 1997).

The establishment of spruce in the area may also be a factor contributing to the inferred acidification. Although the direct effects of spruce colonization on lake-water acidity have been difficult to ascertain, there is clear evidence of the acidifying effects of spruce on forest soil (Korsman et al. 1994). Spruce establishment is often assumed to have occurred ca. 3500 years ago in southern Finland (e.g., Sarmaja-Korjonen 1992), although recent research suggests a far earlier date (Heikkilä & Seppä 2003, Giesecke & Bennett 2004, Seppä et al. 2009). The declining pH trend is, however, evident already at the base of the core corresponding to ca. 4500 cal yr BP and could indicate a steady succession towards increased acidity as spruce invaded the mixed boreal forest. The simultaneous increase in the relative abundance of the input of allochthonous organic material into the lake (Figure 17) might support this notion since the colonization of spruce can increase the accumulation of raw humus (Korsman et al. 1994).

Peatland development near the lake, initiated or accelerated by the changes in temperature and effective precipitation at the onset of the late Holocene, may also have been one factor behind decreasing pH values. LOI and C/N ratio in the sediment profile (Figure 17) indicate that loading of organic material from the catchment was relatively low, but the increasing trend associated with the transition to the late Holocene may well have been strongly enhanced by input of organic material from surrounding peat deposits. Peatland formation is common in the catchments of Fennoscandian lakes, and peat deposits in the catchment can account for a significant fraction of dissolved and particulate organic matter deposited in lakes (Korsman 1999).

The elevated chironomid-inferred hypolimnetic oxygen levels at the bottom of the core (Figure 17) could be related to the low organic content of the sediment and consequent reduced intensity of decomposition processes. High temperatures, particularly when accompanied with high amount of humic compounds in the water column, might induce well developed stratification and vulnerability to oxygen depletion (Cohen 2003, Luoto & Salonen 2010). Since the reconstructed oxygen profiles indicate the opposite, it is possible that the mixing of the water column was, in turn, enhanced by, for example, lower water level as discussed above, allowing effective oxygenation of the hypolimnion by turbulent mixing. The PCA diagram describing temporal variation and relationships between the physicochemical variables in relation to changes in the diatom assemblages also shows that high values of hypolimnetic oxygen are distinctly linked with the base of the core (Figure 11).

With regard to the sediment geochemistry, apart from the previously discussed C/N ratio, the high content of sedimentary phosphorus at the base of the core is a distinct feature in the stratigraphy (Figure 6). In the PCA diagram P is, in effect, most strongly linked with the lowermost samples together with hypolimnetic oxygen (Figure 11). However, due to the high mobility and biological reactivity of P the values do not necessarily give indications of patterns in actual phosphorus loading into the lake (Håkansson & Jansson 2002, Smol 2002, Cohen 2003). Moreover, it is possible that the high concentrations at the base of the core are exaggerated to some extent, or that the overall trend merely reflects the dilution of P concentrations towards the surface due to the changes in the sedimentation rate along the profile (Figure 5b). The gradual and relatively stable decline in sedimentary P throughout the sediment profile would, nevertheless, fit well with the commonly observed natural oligotrophication linked to catchment development. Accordingly, the phosphorus supply into the lake would decline with time due to weathering and depletion of phosphorus-bearing minerals, particularly apatite (Boyle 2007, Norton et al. 2011). In addition to the effects of depletion of phosphates, developing vegetation and soil-formation increase retention of P in the soil by organic compounds and metal-complexes. Secondary hydroxides of Fe and Al precipitated in the lake, along with other compounds effectively binding P, further reduce the availability of phosphorus to organisms and hence affect the productivity of the lake (Fritz & Anderson 2013). Nonetheless, the results should be considered with caution due to the instability of phosphorus and the marked changes in the sedimentation rate in the sediment sequence (Figure 5b). In general, the flux of P between sediment and water column is particularly strongly controlled by redox-conditions, pH and sedimentary iron (Cohen 2003), gradients that have undergone more or less notable changes during the late Holocene development of Lake Storträsk

(Figures 6, 17). The decline in sedimentary P, however, remains relatively stable, which could indicate that the changes in these gradients did not significantly disturb the input of P into the lake and the P flux between water and the sediment.

The distinct species assemblages during this phase support the suggestion that at the time of the termination of the HTM and early stages of the late Holocene environmental conditions were clearly different from those prevailing during the rest of the late Holocene development of Lake Storträsk (Figure 8, Appendices 1−2). The high chironomid productivity (Appendix 1) at the base of the core could be related to the generally accepted relationship between elevated temperatures and productivity (e.g., Adrian et al. 2009, Nevalainen & Luoto 2012, Fritz & Anderson 2013) possibly via prolonged open-water period supporting particularly the primary producers of the lake (Luoto et al. 2008). Diatom productivity was not analyzed but since many chironomid and cladoceran taxa feed on algae it is possible that algal productivity was also enhanced at this time. However, a multiplicity of controlling factors probably enabled the chironomid communities to thrive. Particularly hypolimnetic oxygen concentrations have been shown to have major control over chironomid assemblages in southern Finland (Luoto & Salonen 2010). Similarly, cladoceran productivity (Appendix 2) would have been enhanced by the direct and indirect effects of auspicious climatic conditions and relatively high nutrient levels (de Eyto & Irvine 2001, Luoto et al. 2008, Nevalainen & Luoto 2012).

Elevated chironomid and cladoceran productivity might also support an idea of higher input and biological availability of phosphorus at this time since higher P would greatly benefit primary producers and, through food web associations, also the fauna of the lake. In effect, the high correlations between sedimentary P and primary axes in the PCAs based on diatom and cladoceran assemblages could imply that long-term succession in the algal and zooplankton communities was driven by catchment development processes and nutrient leaching, although it is clear that other environmental gradients have also affected the patterns. The physical features of the sediment, namely LOI (Figure 17), suggest quite the contrary in relation to lake productivity as LOI is generally strongly related to system productivity. Nonetheless, as discussed above, for example the dominance of minerogenic matter in the allochthonous material input from the catchment, enhanced decomposition of organic matter or possibly lower water level could explain the relatively low proportion of organic material in contrast to the rest of the core (Figure 17). It would seem unlikely that Storträsk was highly eutrophic at this stage, but that the prevailing climatic and limnological conditions were able to support relatively high productivity which began to decline gradually already during, and particularly after, this phase.

The low diversity in the assemblages at the bottom of the core (Figures 14−16), could be related to, for instance, low diversity of habitats within the lake. Particularly benthic, vegetation associated taxa are absent of scarce in the lowermost samples (Figure 8, Appendix 2). For one, limited extent and lower depth of the lake would have reduced the availability of littoral habitats, or it may be that environmental conditions at this time could not support an abundant and diverse aquatic macrophyte community reducing the availability of different substrate types for aquatic organism.

