Biogeosciences, 12, 307–322, 2015
www.biogeosciences.net/12/307/2015/
doi:10.5194/bg-12-307-2015

Historical TOC concentration minima during peak sulfur deposition in two Swedish lakes

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Received: 16 October 2013 – Published in Biogeosciences Discuss.: 19 December 2013
Revised: 20 November 2014 – Accepted: 25 November 2014 – Published: 19 January 2015

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, regional variations in sulfur, and nitrogen deposition and climate variations 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 inferred lake-water TOC concentrations were generally high prior to AD 1900, with additional variability coupled mainly to changes in forest cover and agricultural land-use intensity. The last century showed significant changes, and unusually low TOC concentrations were inferred at AD 1930–1990, followed by a recent increase, largely consistent with monitoring data. Variations in sulfur emissions, with an increase in the early 1900s to a peak around AD 1980 and a subsequent decrease, were identified as an important driver of these dynamics at both sites, while processes related to the introduction of modern forestry and recent increases in precipitation and temperature may have contributed, but the effects differed between the sites. The increase in lake-water TOC concentration from around AD 1980 may therefore reflect a recovery process. Given that the effects of sulfur deposition now subside and that the recovery of lake-water TOC concentrations has reached pre-industrial levels, other forcing mechanisms related to land management and climate change may 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 for 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 agreement on the mechanisms controlling DOC and colour variations in lake water during recent decades may partly reflect 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 sulfur emissions that have occurred during the last century. Correspondingly, long-term changes in vegetation, land use and climate have also 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 lake waters consists of DOC, usually defined as organic matter not retained by a filter of 0.45 µm in nominal pore size (Wetzel, 2001). The remaining fraction is particulate organic carbon (POC), which consists of larger organic compounds. 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 DOC pool in boreal lakes (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 concentration 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, which are 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 primary 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 concentration 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 years. One of them (Åbodasjön) is oligotrophic mesohumic with a mosaic landscape in its catchment area and with a long history of anthropogenic disturbance. The other lake (Lindhultsgöl) 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., 2013). We applied a combination of palaeolimnological methods to the sediment sequences, including reconstruction of lake-water TOC concentration based on VNIRS (Rosén, 2005), diatom analysis to determine water pH, and pollen analysis and the Landscape Reconstruction Algorithm approach for reconstruction of catchment land-cover change (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, sulfur and nitrogen deposition, and climate to long-term trends in lake-water TOC concentration since AD 1200. 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 timescales 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 north-west 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–

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. (µg 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

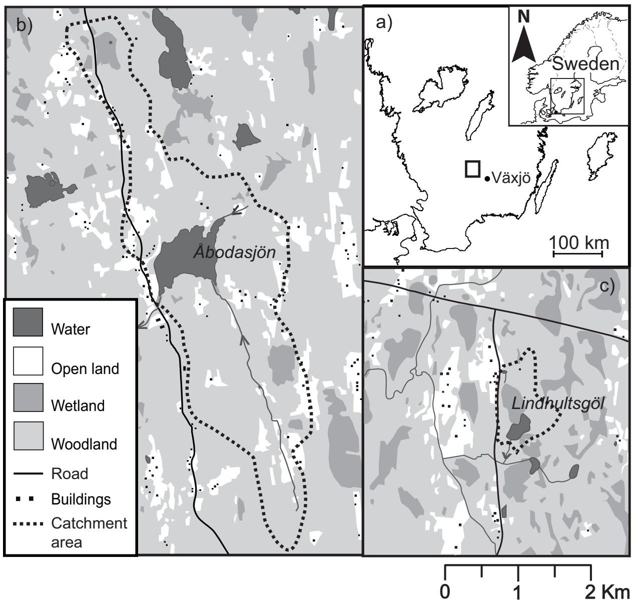

Figure 1. Location of study sites. **(a)** Map of Scandinavia and southern Sweden. The study area is marked by a square, and the closest city is Växjö. **(b)**, **(c)** Maps of the studied lakes and the present-day land cover in their surroundings.

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 et al., 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 industrialization.

Åbodasjön (Table 1, Fig. 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.

Lindhultsgöl (Table 1, Fig. 1) is an oligotrophic polyhumic lake with no visible inlet streams. 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.

3 Methods

3.1 Fieldwork, subsampling and dating

In early spring 2008 sequences of surface sediments were obtained from Åbodasjön and Lindhultsgöl at water depths of 8.6 and 5.2 m, respectively, 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 lead (Pb) pollution 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 to 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 the Zrax mounting medium (refractive index $= \sim 1.7+$). At least 400 diatom valves per sample were counted under a light microscope at 1000 × magnification, using phase-contrast optics and identification keys (Krammer and Lange-Bertalot, 1986, 1988, 1991a, 1991b; Lange-Bertalot and Krammer 1989; 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 decreased light penetration reduces benthic growth.

