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# LUNDQUA Thesis 71

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## A palaeolimnological study of the anthropogenic impact on dissolved organic carbon in South Swedish lakes

*Petra Bragée*

### **Avhandling**

Att med tillstånd från Naturvetenskapliga Fakulteten vid Lunds Universitet för avläggande av filosofie doktorsexamen, offentligen försvaras i Geocentrum IIs föreläsningssal Pangea, Sölvegatan 12, fredagen den 1 november 2013 kl. 13.15.

Fakultetens opponent: Prof. Dr. Richard Bindler, Institutionen för ekologi, miljö och geovetenskap, Umeå Universitet, Umeå, Sverige

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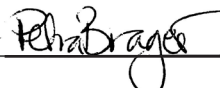
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<b>Abstract</b> <p>During the past three decades, increases have been observed in dissolved organic carbon (DOC) concentrations and colour in the surface waters of lakes and rivers in parts of Europe and North America, raising concern about the effects on the quality of aquatic environments with consequences for biodiversity, resource availability and recreational use. Various hypotheses have been put forward to explain the recent increases in DOC concentration and numerous studies have been published linking them to declining anthropogenic atmospherically deposited sulphur. Others have argued that increases in DOC content are a consequence of changes in climate, land use or land management practices.</p> <p>The work presented in this thesis concentrates on identifying the major forcing mechanisms behind observed increases in DOC concentration in the upland area of southern Sweden during recent decades, by comparing variations in the total organic carbon (TOC) concentration in lake water inferred from lake sediments, in response to changes in land use, sulphur deposition and climate during the past eight centuries. Two small lakes with different catchment properties were selected for the study; one dominated by woodland with abundant peat deposits, and another located nearby with patches of agricultural land in an otherwise mainly forested terrain. A number of palaeolimnological methods were applied to the sediment sequences; decadal-scale variations in TOC concentration in the lakes were reconstructed based on visible-near infrared spectroscopy (VNIRS) of sediment successions, high-resolution (20-y) pollen-based reconstructions of local land use were quantified using the Landscape Reconstruction Algorithm (LRA) and the model Local Vegetation Estimates (LOVE), geochemical records provided further information on environmental changes in the lakes and their catchment areas, and changes in pH in the lakes were inferred from diatom analysis. Comparisons were made with population density data and climate records.</p> <p>The results obtained with the LRA and LOVE models revealed a dynamic land-use pattern, with agricultural expansion from AD 1500 to the end of the 1800s, when population growth and the related increase in the exploitation of the surrounding land had a major impact on catchment erosion and input of terrestrial inorganic and organic matter to the lakes. Evidence also exists of a period of agricultural expansion around AD 1200-1300, followed by partial abandonment of the landscape, which can probably be attributed to the Black Death pandemic. A transition from traditional to modern land use following the industrial revolution took place during the past century, and a concurrent shift in most of the proxy records at around AD 1900 suggests a marked change in external forcing mechanisms common to both lakes, related to a major decrease in population density and the introduction of modern land use. The results revealed generally high TOC concentrations in the lakes prior to AD 1900, with second-order variations associated mainly with changes in the intensity of agricultural land use. The TOC concentrations in the lakes started to decrease around AD 1900, and unusually low TOC concentrations were recorded in the period AD 1930-1990, followed by a recent increase. The variation in sulphur emissions, with an increase in the early 1900s to a peak around AD 1980 followed by a significant decrease, was probably the main driver of lake-water TOC dynamics during the past century. This demonstrates that declining atmospheric sulphur deposition is the most probable driver of the increase in TOC concentration during the past three decades and that these lakes may be recovering to their naturally high-TOC pre-depositional states. The results also demonstrate regional versus local forcing of environmental change and indicate broadly similar regional sensitivities to anthropogenic impact, although responses were site-specific due to the different properties of the catchment areas. Given the reduction in atmospheric sulphur emission during recent decades, it is likely that previously suppressed or masked effects of changes in land use, land management and climate during the past century will become progressively more important drivers of TOC concentrations in lake water in the future. Long-term records of environmental history on decadal to millennial time scales enabled the assessment of ecosystem variability and responses to past anthropogenic disturbance, and may be a useful tool for the development of future environmental management strategies.</p>		
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# A palaeolimnological study of the anthropogenic impact on dissolved organic carbon in South Swedish lakes

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This thesis is based on the four papers listed below, which have been appended to the thesis. Paper I is in press in the journal indicated. Paper II, Paper III and Paper IV have been submitted to the indicated journals for consideration.