On the other hand, sediment-dwelling taxa seem to have thrived in the benthos. The relatively high abundance of *Neidium affine* and *N. ampliatum*, restricted only to this phase (Figure 8), would imply rich epipelic communities as taxa from the genus *Neidium* are commonly known to dwell in epipelic habitats (Round 1990). Taxa from the genus *Pinnularia* are also common to the epipelon (Round 1990) and are, accordingly, relatively well represented in the lowermost samples, though not particularly diverse. Abundance of periphytic diatoms may also have contributed to the high abundance of chironomids through food web linkages. In the cladoceran assemblages, the anomalously high occurrence of *Disparalona rostrata* and *Pleuroxus uncinatus* (Appendix 2), both of which dwell in the sediment-water interface, further supports this suggestion. Due to their habitat preference, the high benthic oxygen levels at this stage were most likely an important benefit for the species. These taxa have also been associated with meso-eutrophy (Luoto et al. 2008, Bjerring et al. 2009) and the latter also with high thermal preference (Luoto et al. 2008), and hence their presence gives further support to the suggested trends in temperature as well as nutrient and oxygen status. Furthermore, the large-sized *Alona affinis* and *Alona qudrangularis* are slightly better represented in the lowermost samples. Both species inhabit the sediment (Nevalainen 2011, Zawiska et al. 2013) and are thus also dependent on oxygen availability. These taxa have also been shown to tolerate wide range of ecological conditions (Luoto et al. 2008, Zawiska et al. 2013) and may also simply have gained competitive advantage in an environment where many other taxa could not thrive.

The relatively high amount of planktonic diatom taxa (Figure 8), particularly the acidophilous, euplanktonic *A. distans* (Virkanen et al. 1997, Turkia et al. 1998, Wolfe 2002), and the notable dominance of planktonic *Eubosmina* in the cladoceran assemblages (Appendix 2) are also peculiar features inherent to this phase and could be related to, for example, extended ice-free period due to warmer temperatures. Planktonic diatoms are highly dependent on turbulent mixing of the water column and would thus greatly benefit from longer open-water season (Bigler et al. 2002, Koinig et al. 2002, Weckström et al. 2006, Adrian et al. 2009). Effective mixing of the water column due to, for example, lower water depth could similarly improve the conditions for these taxa. Furthermore, planktonic diatoms often thrive in lakes with higher nutrient status (Pienitz et al. 2006, Davidson et al. 2010, Fritz & Anderson 2011). *Aulacoseira ambigua* is associated with alkaline waters (Meriläinen et al. 2003) and has a relatively high pH optimum also in the calibration data set (pH 6.5) but, for example, elevated nutrient status could have benefited the taxon to such extent that it was able to thrive during this phase (Figure 8) since it is commonly found in meso-eutrophic waters (Turkia 1998, Kauppila et al. 2002, Pienitz et al. 2006, Poister et al. 2012). The planktonic *Eubosmina* in the cladoceran community (Appendix 2) would also have benefited from increased phytoplankton productivity (Jensen et al. 2012), albeit it remains highly abundant throughout the lower half of the core and well represented also in the upper parts of the profile, whereas planktonic diatoms decline notably after this phase. *Eubosmina* has been shown to favor oligotrophic, acidic habitats (Walseng & Schartau 2001, Bjerring et al. 2009, Nevalainen et al. 2011) and its high presence in the sediment of Storträsk is likely linked to the relative acidity and oligo-dystrophic status of the lake, which seem to have persisted through most of the late Holocene.

Large-sized cladocerans, such as *Eubosmina*, are preferable prey for fish (Brooks & Dodson 1965, Davidson et al. 2010, Nevalainen et al. 2011) and their high numbers may give insights on the general food web structure suggesting scarcity of fish in Storträsk at the termination of the HTM and early stages of the late Holocene. Lower abundance of the phantom midge *Chaoborus flavicans* may also partially explain the substantial dominance of *Eubosmina* in the lowermost samples (Appendices 1−2) since it may feed on this large-sized *Cladocera* (Nevalainen & Luoto 2013). *Chaoborus flavicans* has been also linked with low presence of fish, although various other controls affect its distribution (Luoto & Nevalainen 2009, 2013, Korosi et al. 2013). In general, *C. flavicans* is a common inhabitant of small, shallow lakes in southern Finland, and thrives particularly well in dystrophic lakes with high macrophyte abundance and absence of fish (Luoto & Nevalainen 2009). The lower abundance of this planktonic predator at the bottom of the core, however, is likely related to the high oxygen status as *C. flavicans* is commonly known to prefer low oxygen habitats (Luoto et al. 2008, Luoto & Nevalainen 2009, Quinlan & Smol 2010) because it needs anoxic refugees to avoid fish predation (Liljendahl-Nurminen et al. 2002). Whether or not the species composition bears signals related to fish abundance, it is very possible that the relatively high acidity and high humic content of the lake have been suppressing the abundance of fish throughout the lake’s late Holocene development. Fish are often scarce in brown-water lakes due to restricted temperature, oxygen and light regimes (Wissel & Boeing 2003).

The dominance of the chironomid *Zalutschia zalutschicola* at the bottom of the core (Appendix 1) might give indications on the humic content of the lake as the taxon has been shown to prefer oligotrophic and particularly dystrophic lakes (Sæther 1979, Luoto 2010). *Zalutschia zalutschicola* has also been associated with relatively high temperatures and shallow waters (Luoto 2011). Whether the high abundance of the species was related to overall high content of humic substances in the water column, or possibly to the shore being partly overgrown by *Sphagnum* providing suitable habitats for the taxon, remains unclear. The diatom *Aulacoseira distans* was not identified on subspecies level but its high abundance in the topmost samples (Figure 8), representing the modern diatom assemblages, might suggest that, at least in the upper part of the core, the variety was *A. distans* var. *tenella* which is often found in humic waters (Virkanen et al. 1997, Turkia et al. 1998, Fallu et al. 2002). The comparatively high abundance of *Pinnularia* may be more related to the availability of substrata than humic content but *P. legumen*, for one, is also known to prefer shallow and dystrophic waters (Weckström et al. 1997). In addition, periphytic *Cymbella elginensis* shows maximum abundance in the lowermost samples, and this species has also been shown to favor shallow and dystrophic waters (Weckström et al. 1997). The signals of humic content of the lake at this stage are not unambiguously clear, although, based on the overall taxonomic composition (Figure 8, Appendices 1–2) the lake was most likely at least mesohumic. The low amount of allochthonous organic material (Figure 17) would not support an idea of extensive loading of humic substances from the catchment, albeit if the adjacent peat bog was already developing and acted as a major source for organic compounds, the material would probably not have been extensively depleted of N. High humic content would also have restricted the abundance of submerged macrophytes via reduced light availability, and hence the abundance of epiphytic taxa which are relatively sparse at this phase. Since lakes with high humic content suffer frequently from anoxic hypolimnion (Wetzel 2001), a prerequisite for the suggestion would be that the lake level was adequately low to prevent thermal stratification from forming and to ensure efficient mixing of the water column.

In general, the lowermost samples from 42 to 38 cm (~4500–3300 cal yr BP) show distinct features in most of the studied abiotic and biotic parameters when compared with the rest of the profile. The phase is also seen as a more or less distinct separate cluster in all of the PCA ordination diagrams (Figures 10–13) indicating that the environmental conditions during this phase were likely markedly anomalous in comparison with the rest of the profile. Large-scale climatic shift from the warm and dry HTM to the cooler and increasingly humid late Holocene seems to have affected the lake profoundly. Moreover, post-glacial catchment development likely bore substantial significance on the composition of material derived from the catchment, thus greatly affecting the physical features of the sediment as well as the flora and fauna of the lake.