Changes in pH were 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 locally weighted averaging and inverse deshrinking (Juggins and Birks, 2012). The model performance of the transfer function applied to Åbodasjön and Lindhultsgöl was assessed by leave-one-out cross-validation, which showed an R^2 value of 0.8 and an RMSEP of 0.4 pH units.

3.4 Carbon and nitrogen elemental analyses

The carbon–nitrogen (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 ($TOC_{sediment}$) and total nitrogen (TN) contents by combustion using a Costech ECS 4010 elemental analyser. The samples were pre-treated with 10 % HCl at 90 °Cfor 5–7 min for removal of potential trace amounts of $CaCO_3$. Elemental C / N ratios were converted to atomic ratios by multiplication by 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 phosphorus (P), 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 LRA (Sugita, 2007a; b) based on pollen counts of dominant taxa in the sediment records from the two study sites and an additional lake (needed for the LRA calculation). A minimum of 1000 pollen grains of modelled arboreal and non-arboreal taxa were counted for contiguous 0.5 cm samples (1–10 samples) covering 20-year time spans.

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-year 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 covers of individual taxa are grouped into five different categories of land use according to Mazier et al. (2014): coniferous woodland, deciduous woodland, grassland, cropland and wetland. 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.

3.7 Multivariate analyses

To explore the impact of various potential driving forces on the lake environment as reflected in the sediment record, we carried out canonical ordinations. The palaeolimnological parameters (VNIRS-inferred TOC concentration, sediment TOC_{sed} and TN, C / N ratio, sediment P content (ppm), Zr / Ti ratio and diatom inferred pH) were used as response variables. As potential forcing variables we used the pollen-inferred land-use categories (coniferous woodland, deciduous woodland, grassland, cropland and wetland) and in addition the cover of individual tree species (spruce and pine), total woodland cover (coniferous and deciduous), and the sum of cropland and grassland.

For the entire period after AD 1200, 20-year time slices were used for the analysis, using land cover as forcing. A mean value for each sedimentary variable was calculated over each 20-year time slice. A few time slices lacked measurements of P content and Zr / Ti ratio and were therefore left out of the analysis. For Di-pH, the analytical resolution was lower than 20 years (see Fig. 2), so linear interpolations between the available estimates were used to calculate average values for each 20-year time slice. A separate analysis was carried out for the period after AD 1880 including data on atmospheric deposition of sulfur (S), ammonium (NH_4) and nitrogen oxides (NO_x), and monitoring records of temperature and precipitation as potential forcing factors in addition to land cover. Annual climate data are available for the region from AD 1860, and deposition data are available at 5-year intervals from AD 1880. The temporal resolution of this ordination analysis was determined by the resolution of the inferred VNIRS-TOC reconstruction (see Fig. 3). Land cover was considered constant for each 20-year interval. For the other sediment parameters and for atmospheric deposition, linear interpolation was used to derive an estimated value for the year corresponding to each VNIRS-TOC sample. For the climatic variables (annual mean temperature and total annual precipitation), the value measured in the sample year and the mean value for the 10 years up to and including the sample year were both included in the analysis.

Ordinations were carried out using CANOCO v4.51. For all analyses, preliminary detrended canonical correspondence analysis showed the response data set had a gradient length < 1 standard deviation units, implying that linear based ordination techniques such as redundance analysis (RDA) were most suitable for these data sets (ter Braak and Smilauer, 1998).

Land-cover percentages were square-root-transformed, while the limnological parameters (which are measured in different units) were centred and standardized. Time was used as a co-variable to remove co-varying effects between, for example, changes in land use and atmospheric deposition. Manual forward selection was used to explore the explanatory power of the different forcing variables, and Monte Carlo tests with 999 unrestricted permutations were run to check their statistical significance in order to select the best explanatory variables for further analysis. The selected variables were checked for collinearity by inspecting their variance inflation factors, which were in all cases < 10, which indicates that the selected parameters are not too closely correlated (Oksanen, 2011).

4 Results

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

The diatom-inferred pH varies between 6.2 and 6.7, with a sample-specific standard error between 0.32 and 0.45. (Fig. 2 and Supplement). Periods of slightly 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 increases to a peak around AD 1400, followed by a decrease to relatively stable values and a second decrease after AD 1850. The planktonic diatom taxa vary between 40 and 70 % of the diatom assemblage, and slightly elevated P / B ratios were recorded at AD 1250–1500 and in the top sample.

