Biogeosciences Discuss., 10, 19969–20003, 2013
www.biogeosciences-discuss.net/10/19969/2013/
doi:10.5194/bgd-10-19969-2013

Biogeosciences
Discussions
Open Access

Forcing mechanisms behind variations in total organic carbon (TOC) concentration of lake waters during the past eight centuries – palaeolimnological evidence from southern Sweden

P. Bragée[1], F. Mazier[2], P. Rosén[3], D. Fredh[1], A. Broström[1,*], W. Granéli[4], and D. Hammarlund[1]

[1]Department of Geology, Quaternary Sciences, Lund University, Lund, Sweden
[2]GEODE, UMR 5602, University of Toulouse-Le Mirail, Toulouse, France
[3]Department of Ecology and Environmental Science, Umeå University, Umeå, Sweden
[4]Department of Biology, Aquatic Ecology, Lund University, Lund, Sweden
[*]now at: Swedish National Heritage Board, Contract Archaeology Service, Lund, Sweden

Received: 16 October 2013 – Accepted: 27 November 2013 – Published: 19 December 2013

Correspondence to: P. Bragée (petra.bragee@geol.lu.se)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Decadal-scale variations in total organic carbon (TOC) concentration in lake water since AD 1200 in two small lakes in southern Sweden were reconstructed based on visible-near infrared spectroscopy (VNIRS) of their recent sediment successions. In order to assess the impacts of local land-use changes and regional variations in sulphur deposition and climate on the inferred changes in TOC concentration, the same sediment records were subjected to multi-proxy palaeolimnological analyses. Changes in lake-water pH were inferred from diatom analysis, whereas pollen-based land-use reconstructions (Landscape Reconstruction Algorithm) together with geochemical records provided information on catchment-scale environmental changes, and comparisons were made with available records of climate and population density. Our long-term reconstructions reveal that TOC concentrations were generally high prior to AD 1900, with second-order variations coupled mainly to changes in agricultural land-use intensity. The last century showed significant changes, and unusually low TOC concentrations were recorded in 1930–1990, followed by a recent increase. Variations in sulphur emissions, with an increase in the early 1900s to a peak around AD 1980 and a subsequent decrease, were most likely the main driver of these dynamics, although processes related to the introduction of modern forestry and recent increases in precipitation and temperature may have contributed. The increase in lake-water TOC concentration from around AD 1980 may therefore reflect a recovery process. Given that the effects of sulphate deposition now subside, other forcing mechanisms related to land management and climate change will possibly become the main drivers of TOC concentration changes in boreal lake waters in the future.

1 Introduction

Several studies have demonstrated increases in dissolved organic carbon (DOC) concentrations and colour in surface waters across large parts of Europe and North America over the last three decades (Stoddard et al., 2003; Hongve et al., 2004; Evans et al., 2005; Worrall and Burt, 2007; Erlandsson et al., 2008; Arvola et al., 2010). These trends have raised concerns about drinking water quality as contaminants and toxic compounds may be associated with DOC (Ledesma et al., 2012). This may lead to increased demands of chemical pre-treatment in drinking water plants. Increased DOC export to surface waters may also have major consequences for aquatic ecosystems (Karlsson et al., 2009) and recreational values, as well as the role of lakes as carbon sources to the atmosphere (Cole et al., 2007).

A number of hypotheses have been put forward as explanations of the recent increase in DOC concentration. Several studies have proposed a link to declining atmospheric acid deposition (Evans et al., 2006; Vourenmaa et al., 2006; Monteith et al., 2007), while others have coupled enhanced leaching of DOC from soils to changes in climate (Freeman et al., 2001; Hongve et al., 2004; Worrall and Burt, 2007; Haaland et al., 2010) or nitrogen deposition (Findlay, 2005). Local-scale land-use and land management practices have also been demonstrated to influence DOC concentrations (Corell et al., 2001; Mattsson et al., 2005; Armstrong et al., 2010; Yallop et al., 2011). The lack of scientific agreement on the mechanisms controlling DOC and colour variations in lake water during recent decades may reflect partly that many studies have been performed on catchment areas with heterogeneous types of land use, making it difficult to distinguish between co-existing forcing factors. Moreover, most studies have been based on monitoring data covering only a few decades, and have therefore failed to place the recent DOC trends in the perspective of the pronounced dynamics of anthropogenic atmospheric sulphur emissions that have occurred during the last century. Correspondingly, long-term changes in vegetation, land use and climate also have not been considered.

One way of gaining an increased understanding of this important environmental problem is to obtain long-term records of past changes in total organic carbon (TOC) concentration in lake water by using inference models derived from visible-near-infrared spectroscopy (VNIRS) of lake sediments (Rosén, 2005; Cunningham et al., 2011;

Rosén et al., 2011). Following methodological development, this palaeolimnological approach has recently gained increased attention as a trustworthy proxy for ambient variations in lake-water DOC concentrations, building on the fact that the dominant fraction (> 95 %) of TOC in Scandinavian surface waters consists of DOC, usually defined as organic matter not retained by a filter of 0.45 µm in nominal pore size (Wetzel, 2001). In boreal forested catchment areas, DOC is primarily allochthonous, originating from leaching of terrestrial soils. Additional autochthonous DOC may be produced in lakes by phytoplankton and aquatic macrophytes, although this part commonly constitutes only a minor fraction of the TOC pool (Bade et al., 2007). The composition and quantity of DOC may differ between sites depending on climate and catchment properties such as vegetation, hydrology and soil properties (e.g. Clark et al., 2010). Lake-water DOC concentrations and colour often show strong correlations (Pace and Cole, 2002; von Einem and Granéli, 2010) and their mutual increases over recent decades have been referred to as brownification (Granéli, 2012). Surface waters are variably coloured by humic substances formed by terrestrial humification during degradation of soil organic matter and may comprise 50–75 % of the DOC pool (McDonald et al., 2004). Humic substances absorb solar radiation, especially UV and short-wavelength visible radiation, and hence affect water temperature and aquatic productivity, with consequences for lake stratification and ecosystem functioning (Snucins and Gunn, 2000; Diehl et al., 2002; von Einem and Granéli, 2010). However, some studies have reported clear discrepancies between DOC concentrations and colour in lake water (Erlandsson et al., 2008; Kritzberg and Ekström, 2012), indicating that the composition of DOC at the molecular level may be equally important for changes in water colour.

Here we present a detailed multi-proxy study based on well-dated sediment successions from two small nearby lakes in southern Sweden spanning the last approximately 800 yr. One of them is oligotrophic mesohumic with a mosaic landscape in its catchment area and with a long history of anthropogenic disturbance. The other lake is oligotrophic polyhumic with a catchment area dominated by forest and wetlands, and is historically less influenced by anthropogenic disturbance (Bragée et al., 2013; Fredh et al., 2014). We applied a combination of palaeolimnological methods to the sediment sequences, including reconstructions of lake-water TOC concentration based on VNIRS (Rosén, 2005), diatom analysis to determine water pH and catchment land cover based on pollen analysis and the Landscape Reconstruction Algorithm approach (Sugita, 2007a, b). The aim of this study is to identify the major forcing mechanisms behind observed increases in TOC concentration in lakes of the upland area of southern Sweden during recent decades by comparing the impacts of changes in land use, sulphur deposition and climate to long-term trends in lake-water TOC concentration since AD 1200 inferred from proxy records. Particular focus is placed on the effects of differences in catchment characteristics and the degree of land-use intensity between the two study lakes. Ultimately, our findings may contribute to an enhanced understanding of lake-water TOC dynamics generally, on time-scales beyond monitoring series, and to prediction of the future development of lake-water quality in boreal environments.

2 Study area and site descriptions

The two study lakes, Åbodasjön and Lindhultsgöl, are situated 6 km apart, about 30 km northwest of Växjö in the province of Småland, southern Sweden (Fig. 1). The crystalline bedrock is dominated by granite and gneiss (Wikman, 2000) and covered by sandy till of various thicknesses and scattered peat deposits (Daniel, 2009). The area is part of the boreo-nemoral zone characterized by mixed coniferous and deciduous forest (Sjörs, 1963; Gustafsson, 1996). The climate is generally maritime with a mean annual temperature of 6.4 °C (January 2.7 °C, July 15.9 °C) and an annual precipitation of 651 mm (January 52 mm, July 75 mm), based on reference normals from Växjö for 1961–1990 (Alexandersson et al., 1991). The lakes are situated within the area of Sweden most significantly affected by increasing DOC concentrations since the 1990s (Löfgren, 2003). Lake size was also taken into account at the selection of study sites to enable reconstructions of local-scale land use based on fossil pollen records. The lakes are situated in the parish of Slätthög, established around AD 1000, and the first local population data are available from AD 1571, revealing 301 inhabitants (Andersson Palm, 2000). During the 1700s the population started to increase rapidly and a population peak was reached in the end of the 1800s, followed by a decrease in rural population due to industrialisation.

Åbodasjön (Table 1) is an oligotrophic mesohumic lake fed by two inlet streams, situated in the south and north-east, and with an outlet in the south-west. The village of Åboda (40 residents in 2004) is situated west of the lake, and the area around the lake margin is semi-open with mainly deciduous trees, grassland and cropland. The vegetation cover within the catchment area is dominated by managed coniferous woodland, wetlands and patches of grassland and cropland (Fig. 1).

Lindhultsgöl (Table 1) is an oligotrophic polyhumic lake with no natural inlets. At least two artificial ditches drain into the lake from nearby wetlands and woodland, and there is an outlet consisting of an artificial ditch in the south. The catchment area is covered by managed coniferous forest and wetlands with shrubs and scattered pine trees (Fig. 1).