6.2 Late Holocene, early catchment disturbances and human activities

After the termination of the HTM, which left distinct marks in the physicochemical and biological features of the sediment of Lake Storträsk, and the early stages of the late Holocene, a more steady succession seems to have taken place around three millennia ago. The gradually increasing effective precipitation and long-term cooling following the warm and dry HTM (e.g., Korhola et al. 2000, Heikkilä & Seppä 2003, Väliranta et al. 2007) were likely significant forcing factors driving changes in environmental conditions after the HTM. Studies from southern Finland have demonstrated also trends of generally increasing water levels towards the present during the late Holocene likely driven by changes in the climate (effective precipitation) (Sarmaja-Korjonen 2001, Hakala et al. 2004, Luoto & Nevalainen 2009, Luoto et al. 2010). Closer to the present, smaller-scale climatic events, such as the Medieval Climate Anomaly (MCA), have been shown to distract this pattern to some extent and were possibly partially behind the more intensive fluctuation shown by many of the studied environmental gradients in the sediment of Lake Storträsk (Figure 17). Signs of the MCA have been recorded in lake sediments from various sites in Finland (e.g., Weckström et al. 2006, Luoto et al. 2010, Luoto & Helama 2010), and this period of relatively warm temperatures and dry climate occurred in Finland between ca. 900–1300 AD.

The increase in LOI values from ca. 2800 cal yr BP onwards (Figure 17) depicts a significant increase in the loading of organic matter into the lake bottom (from ~55 to 75 % between ~2800–2200 cal yr BP), which may have been even more pronounced given the increase in the sedimentation rate. The simultaneous rise in C/N ratio (Figure 17) suggests that the increase in the accumulation of organic matter was mostly caused by increased loading of organic material from the catchment rather than an increase in in-lake productivity (Wetzel 2001). Such a trend could well have been caused by the increased precipitation and enhanced surface water runoff at the onset of the late Holocene. The continuity of these patterns, distracted only by minor fluctuations, up to the middle of the core would suggest that long-term climatic patterns were significant factors driving changes in the limnology of Lake Storträsk during this phase.

The cessation of the decline in lake-water pH (Figure 17) and subsequent acidic phase prevailing near a millennium (~2800–1900 cal yr BP) likely posed an important control particularly on the flora and fauna of the lake. Given that the colonization of spruce reached its full extent around 3500 years ago, as suggested by the pollen records from Storträsk and adjacent lakes (Sarmaja-Korjonen 1992), the aforementioned changes in water acidity as well as input of organic material may have been related to the processes occurring in the young *Picea* dominated forest (Korsman et al. 1994). Furthermore, the long-term acidification associated with catchment development may still have been contributing to the acidity (Norton et al. 2011, Rosen et al. 2011). The slightly elevated concentrations of some of the metals, particularly Fe, As, Cu and Cr (Figure 6), during this phase could be related to increased mobilization of these elements by acidic surface water runoff. In effect, many of the metals seem to follow the development in the diatom-inferred pH values (Figures 6, 17) likely due to the significance of acid-base status in controlling the mobility of these elements (Lahermo et al. 1996, Verta et al. 1990).

Given the relatively constant development in other environmental gradients (Figure 17), it is possible that the slow increase in pH after the minimum values between ~36 and 30 cm (~1900 cal yr BP onwards) was driven by long-term climate change and catchment development processes as they strongly affect the acid-base status of lakes (Renberg et al. 1993a, Rosen et al. 2011, Fritz & Anderson 2013). The increasing trend, rather than a progressive decline, however, is curious as the usual long-term development in boreal lakes with base-poor bedrock is towards increased acidity (Norton et al. 2011, Rosen et al. 2011). A similar pattern; long-term decline followed by an increase in pH around two millennia ago, has been distinguished from multiple southwestern Swedish lakes most of which are acidic today (Renberg et al. 1993a, 1993b). In these studies, the gradual or abrupt pH rises were associated with land use and consequent increase in the flux of nutrients and base cations from the catchment due to, for example, forest burning, cultivation and other catchment disturbances. Some of the studied Swedish lakes are located in rocky catchments restricting cultivation practices, which suggests that direct influence of cultivation was not needed to force the changes in lake-water pH. According to Sarmaja-Korjonen (1992), anthropogenic activities around Storträsk at this time were infrequent. Hence, it seems unlikely that early clearances would have solely caused the gradual long-term increase in lake-water pH, though they may well have contributed to the observed pattern.

Furthermore, changes in temperature have been linked to variation in lake-water pH, particularly related to the extent and timing of ice-cover and carbon dioxide (CO_2) saturation as well as CO_2 uptake linked to productivity (Wolfe 2002, Fritz & Anderson 2013). This linkage has been recorded from multiple sites particularly from high-alpine and high-latitude lakes which have been shown to be sensitive to climate-driven changes in lake-water pH (Psenner & Schmidt 1992, Sommaruga-Wögrath et al. 1997, Koinig et al. 1998, Wolfe 2002). Though decreasing temperatures could have contributed to the declining pH in the lower part of the core, they do not provide answers for the subsequent increase supposing that the general decline in temperatures was still continuing. Anthropogenic disturbances may often diminish the linkage between climate and pH (Wolfe 2002) but, again, the low intensity and frequency of human activities in the area (Sarmaja-Korjonen 1992) make such suggestions dubious. Even if direct effects of climate change were not behind the observed changes, indirect climate-related mechanisms could well have contributed to the pattern in the diatom-inferred lake-water pH. Such mechanism could be related to, for example, changes in residence time and catchment weathering (Psenner & Schmidt 1992, Korhola et al. 1996, Wolfe 2002, Larsen et al. 2006). The dilute effects of rain and an increase in the lake volume could perhaps also have affected the acid-base equilibrium of the lake. Moreover, it is possible that the long-term increase in pH after the low values between ~36 and 30 cm, forced upon by simultaneous acidifying effect of multiple pressures, simply represents a recovery towards more natural pH status of the lake.

The declining oxygen levels during this phase demonstrated by both chironomid-based hypolimnetic oxygen and *Chaoborus*-based anoxia inferences (Figure 17) also support the suggestion of increased organic input since accelerated bacterial decomposition increases the depletion of oxygen. Increasing water level and subsequent enhanced stratification could also be a factor suppressing oxygen levels, though this is not as likely a cause in the studied lake. The two oxygen reconstructions seem to be somewhat synchronous until the depth of 28 cm (ca. 1600 cal yr BP) after which general fluctuation and deviation seem to have increased. The peaks in anoxia at 28 cm (ca. 1600 cal yr BP) and in hypolimnetic oxygen at 22 cm (ca. 1000 cal yr BP) are not accompanied by any particular patterns in other environmental gradients preventing further speculations. Nevertheless, the general trend shows that the oxygen deterioration reached its highest extent around 1600 years ago (at ~28 cm) after which the overall trend shows a slight increase in hypolimnetic oxygen concentrations towards the surface.