Sediment total organic carbon content (TOC_{sed}) and TN show slightly elevated values at AD 1250–1350, followed by a slight transient decrease and a gradual increase after AD 1450. In the 1800s TOC_{sed} content stabilizes at maximum values. The C / N ratio increases in AD 1200 to ca. 1300, followed by a slight decrease and a continuous increase from around AD 1450 to a sequence maximum at AD 1850–1900. There is a shift towards substantially lower TOC_{sed} and TN content, and C / N ratios at ca. AD 1850 (TN) and 1900 (TOC_{sed} and C / N). Thereafter, increasing trends in both TOC_{sed} and TN content is recorded from ca AD 1970 to the present, and C / N ratios after AD 1990.

P concentration decreases gradually from the beginning of the sequence interrupted by a shift to higher values at ca. AD 1440 and thereafter followed by continuously decreasing concentrations. The onset of the 1900s is characterized by an increase in P concentration peaking shortly after AD 1950. 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 varies between 33 and 80 % since AD 1200. The cover of grassland and cropland together is 40–50 % at AD 1240–1400, followed by a decrease to a minimum of 15 % at AD 1520–1540, when deciduous and coniferous woodland reaches a peak in cover. After around AD 1540 grassland and cropland cover increases and reaches maxima of ca. 60 and 12 %, respectively, between AD 1820 and 1900. During the 1900s coniferous woodland, dominated

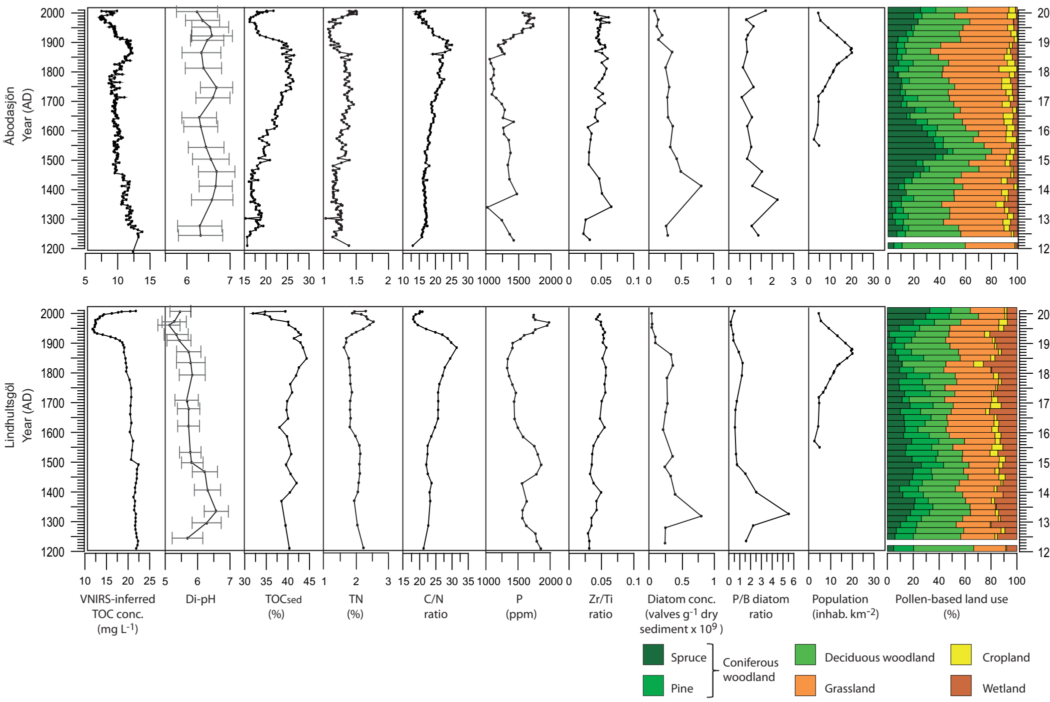

Figure 2. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration, diatom-inferred pH (Di-pH) (horizontal error bars represent ±1 SD), sediment total organic carbon ($\mathrm{TOC_{sed}}$) and total nitrogen (TN) content, atomic carbon : nitrogen (C / N) ratio, elemental phosphorus (P) content, elemental zirconium : titanium (Zr / Ti) ratio, diatom valve concentration, 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).