**Paper I:** Bragée P, Choudhary P, Routh J, Boyle JF, Hammarlund D (2013) Lake ecosystem responses to catchment disturbance and airborne pollution: an 800-year perspective in southern Sweden. *Journal of Paleolimnology* (in press)

**Paper II:** Mazier F, Broström A, Bragée P, Fredh D, Stenberg L, Thiere G, Sugita S, Hammarlund D. Two hundred years of land-use change in the South Swedish Uplands: comparison of historical map-based estimates with pollen-based reconstruction using the Landscape Reconstruction Algorithm. *Review of Palaeobotany and Palynology* (in review)

**Paper III:** Bragée P, Mazier F, Rosén P, Fredh D, Broström A, Granéli W, Hammarlund D. Forcing mechanisms behind variations in total organic carbon (TOC) in lake water during the past eight centuries – Palaeolimnological evidence from southern Sweden. Submitted to *Biogeosciences*

**Paper IV:** Fredh D, Mazier F, Bragée P, Lagerås P, Rundgren M, Hammarlund D, Broström A. The effect of local land-use change on floristic diversity around two lakes in southern Sweden, AD 1000-2008. Submitted to *The Holocene*



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## 1. Introduction

### 1.1 Background and objectives

During the past thirty years, increases in dissolved organic carbon (DOC) concentrations and the colour of lakes and running water have been observed in many areas, including Scandinavia (Hongve et al. 2004; Skjelkvåle et al. 2005; Vuorenmaa et al. 2006; Erlandsson et al. 2008; 2010; Sarkkola et al. 2009; Arvola et al. 2010), the Baltic states (Pärn and Mander 2012), the UK (Evans et al. 2005; Worrall and Burt 2007; Chapman et al. 2010), Central Europe (Hejzlar et al. 2003; Oulehle and Hruška 2009) and parts of North America (Stoddard et al. 2003; Findlay 2005; SanClements et al. 2012). This has led to concern regarding the effects on the function of the aquatic environments with consequences for biodiversity, resource availability and recreational use. DOC is operationally defined as the fraction of organic matter that passes through a filter with a nominal pore size of 0.45 µm (Wetzel 2001). Due to the dark colour of many DOC compounds, DOC affects the light regime, thermal structure and mixing depth of lakes, affecting the aquatic primary and secondary production (Cole et al. 2007; Karlsson et al. 2009; von Einem and Granéli, 2010). DOC can also cause acidification of surface waters (Kortelainen 1995) and influence the transport of trace metals and organic contaminants (Lawlor and Tipping 2003). Furthermore, the export of DOC from the terrestrial and aquatic ecosystems to the coasts constitutes an important part of global carbon cycling through local landscapes and continents (Tranvik et al. 2009), and contributes to CO<sub>2</sub> emissions to the atmosphere (Cole et al. 2007; Weyhenmeyer et al. 2012). Concern has also been raised about drinking water quality, as increasing DOC concentrations can enhance bacterial growth and the transport of contaminants and toxic compounds with potential carcinogenic and mutagenic properties (Ledesma et al. 2012). DOC can also affect recreational activities, such as fishing and swimming. There is general concern that an increase in DOC constitutes a hazard to the environment, and methods of controlling or preventing this trend must be found.

In Sweden, the observed increase in DOC content and thus colour in lakes, rivers and coastal waters, also referred to as “brownification” (Granéli 2012), has recently been recognized as an environmental problem and a future challenge (Swedish Agency for Marine and Water Management).