Magnetic susceptibility fluctuates strongly in the sediment profile and, since it is affected by a variety of factors, it should generally be interpreted with caution and in reference to other environmental variables. Increase in the magnetic susceptibility of the sediment could, nevertheless, provide insight on, for example, phases of intensified mineral erosion (Thompson et al. 1975, Oldfield et al. 1983). At the depth of 32 cm (ca. 2200 cal yr BP) a minor, simultaneous increase in the organic content of the sediment, input of allochthonous organic material and magnetic susceptibility (Figure 17) could, for example, indicate more intensive and frequent forest fires in the vicinity of the lake

(Rummery 1983), albeit the changes are diminutive and difficult to interpret due to the sample and time resolutions. Nevertheless, intensive forest fires could have accounted for the increase in magnetic susceptibility and the amount of allochthonous organic material washed into the lake basin via increased erosion rate (Rummery 1983, Sarmaja-Korjonen 1992). Although the effects of forest fires are usually local in scale, fire can be considered an important factor affecting the northern hemisphere high-latitude aquatic ecosystems (Korhola et al. 1996). However, the spatial extent and magnitude of a fire in addition to catchment characteristics, lake bathymetry and climate all determine the extent of material transported to the lake, and thus the effects of forest fires can vary significantly (Korhola et al. 1996). On the other hand, the area around Lake Storträsk was briefly influenced by human activities ca. 2300 years ago (Sarmaja-Korjonen 1992), and the short period of slash-and-burn cultivation might also have accounted for the aforementioned changes since fire was used to clear the land for cultivation (Thompson 1975, Renberg et al. 1993b, Rosen et al. 2011).

Another simultaneous peak in LOI, C/N ratio and magnetic susceptibility (Figure 17) may be distinguished near the depth of 19 cm (ca. 800 cal yr BP, 1100 AD). These patterns are accompanied with a distinct increase in lake-water pH (Figure 17), which could have been caused by forest fires releasing base cations formerly bound in biomass and destroying acidic humus (Renberg et al. 1993a, Korhola et al. 1996, Virkanen et al. 1997). The timing falls within the suggested timing of the MCA in the region, although the resolution of this study is probably not adequate enough to make further interpretations about possible connections between the MCA and possible increase in forest fires in the area. Effects of increased magnetic susceptibility in lake sediments in eastern Finland, likely caused by intensive forest fires during the MCA megadrought (Helama et al. 2009), have, however, been observed by, for example, Luoto & Helama (2010). Moreover, extensive agriculture began in the area around 1200 years ago (Sarmaja-Korjonen 1992), and hence it is possible that the aforementioned changes could have been related to early land use practices in the area.

The low productivity of chironomids prevailed over centuries after the markedly higher productivity associated with the termination of the HTM (Appendix 1), and hence it is possible that the change was at least partially driven by direct or indirect effects of the late Holocene climate changes. The mechanism behind climate-induced change might be related to the decreasing oxygen levels in the hypolimnion possibly caused by increased input of organic material or enhanced stratification of the water column. In addition, leaching of nutrients from the catchment soils could have been contributing to the pattern. Chironomid productivity in Storträsk correlates particularly strongly with the measured sedimentary P ($p < 0.001$, $R_S = 0.70$) but also the C/N ratio ($p < 0.001$, $R_S = -0.56$). Furthermore, temperature is also a significant factor influencing chironomid communities (e.g., Luoto et al. 2008, Luoto & Helama 2010), and the long-term decline in productivity could have been enhanced by the late Holocene cooling. The decline in cladoceran productivity (Appendix 2) is not as evident due to the increase in productivity between 32 and 30 cm (~2200–1900 cal yr BP). The species composition was not much changed at this point thus not providing information on the possible cause of the shift. However, the timing coincides with the pattern of increased MS, LOI and C/N discussed above suggesting that the brief disturbance, possibly caused by early clearances or forest fires, benefited the cladoceran community. Elevated productivity in both chironomid and cladoceran communities can also be detected near the depths of 18 and 17 cm (~700–600 cal yr BP, ~1200–1300 AD). Based on the notion that warming climate can increase lake productivity, the pattern could possibly be related to a period of elevated temperatures during the MCA. Alternatively, land use practices (Sarmaja-Korjonen 1992) could have led to enhanced nutrient input.

Increase in the species diversity (Figures 14−16), particularly evident in the diatom assemblages, could indicate an increase in the abundance and diversity of macrophytes providing more diverse substrata and habitats for aquatic organisms. This could be related to, for example, increasing light penetration improving the growth of submerged macrophytes (Jensen et al. 2012) caused by declining humic content of the water column. Epiphytic and periphytic diatom taxa, particularly from the genera *Eunotia* and *Cymbella*, became more abundant after the turn into the late Holocene and have continued to thrive to the present (Figure 8). In addition, the littoral-associated, tychoplanktonic *Aulacoseira lirata* var. *lacustris* appeared in the stratigraphy, though only after the most acidic phase since the species has a relatively high pH optimum in the training set (pH 6.0). The marked increase in the abundance of *Tabellaria flocculosa* is difficult to associate with any particular change in environmental conditions. The most common form of *T. flocculosa* is, however, benthic-epipsammic (Round 1990) and it has been associated with oligotrophic (Pienitz et al. 2006) to mesotrophic (Kauppila et al. 2002) conditions. The cladoceran communities also showed a clear increase in the diversity of benthic taxa, although *Eubosmina* remained by far the dominant species (Appendix 2). Particularly the vegetation-associated acidity-tolerant *Alonella nana*, *Alonella excisa* and *Acroperus harpae* increased distinctly after the initial phase. *Alonella nana* and *A. harpae* are also cold-tolerant species and may have benefited from the declining temperatures (Nevalainen et al. 2013b). The most abundant taxon in the benthic cladoceran communities, *A. nana*, is generally associated with oligotrophic (Luoto et al. 2013) and oligo-dystrophic (Nevalainen & Sarmaja-Korjonen 2008) lakes.

The PCAs on diatom data (Figures 10−11) separate the phase between ~36 and 30 cm (~2800–1900 cal yr BP) as a clearly separate cluster in the biplot and it is probable that this pattern is related to the low pH values inherent to the phase. Overall, the grouping of samples into distinct clusters and detailed patterns within the clusters in the diatom PCA seem to follow a pattern connected to the inferred pH values in each sample, which suggests that acid-base status has had a strong control over the diatom assemblages in Storträsk. Such a feature is lacking from the cladoceran and chironomid PCA ordination diagrams (Figures 12−13) suggesting that other factors were probably driving changes in the zooplankton and benthic fauna. Lake-water pH was most likely a significant factor controlling the diatom assemblage composition, yet the maximum diatom diversity at this point may also simply reflect a shift from one stable state to another characterized by marked changes in almost all of the environmental gradients (Figure 17). The phase marks the disappearance and appearance of many diatom taxa with few species inherent only to this particular phase (Figure 8). Nonetheless, the scarcity of planktonic taxa during this phase could well be related to the increased acidity since it often limits the abundance of diatoms in the plankton (Round 1990, Korsman et al. 1994, Virkanen et al. 1997).

The gradual increase in the abundance of the planktonic, alkaliphilous *Aulacoseira ambigua* and *Asterionella formosa* beginning near the depth of 26 cm (ca. 1400 cal yr BP) is very likely a significant factor behind the slowly increasing trend in the diatom-inferred pH reconstruction (Figures 8−9). Both taxa have also been often associated with nutrient enrichment caused by early human settlements as well as recent human activities (Turkia et al. 1998, Bigler et al. 2002, Pienitz et al. 2006). Hence, it is also possible that increased input of nutrients and base cations from the catchment caused by human activities, forest fires, other catchment disturbances, or a combination of the three, improved the conditions for these alkaline taxa and was reflected as an increase in the diatom-inferred lake-water pH.