3 Methods

3.1 Fieldwork, subsampling and dating

In early spring 2008 sequences of surface sediments were obtained from Åbodasjön and Lindhultsgöl, respectively, at water depths of 8.6 and 5.2 m, using a gravity corer and a 1 m-long russian peat corer. Correlations between core segments and surface sediments were based on mineral magnetic properties and X-ray fluorescence (XRF) measurements of element compositions. The uppermost 1 m parts of the sequences were subsampled into 0.5 cm contiguous sections for stratigraphic analyses. Age-depth models were based on ^{210}Pb dating along with ^{137}Cs, supplemented by radiocarbon dating of terrestrial plant remains and pollution lead (Pb) concentration variations (Bragée et al., 2013).

3.2 Visible-near infrared spectroscopy (VNIRS)

Past changes of TOC concentration in the lake waters were reconstructed using a calibration model based on visible-near infrared spectroscopy (VNIRS) of surface sediments from 140 Swedish lakes covering a TOC gradient from 0.7 to 24 mg L^{-1} (Cunningham et al., 2011). The inferred TOC concentrations from Lindhultsgöl exceeded the range within the calibration set and an additional set of 160 Canadian lakes with a DOC range of 0.6–39.6 mg L^{-1} was also used (Rouillard et al., 2011). The model performance of the combined Swedish and Canadian calibration set is similar to the Swedish calibration set with an R^2 value of 0.6 between measured and predicted TOC concentration and a root mean squared error of prediction (RMSEP) of 4.1 mg L^{-1} (10.5 % of the gradient).

3.3 Diatom analysis

Past changes in lake-water pH were reconstructed based on diatom assemblages in the sediment records. Diatom samples were prepared following standard methods (Battarbee et al., 2001). Following oxidization of freeze-dried sediment samples (0.01 g) with 15 % H_2O_2 solution for 24 h, 30 % H_2O_2 was added to digest organic matter using the water-bath technique described by Renberg (1990). For some samples HNO_3 was added to digest the remaining organic matter. To estimate diatom concentrations, known quantities of DVB (divinylbenzene) microspheres were added to the digested and cleaned samples (Battarbee and Kneen, 1982; Wolfe, 1997). Samples of 0.2 mL of the mixtures were evaporated onto cover slips and mounted onto microscope slides using ZRAX (refractive index = ~ 1.7+). At least 400 diatom valves per sample were counted under a light microscope at 1000 ×, using phase-contrast optics. The main taxonomic sources were Krammer and Lange-Bertalot (1986–1991), Lange-Bertalot and Krammer (1989) and Krammer (1992). The diatom counts were expressed as relative abundances of each taxon. Diatoms were grouped into planktonic and benthic taxa for calculation of planktonic/benthic (P/B) ratios, indicative of light availability, as deceased light penetration reduces benthic growth.

Changes in pH was inferred from sedimentary assemblages (Di-pH) using a transfer function set, the online combined pH training set in the European Diatom Database (http://craticula.ncl.ac.uk/Eddi/jsp/). The calibration set for the model consists of 627 lakes with a pH range of 4.3–8.4. The diatom-inferred pH was based on a locally-weighted weighted average model and inverse deshrinking.

3.4 Carbon and nitrogen elemental analyses

The C/N ratio of lake sediments gives an indication of the source (terrestrial and aquatic) of organic matter (Meyers and Lallier-Vergès, 1999). Acid-treated and freeze-dried sediment samples were analysed for sedimentary total organic carbon (TC) and total nitrogen (TN) contents by combustion, using a Costech ECS 4010 elemental analyser. The samples were pre-treated with 10 % HCl at 90 °C for 5–7 min for removal of potential trace amounts of $CaCO_3$. Elemental C/N ratios were converted to atomic ratios by multiplication with 1.167.

3.5 Trace element concentrations and x-ray fluorescence analysis (XRF)

Enhanced catchment erosion may be reflected by elevated concentrations of lithogenic elements in the sediment profile (Engstrom and Wright, 1984). Concentrations of zirconium (Zr) and titanium (Ti), in the sediments were measured by X-ray fluorescence (XRF) analysis (Boyle, 2000) followed by calculation of elemental Zr/Ti ratios for estimation of mineral grain-size variations within the lake sediments as Zr is commonly associated with silt particles and Ti often occurs in the fine silt and clay fractions (Koinig et al., 2003; Taboada et al., 2005). Freeze-dried samples at 2–5 cm intervals of the sediment sequences were measured, using an S2 Ranger XRF spectrometer for total concentrations of 35 different major and trace elements. The spectrometer was calibrated using certified reference materials. Mass attenuation correction was based on theoretical alpha coefficients, with calculations taking organic matter concentrations into account.

3.6 Pollen analysis and Landscape Reconstruction Algorithm (LRA)

Changes in land use were quantified using the Landscape Reconstruction Algorithm (LRA) based on pollen counts of dominant taxa (Sugita, 2007a, b) in the sediment records from the two study sites and from the larger, nearby lake Fiolen (Fredh et al., 2013). Pollen samples were prepared according to the standard acetolysis method, (Berglund and Ralska-Jasiewiczowa, 1986) using 1 cm^3 samples at 0.5 cm intervals. A minimum of 1000 pollen grains of 26 taxa were counted for contiguous 0.5 cm samples (1–10 samples) covering 20 yr time spans, a temporal resolution that averages out weather-induced year-to-year variations in pollen productivity (Fredh et al., 2012, 2013). The identification was based on pollen keys (Moore et al., 1991; Beug, 2004; Punt et al., 1976–2009) and the reference pollen collection at the Department of Geology, Lund University.

The LRA allows the estimation of changes in the spatial coverage of 26 target taxa at regional and local scales. The pollen data, the LRA approach with its associated parameters, and the reconstructions of land use were described in detail by Fredh et al. (2013, 2014) and Mazier et al. (2014). In this paper, we focus on local land-use dynamics at 20 yr intervals since AD 1200 at a spatial scale (modelled area) identified by Mazier et al. (2014) as a radius of 1740 m around Åbodasjön and 1440 m around Lindhultsgöl. The inferred cover of individual taxa are expressed as percentages and grouped into five different categories of land use according to the pollen/land use translation scheme in Mazier et al. (2014): grassland, cropland, wetland, coniferous woodland and deciduous woodland. Although the LRA approach provides no information on the spatial distribution of the types of land use within the modelled areas – larger than the actual catchment areas – we assume that the changes in land use within the modelled areas broadly reflect catchment-scale vegetation changes.

4 Results

Åbodasjön (Fig. 2): the inferred TOC reconstruction shows maximum inferred values of 14 $\mathrm{mg\,L^{-1}}$ around AD 1250, followed by a decrease to rather stable values at 9–10 $\mathrm{mg\,L^{-1}}$after AD 1450. Around AD 1800 a significant increase was recorded, reaching peak values at c. 12 $\mathrm{mg\,L^{-1}}$ between AD 1860 and 1910, followed by a substantial decrease to a sequence minimum of c. 7 $\mathrm{mg\,L^{-1}}$ in the 1980s. After AD 1990 an increase to 9–10 $\mathrm{mg\,L^{-1}}$ was recorded.

The diatom-inferred pH varied between 6.2 and 6.7. Periods of elevated pH were recorded at AD 1350–1500 and AD 1700–1780, while lower values were recorded at AD 1520–1670 and after AD 1970. The diatom concentration increased to a peak around AD 1450, followed by a decrease to relatively stable values and a second decrease after AD 1900. The planktonic diatom taxa varied between 40 and 70 % of the diatom assemblage, and elevated P/B ratios were recorded at AD 1250–1500 and in the top sample

Sediment total organic carbon content (TC) shows slightly elevated values at AD 1250–1350, followed by a slight transient decrease and a gradual increase after AD 1450 to stabilization at maximum TC content of c. 25 % in the 1800s. The C/N ratio increased from 13 in AD 1200 to c. 17 around AD 1300, followed by a slight decrease and a continuous increase from around AD 1450 to a sequence maximum of c. 23 at AD 1850–1900. Thereafter, a persistent decrease to c. 14 was recorded, followed by slight increases in both TC content and C/N ratios after AD 1990. The Zr/Ti ratio record shows a period of elevated values at AD 1320–1450, followed by a temporary decease and continuously elevated values at AD 1600–1900. After around AD 1950 a slight decrease was recorded.

The LRA-inferred woodland (coniferous and deciduous) cover around Åbodasjön varied between 33 and 80 % since AD 1200. The cover of grassland and cropland together was 40–50 % at AD 1240–1400, followed by a decrease to a minimum of 15 % at AD 1520–1540 when deciduous and coniferous woodland reached a peak in cover.

After around AD 1540 grassland and cropland cover increased and reached their maxima of c. 60 and 12 %, respectively, between AD 1820 and 1900. During the 1900s coniferous woodland, dominated by spruce, increased from 10 % to 30 %. Coniferous and deciduous woodland covers c. 60 % of the lake catchment today.

Lindhultsgöl (Fig. 2): the VNIRS-inferred TOC concentration exhibits high and stable values (21–22 $\mathrm{mg\,L^{-1}}$) at AD 1200–1500, followed by a small but sudden decrease to values around 20 $\mathrm{mg\,L^{-1}}$. After AD 1780 a gradual decrease was recorded, followed by a substantial decrease in AD 1900 to minimum values (12 $\mathrm{mg\,L^{-1}}$) around AD 1930. An increase was recorded at AD 1980, which was accentuated after AD 1990, and reached pre-1900 values in the surface sediments.