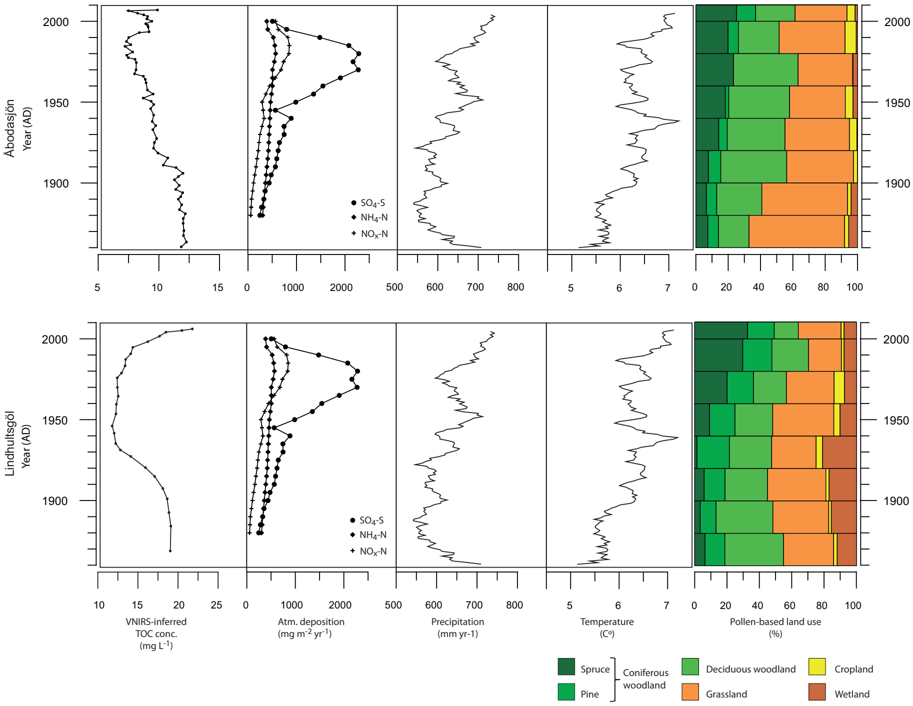

Figure 3. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration and pollen-based land use since AD 1900 from Åbodasjön and Lindhultsgöl plotted together with atmospheric sulfur (sulfate SO_4) and nitrogen (ammonium (NH_4) and nitrogen oxides (NOx)) deposition (from the Swedish Environmental Research Institute MAGIC model) and climate data from Växjö (annual precipitation and temperature expressed as 10-year running averages).

by spruce, increases from 10 to 30 %. Coniferous and deciduous woodland covers ca. 60 % of the lake catchment today.

RDA was used to describe the major gradients in the limnological data set and relate these patterns to the land-use variables during the last 800 years. Total woodland cover was identified as the most significant land-cover factor, explaining a statistically significant 13 %. Other land-cover variables that were significant when analysed on their own were spruce and cropland cover, each explaining 12 %; wetland and coniferous woodland, 10 %; and deciduous woodland, 6 %. When total woodland cover was included in the RDA analysis, deciduous woodland cover could still explain an additional 6 % of the variation, while no other land-cover parameters were statistically significant at the $P < 0.05$ level, as most of the variation they could explain was captured by the relationship to total woodland cover.

The ordination results are presented as a so-called triplot (Fig. 4a) showing the RDA scores for both the palaeolimnological response variables and the selected forcing variables, as well as the trajectory of down-core sample scores over time, along the first and second RDA axes. The figure indicates that the VNIRS-inferred TOC concentration along with sediment $\mathrm{TOC_{sed}}$, C / N and Zr / Ti are all negatively related with woodland cover, which is correlated with the first RDA axis ($r = 0.71$). Deciduous tree cover is negatively correlated with the second axis ($r = -0.55$), along with diatom-inferred pH. Both seem to be negatively correlated with sediment TN.

For the period after AD 1880, five significant drivers were retained on the basis of forward selection. $\mathrm{NH_4}$ deposition was identified as the main driver and explained 21 % of the variance, $\mathrm{NO_x}$ deposition (additional 12 %), 10-year mean annual precipitation (additional 3 %), sulfur deposition (additional 2 %) and 10-year mean annual temperature (additional 2 %). While some of the land-cover categories had significant effects if analysed individually, such as deciduous tree cover (18 %) and grassland cover (14 %), they were less important than the depositional and climatic parameters, and did not add significantly to the combined analysis. The RDA plot (Fig. 4b) indicates that Di-pH is negatively correlated with sediment $\mathrm{TOC_{sed}}$ and VNIRS-inferred TOC, which are both negatively related to $\mathrm{NH_4}$- and S deposition and positively related to precipitation.