Most notable changes in the chironomid species composition seem to be linked with oxygen status and humic content of the lake (Figure 17, Appendix 1). The decline in the dominance of *Zalutschia zalutschicola*, associated with dystrophic waters (Sæther 1979, Luoto 2010) and also moderate warmth and oxygen status (Luoto 2011) is accompanied with the simultaneous increase in *Sergentia coracina*-type which, in turn, can tolerate even anoxic conditions but is not found in dystrophic lakes (Nevalainen & Luoto 2012). The clear pattern strongly suggests that the humic content of the lake declined during this stage. *Sergentia coracina*-type is known to dwell in the profundal zone (Meriläinen et al. 2003) and has been associated with increasing water depth (Luoto 2010) suggesting a possibility that the water level increased slightly after the warm and dry HTM. The increase in *Chaoborus flavicans* follows a similar pattern, though the increase in the abundance of this species is probably most significantly driven by the declining oxygen concentrations. The taxon has also been observed to prefer deeper waters (Luoto & Nevalainen 2009, Luoto 2010). Another feature of interest is the occurrence of *Tanytarsus lugens*-type at the time of the dominance of *S. coracina*-type. This cold-stenothermic species is most often found in arctic lakes or very clear watered lakes in southern Finland and has been associated with deep, vegetation-free habitats (Luoto 2010, 2011). Accordingly, its co-occurrence with *S. coracina*-type could be related to a decline in the humic content of the water column and possible changes in water level. The PCA based on chironomid assemblage data (Figure 12) shows a steady decline along axis 1 until the depth of 28 cm (ca. 1500 cal yr BP), which could well reflect the gradual decline in oxygen concentrations in the lower parts of the stratigraphy (Figure 17). The pattern in the PCA diagram becomes more dispersed in the upper parts of the stratigraphy, yet so does the variation in the inferred hypolimnetic oxygen.

In general, this phase seems to have been somewhat stable as suggested by the centennial-scale gradual development in the studied environmental gradients. The long-term trends in the physicochemical gradients have most likely been essentially climate-driven and these changes affected also the biota of the lake. More temporary catchment disturbances such as periods of more frequent forest fires or the early clearance periods in the area may also have left a record in the sediment, albeit the temporal resolution of this study and scarcity of reference studies does not allow for more detailed examination of the patterns.

6.3 Recent lake development and human disturbances

The warm and dry MCA was followed by the Little Ice Age (LIA) around 1500–1850 AD characterized by cooler and wetter climate in southern and eastern Finland (Luoto et al. 2008, Luoto & Helama 2010). The post-industrial warming has also been recorded from several sites (e.g., Korhola et al. 2000, Weckström et al. 2006), but the resolution in this study is not detailed enough to clearly distinguish such signals.

The general increase in the frequency of fluctuation in many of the studied environmental parameters (Figures 6, 17) in the upper parts of the profile is obviously affected by the sample frequency, but the magnitude of variation is also higher suggesting an increase in environmental stressors affecting the status of Lake Storträsk. It seems that the millennial-scale steady increase in the relative proportion of allochthonous organic material in the sediment was suddenly disturbed for a few centuries between ~18 and 9 cm (~1200–1800 AD) after which the C/N ratio began to increase again (Figure 17). The long-term development towards lower relative proportion of autochthonous organic material could perhaps be explained by the leaching of nutrients from the catchment leading to declined productivity, or possibly the increase in precipitation and surface water runoff increasing the relative proportion of allochthonous organic material. Apart from the drop in the C/N ratio between ~18 and 9 cm, the C/N curve seems to, in fact, depict a trend reverse to that of sedimentary P (Figures 6–7), although, probably due to the temporary decline in C/N, the correlation between the two variables remains moderate ($p = 0{,}01$, $R_S = -0{,}45$). Furthermore, as noted earlier, the pattern in the concentrations of P in the sediment profile might not reflect actual changes in the accumulation of phosphorus into the lake.

The minimum values in C/N ratio precede the marked increase in LOI values at the depth of 10 cm (ca. 1750 AD) and are, in fact, reflected as a drop in the overall organic content of the sediment before the peak value suggesting that the decline in C/N ratio was caused by decreased input of allochthonous organic material from the catchment rather than relative increase in productivity. The maximum value in loss-on-ignition (92 %) suggests a brief phase during which the loading of organic material increased markedly, and since C/N ratio increases simultaneously it is likely that the pattern in LOI was at least partly related to increased input of organic material from the catchment. Since continuous human activities began in the area already around 1200 years ago (Sarmaja-Korjonen 1992), it seems likely that these anomalies were brought upon by agricultural activities in the area, even though the catchment of Storträsk itself could not have been deployed for cultivation (Figures 1a, 2). The distinctly elevated pH values between 15 and 9 cm (~1500–1800 AD) coincide with the anomalies in LOI and C/N and are probably also partly related to human activities in the area (Figure 17).

One possible explanation for the aforementioned anomalous patterns could be related to retting and soaking of fiber plants which has been shown to significantly affect the trophic status and pH of the lakes used for this purpose (Grönlund et al. 1986, Hakala et al. 2004). In the study of Grönlund et al. (1986), intensive fiber-plant retting was associated with eutrophication of the studied southern Finnish lake in addition to increased sedimentation rate, periodical oxygen depletion and increase in pH status. Sarmaja-Korjonen (1992) found possible evidence of cultivation of *Cannabaceae* in the area recorded in the pollen stratigraphy of Lake Storträsk and nearby Lake Hampträsk. Though no detailed chronology is available, the pollen curves show that the possible cultivation of fiber plants was associated with the general intensification of agricultural practices around 1200 years ago. The highest pollen abundance in Storträsk was slightly delayed in comparison with Hampträsk, and in both the signal was strongest only for a relatively short period of time and declined towards the present. Due to lack of time reference and historical data, no clear evidence can be presented but it seems possible that fiber plant processing in the lake could have left a distinct record in the sediment of the otherwise relatively undisturbed lake, reflected as increased LOI, C/N ratio and lake-water pH.

The anomalously high LOI value of 92 % at 10 cm (18th century) was followed by a decline in oxygen concentrations, as expressed by both hypolimnetic oxygen and anoxia curves (Figure 17). After this event, the oxygen status of the lake seems to have improved to some extent simultaneously with decreasing LOI values. The decline in

LOI is likely related to the continuous rapid increase in the relative proportion of allochthonous organic material input. The increase in C/N ratio after the low values discussed above might suggest increased sedimentation from the catchment and an increase in minerogenic material washed into the lake causing a decline in the relative proportion of organic material in the sediment. A likely reason for this kind of development could be changes in land use in the area around the lake.

The marked increase in the diatom-inferred lake-water pH from 5.7 at 16 cm to 6.2 at 12 cm (~1450–1650 AD) and subsequent decline occurs simultaneously with the anomalies in LOI and C/N ratio (Figure 17), as noted. The overall change in pH is 0.5 pH units, and hence likely ecologically relevant since it clearly stands out from the rest of the variation. It would seem very feasible to assume that the change was, at least to some extent, promoted by more intensive human activity in the vicinity of the lake, for example the fiber plant retting practices suggested above. However, since this increase in pH seems to be part of a continuum which already originated nearly a millennium earlier the extent to which natural factors and human disturbances have been driving the observed changes is difficult to determine. The rapid decline towards values similar to those at the base of the core (~pH 5.7) after the anomalously high pH values between 15 and 9 cm (~1500–1800 AD) could, nevertheless, indicate a recovery due to decline in land use practices in the area as the values remain low and stable up to the surface.