Diatom-inferred pH varied between 5.0 and 6.8. The highest value was recorded following an increase around AD 1250 to above 6 between AD 1300 and 1450. The period between AD 1500 and 1800 shows rather stable values around 5.8. In the 1900s, pH decreased to a minimum of 5.0 around AD 1960, followed by a slight increase until AD 2008. The pH reconstruction for Lindhultsgöl was influenced by a few dominating diatom taxa. The high values inferred in the lower parts were associated with the high abundance of the alkaliphilous ($\mathrm{pH} > 7$) diatom taxon *Aulacoseira ambigua* ($< 60\,\%$), and the low pH in the 1900s was affected by the dominant acidophilous ($\mathrm{pH} < 7$) taxon *Frustulia rhomboides* ($< 60\,\%$). The diatom concentration was high in the 1300s, followed by stable values until around AD 1900 when concentrations decreased. The planktonic diatom taxa varied between 15 and 85 % and the maximum in P/B ratio recorded in the 1300s was followed by rather stable ratios with a slight increase in the 1800s. Lowered P/B ratios were recorded after around AD 1900.

Relatively stable values were recorded for TC content at c. AD 1200–1700 and for C/N ratios at c. AD 1200–1550. This was followed by increasing values, peaking in the late 1800s. At c. AD 1900 significant decreases in both TC content and C/N ratio to minima in the 1980s were followed by reversed trends after c. AD 1990 (TC) and c. AD 2000 (C/N ratios) towards the top.

The Zr/Ti ratio shows a peak around AD 1350 following a gradual increase from c. AD 1250. After a subsequent decrease, the Zr/Ti ratio increased around AD 1500 to rather stable values in the AD 1800s, followed by a decrease after AD 1930.

The woodland (coniferous and deciduous) cover around Lindhulsgöl varied between 44 and 70 % during the last 800 yr. In contrast to Åbodasjön, wetlands covered more than 20 % during most of the period and decreased to less than 10 % after AD 1960. Grassland and cropland varied between 20 and 30 % at AD 1200–1580, followed by an increase to c. 40 %. During the 1900s coniferous woodland increased, and this land-use category covers c. 50 % of the lake catchment today.

5 Discussion

5.1 Impacts of land-use changes prior to AD 1900

In Åbodasjön, the highest TOC concentration was recorded at the beginning of the record around AD 1200, and decreased during the following century, while human impact increased (Fig. 2). From AD 1260 the pollen record indicates an agricultural expansion with increased extent of croplands, meadows and pastures in the catchment (Fredh et al., 2014). This expansion probably resulted in increased erosion and input of coarse lithogenic material, as indicated by elevated Zr/Ti ratios in the sediments. Also, elevated pH and diatom concentration suggest that more base cations and nutrients were released from the catchment. However, despite increased erosion, TOC concentration decreased in the lake water during the 1200s, which may be explained by increased openness, which lowered the biomass production from where a large portion of the TOC in lake water originates (Rosén et al., 2011).

At Lindhultsgöl, increasing anthropogenic impact was recorded during the 1200s by enhanced Zr/Ti ratios (Fig. 2) and increased charcoal concentrations (Fredh et al., 2014), reflecting increased erosion and land clearance by fire, respectively. During this time, the pH increased from 5.6 to 6.6, which was most likely caused by release of bases and nutrients from burning and grazing in the landscape (Renberg et al., 1993; Boyle, 2007). Moreover, the diatom concentration peaked, indicating temporarily enhanced aquatic productivity (cf. Rosén et al., 2011). However, there are no indications of major increases in open land cover around the lake. The persistently high TOC concentrations in the lake were probably related to the large proportion of wetlands within the catchment, which are often associated with substantial supplies of DOC to nearby lakes (Xenopoulos et al., 2003; Laudon et al., 2004; Mattsson et al., 2007). The lack of any response in lake-water TOC concentrations to catchment disturbance at Lindhultsgöl during the period of increased anthropogenic impact and alkalinization may be related to the unchanged proportion of open land, indicative of stable biomass production with a continuously high supply of DOC to the lake.

From c. AD 1350 there was a reduction of human-induced catchment disturbance at Åbodasjön, as indicated by a decline in cropland and grassland cover (Fig. 2), and coniferous woodland, in particular spruce, increased substantially around AD 1400. This agricultural regression was followed by decreasing catchment erosion and stabilization of TOC concentrations in the lake water, an event that may be related to the Black Death pandemic, which struck Sweden in AD 1350. This was followed by several outbreaks throughout the 1400s, and as much as 60–70 % of the farms in the region were abandoned (Lagerås, 2007; Myrdal, 2012). At c. AD 1450 there was a shift to lower TOC concentrations, accompanied by decreasing Zr/Ti ratios and diatom concentration. At Lindhultsgöl the regression led to decreases in catchment erosion, inferred pH and diatom concentration from c. AD 1400 in response to increased cover of coniferous woodland.

From c. AD 1450 to 1800 TOC concentrations in Åbodasjön were relatively stable, with only minor variations, despite major changes in land use. Following the increase at c. AD 1350, coniferous woodland reached maximum cover of c. 50 % around AD 1550, followed by a decrease related to the onset of a second agricultural expansion in the region (Lagerås, 2007). The pollen records from both lakes showed a gradual increase in cropland, meadows and pasture, more pronounced at Åbodasjön, together with enhanced erosion as reflected by increasing C/N and Zr/Ti ratios. Although these changes were not reflected in any of the inferred TOC concentration records, as would be expected, the slight decrease after AD 1650 at Åbodasjön may be a response to intensified agricultural activity, which also led to an increase in inferred pH.

A substantial increase in TOC concentration was recorded in Åbodasjön from c. AD 1800, peaking at AD 1860–1900, simultaneously with a substantial increase in population density (Fig. 2). The increase in rural population led to increased demands of land for crop cultivation, meadows and grazing, and areas previously regarded as less suitable for agriculture were cleared and drained (Myrdal, 1997). The pollen record shows a dominance of open-land taxa, and the open land cover, predominantly grassland, reached a maximum of c. 60 %. These changes were accompanied by maximum C/N ratios and TC values, reflecting an elevated input of terrestrial organic matter to the lake. From c. AD 1700 improvements of the agrarian management in Sweden enhanced food productivity through a number of reforms, such as land-divisions, crop rotation, irrigation, marling and better management of manure and urine (Emanuelsson, 2009). The introduction of agriculture in lake catchments, even at low proportions, is commonly associated with elevated DOC export and lake-water concentrations (Correll et al., 2001; McTiernan et al., 2001; Mattsson et al., 2005). In contrast to the decrease in the inferred TOC concentrations in response to the early agricultural expansion, the new agricultural management in the 1800s improved organic productivity through the application of manure and fertilizers, leading to increased leaching of DOC to the lake water (cf. McTiernan et al., 2001). These agricultural reforms, in combination with the general increase in land-use pressure, may hence explain the substantial increase of TOC concentration in Åbodasjön. This, in turn, most likely led to decreased pH in the lake as a result of enhanced organic acidity.

At Lindhultsgöl broadly similar trends in C/N ratio and TC from the late 1700s to c. AD 1900 as compared to Åbodasjön suggest increased land-use pressure and disturbance within the lake catchment. Coniferous woodland decreased, especially after AD 1800 and was partly replaced by deciduous woodland, indicating increased logging and expanding semi-open grazing areas. However, the high proportion of wetlands made the catchment less suitable for crop cultivation and resulted in a strikingly different development as compared to Åbodasjön. In the more marginal, forest-dominated area around Lindhultsgöl the increase in anthropogenic impact resulted in an increase in pH and a corresponding decrease TOC concentration in the lake around AD 1800, probably reflecting decreased catchment biomass in a gradually more open woodland.

5.2 Forcing mechanisms during the last century

Around AD 1900 pronounced decreases of TOC concentrations were recorded in both studied lakes (Figs. 2 and 3). In Åbodasjön the decrease was slightly more gradual, reaching minimum values in the 1980s, while the inferred values in Lindhultsgöl declined rapidly to a sequence minimum around AD 1940 (Fig. 3). At AD 1980–1990 increasing trends were initiated at both lakes. These inferred variations in TOC concentration during the last century are in general inversely correlated with historically documented trends in sulphur deposition regionally in southern Sweden (Fig. 3). Sulphur deposition started to increase at the onset of industrialisation at the end of the 1800s, which led to acidification of soils and surface waters across large parts of Europe (Rohde et al., 1995). Thereafter, sulphur deposition increased significantly in the 1940s, peaking at AD 1980–1995 (Schöpp et al., 2003), followed during recent decades by progressively decreasing deposition and wide-spread recovery from acidification through decreasing sulphate concentrations in lakes and streams throughout Europe and North America (Evans et al., 2001; Skjelkvåle et al., 2003). The timing of this recovery is largely consistent with the increasing TOC concentrations at our two study lakes (Fig. 3) as well as with other monitoring studies in Sweden (Erlandsson et al., 2010). Increases in lake-water DOC concentration have been linked to increased solubility of soil organic matter in response to declining acid deposition (Evans et al., 2006; Monteith et al., 2007), and conversely, elevated sulphur deposition usually results in reduced transport of soil organic matter. In our lakes declining inferred TOC concentrations were accompanied by decreasing C/N ratios, which suggests a reduction of terrestrial organic matter with increased acid deposition. Decreasing values of inferred pH from the late 1800s to minimum values around AD 1960 at Lindhultsgöl also provide evidence of the acidification history. Although the diatom-based pH reconstruction indicates continued acidification until the 1960s, the minimum in inferred TOC concentration was reached already around AD 1940, a few decades before sulphur deposition peaked. This may be explained by the high proportion of wetlands in the catchment of Lindhultsgöl. Evans et al. (2012) showed that already acidic soils may exhibit limited responses to enhanced acid deposition as DOC leaching stabilizes at a certain pH, below which no further decrease in DOC concentration occurs.