Various hypotheses have been put forward to explain the recent increases in DOC concentration (see Evans et al. 2005; Porcal et al. 2009), and numerous studies have been published linking them to declining anthropogenic atmospherically deposited sulphur (SO<sub>4</sub><sup>2-</sup>) (Evans et al. 2005; 2006; 2012; Vuorenmaa et al. 2006; Monteith et al. 2007; Erlandsson et al. 2008; Oulehle and Hruška 2009; Arvola et al. 2010; Clark et al. 2010; Ekström et al. 2011; SanClements et al. 2012). Others have argued that increases in DOC content are a consequence of changes in climate (Freeman et al. 2001; Hudson et al. 2003; Hejzlar et al. 2003; Hongve et al. 2004; Worrall and Burt 2007; Eimers et al. 2008; Lepistö et al. 2008; Sarkkola et al. 2009). In other studies, elevated DOC content has been linked to land use and land management practices, such as nitrogen deposition and fertilization (Findlay 2005; Correll et al. 2001), heather burning (Yallop et al. 2010) and drainage (Armstrong et al. 2010). However, there is no overall scientific consensus on the mechanisms controlling the observed variations in DOC and colour in lakes over recent decades. This suggests that individual lakes respond differently to the suggested forcing mechanisms, and that brownification can not be ascribed to a single forcing mechanism.

Previous studies have been based on experiments or data covering a few decades, and provide only a snapshot of the short-term and present-day conditions. There is, therefore, a lack of information on the natural variability and temporal evolution of DOC on the centennial and millennial scales, i.e. beyond monitoring data. Many of the suggested drivers of brownification are caused by human activities during the past century, and it is necessary to adopt a long-term perspective to distinguish between anthropogenic and natural changes. Palaeolimnology and lake sediment analysis can provide unique information on the environmental history of lacustrine ecosystems and their terrestrial



surroundings, providing evidence of the nature and timing of environmental change. This enables the causes of change to be identified in many cases. Such information is invaluable for projections of future ecological trends and the development of strategies for environmental management (Renberg et al. 2009). Palaeolimnological studies have successfully helped identify the effects of anthropogenic impacts on the lacustrine ecosystem, for example, eutrophication (Fritz 1989; Bradshaw et al. 2005), pollution (Renberg et al. 2000; Bindler et al. 2008) and acidification (Renberg 1990a).

This project was initiated to explore possible forcing mechanisms behind the observed brownification and increase in DOC concentration in lakes by comparing the impact of changes in land use, sulphur deposition and climate on long-term (centennial to millennial) variations in the total organic carbon (TOC) concentration in lake water. The TOC content of Scandinavian surface waters in lakes is dominated by DOC (>95%) (Mattsson et al. 2005), the remaining fraction being particulate organic carbon (POC). A number of palaeolimnological methods were applied to sediment sequences, including reconstruction of TOC concentration based on visible-near-infrared spectroscopy (VNIRS), diatom analysis to determine water pH, and pollen analysis to determine catchment land cover using the Landscape Reconstruction Algorithm (LRA), and geochemistry. Two lakes, Åbodasjön and Lindhultsgöl, in southern Sweden were chosen for this study because of their observed increase in TOC content in recently deposited sediments, in spite of their different catchment properties. Comparison of companion sediment records in different settings will allow the identification of regional and local forcing mechanisms of TOC variations. A third lake, Fiolen, was selected to reconstruct the regional vegetation, which was used in the pollen-based LRA to quantify local vegetation and land use, in a joint project investigating the impact of past changes in land use on floristic diversity.

The aims of this study were:

- to explore possible forcing mechanisms behind the observed present-day increase in DOC content in two southern Sweden lakes by comparing

changes in land use, acid deposition and climate with long-term reconstructions of TOC concentration inferred from lake sediments (Paper III),

- to quantify long-term changes in land use on a local scale using the LRA based on fossil pollen records and comparison with historical maps to assess the catchment scale forcing on TOC variations (Papers II and IV),

- to assess how the aquatic ecosystems of two lakes with contrasting catchment characteristics have been affected by anthropogenic activities and climate change during the past 800 years (Papers I and III), and

- to introduce the potential future change in lake-water TOC concentration as an important variable in land management and policy decisions affecting drinking water (Paper III).