Furthermore, the observed decline in lake-water pH could be related to increase in atmospheric loading of acidic substances related to industrial expansion, or possibly the combined effect of both. The change from agricultural to industrial economy occurred in Europe mostly during the last 200 years leading to notable changes in land use patterns and profound increase in atmospheric loading of pollutants and acidic substances (e.g., Renberg et al. 1993a, 1993b). A clear link between lake acidification and acid deposition has been established in multiple studies (e.g., Battarbee et al. 1990, Renberg et al. 1993a, Rosen et al. 2011) and it is probable that a record was left also in the sediment of Lake Storträsk due to its location and general characteristics making it sensitive to the effects of atmospheric loading. Most pronounced sulphate deposition occurred generally around the 1970’s in Finland and elsewhere in Europe after which the emissions began to decline (e.g., Mannio 2001, Vestreng et al. 2007). An increase in the S content of the sediment is already evident at 11 cm (Figure 17) corresponding to ca. 1700 AD. The next distinct rise can be detected near the depth of 6 cm (~early 20th century) and the most pronounced peak is seen at the depth of 3 cm representing the late 20th century (Figure 5a) and is followed by a slight decline.

A similar trend can be observed from nearly all of the trace metals (Figure 6) many of which are related to the burning of fossil fuels and industrial processes which were most distinctly intensified around the 1960’ (Virkanen et al. 1997, Moser et al. 2010). It is possible that these trends reflect depositional processes rather than increase in accumulation, but if the latter was considered, the likely source of increased loading would be atmospheric depositions as there are no records of major point sources for trace metals or S in the vicinity of the lake. The peak concentrations near the depth of 3 cm (late 20th century, Figure 5a) would strongly suggest that the patterns were related to industrial activities. Considering the clear increase in sedimentation rate from the base of the core towards the surface (Figure 5b), the annual accumulation of trace metals into the lake could have been even more pronounced in relation to post-industrial times suggesting that Lake Storträsk is very sensitive to changes in atmospheric loading.

A striking feature in the sediment geochemistry is the increase in sedimentary Pb concentrations in the upper half of the profile when compared with the near-zero values of the lower core (Figure 6). In addition, Zn and As contents in the sediment seem to follow a somewhat similar trend, although the rise begins later and is not as pronounced. The distribution of Pb in southern Finland is little affected by the composition of underlying bedrock and is most often related to anthropogenic activities (Lahermo et al. 1996). Early human activities known to increase hemispheric-scale atmospheric lead deposition span as far back in time as the Greco-Roman civilization around two millennia ago (Hong et al. 1994, Mannio et al. 2001, Renberg et al. 2001). During the Middle Ages, the usage of lead was less pronounced but increased abruptly in association with the industrial revolution beginning in the 18th century (Lahermo et al. 1996). For example, Hakala and Salonen (2004) found evidence of early Pb emissions and deposition in the sediment deposits of a small, undisturbed lake in southwestern Finland accompanied with signs of medieval Zn deposition. Due to the sensitivity of Lake Stroträsk to the effects of atmospheric loading, it is not surprising to find evidence of these early human activities recorded in the Pb stratigraphy of the sediment. Accordingly, the Pb content of the sediment increases very slowly at the bottom of the core but is slightly accelerated after 30 cm (ca. 100 AD) which coincides with the time of the highest extent of the Roman Empire. The more evident increase at 18 cm (ca. 1200 AD) could be related to the medieval industry (Renberg et al. 2001), whereas the most substantial increase at the depth of 8 cm with the estimated age of ca. 1850 AD is likely related to the Industrial Revolution in Europe.

The connection between chironomid and cladoceran productivity seems to diminish during this phase as the former remains low in the rest of the profile and the latter fluctuates strongly. The continuous decline in chironomid productivity (Appendix 1) after the HTM, and the fact that the brief increase in productivity occurs during the general timing of the MCA, would support an idea of temperature as a significant forcing factor controlling chironomid productivity. As discussed earlier, the trend could also be related to the long-term leaching and depletion of nutrients from the catchment. Cladocerans, however, seem to respond to entirely different environmental gradients at this stage. The anomalies in LOI, C/N and pH at the beginning of this phase seem to be associated with a slight decline in the cladoceran productivity suggesting that, whatever mechanism was behind the changes, the cladoceran community did not benefit from it (Figure 17, Appendix 2). Moreover, the PCAs on all biological assemblages (Figures 10–13) separate the samples with highest diatom-inferred pH values between 13 and 9 cm (~1600–1800 AD) as more or less distinct clusters, which suggests that the algal, zooplankton and benthic communities were affected by the disturbances altering the acid-base equilibrium of the lake. Minimum productivity in both cladoceran and chironomid assemblages, reached at the depth of 6 cm (~early 20th century), is probably related to the event which drove marked changes in the community structure of both groups as will be discussed below.

The anomalies in various environmental gradients between ~16 and 9 cm (~1450–1800 AD) seem not to have affected the diversity of cladocerans in any particular way but chironomid diversity was clearly increased, whereas diatom assemblages became less diverse (Figures 14–16). The increase in the diversity of chironomids is seen in the species composition level as a decline of the dominant taxa *Z. zalutschicola* and *S. coracina*-type in favor of several taxa which are scarce or absent in the rest of the core, but seem to thrive in the upper half of the profile (Appendix 1). The increased pH probably kept the abundance of *Z. zalutschicola* low since the species is generally restricted to humic, acidic lakes (Sæther 1979, Luoto et al. 2010), but it may also be that the changed environmental conditions were merely auspicious to a larger variety of species decreasing the relative dominance of the dominant taxa. The anomaly in diatom-inferred lake-water pH might be behind the increase of *Cricotopus cylindraceus*-type and *Glyptotendipes pallens*-type, both of which have been associated with higher pH habitats (Luoto 2011). On the other hand, some taxa were undoubtedly affected by the fluctuations in oxygen status and, for example, the brief but marked drop in both oxygen reconstructions at 9 cm is probably behind the absence of *Psectrocladius sordidellus*-type which prefers higher levels of oxygen (Luoto & Salonen 2010, Luoto 2011). Some eurytopic species may simply have benefited from large fluctuations in environmental conditions restricting the abundance of other taxa. For instance, *Procladius* and *Dicrotendipes nervosus*-type, thriving during the disturbance period, have been shown to be tolerant of a wide range of environmental conditions (Sæther 1979, Luoto 2010, Nevalainen & Luoto 2012).