Despite the general anti-correlation between sulphur deposition and inferred TOC concentration at our study sites, major changes in land use during the last century may also have had important effects on DOC export to the lakes. The onset of industrialisation in the late 1800s led to urbanization and the documented decrease in rural population. Traditional types of land use were abandoned, in particular meadows and pastures, which were typically converted into spruce plantations and cultivated fields (Antonsson and Jansson, 2011). This development is clearly reflected in our pollen records as concomitant decreases in grassland and increases in coniferous woodland in the 1900s (Fig. 3). A significant reduction in the supply of terrestrial organic matter, as indicated by decreasing C/N ratios at both lakes, may be partly explained by the increase in sulphur deposition, which suppressed leaching of soil organic matter, although reduced catchment erosion may also have been a direct effect of stabilization of previously disturbed soils following the rural population decrease and woodland succession.

At the transition to commercial forestry and crop cultivation around AD 1900, new management practices with possible effects on lake-water TOC concentration, such as ditching, drainage and clear-cut harvesting were introduced. However, ditching and drainage of forests and crop cultivations may involve complex responses of surface-water DOC concentrations as some studies report increases (Ecke, 2008), while other provide evidence of decreases (Åström et al., 2001; McTiernan et al., 2001). In Sweden, ditching and drainage operations started in the late 1800s with substantial increases from AD 1900 to the 1930s (Hånell et al., 2009). A major artificial deepening of the inlet stream at Åbodasjön in the early 1920s resulted in enhanced export of lithogenic material from the adjacent croplands for a few decades (Bragée et al., 2013). The interruption of decreasing TOC concentrations in the lake at c. AD 1920–1960 (Fig. 3) was probably associated with enhanced supply of base cations, which led to a temporary alkalinization and a slight increase in pH (Fig. 2), possibly reinforced by increased supply of soil-derived DOC from the inlet surroundings. Crop cultivation along the inlet was abandoned in the 1950s, which led to decreased supply of lithogenic material (Bragée et al., 2013) and an accelerated decrease of TOC concentration in the lake. Ditching activities were very common until the 1990s, followed by a significant decrease, although new regulations no longer have to be reported to the authorities (Hånell et al., 2009). Hence, it is difficult to estimate the potential effects of ditching after the 1990s. Previous studies have attributed changing forestry practices to variations in the release of DOC to surface waters, and clear-cutting can significantly affect stream-water DOC levels in boreal forests (Lepistö et al., 2008; Laudon et al., 2009). Considering the increased areal distribution of woodland and forestry activities within the catchments during the last century, this may constitute a potential source of increased DOC supply. Given the increase in the extent of clear-cuts between AD 1946 and 2005, from 1 to 20 % at Åbodasjön and from 0 to 13 % at Lindhultsgöl (Mazier et al., 2014), this process may have contributed to the elevated TOC concentrations in the lakes in the 1990s. However, ditching and clear-cutting usually result in only temporary increases in the supply of DOC (Laudon et al., 2009) and may therefore be difficult to distinguish in palaeolimnological records.

The increase in inferred TOC concentration at both lakes around AD 1990 is most likely linked to the recovery from acidification. The low sample resolution in the uppermost parts of the diatom records precludes detailed evaluation of recent changes in pH in response to decreased sulphur deposition, although the slight increase in the uppermost part of the record from Lindhultsgöl indicates a recent recovery. However, pH is not a straightforward measure of recovery from acidification (Skjelkvåle et al., 2003; SanClements et al., 2012), and the inconsistent responses in our records may be explained by the contemporary increase in lake-water TOC concentration as organic acids usually have an acidifying effect (Evans et al., 2001). Soil conditions are important for the solubility of organic matter, and the high proportion of coniferous woodland at both lakes and wetlands at Lindhultsgöl, typically associated with organic-rich soils may have induced increased leaching of DOC in response to decreasing sulphur deposition during recent decades (Evans et al., 2012). Site-specific catchment soil properties may therefore be important for explaining the observed increases in TOC concentration in our study lakes after AD 1990 compared to other lakes in the region that show unchanged or even decreasing trends in DOC concentration (von Einem and Granéli, 2010). In addition, wetland areas in the catchments of both lakes have been treated by liming on a yearly basis to mitigate acidification, starting in AD 1984 at Åbodasjön and in AD 1993 at Lindhultsgöl, which may have contributed to the effects of declining sulphur deposition by accelerated leaching of DOC to the lakes (cf. Hindar et al., 1996).

In addition to changes in sulphur deposition and land management practices, climate may affect DOC concentration in lake water through a variety of processes, including temperature-driven soil organic productivity and decomposition as well as precipitation-driven water table fluctuations and transport of organic carbon from terrestrial soils (e.g. Sobek et al., 2007). Increases in precipitation and temperature have been brought forward as potential causes of observed increases in DOC concentration in lake water during the last three decades in several studies (Freeman et al., 2001; Hongve et al., 2004; Sarkkola et al., 2009). Future climate predictions for northern Europe include higher seasonal amounts and intensity of precipitation, and increasing mean annual air temperatures (Alcamo et al., 2002), which may result in continued increases in DOC export to lake waters (Larsen et al., 2010). Available meteorological data from Växjö (Fig. 1), reaching back to AD 1860, show an increase in annual precipitation from c. AD 1980 and an increase in mean annual temperature from c. AD 1990 (Fig. 3). Hence, climate change may have contributed to the observed and reconstructed increases in lake-water TOC concentration over recent decades, although changes in land use and sulphur deposition have probably played more important roles at the centennial time-scale (Fig. 4). This demonstrates the importance of applying a long-term perspective on lake-water DOC dynamics in order to differentiate between causal relationships.

5.3 Recent brownification and future implications

Our reconstructions indicate that TOC concentrations in the lakes were generally high during the past eight centuries, reaching similar or higher concentrations than those observed during recent decades. Commonly, there is a correlation between colour (usually measured as absorbance at c. 300 nm or using the platinum scale) and DOC concentration in lake water. However, colour is strongly influenced by the composition of DOC, and a recent study has demonstrated that declining acidification has led to increased leaching of mobile, hydrophobic and aromatic DOC from soils in southern Sweden containing relatively large and strongly coloured molecular compounds (Ekström et al., 2011). Moreover, iron has a strong influence on water colour, and elevated iron concentrations have been observed with the recent brownification in the UK (Neal et al., 2008) as well as in Sweden (Huser et al., 2011; Kritzberg and Ekström, 2012). Therefore, the inferred changes in TOC concentration in our two study lakes may not necessarily reflect changes in colour, although monitoring data from Åbodasjön indicate that this was indeed the case during recent decades, consistent with increases in water colour observed in several other lakes in the study region. This is supported by high abundance of the diatom *Aulacoseira tenella* (> 20 %) in surface sediments from Åbodasjön, a species often associated with high DOC concentrations and strongly coloured lake waters (Huttunen and Turkia, 1994).

In contrast, the elevated TOC concentrations recorded in Åbodasjön during the late 1800s were most likely not associated with a corresponding increase in colour, as indicated by unchanged diatom P/B ratios. Benthic and planktonic diatom communities are likely to respond to changes in the input of terrestrial organic matter through associated effect on the transparency of the water column, as the benthic community is primarilary limited by light in nutrient-poor lakes (Rosén et al., 2009; Karlsson et al., 2009). The pronounced increase in agricultural intensity in the late 1800s probably resulted in enhanced export of DOC characterized by relatively low molecular weight (cf. Cronan et al., 1999; Dalzell et al., 2011). A dominance of this type of DOC would not result in any significant increase in water colour as DOC derived from agricultural areas is in general structurally less complex and less colored than DOC from forest soils (Wilson and Xenopoulos, 2009).

The early agricultural expansion in the 1200s resulted in a change in the diatom community towards elevated P/B ratios and a dominance of planktonic taxa typically favoured by high pH (Fig. 2). Hence, this change was not associated with any major increase in water colour caused by increased input of terrestrial organic carbon. Possibly, the increase in diatom productivity and concentration led to reduced light penetration and decreased proportions of benthic diatoms (cf. Chandra et al., 2005).

At Lindhultsgöl minimum P/B ratios were recorded during the period of maximum sulphur emissions at AD 1950–1990, which indicates a decrease in water colour associated with the corresponding minima in TOC concentration and pH. However, in Åbodasjön the minimum in TOC concentration was not associated with any corresponding decrease in P/B ratio. This may be explained by the difference in the extent of the inferred decrease in TOC concentration, with the most dramatic change recorded at Lindhultsgöl, probably resulting in a larger effect on the diatom assemblage.

Based on our results we can conclude that the increases in TOC concentration and colour in our study lakes during the past three decades have been driven mainly by declining atmospheric sulphur deposition. This suggests a recovery from the phase of maximum sulphur emissions, which resulted in exceptionally low TOC concentrations in the lakes at c. AD 1930–1990. However, our long-term records also demonstrate that the TOC concentrations of the study lakes were strongly influenced by changes in agricultural practices and general land-use pressure during the last 800 yr (Fig. 4). Site-specific catchment characteristics and land-use dynamics are of great importance for lake-water DOC variations, as exemplified by the sediment-based records from our two study lakes, which have experienced clear differences historically in the extent of agricultural activity in their catchments. The recently initiated increase in TOC concentration in the lakes may continue in the near future depending on the quantity of organic carbon stored in catchment soils due to suppression of DOC leaching during the acidification episode. However, the recovery of TOC concentrations has now reached historically "normal" levels, which may lead to a leveling off of the increasing trend. Given the reduction of atmospheric sulphur emissions during recent decades it is likely that previously suppressed or masked effects of land management and climate during the last century will become progressively more important drivers of TOC concentration in lake water in the future.