## *1.2 DOC content and colour of lake water*

The DOC content in boreal forested catchment areas typically originates from organic breakdown of plant material and the leaching of decayed dead organisms from terrestrial soils. A minor fraction of the DOC pool arises from aquatic sources such as leachate from dead organisms, phytoplankton and aquatic macrophytes (Bade et al. 2007). DOC can be divided into a humic fraction, consisting of organic compounds of high molecular weight and refractory organic matter, generally characterized as being yellow to black in colour, and a colourless non-humic fraction, consisting, for example, of lipids, carbohydrates and amino acids. Humic substances are ubiquitous in water, soil and sediments, and usually comprise 50-75% of the DOC in water, but may exceed 95% in very coloured lakes (McDonald et al. 2004).

As humic substances constitute a major part of the DOC, DOC concentrations in lakes are usually strongly correlated to water colour (Pace and Cole 2002; von Einem and Granéli 2010). However, some studies have reported clear discrepancies between DOC concentrations and the colour of 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. Moreover,

Table 1. Classification of colour, absorbance and total organic carbon (TOC) in lakes and running water (Swedish Environmental Protection Agency)

	Colour (mg Pt L <sup>-1</sup> )	Absorbance (filtered)	TOC (mg L <sup>-1</sup> )
Not or slightly coloured	< 10	< 0.02	< 4
Slightly coloured	10-25	0.02-0.05	4-8
Moderately coloured	25-60	0.05-0.12	8-12
Significantly coloured	60-100	0.12-0.20	12-16
Highly coloured	> 100	> 0.20	> 16

iron (Fe) and manganese (Mn) also have a strong influence on water colour (Maloney et al. 2005). Elevated iron concentrations have been observed during the past few decades in the UK (Neal et al. 2008), as well as in Sweden (Huser et al. 2011; Kritzberg and Ekström 2012). Kritzberg and Ekström et al. (2012) found a strong correlation between water colour and iron concentration, indicating that brownification of some waters in Sweden was more likely to be related to increases in iron than organic carbon.

The amount of DOC leaching from terrestrial soils into lakes is driven by factors affecting the production, solubility and transport of DOC (see Clark et al. 2010). Biological processes involved in organic production and decomposition are controlled by factors affecting plant growth and soil organisms (i.e. climate and nutrient availability). Chemical processes regulate the solubility of organic carbon through pH and the ionic strength of the soil solution, while precipitation and hydrology control the export of DOC to surface waters via runoff and water flow in soils. Hence, the composition and quantity of DOC may differ between sites depending on climate and catchment properties such as vegetation, hydrology and soil properties, which may be altered by natural or human-induced changes in conditions. The DOC concentration is further regulated by gradual degradation by mineralization, photooxidation and sedimentation within the lake (Tranvik et al. 2009). In boreal lakes, this degradation results in a loss of carbon through sedimentation and CO<sub>2</sub> emission to the atmosphere, which have similar magnitudes (von Wachenfeldt and Tranvik 2008).

Water colour is usually determined by measurements of absorbance at wavelengths around 400 nm, or by using a platinum salt solution as

reference, giving the colour as mg Pt L<sup>-1</sup>. Water colour can also be assessed by DOC and TOC concentrations (Table 1). DOC concentrations in lake water are dynamic and show seasonal and long-term variations over several years or decades (see Clark et al. 2010). Generally, DOC concentrations in freshwater vary from 1 to 30 mg L<sup>-1</sup> and are highest in lakes from forested and peaty catchment areas with soils associated with high organic matter content, i.e., nutrient-poor systems, where organic production is greater than decomposition (Aitkenhead and McDowell 2000). The TOC content of Scandinavian lakes ranges from very low (< 1 mg L<sup>-1</sup>) in the mountainous regions, to above 20 mg L<sup>-1</sup> in Finland and the south-east of Sweden.