In the diatom community, there is a clear shift in the relative abundances of planktonic and benthic taxa (Figure 8) mostly resulting from the increase in the abundance of alkaliphilous, planktonic *Aulacoseira ambigua* between 15 and 9 cm (1500–1800 AD). This increase is likely a major factor behind the simultaneous increase in the diatom-inferred pH (Figure 9). A sudden increase in lake-water pH may have been the dominant factor making the conditions suitable for *A.ambigua* to thrive, and the abundance of the species does seem to follow the inferred pH closely throughout the stratigraphy. However, in addition to the species alkaline preferences, increased abundance of *A. ambigua* has been often associated with enhanced nutrient status (Kauppila et al. 2000, Pienitz et al. 2006, Luoto et al. 2012). The slight increase in the abundance of *Asterionella formosa* could also be related to increased alkalinity since it has been associated with more alkaline habitats (Turkia et al. 1998). On the other hand, the occurrence of this species has also been clearly linked with nutrient enrichment (Turkia et al. 1998, Bigler 2002). The most distinct simultaneous decline is seen in the abundance of the two most dominant *Eunotia* taxa (namely, *E. incisa* and *E. serra*), both of which are clearly acidophilous in the training set (pH optima of 5.1 and 5.4, respectively) and the former also associated with acidity in various previous studies (Round 1990, Turkia et al. 1998, Korsman 1999). In the cladoceran community, the coincident sudden increase in the abundance of *Chydorus* cf. *sphaericus* could possibly provide additional information on the nature of the change (Appendix 2). The species is tolerant to a variety of environmental conditions (Korhola & Rautio 2001, Sarjama-Korjonen 2001), but is often found in abundance in lakes with higher pH (de Eyto & Irvine 2001) also in southern Finland (Nevalainen 2008). The species has, however, been associated with high nutrient status, as well (de Eyto & Irvine 2001). Moreover, it is possible that, rather than through direct effects of environmental change, *C. sphaericus* may have achieved competitive advantage by moving to the pelagic zone by attaching to filamentous algae (de Eyto & Irvine 2001), for example, *A. ambigua* filaments. It is possible that an actual shift towards more alkaline conditions did occur at this time and was likely either promoted or caused by fiber plant retting or other adjacent human activities. Nonetheless, the confounding effects of increased nutrient input should not be disregarded.

One curious and very likely pH-related shift near the surface is the sudden increase in the abundance of *Achnantes minutissima*. The increase of this alkaliphilous species has been associated with liming-induced increase in alkalinity (Renberg et al. 1993a), and hence it is probable that the adding of carbonates into the lake during the recent years is the cause for this shift. There are no clear signs of the liming practices in the sediment profile probably due to the resolution of the study, although, for example the distinct increase in Fe concentration in the topmost sample (Figure 6) could be related to increased pH caused by liming (Wällstedt 2005).

The pH development in the lake during the mid-Holocene is not revealed based on the obtained sediment core but the data does, however, imply that Storträsk has been relatively acidic for much of its history. The inferred pH in the topmost sample (5.67) is slightly higher than the pH of 5.40 observed from the epilimnion in May 2012. The difference does not exceed the standard error of estimate (0.34 pH units) suggesting that the pH reconstruction is relatively accurate. However, it must be noted that the values do not necessarily represent the natural acid-base equilibrium of the lake due to the liming of the lake. In effect, measurements from the epilimnion in 2011 are notably higher; 6.9 in August and 5.9 in November 2011. Due to scarce and infrequent contemporary measurements and lack of comprehensive records of the liming practices, no detailed presumptions can be attained of the effects of liming on the acid-base equilibrium of Lake Storträsk.

Vegetation-associated cladocerans remain abundant in the upper half of the core and show no major changes in their relative proportions. However, the slight increase of *Camptocercus rectirostris* and *Graptoleberis testudinaria* (Appendix 2), both of which are strongly affiliated with macrophytes (Nevalainen 2011, Zawiska et al. 2013) and thrive in moss environments (Sarjama-Korjonen 2001), could indicate changes in the macrophyte composition of the lake during this phase. The epiphytic diatom taxa also remain well represented in the upper half of the core (Figure 8) apart from the inferences caused by the drastic changes in environmental conditions between ~16 and 9 cm (~1500–1800 AD) (Figure 17). Hence, it seems likely that the lake had an abundant macrophyte cover at this time, although the plant species composition may have varied.

The chironomid *Zalutschia zalutschicola* seems to have regained an infrequent dominance in the upper half of the core (Appendix 1) possibly bearing implications on changes in the humic content of the lake. The incongruity of *Z. zalutschicola* and *S. coracina*-type is not as distinct during this phase when compared with the lower half of the stratigraphy, and both species seem to have been able to sustain relatively abundant populations. It is thus possible that, for example, a well-developed *Sphagnum* rim in the shore provided habitats for *Z. zalutschicola* whereas *S. coracina*-type flourished in the deeper waters. The fact that *Z. zalutschicola* is more dominant in the topmost samples agrees well with the current polyhumic status of the lake.

Aside from the relatively stable development in the benthic cladoceran communities, the pelagic cladoceran taxa underwent major changes particularly closer to the surface (Appendix 2). The environmental changes between 16 and 9 cm (~1450–1800 AD) seem not to have influenced the abundance of *Eubosmina* greatly since its dominance had already been declining gradually from the base of the core. However, the drop in the abundance of *Eubosmina* at the depth of 6 cm (~early 20th century) and concurrent pronounced rise in the abundance of *Bosmina longirostris* must have been caused by some more temporary stressor affecting the planktonic cladoceran community most thoroughly without much effect on the benthic fauna or the physicochemical features of the sediment. Changes in cladoceran communities are often associated with alteration in the aquatic food web structure as zooplankton are sensitive to both vertebrate and invertebrate predation and changes in primary production via food availability (Korosi et al. 2013). It is thus possible that changes in food web structure were behind the observed changes. The appearance and high abundance of *Bosmina longirostris* may be related to increased predation pressure of zooplankton by visual predators since this small species is not as sensitive to size-selective predation (Davidson et al. 2010, Korosi et al. 2013). *Eubosmina*, on the other hand, is large in size and preferred food for many predators. Since macroinvertebrate predators such as *Procladius* and *Chaoborus flavicans* are scarce or even absent in the topmost samples (Appendix 1), increased fish predation seems like a plausible explanation for the observed pattern. No historical records are available, but it is possible that fish were stocked in the lake at this time causing major changes in the food web structure of the lake. *Bosmina longirostris* has also been strongly connected with climatic warming and elevated nutrient status in various studies (Jensen et al. 2012, Nevalainen & Luoto 2012, Nevalainen & Luoto 2013, Nevalainen et al. 2013b). Given the substantial increase in the abundance of the species, there might be some inferences with regard to climate warming or changes in nutrient status which were not clearly recorded by the other studied proxies.

The absence of *Chaoborus flavicans* at 5 and 4 cm (~mid-20th century) is probably related to the marked decline in *Eubosmina* abundance since both taxa are easily consumed by fish. In most of the profile, the distribution of this predator seems to follow reversely the general pattern in the reconstructed hypolimnetic oxygen (Figure 17, Appendix 1), but the sudden disappearance is hard to explain solely on the basis of oxygen status. In fact, hypolimnetic oxygen is relatively low at this stage, and hence the disappearance of *C. flavicans* was more likely caused by food web related changes as it is known to be sensitive to the presence of fish (Luoto & Nevalainen 2009).

Generally, after the middle part of the sediment sequence (ca. 1000 AD) many of the environmental gradients have experienced pronounced fluctuation. Particularly beginning around the 15th century, environmental perturbation seems to have increased as expressed by the increase in the magnitude of variation in the studied environmental parameters. These changes, more rapid and extensive in scale, were most likely related to intensive agricultural practices in the area beginning around 1200 years ago, and the lake seems to have responded to these disturbance very sensitively as the catchment of the lake is not suitable for cultivation. The biological communities in the lake were likely affected by both large-scale environmental changes and human activities, and particularly the topmost samples show evidence of disturbances, possibly human-induced, which bore distinct impact on the balance and structure of the biological communities in the lake. Furthermore, increased atmospheric loading has been distinctly recorded in the sediment core.