Acknowledgements. The authors are grateful to Shinya Sugita for input on the quantitative vegetation reconstructions, and Sofia Holmgren, Linda Randsalu-Wendrup and Christian Bigler for helpful support with diatom preparation and categorisation. We acknowledge the Swedish Meteorological Institute (SMHI) for precipitation and temperature data, and the Swedish Environmental Research Institute and MAGIC for sulphur deposition data. This work was funded by the Swedish Research Council Formas (grant to W. Granéli).

References

Alcamo, J., Mayerhofer, P., Guardans, R., van Harmelen, T., van Minnen, J., Onigkeit, J., Posch, M., and de Vries, B.: An integrated assessment of regional air pollution and climate change in Europe: findings of the AIR-CLIM Project, Environ. Sci. Policy, 5, 257–272, 2002.

Alexandersson, H., Karlström, C., and Larsson-McCann, S.: Temperature and precipitation in Sweden, 1961–90, Reference normals, SMHI report 81, SMHI, Norrköping, 1991.

Andersson Palm, L.: Folkmängden i Sveriges Socknar Och Kommuner 1571–1997, Books-on-Demand, Göteborg, 2000.

Antonsson, H. and Jansson, U. (Eds.): Agriculture and Forestry in Sweden Since 1900, The Royal Swedish Academy of Agriculture and Forestry, Stockholm, 512 pp., 2011.

Armstrong, A., Holden, J., Kay, P., Francis, B., Foulger, M., Gledhill, S., McDonald, A. T., and Walker, A.: The impact of peatland drain-blocking on dissolved organic carbon loss and discolouration of water; results from a national survey, J. Hydrol., 381, 112–120, 2010.

Arvola, L., Rask, M., Ruuhijärvi, J., Tulonen, T., Vuorenmaa, J., Ruoho-Airola, T., and Tulonen, J.: Long-term patterns in pH and colour in small acidic boreal lakes of varying hydrological and landscape settings, Biogeochemistry, 101, 269–279, 2010.

Åström, M., Aaltonen, E.-K., and Koivusaari, J.: Effect of ditching operations on stream-water chemistry in a boreal forested catchment, Sci. Total Environ., 279, 117–129, 2001.

Bade, D. L., Carpenter, S. R., Cole, J. J., Pace, M. L., Kritzberg, E., Van de Bogert, M. C., Cory, R. M., and McKnight, D. M.: Sources and fates of dissolved organic carbon in lakes as determined by whole-lake carbon isotope additions, Biogeochemistry, 84, 115–129, 2007.

Battarbee, R. W. and Kneen, M. J.: The use of electronically counted microspheres in absolute diatom analysis, Limnol. Oceanogr., 27, 184–188, 1982.

Battarbee, R. W., Jones, V. J., Flower, R. J., Cameron, N. G., Bennion, H., Carvalho, L., and Juggins, S.: Diatoms, in: Tracking Environmental Change Using Lake Sediments, Vol. 3: Terrestrial, Algal and Siliceous Indicators, edited by: Smol, J. P., Birks, H. J. B., and Last, W. M., Kluwer Academic, Dortrecht, 155–202, 2001.

Berglund, B. and Ralska-Jasiewiczowa, M.: Pollen analysis and pollen diagrams, in: Handbook of Holocene Palaeoecology and Palaeohydrology, edited by: Berglund, B. E., John Wiley & Sons Ltd, Chichester, New York, Brisbane, Toronto, Singapore, 155–484, 1986.

Beug, H.-J. (Ed.): Leitfaden der Pollenbestimmung für Mitteleuropa und angrenzende Gebiete, Verlag Dr Friedrich Pfeil, Munich, 542 pp., 2004.

Boyle, J. F.: Rapid elemental analysis of sediment samples by isotope source XRF, J. Paleolimnol., 23, 213–221, 2000.

Boyle, J. F.: Loss of apatite caused irreversible early-Holocene lake acidification, Holocene, 17, 543–547, 2007.

Bragée, P., Choudhary, P., Routh, J. Boyle, J. F., and Hammarlund, D.: Lake ecosystem responses to catchment disturbance and airborne pollution: an 800-year perspective in southern Sweden, J. Paleolimnol., 50, 545–560, 2013.

Chandra, S., Vander Zanden, M. J., Heyvaert, A. C., Richards, B. C., Allen, B. C., and Goldman, C. R.: The effects of cultural eutrophication on the coupling between pelagic primary producers and benthic consumers, Limnol. Oceanogr., 50, 1368–1376, 2005.

Clark, J. M., Bottrell, S. H., Evans, C. D., Monteith, D. T., Bartlett, R., Rose, R., Newton, R. J., and Chapman, P. J.: The importance of the relationship between scale and process in understanding long-term DOC dynamics, Sci. Total Environ., 408, 2768–2775, 2010.

Cole, J. J., Prairie, Y. T., Caraco, N. F., McDowell, W. H., Tranvik, L. J., Striegl, R. G., Duarte, C. M., Kortelainen, P., Downing, J. A., Middelburg, J. J., and Melack, J.: Plumbing the global carbon cycle: integrating inland waters into the terrestrial carbon budget, Ecosystems, 10, 172–185, 2007.

Correll, D. L., Jordan, T. E., and Weller, D. E.: Effects of precipitation, air temperature, and land use on organic carbon discharges from Rhode River watersheds, Water Air Soil Poll., 128, 139–159, 2001.

Cronan, C. S., Piampiano, J. T., and Patterson, H. H.: Influence of land use and hydrology on exports of carbon and nitrogen in a Maine river basin, J. Environ. Qual., 28, 953–961, 1999.

Cunningham, L., Bishop, K., Mettävainio, E., and Rosén, P.: Paleoecological evidence of major declines in total organic carbon concentrations since the nineteenth century in four nemoboreal lakes, J. Paleolimnol., 45, 507–518, 2011.

Dalzell, B. J., King, J. Y., Mulla, D. J., Finlay, J. C., and Sands, G. R.: Influence of subsurface drainage on quantity and quality of dissolved organic matter export from agricultural landscapes, J. Geophys. Res., 116, G02023, doi:10.1029/2010JG001540, 2011.

Daniel, E.: Beskrivning Till Jordartskartan 5E Växjö NV, K 168, Sveriges Geologiska Undersökning (SGU), 2009.

Diehl, S., Berger, S., Ptacnik, R., and Wild, A.: Phytoplankton, light, and nutrients in a gradient of mixing depths: field experiments, Ecology, 83, 399–411, 2002.

Ecke, F.: Drainage ditching at the catchment scale affects water quality and macrophyte occurrence in Swedish lakes, Freshwater Biol., 54, 119–126, 2008.

Ekström, S. M., Kritzberg, E. S., Kleja, D. B., Larsson, N., Nilsson, P. A., Graneli, W., and Bergkvist, B.: Effect of acid deposition on quantity and quality of dissolved organic matter in soil–water, Environ. Sci. Technol., 45, 4733–4739, 2011.

Emanuelsson, U. (Ed.): The rural landscapes of Europe: how man has shaped European nature, Swedish Research Council Formas, Stockholm, Sweden, 384 pp., 2009.

Engstrom, D. R. and Wright, H. E. J.: Chemical stratigraphy of lake sediments as a record of environmental change, in: Lake Sediments and Environmental History, edited by: Haworth, E. Y. and Lund, J. W. G., Leicester University Press, Bath, 11–68, 1984.

Erlandsson, M., Buffam, I., Fölster, J., Laudon, H., Temnerud, J., Weyhenmeyer, G. A., and Bishop, K.: Thirty-five years of synchrony in the organic matter concentrations of Swedish rivers explained by variation in flow and sulphate, Glob. Change Biol., 14, 1191–1198, 2008.

Erlandsson, M., Cory, N., Köhler, S., and Bishop, K.: Direct and indirect effects of increasing dissolved organic carbon levels on pH in lakes recovering from acidification, J. Geophys. Res., 115, G03004, doi:10.1029/2009JG001082, 2010.

Evans, C. D., Cullen, J. M., Alewell, C., Kopácek, J., Marchetto, A., Moldan, F., Prechtel, A., Rogora, M., Veselý, J., and Wright, R.: Recovery from acidification in European surface waters, Hydrol. Earth Syst. Sci., 5, 283–298, doi:10.5194/hess-5-283-2001, 2001.

Evans, C. D., Monteith, D. T., and Cooper, D. M.: Long-term increases in surface water dissolved organic carbon: observations, possible causes and environmental impacts, Environ. Pollut., 137, 55–71, 2005.

Evans, C. D., Chapman, P. J., Clark, J. M., Monteith, D. T., and Cresser, M. S.: Alternative explanations for rising dissolved organic carbon export from organic soils, Glob. Change Biol., 12, 2044–2053, 2006.

Evans, C. D., Jones, T. G., Burden, A., Ostle, N., Zieliñski, P., Cooper, M. D., Peacock, M., Clark, J. M., Oulehle, F., Cooper, D., and Freeman, C.: Acidity controls on dissolved organic carbon mobility in organic soils, Glob. Change Biol., 18, 3317–3331, 2012.