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 varies between 5.0 and 6.8, with sample-specific standard errors between 0.31 and 0.47. 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 decreases 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 (pH > 7) diatom taxon *Aulacoseira ambigua* (< 60 %), and the low pH in the 1900s was affected by the dominant acidophilous (pH < 7) taxon *Frustulia rhomboides* (< 60 %). Inference models are always associated with uncertainties and diatoms may respond to other variables than pH (Juggins, 2013). Therefore caution is necessary when interpreting the reconstructed pH data. The diatom concentration is high in the 1300s, followed by stable values until around AD 1850, when concentrations decrease. The planktonic diatom taxa vary 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 $\mathrm{TOC_{sed}}$ content at ca. AD 1200–1700 and for C / N ratios at ca. AD 1200–1550, followed by increasing values, peaking in the late 1800s. The total nitrogen (TN) content showed slightly decreasing values until AD 1900. At ca. AD 1900 significant decreases in both $\mathrm{TOC_{sed}}$ content and C / N ratio to minima in the 1980s to 1990s together with a subsequent increase in TN content to a coherent maxima in the 1980s were recorded. This was followed by reversed trends and a coherent increase after ca. AD 1990–2000 towards the top.

P concentration decreases gradually from the beginning of the sequence interrupted by a shift to higher values in the 1400s and thereafter followed by continuous decreasing concentrations. The onset of AD 1900 is characterized by an increase in P concentration peaking shortly after AD 1950. The Zr / Ti ratio shows a peak around AD 1350, following a gradual increase from ca. AD 1250. After a subsequent decrease, the Zr / Ti ratio increases 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 varies between 44 and 70 % during the last 800 years. In contrast to Åbodasjön, wetlands cover more than 20 % during most of the period and decreases to less than 10 % after AD 1960. Grassland and cropland varies between 20 and 30 % at AD 1200–1580, followed by an increase to ca. 40 %. During the 1900s, coniferous woodland increases, and this land-use category covers ca. 50 % of the lake catchment today.

In the RDA analysis for the last 800 years, the forward selection for this site showed that spruce cover, the main explanatory variable, explains 20 % of the variance. After its inclusion in the RDA model, two other variables were found significant – cropland and wetland covers, explaining 7 % respectively 4 % of additional variance.

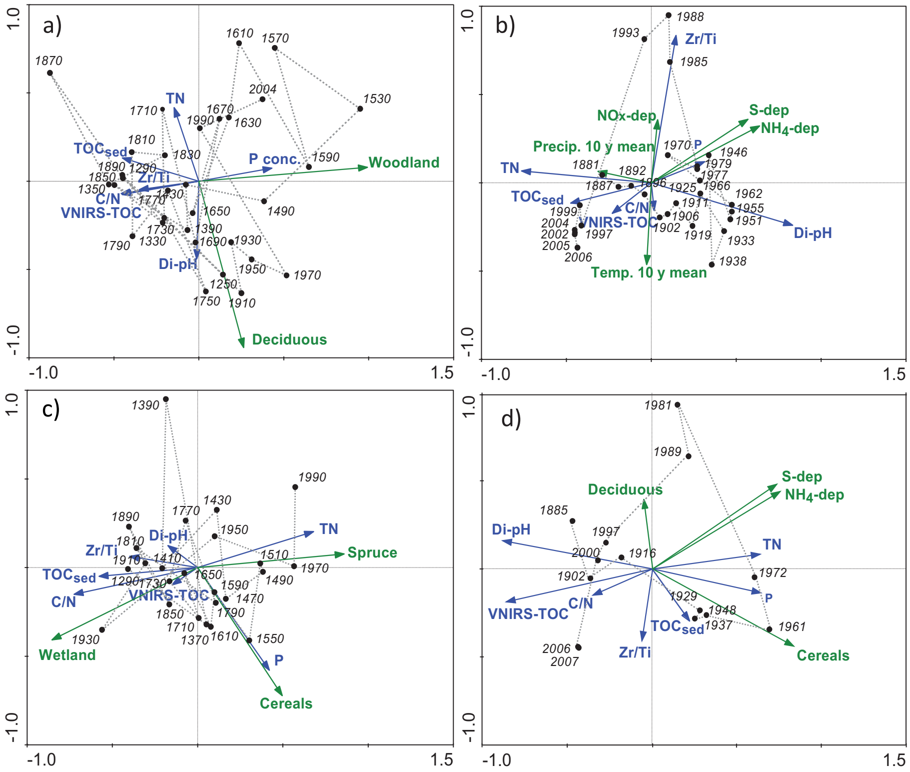

Figure 4. Scores for samples (black circles), palaeolimnological parameters (blue arrows) and driving forces (green arrows) on the first and second axes of the redundancy analyses for **(a)** Åbodasjön AD 1200–present, **(b)** Åbodasjön AD 1880–present, **(c)** Lindhultsgöl AD 1200–present and **(d)** Lindhultsgöl AD 1880–present. Sample ages represent the midpoint of each 20-year time slice.