### 1.3 Long-term DOC trends in lake water

Recent changes in DOC concentrations have raised questions about the natural variability and temporal evolution of DOC on centennial and millennial scales. According to the European Union Water Framework Directive (EU WFD) (European Union 2000) all member countries are obliged to achieve “good” water status for relevant water bodies by AD 2015, defined as “conditions deviating only slightly from undisturbed conditions with low levels of anthropogenic disturbance”. To achieve this, it is important to understand the reference conditions for DOC in lake water, i.e., the “background conditions with no, or minimal anthropogenic stress” (Bennion et al. 2004). The date for reference conditions has generally been set to around AD 1850 (Bennion et al. 2004; Bjerring et al. 2008; Erlandsson et al. 2011), immediately before the onset of industrialisation. However, this date has been debated since different countries, regions and individual catchment areas often have different

histories regarding human influence (Bennion et al. 2011). It has been shown in palaeolimnological studies that Swedish lakes have been significantly affected by anthropogenic activities in pre-industrial times, about 4000 years ago, through settlements, agriculture and pollution (Renberg et al. 2000; Berglund et al. 2002). It may sometimes be difficult to differentiate between natural and anthropogenic changes to the lake ecosystem, as several processes could be due to natural or anthropogenic forcing. For example, sediment evidence of eutrophication may be identical to anthropogenic nutrient pollution (Jeppesen et al. 2010).

Available long-term DOC records are based on either VNIRS-inferred TOC reconstructions (Rosén 2005) or diatom-based calibration models providing quantitative measures of the DOC content in lake water (Kingston et al. 1990). The VNIRS approach has recently gained increasing attention as a proxy for ambient variations in DOC concentrations in lake water. Compared to the diatom-based approach, VNIRS models generally show better statistical performance, as diatoms respond to several environmental variables and may be influenced by a stronger pH signal.

A long-term study on four lakes in southern Sweden, covering about 300 years, revealed considerable decreases in VNIRS-inferred TOC concentrations in lake water over the past two centuries (Cunningham et al., 2010), in contrast to the reported present-day high DOC contents. On an even longer timescale (12,500 BP), Rosén et al. (2011) reported predominantly higher VNIRS-inferred TOC during most of the Holocene in a south Swedish lake and decreasing TOC concentrations during the past millennium. This is similar to findings in other long-term studies in boreal forest and subarctic lakes in northern Sweden (Rosén, 2005; Rosén and Hammarlund, 2007; Reuss et al., 2010). Rosén et al. (2011) suggested that the decrease in TOC content was caused by reduced transport of organic carbon from the catchment area due to early anthropogenic activities including agriculture, and man-made fires for clearance, resulting in more open land with lower biomass production. Limited soil carbon pools were also used to explain initial low TOC concentrations during early soil and vegetation development

following deglaciation. Variations in long-term TOC content recorded in more pristine areas of northern Sweden with little or no anthropogenic impact have been explained by changes in climate, especially humidity, mire development and fire frequency (Rosén and Hammarlund 2007). In a study carried out in North America, decreasing diatom-inferred DOC concentrations were observed concurrently with decreasing pH in some recently acidified lakes (Kingston et al. 1990).

These studies suggest that the “reference level” of DOC in lake water was generally higher than today, and that the observed decrease over the past millennium and centuries in southern Sweden may be due to anthropogenic activities. This clearly illustrates the importance of adopting a long-term perspective in our attempts to understand the historical development of DOC levels, and to achieve conditions in lakes close to undisturbed conditions according to the EU WFD (European Union 2000).

#### *1.4 Brownification, regional trends*

Both the lakes studied in the present work are situated in the part of Sweden subject to the most significant increases in TOC concentrations over the period AD 1990-1999, according to monitoring data from 344 lakes in Scandinavia (Löfgren et al. 2003). Long-term individual time series from southern Sweden starting earliest AD 1960 (<http://miljodata.slu.se/mvm/>) show some variability in colour and TOC concentration patterns in lakes (Fig. 1). Generally, most of the lakes showed an increasing trend in colour and TOC concentration since the beginning of the measurements. However, some of the lakes showed no distinct trends in colour during the period, notably Nässjön and V. Skälsjön. One lake, Farstusjön, showed decreasing trends from AD 2000. The most significant changes were observed in the lakes with the highest baseline colour, a relationship observed by others (Worall and Burt 2004a). Generally, lake colour and TOC content exhibit the same trends, with a few exceptions, for example, Gyslättsjön and Skärilen.