Findlay, S. E.: Increased carbon transport in the Hudson River: unexpected consequence of nitrogen deposition?, Front. Ecol. Environ., 3, 133–137, 2005.

Fredh, D., Broström, A., Zillén, L., Mazier, F., Rundgren, M., and Lagerås, P.: Floristic diversity in the transition from traditional to modern land-use in southern Sweden, A.D. 1800–2008, Veg. Hist. Archaeobot., 21, 439–452, 2012.

Fredh, D., Broström, A., Rundgren, M., Lagerås, P., Mazier, F., and Zillén, L.: The impact of land-use change on floristic diversity at regional scale in southern Sweden 600 BC–AD 2008, Biogeosciences, 10, 3159–3173, doi:10.5194/bg-10-3159-2013, 2013.

Fredh, D., Mazier, F., Bragée, P., Lagerås, P., Rundgren, M., Hammarlund, D., and Broström, A.: The effect of local land-use on floristic diversity during the past 1000 years in southern Sweden, Holocene, submitted, 2014.

Freeman, C., Evans, C. D., Monteith, D. T., Reynolds, B., and Fenner, N.: Export of organic carbon from peat soils, Nature, 412, 785, doi:10.1038/35090628, 2001.

Granéli, W.: Brownification of lakes, in: Encyclopedia of Lakes and Reservoirs, edited by: Bengtsson, L., Herschy, R. W., and Fairbridge, R. W., Springer Science, New York, 117–119, 2012.

Gustafsson, L.: Geographical classifications of plants and animals, in: National Atlas of Sweden. Geography of Plants and Animals, edited by: Gustafsson, L. and Ahlén, I., SNA publishing, Stockholm, 25–28, 1996.

Haaland, S., Hongve, D., Laudon, H., Riise, G., and Vogt, R. D.: Quantifying the drivers of the increasing colored organic matter in boreal surface waters, Environ. Sci. Technol., 44, 2975–2980, 2010.

Hindar, A., Kroglund, F., Lydersen, E., Skiple, A., and Høgberget, R.: Liming of wetlands in the acidified Lake Røynelandsvatn catchment in southern Norway: effects on stream water chemistry, Can. J. Fish. Aquat. Sci., 53, 985–993, 1996.

Hongve, D., Riise, G., and Kristiansen, J.: Increased colour and organic acid concentrations in Norwegian forest lakes and drinking water – a result of increased precipitation?, Aquat. Sci., 66, 231–238, 2004.

Huser, B. J., Köhler, S. J., Wilander, A., Johansson, K., and Fölster, J.: Temporal and spatial trends for trace metals in streams and rivers across Sweden (1996–2009), Biogeosciences, 8, 1813–1823, doi:10.5194/bg-8-1813-2011, 2011.

Huttunen, P. and Turkia, J.: Diatoms as indicators of alkalinity and TOC in lakes: estimation of optima and tolerances by weighted averaging, in: Proceedings of the 11th International Diatom Symposium, San Francisco, USA, 12–17 August 1990, Memoirs of the California Academy of Sciences, No 17, edited by: Kociolek, J. P., 649–658, 1994.

Hånell, B.: Möjlighet Till Höjning av Skogsproduktionen i Sverige Genom Dikesrensning, Dikning Och Gödsling av Torvmarker, in: Skogsskötsel För Ökad Tillväxt, edited by: Fahlvik, N., Johansson, U., and Nilsson, U., Faktaunderlag Till MINT-Utredningen, Rapport, Bilaga 4, SLU, 1–28, 2009.

Karlsson, J., Bystrom, P., Ask, J., Ask, P., Persson, L., and Jansson, M.: Light limitation of nutrient-poor lake ecosystems, Nature, 460, 506–509, 2009.

Koinig, K. A., Shotyk, W., Lotter, A. F., Ohlendorf, C., and Sturm, M.: 9000 years of geochemical evolution of lithogenic major and trace elements in the sediment of an alpine lake – the role of climate, vegetation, and land-use history, J. Paleolimnol., 30, 307–320, 2003.

Krammer, K.: Pinnularia: Eine Monographie der Europäischen Taxa, Bibliotheca Diatomologica, Vol. 26, J. Cramer, Stuttgart, 353 pp., 1992.

Krammer, K. and Lange-Bertalot, H.: *Bacillariophyceae*, 1. *Naviculaceae*, in: Süsswasserflora von Mitteleuropa, Vol. 2, edited by: Ettl, H., Gärtner, G., Gerloff, J., Heynig, H., and Mollenhauer, D., Gustav Fischer Verlag, Stuttgart, 876 pp., 1986.

Krammer, K. and Lange-Bertalot, H.: *Bacillariophyceae*, 2. *Bacillariaceae*, in: Süsswasserflora von Mitteleuropa, Vol. 2, edited by: Ettl, H., Gärtner, G., Gerloff, J., Heynig, H., and Mollenhauer, D., Gustav Fischer Verlag, Stuttgart, 596 pp., 1988.

Krammer, K. and Lange-Bertalot, H.: *Bacillariophyceae*, 3. *Centrales*, *Fragilariaceae*, *Eunotiaceae*, in: Süsswasserflora von Mitteleuropa, Vol. 2, edited by: Ettl, H., Gärtner, G., Gerloff, J., Heynig, H., and Mollenhauer, D., Gustav Fischer Verlag, Stuttgart, 576 pp., 1991a.

Krammer, K. and Lange-Bertalot, H.: *Bacillariophyceae*, 4. *Achnanthaceae*, Kritische Ergänzungen zu *Navicula* (*Lineolatae*) und *Gomphonema*, in: Süsswasserflora von Mitteleuropa, Vol. 2, edited by: Ettl, H., Gärtner, G., Gerloff, J., Heynig, H., and Mollenhauer, D., Gustav Fischer Verlag, Stuttgart, 437 pp., 1991b.

Kritzberg, E. S. and Ekström, S. M.: Increasing iron concentrations in surface waters – a factor behind brownification?, Biogeosciences, 9, 1465–1478, doi:10.5194/bg-9-1465-2012, 2012.

Lagerås, P.: The Ecology of Expansion and Abandonment – Medieval and Post-Medieval Land-Use and Settlement Dynamics in a Landscape Perspective, National Heritage Board, Stockholm, 256 pp., 2007.

Lange-Bertalot, H. and Krammer, K.: *Achnanthes*, eine Monographie der Gattung: mit Definition der Gattung *Cocconeis* und Nachträgen zu den *Naviculaceae*, Bibliotheca Diatomologica, Vol. 18, J. Cramer, Stuttgart, 393 pp., 1989.

Larsen, S., Andersen, T. O. M., and Hessen, D. O.: Climate change predicted to cause severe increase of organic carbon in lakes, Glob. Change Biol., 17, 1186–1192, 2010.

Laudon, H., Köhler, S., and Buffam, I.: Seasonal TOC export from seven boreal catchments in northern Sweden, Aquat. Sci., 66, 223–230, 2004.

Laudon, H., Hedtjärn, J., Schelker, J., Bishop, K., Sørensen, R., and Ågren, A.: Response of dissolved organic carbon following forest harvesting in a boreal forest, AMBIO, 38, 381–386, 2009.

Ledesma, J. L. J., Köhler, S. J., and Futter, M. N.: Long-term dynamics of dissolved organic carbon: Implications for drinking water supply, Sci. Total Environ., 432, 1–11, 2012.

Lepistö, A., Kortelainen, P., and Mattsson, T.: Increased organic C and N leaching in a northern boreal river basin in Finland, Global Biogeochem. Cy., 22, GB3029, doi:10.1029/2007GB003175, 2008.

Löfgren, S., Forsius, M., and Andersen, T.: The color of water – climate induced water color increase in Nordic lakes and streams due to humus, Nordic Council of Ministers Brochure, Copenhagen, 12 pp., 2003.

Mattsson, T., Kortelainen, P., and Räike, A.: Export of DOM from boreal catchments: impacts of land use cover and climate, Biogeochemistry, 76, 373–394, 2005.

Mattsson, T., Kortelainen, P., Lepistö, A., and Räike, A.: Organic and minerogenic acidity in Finnish rivers in relation to land use and deposition, Sci. Total Environ., 383, 183–192, 2007.

Mazier, F., Broström, P., Bragée, P., Fredh, D., Stenberg, L., Thiere, G., Sugita, S., and Hammarlund, D.: Two hundred years of land-use change in south Swedish uplands: comparison of historical map-based estimates with pollen-based reconstruction using the Landscape Reconstruction Algorithm, Rev. Palaeobot. Palyno., in review, 2014.

McDonald, S., Bishop, A. G., Prenzler, P. D., and Robards, K.: Analytical chemistry of freshwater humic substances, Anal. Chim. Acta, 527, 105–124, 2004.

McTiernan, K. B., Jarvis, S. C., Scholefield, D., and Hayes, M. H. B.: Dissolved organic carbon losses from grazed grasslands under different management regimes, Water Res., 35, 2565–2569, 2001.

Meyers, P. A. and Lallier-Verges, E.: Lacustrine sedimentary organic matter records of Late Quaternary paleoclimates, J. Paleolimnol., 21, 345–372, 1999.

Monteith, D. T., Stoddard, J. L., Evans, C. D., De Wit, H. A., Forsius, M., Hogasen, T., Wilander, A., Skjelkvale, B. L., Jeffries, D. S., Vuorenmaa, J., Keller, B., Kopacek, J., and Vesely, J.: Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry, Nature, 450, 537–540, 2007.

Moore, P. D., Webb, J. A., and Collinson, M. E.: Pollen Analysis, 2nd edn., Blackwell Scientific Publications, Oxford, 216 pp., 1991.