7. CONCLUSIONS

Lake Storträsk has been relatively acidic and humic through most of its history, yet the development of the lake has been far from stable. Long-term climatic shifts have clearly been the most significant factors driving changes in the water quality and limnoecological status of Storträsk and are readily reflected in the sedimentary features of the lake. In spite of the unsuitability of the rocky catchment of Storträsk for agricultural activities, the phase of intensive slash-and-burn agriculture in the surrounding area has left distinct signs of limnological perturbation. The most obtrusive human activities within the lake's vicinity have apparently receded towards the present, yet the impacts of industrial atmospheric loading have been sensitively recorded in the sediment. The effects of the most recent liming practices and aquaculture could not be clearly recognized from the sediment proxies most likely due to the low temporal resolution of the study. However, the results suggest that Lake Storträsk is sensitive to anthropogenic disturbances, even to such that do not occur in the instant vicinity of the lake, which highlights the importance of continuous monitoring particularly as human activities in the area will be intensified in the future.

The lowermost part of the stratigraphy around ~4000 cal yr BP depicts the transition from the warm and dry Holocene Thermal Maximum to the late Holocene, and was characterized by relatively high humic content and acidity as well as efficiently mixing water column maintaining a well-oxygenated hypolimnion. Post-glacial vegetation development and soil-forming processes were still ongoing resulting in higher yield of minerogenic matter, nutrients and base cations from the catchment. Planktonic communities thrived due to extended ice-free period and enhanced nutrient input, whereas the benthos was characterized by abundant sediment-dwelling communities.

At the onset of the late Holocene, increased effective precipitation and stabilization of the catchment were causing increased accumulation of organic material and consequent deterioration of oxygen levels. Lake-water pH remained low for a millennium but this phase was followed by a long-term gradual increase apparently driven by indirect climatic mechanisms and supported by early human activities and catchment disturbances in the area. Lake productivity began to decline but the diversity of aquatic organisms was relatively high suggesting diverse littoral and benthic habitats allowing a diversity of epiphytic taxa to emerge and thrive in the benthos. Concurrently, the humic content of the lake water declined.

Since around the 15th century, the long-term gradual development patterns were replaced by increasingly fluctuating environmental gradients. The most distinct changes were elevated lake-water pH and increased organic content linked with more intensive human activities, although the most significant disturbances were temporary allowing the lake to recover to some extent towards the present. The planktonic and dystrophic taxa inherent to the base of the core reattained some of their abundance and the epiphyton remained equally abundant. The food web structure and predation regimes of the lake were disturbed profoundly near the beginning of the 20th century likely related to human activities. Furthermore, increase in the atmospheric loading of trace elements due to industrial activities has been sensitively recorded in the sediments of Storträsk.

The results of this study support the relevance of long-term perspective in assessing reference conditions as the ontogeny of lakes can be highly variable. Hence, this study is relevant to the discussion concerning the determination of reference status which forms an essential basis for the European Water Framework Directive and defines the premises for setting priorities and strategies in water management and protection. Furthermore, the results provide insights on the functioning of small, humic boreal lakes under different sources of pressure and are valuable, since identifying the mechanism and linkages between different environmental variables and lake responses is essential for sound and effective water management practices. The distinct characteristics and particular responses of these types of lakes further stress the importance of employing data from a variety of paleolimnological proxies that provide more robust insights on the processes occurring within the lake and its catchment and reduce the potentialities for misinterpretation. Furthermore, since it is evident that multiplicity of intercorrelated environmental stressors have been driving changes in the biological assemblages of the lake during the late Holocene, combining information from various biotic proxies provides a more detailed and accurate picture of changes in the ecology of the lake. To better understand the effects of anthropogenic stressors on lake ecosystems, more holistic assessments of lake-environment dynamics from varying sites are needed. Especially the impacts of changing climate need to be further unraveled as human-induced climate change may increase the significance of climatic drivers, for example through influencing thermal regimes and input of allochthonous materials, on aquatic ecosystems.

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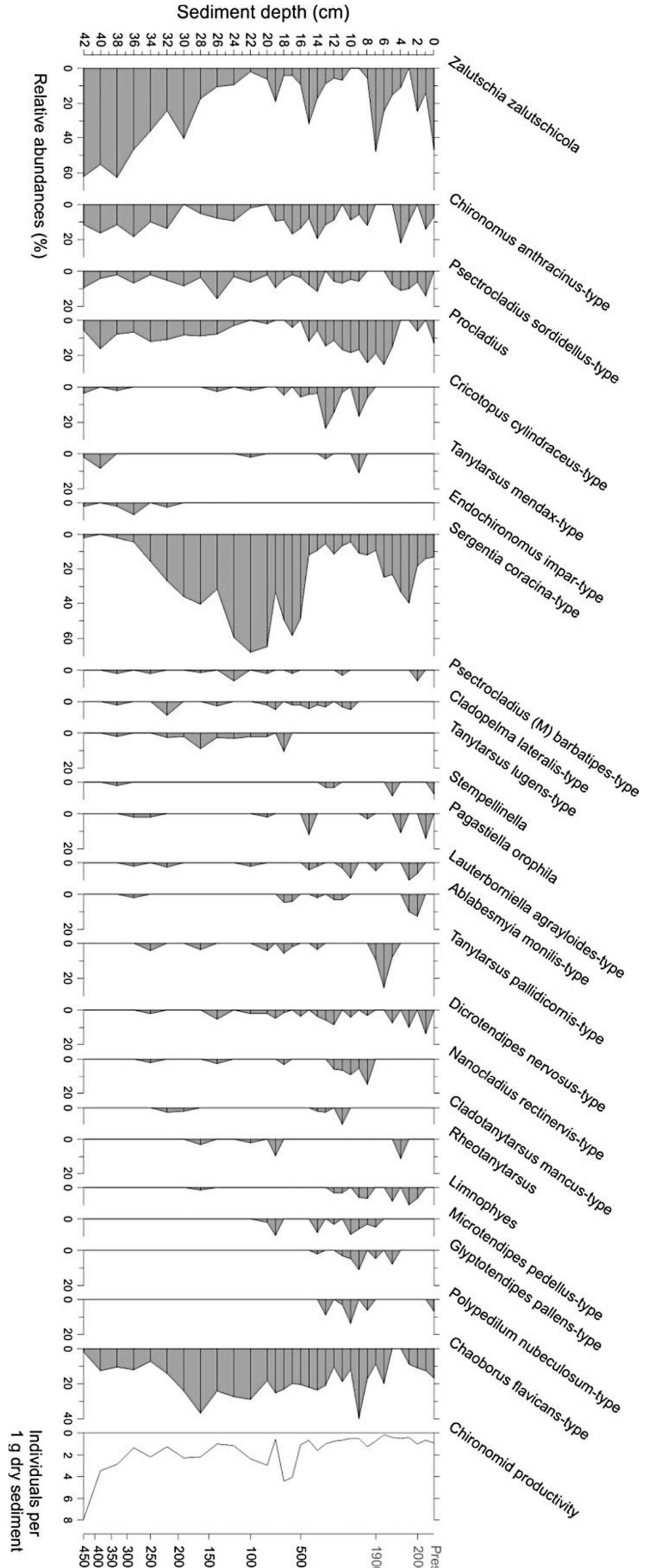
Appendix 1. Relative abundances of most common midge taxa in the sediment of Lake Storträsk and chironomid productivity.



Appendix 2. Relative abundance of most common cladoceran taxa in the stratigraphy of Lake Storträsk and cladoceran productivity.