A triplot showing the first and second RDA axes (Fig. 4c) indicates that TOC_{sed}, C / N and to a lesser extent VNIRS-inferred TOC concentration seem to be positively related to wetland cover, and negatively with spruce cover along the first axis. Sediment TN is positively correlated with spruce cover, while along the second axis sediment P content is correlated with cereal cover.

For the period after AD 1880, the RDA analysis indicates that cereal cover was the most important driver, alone explaining 28 % of the variation in the palaeolimnological data. The stepwise forward selection showed that three further variables could contribute significantly to the explanatory power of the model, i.e. S deposition (which could explain an additional 10 % of the variation if included together with cereal cover), NH_4 deposition (5 %) and deciduous tree cover (3 %). Ten-year mean precipitation has a small significant effect on its own, explaining 14 %, but is not significant once crop cover was included. No other climate parameter seems to have an effect at Lindhultsgöl, being overshadowed by the stronger effect of land use change at this site. The RDA plot (Fig. 4d) indicates that, like at Åbodasjön, VNIRS-inferred TOC concentration is negatively related to NH_4- and S deposition. But at this site, TOC and Di-pH seem to be positively related. The sediment TN and P content at this site are positively related to cereal cover and the atmospheric N deposition.

5 Discussion

5.1 Impacts of land-use changes prior to AD 1900

In Åbodasjön, the highest lake-water TOC concentration was inferred 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 (Fig. 2; 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, thereby reflecting cultural alkalinization (Renberg, 1990; Rosén et al., 2011). However, despite increased erosion, TOC concentration decreased in the lake water during the 1200s, which may be explained by decreased woodland cover, which lowered the terrestrial 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 from 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, the sediment record shows only a slight increase of open land, mainly grassland and cropland, around the lake. The persistently high TOC concentrations in the lake were probably related to the large proportion of wetlands within the catchment, as also indicated by the RDA results. High proportions of wetland are often associated with substantial supplies of DOC to nearby lakes (Rasmussen et al., 1989; Kortelainen, 1993; Xenopoulos et al., 2003; Mattsson et al., 2007). The lack of response in lake-water TOC concentration to catchment disturbance at Lindhultsgöl during the period of increased anthropogenic impact and potential 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 ca. 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 ca. AD 1450 there was a shift to lower lake-water TOC concentrations, accompanied by decreasing Zr / Ti ratio and diatom concentration. At Lindhultsgöl the regression led to decreases in catchment erosion, inferred pH and diatom concentration from ca. AD 1400 in response to increased cover of coniferous woodland.

From ca. 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 ca. AD 1350, coniferous woodland reached maximum cover of ca. 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.

A substantial increase in lake-water TOC concentration was inferred at Åbodasjön from ca. 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 for 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 ca. 60 %. The RDA plot (Fig. 4a) also reflects that both total and deciduous woodland cover reached minimum values around this time. These changes were accompanied by maximum C / N ratios and $\mathrm{TOC_{sed}}$ values, reflecting an elevated input of terrestrial organic matter to the lake. From ca. 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 DOC 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 in the inferred TOC concentration at Åbodasjön.

At Lindhultsgöl broadly similar trends in C / N ratio and $\mathrm{TOC_{sed}}$ from the late 1700s to ca. 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 in 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 in TOC concentrations were recorded in both of the study lakes (Figs. 2, 3). At Åbodasjön the decrease was slightly more gradual, reaching minimum values in the 1980s, while the inferred values at 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 sulfur deposition regionally in southern Sweden (Fig. 3), and