Myrdal, J.: En agrarhistorisk syntes, in: Agrarhistoria, edited by: Larsson, B. M. P., Morell, M., and Myrdal, J., LTs Förlag, Stockholm, 302–322, 1997.

Myrdal, J.: Scandinavia, in: Agrarian Change and Crisis in Europe, 1200–1500, edited by: Kitsikopoulos, H., Routledge, New York, 204–249, 2012.

Neal, C., Lofts, S., Evans, C. D., Reynolds, B., Tipping, E., and Neal, M.: Increasing iron concentrations in UK upland waters, Aquat. Geochem., 14, 263–288, 2008.

Pace, M. L. and Cole, J. J.: Synchronous variation of dissolved organic carbon and color in lakes, Limnol. Oceanogr., 47, 333–342, 2002.

Punt, W., Blackmore, S., Clarke, G. S. C., and Hoen, P. P.: The Northwest European Pollen Flora, Vol. 1–9, Elsevier, Amsterdam, Oxford, New York, 1976–2009.

Renberg, I.: A procedure for preparing large sets of diatom slides from sediment cores, J. Paleolimnol., 4, 87–90,1990.

Renberg, I., Korsman, T., and Birks, H. J. B.: Prehistoric increases in the pH of acid-sensitive Swedish lakes caused by land-use changes, Nature, 362, 824–827, 1993.

Rohde, H., Grennfelt, P., Wisniewski, J., Ågren, C., Bengtsson, G., Johansson, K., Kauppi, P., Kucera, V., Rasmussen, I., Rosseland, B., Schotte, I., and Selldén, G.: Acid Reign '95 – conference summary statement, Water Air Soil Pollut., 85, 1–14, 1995.

Rosén, P.: Total organic carbon (TOC) of lake water during the Holocene inferred from lake sediments and near-infrared spectroscopy (NIRS) in eight lakes from northern Sweden, Biogeochemistry, 76, 503–516, 2005.

Rosén, P., Cunningham, L., Vonk, J., and Karlsson, J.: Effects of climate on organic carbon and the ratio of planktonic to benthic primary producers in a subarctic lake during the past 45 years, Limnol. Oceanogr., 54, 1723–1732, 2009.

Rosén, P., Bindler, R., Korsman, T., Mighall, T., and Bishop, K.: The complementary power of pH and lake-water organic carbon reconstructions for discerning the influences on surface waters across decadal to millennial time scales, Biogeosciences, 8, 2717–2727, doi:10.5194/bg-8-2717-2011, 2011.

Rouillard, A., Rosén, P., Douglas, M. S., Pienitz, R., and Smol, J. P.: A model for inferring dissolved organic carbon (DOC) in lakewater from visible-near-infrared spectroscopy (VNIRS) measures in lake sediment, J. Paleolimnol., 46, 187–202, 2011.

SanClements, M. D., Oelsner, G. P., McKnight, D. M., Stoddard, J. L., and Nelson, S. J.: New insights into the source of decadal increases of dissolved organic matter in acid-sensitive lakes of the Northeastern United States, Environ. Sci. Technol., 46, 3212–3219, 2012.

Sarkkola, S., Koivusalo, H., Laurén, A., Kortelainen, P., Mattsson, T., Palviainen, M., Piirainen, S., Starr, M., and Finér, L.: Trends in hydrometeorological conditions and stream water organic carbon in boreal forested catchments, Sci. Environ., 408, 92–101, 2009.

Schöpp, W., Posch, M., Mylona, S., and Johansson, M.: Long-term development of acid deposition (1880–2030) in sensitive freshwater regions in Europe, Hydrol. Earth Syst. Sci., 7, 436–446, doi:10.5194/hess-7-436-2003, 2003.

Sjörs, H.: Amphi-Atlantic zonation, nemoral to Arctic, in: North Atlantic Biota and Their History, edited by: Löve, A. and Löve, D., Pergamon Press, Oxford, 109–125, 1963.

Skjelkvåle, B. L., Evans, C., Larssen, T., Hindar, A., and Raddum, G. G.: Recovery from acidification in European surface waters: a view to the future, AMBIO, 32, 170–175, 2003.

Snucins, E. and Gunn, J.: Interannual variation in the thermal structure of clear and colored lakes, Limnol. Oceanogr., 45, 1639–1646, 2000.

Sobek, S., Tranvik, L. J., Prairie, Y. T., Kortelainen, P., and Cole, J. J.: Patterns and regulation of dissolved organic carbon: an analysis of 7500 widely distributed lakes, Limnol. Oceanogr., 52, 1208–1219, 2007.

Stoddard, J. L., Kahl, J. S., Deviney, F. A., DeWalle, D. R., Driscoll, C. T., Herlihy, A. T., Kellogg, J. H., Murdoch, P. S., Webb, J. R., and Webster, K. E.: Response of Surface Water Chemistry to the Clean Air Act Amendments of 1990, Report EPA 620/R-03/001, US Environmental Protection Agency, North Carolina, 2003.

Sugita, S.: Theory of quantitative reconstruction of vegetation I: pollen from large sites REVEALS regional vegetation composition, Holocene, 17, 229–241, 2007a.

Sugita, S.: Theory of quantitative reconstruction of vegetation II: all you need is LOVE, Holocene, 17, 243–257, 2007b.

Taboada, T., Cortizas, A. M., García, C., and García-Rodeja, E.: Particle-size fractionation of titanium and zirconium during weathering and pedogenesis of granitic rocks in NW Spain, Geoderma, 131, 218–236, 2005.

von Einem, J. and Granéli, W.: Effects of fetch and dissolved organic carbon on epilimnion depth and light climate in small forest lakes in southern Sweden, Limnol. Oceanogr., 55, 920–930, 2010.

Wetzel, R.: Limnology, 3rd edn.: Lake and River Ecosystems, Academic Press, San Diego, 1006 pp., 2001.

Wikman, H.: Beskrivning Till Berggrundskartorna 5E Växjö NO och NV, Af 201 and 216, Sveriges Geologiska Undersökning (SGU), 2000.

Wilson, H. F. and Xenopoulos, M. A.: Effects of agricultural land use on the composition of fluvial dissolved organic matter, Nat. Geosci., 2, 37–41, 2009.

Wolfe, A. P.: On diatom concentrations in lake sediments: results from an inter-laboratory comparison and other tests performed on a uniform sample, J. Paleolimnol., 18, 261–268, 1997.

Worrall, F. and Burt, T. P.: Trends in DOC concentration in Great Britain, J. Hydrol., 346, 81–92, 2007.

Xenopoulos, M. A., Lodge, D. M., Frentress, J., Kreps, T. A., Bridgham, S. D., Grossman, E., and Jackson, C. J.: Regional comparisons of watershed determinants of dissolved organic carbon in temperate lakes from the Upper Great Lakes region and selected regions globally, Limnol. Oceanogr., 48, 2321–2334, 2003.

Yallop, A. R., Clutterbuck, B., and Thacker, J.: Increases in humic dissolved organic carbon export from upland peat catchments: the role of temperature, declining sulphur deposition and changes in land management, Clim. Res., 45, 43–56, 2011.

Table 1. Morphometric and hydrological characteristics of the two study lakes, sampled in July 2007 (von Einem and Granéli, 2010).

	Åbodasjön	Lindhultsgöl
Altitude (m)	221	212
Lake surface area (km^2)	0.5	0.07
Maximum depth (m)	9	5
Catchment area (km^2)	9.5	0.6
Residence time (yr)	0.5	–
pH	7.0	6.4
Alkalinity ($mEq\,L^{-1}$)	0.56	0.83
Chlorophyll *a* conc. ($\mu g\,L^{-1}$)	7.7	14.9
DOC conc. ($mg\,L^{-1}$)	11.0	23.8
Water colour ($mg\,Pt\,L^{-1}$)	40	960
Liming started	1984	1993

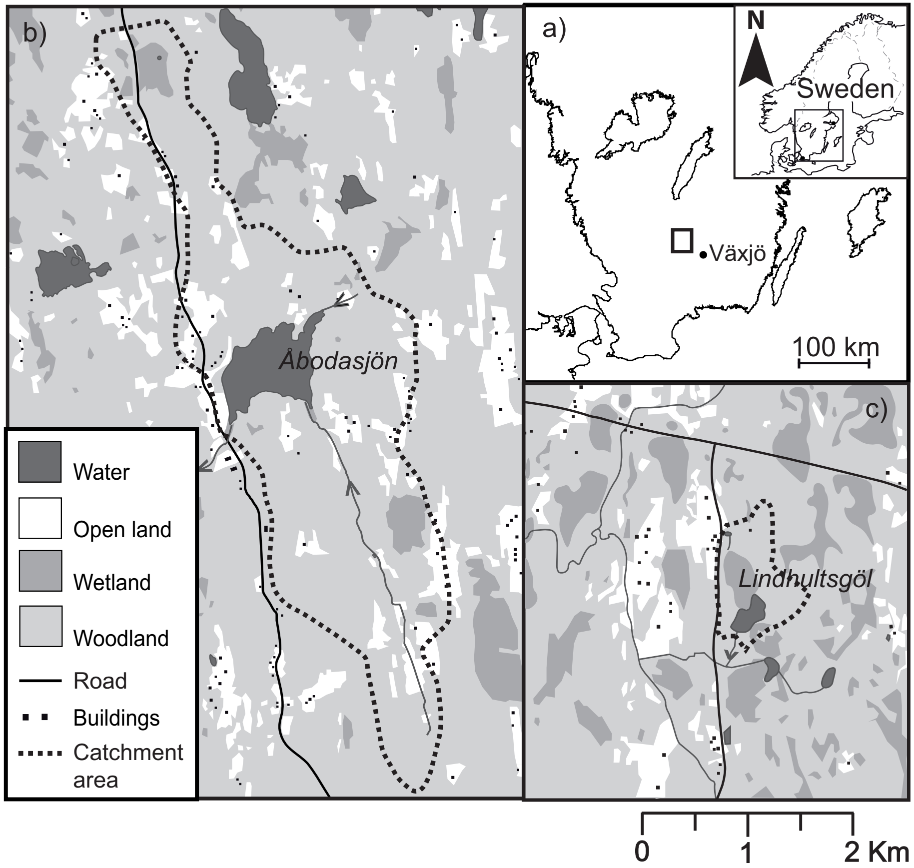

Fig. 1. Location of study sites. **(a)** Map of Scandinavia and southern Sweden. The study area is marked by a square, and the closest city is Växjö. **(b)** and **(c)** maps of the studied lakes and the present surrounding land cover.

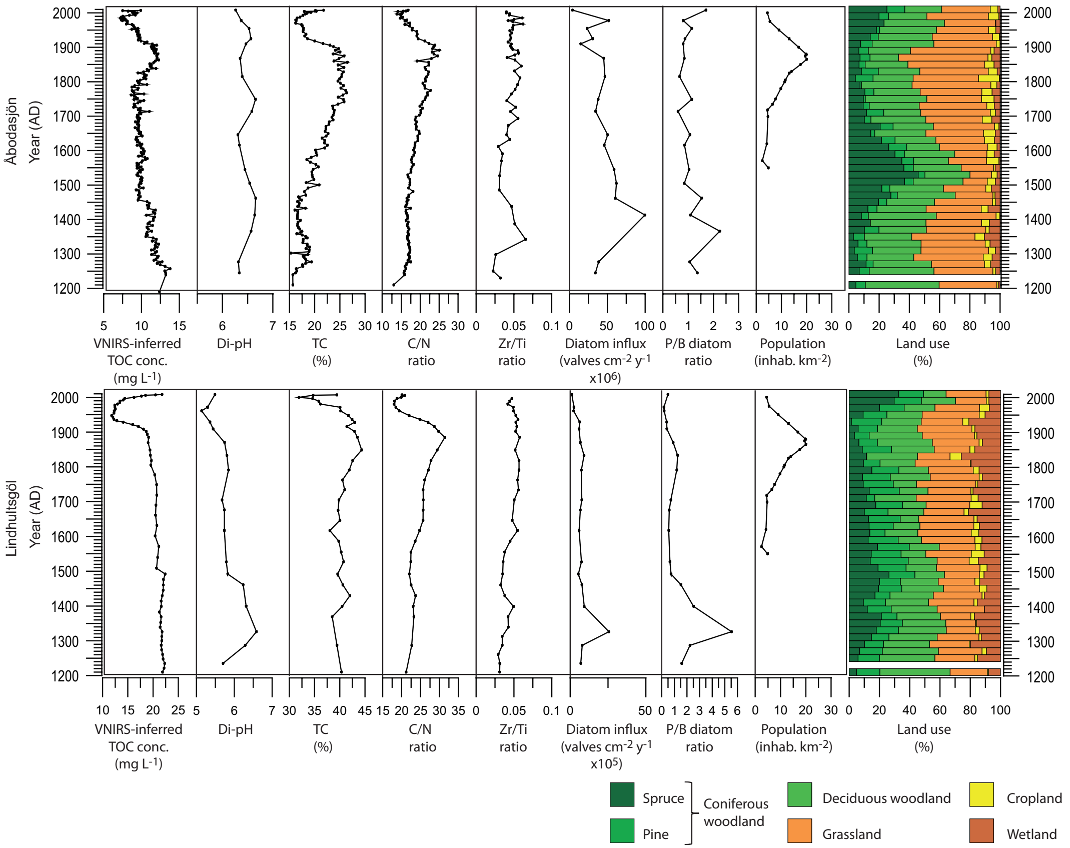

Fig. 2. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration, diatom-inferred pH (Di-pH), sediment total organic carbon (TC) content, atomic carbon: nitrogen (C/N) ratio, elemental zirconium : titanium (Zr/Ti) ratio, diatom valve influx, diatom planctonic : benthic (P/B) ratio, documented population density and pollen-based land use plotted against age from Åbodasjön (upper panel) and Lindhultsgöl (lower panel).

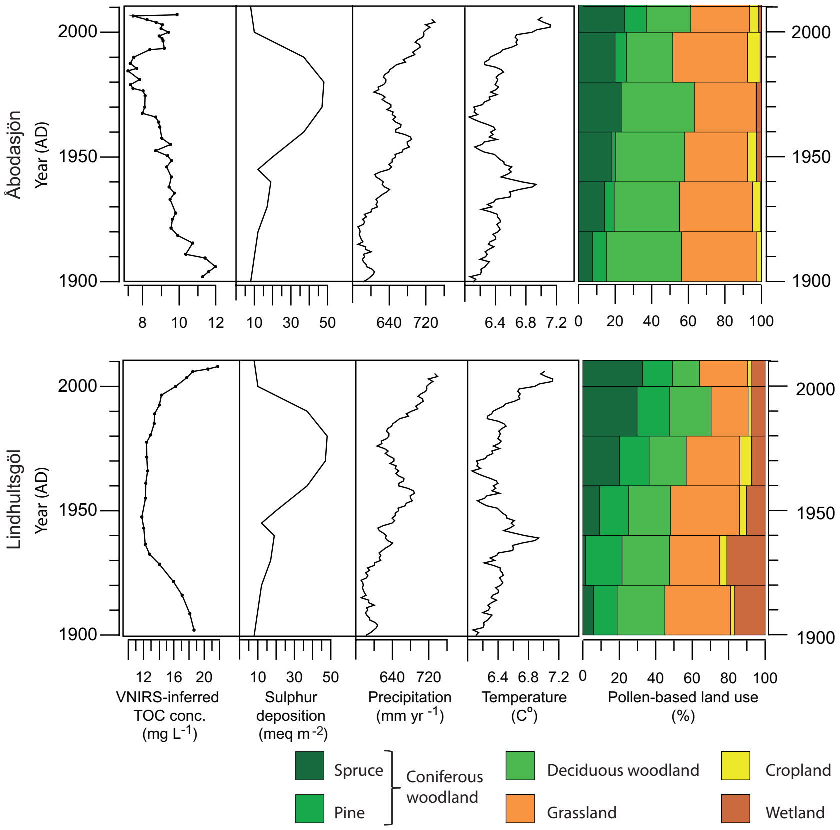

Fig. 3. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration and pollen-based land use since AD 1900 from Åbodasjön and Lindhultsgöl plotted together with sulphur deposition (modified from the Swedish Environmental Research Institute MAGIC-model) and climate data from Växjö (annual precipitation and temperature expressed as 15 yr running averages).

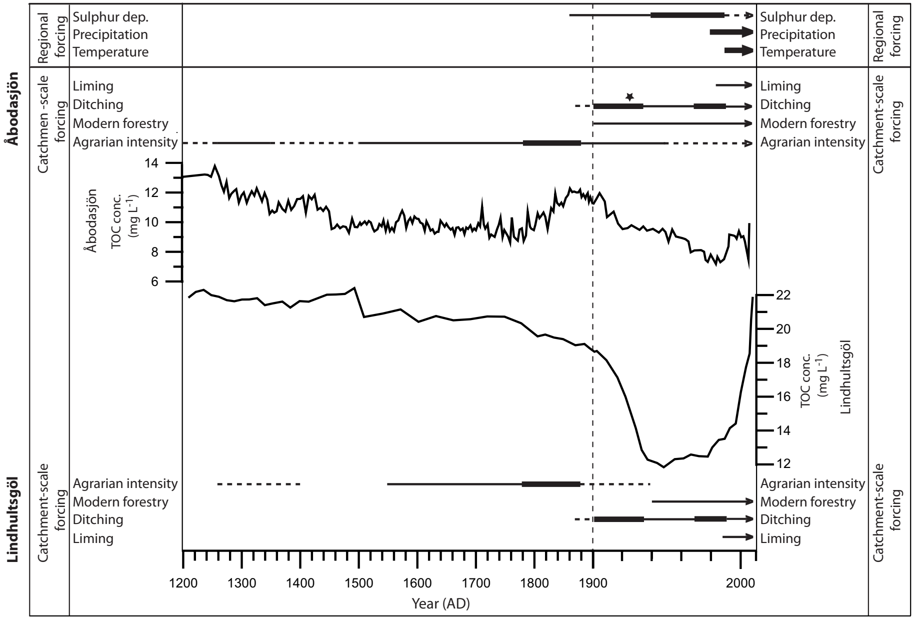

Fig. 4. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration from Åbodasjön (upper graph) and Lindhultsgöl (lower graph) in the perspective of possible regional and catchment-scale forcings of TOC changes. Regional forcings include sulphur deposition, precipitation and temperature (Fig. 3). Local forcings include site-specific liming history, regional trends in ditching (Hånell, 2009) and changes in land use inferred from pollen data (Fig. 2) and historical accounts (agrarian intensity and modern forestry). Horizontal lines represent periods of activity, thick lines represent periods of increase or high intensity, and dashed lines represents periods of decrease or low intensity. Arrows indicate ongoing processes. The star marks a major drainage effort undertaken at the inlet of Åbodasjön in AD 1922. The vertical dashed lines represents AD 1900. Note the different scale in AD 1900–2010.