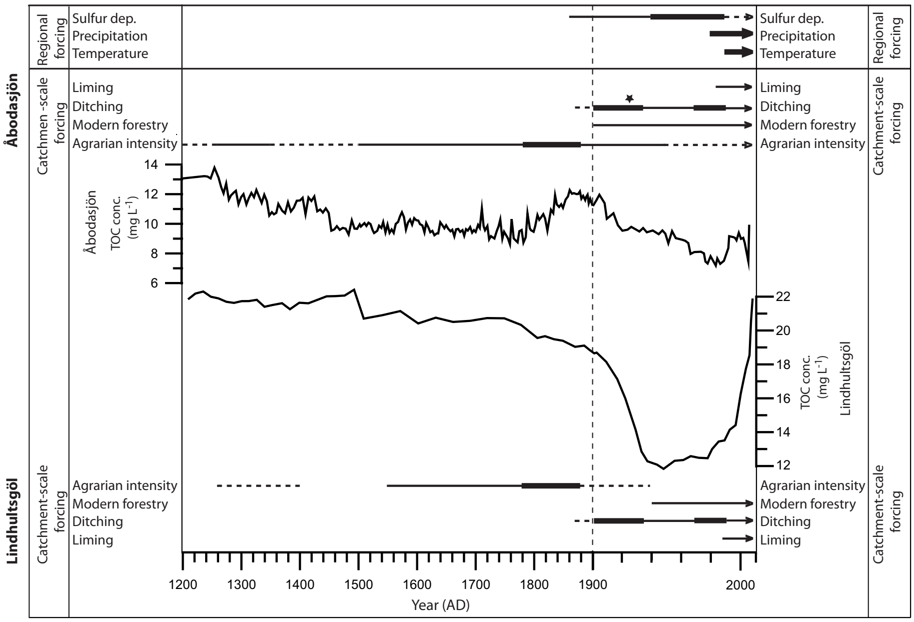

Figure 5. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration from Åbodasjön (upper graph) and Lindhultsgöl (lower graph) in the perspective of possible regional and catchment-scale forcings of TOC changes. Regional forcings include sulfur deposition, precipitation and temperature (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 represent 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 represent AD 1900. Note the different scale for the period AD 1900–2010.

the RDA data also indicate that sulfur deposition is among the significant drivers of limnological changes in both lakes since AD 1880. Sulphur deposition started to increase at the onset of industrialization 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, sulfur deposition increased significantly in the 1940s, peaking at AD 1980–1995 (Schöpp et al., 2003), followed during recent decades by progressively decreasing deposition and widespread recovery from acidification through decreasing sulfate 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 in our two study lakes (Fig. 3) as well as with a study of TOC trends in Swedish rivers (Erlandsson et al., 2010). The deposition of nitrogen oxides, which also contribute to acidification, also showed a dramatic increase during the 1900s, with deposition peaking slightly later than for sulfur deposition, around AD 1990 (Fig. 3). In addition to contributing to acidification, deposition of nitrogen, both in the form of nitrogen oxides and ammonia, may contribute to eutrophication, and therefore can have an impact on limnic ecosystems. It has also been suggested that the response of soil microbial activity to nitrogen deposition may affect the export of humic matter to freshwaters (Findlay, 2005). Our analysis indicates that nitrogen deposition was among the most significant drivers of change in the palaeolimnological record over the last century together with sulfur deposition.

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 sulfur deposition usually results in reduced transport of soil organic matter. In our lakes, declining VNIRS-inferred TOC concentrations were accompanied by decreasing C / N ratios, which suggests a reduction of terrestrial organic matter deposition 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 VNIRS-inferred TOC concentration was reached already around AD 1940, a few decades before sulfur 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 negative correlation between sulfur 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 industrialization 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 cover in the 1900s (Fig. 3). This land-use change is most pronounced at Lindhultsgöl, where grassland, cropland and wetland cover are reduced at the expense of woodland, and the RDA indicates a significant effect of especially the cereal cover reduction at this site. A significant reduction of the supply of terrestrial organic matter, as indicated by decreasing C / N ratios at both lakes, may be partly explained by the increase in sulfur deposition, which suppressed leaching of soil organic matter. However, 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 others 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 ca. AD 1920–1960 (Fig. 3) was possibly reinforced by increased supply of soil-derived DOC through enhanced export 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 in TOC concentration in the lake. Previous studies have attributed variations in the release of DOC to surface waters to changing forestry practices, 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 % of the modelled land-use area 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 probably result in only temporary increases in the supply of DOC, affecting at least the following growth season (Laudon et al., 2009) and may therefore be difficult to distinguish in palaeolimnological records.

The increase in VNIRS-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 sulfur 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 sulfur 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 sulfur deposition by accelerated leaching of DOC to the lakes (cf. Hindar et al., 1996).

In addition to changes in sulfur deposition and land management practices, climate may affect DOC concentration of lake waters 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 waters 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, as well as 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 ca. AD 1980 and an increase in mean annual temperature from ca. AD 1990 (Fig. 3). Hence, climate change may have contributed to the observed and reconstructed increases in lake-water TOC concentration over recent decades, and the RDA data indicate that, at least at Åbodasjön, both precipitation and temperature have had an impact on the lake over the last century, while these effects seem to be less important at Lindhultsgöl. A possible explanation may be the larger catchment of Åbodasjön, making it more sensitive to changes in run-off, erosion and transport of terrestrial organic matter. The large proportion of wetland around Lindhultsgöl may also have a dampening effect on increased precipitation. At Lindhultsgöl, changes in land use have played a more important role at the centennial timescale. Changes in sulfur deposition during the last century have been a main driver for limnological change at both sites, despite their different land use and catchment characteristics (Figs. 3, 4), which supports the interpretation that this is a key factor behind the regional changes observed in lake-water TOC concentrations. 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 water colour (usually measured as absorbance at ca. 420–436 nm or using the platinum scale) and DOC concentration in lake waters. However, colour is strongly influenced by the composition of DOC, and a recent study has demonstrated that declining acidification in southern Sweden has led to increased leaching from soils of mobile, hydrophobic and aromatic DOC that contains 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 VNIRS-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 (County Administrative Board of Kronoberg, unpublished data), consistent with increases in water colour observed in several other lakes in the study region. This is supported by high abundances 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 water colour, as indicated by unchanged diatom planktonic : benthic (P / B) ratios. Benthic and planktonic diatom communities are likely to respond to changes in the input of terrestrial organic matter through associated effects on the transparency of the water column, as the benthic community is primarily limited by light in nutrient-poor lakes (Rosén et al., 2009; Karlsson et al., 2009). At this site, the pronounced increase in agricultural intensity in the late 1800s probably resulted in enhanced export of DOC compounds with relatively low molecular weights, which are typically associated with agriculture (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 coloured 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 diatom response to increased nutrient transport to the lake was most likely associated with early land use and not with any major increase in water colour caused by increased input of terrestrial organic carbon.

At Lindhultsgöl, minimum P / B ratios were recorded during the period of maximum sulfur emissions at AD 1950–1990, which indicates a decrease in water colour associated with the corresponding minima in inferred lake-water TOC concentration and pH.

Based on our results we can conclude that the increases in TOC concentration and water colour in our study lakes during the past three decades have been driven mainly by declining atmospheric sulfur deposition. This suggests a recovery from the phase of maximum sulfur emissions, which resulted in exceptionally low TOC concentrations in the lakes at ca. AD 1930–90. The RDA data obtained from the palaeolimnological records over the period since AD 1880 also indicate a recovery. At both sites, the temporal development of the RDA scores during this period (Fig. 4b and d) show that the youngest samples fall near the oldest, indicating a return to pre-industrial conditions, following a time of highly anomalous conditions. At Åbodasjön, there was first a period of low inferred lake-water TOC concentration and high pH in the 1930s–1960s, followed by decreasing pH but high Zr / Ti ratios (perhaps indicating enhanced erosion) in the 1980s and early 1990s, and then increasing lake-water TOC concentration and low Zr / Ti ratios towards the present. At Lindhultsgöl, the temporal development of the RDA scores also illustrates a partial recovery with similar scores, especially on the first axis, for young and old samples, while the period 1930–1980 was characterized by high first axis scores, associated with sedimentary indicators of low pH and VNIRS-inferred TOC concentration, while the driving forces were characterized by high sulfur and nitrogen deposition values and relatively high cereal cover in the catchment.

Our long-term records demonstrate that the TOC concentrations of the study lakes were strongly influenced by changes in agricultural practices, general land-use pressure, and associated variations in forest cover during the last 800 years (Fig. 5). The historical differences in the extent of agricultural activity at the sites establish that site-specific catchment characteristics and land-use dynamics are of great importance for lake-water DOC variations. 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 lake-water TOC concentrations has now reached levels that are comparable to the situation before the onset of 20th century acidification, which may lead to a levelling-off of the increasing trend. Given the reduction of atmospheric sulfur emissions during recent decades, it is likely that previously suppressed or masked effects of changes in land management and climate during the last century will become progressively more important drivers of lake-water DOC concentrations in the future.

The Supplement related to this article is available online at doi:10.5194/bg-12-307-2015-supplement.

Acknowledgements. This work was funded by the Swedish Research Council Formas (grant to W. Granéli). 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 categorization. We are very grateful to all members of the NordForsk network LANDCLIM (coordinated by M. J. Gaillard, Linnaeus University, Sweden) for useful and inspiring discussions during the numerous workshops (2009–2011). We acknowledge the Swedish Meteorological Institute (SMHI) for precipitation and temperature data, and the Swedish Environmental Research Institute and MAGIC for sulfur and nitrogen deposition data. Constructive comments by the reviewers improved the final presentation.

Edited by: C. P. Slomp

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