A survey of 38 lakes in the southern Swedish uplands, within an area of approximately 150 x 100 km, was conducted in July 2007, and included the

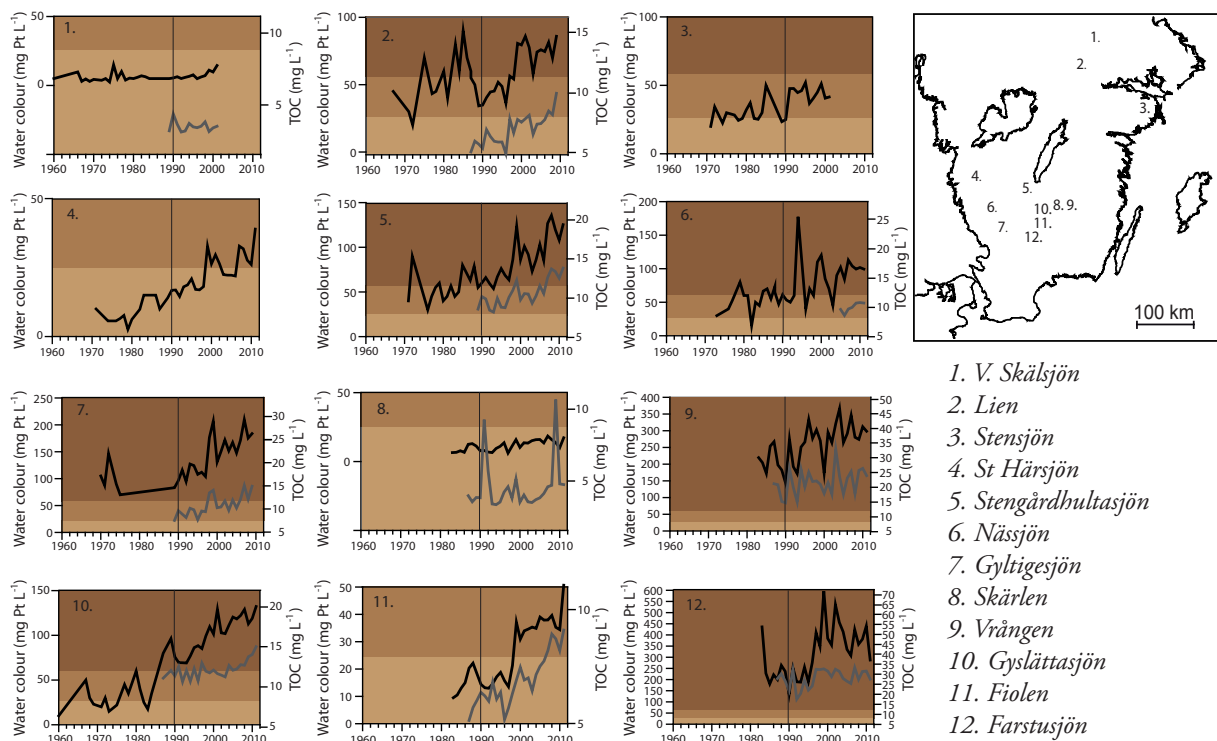


Figure 1. Long-term monitoring data from twelve lakes in southern Sweden. the map show the location of the lakes. Lake-water colour ( $\text{mg Pt L}^{-1}$ ) is shown by the black curve and total organic carbon (TOC) concentration ( $\text{mg L}^{-1}$ ) by the grey curve for each lake, as a function of time. Measurements of water colour were generally replaced by measurements of absorbance (400 nm) in the AD 1980-1990s, and have been converted here to water colour by multiplying by 500, according to the standard method (Swedish Environmental Protection Agency, 1999). The vertical lines in each diagram indicate the year AD 1990. The shaded areas represent different colour regimes in each lake according to the classification in Table 1. Light brown: not or slightly coloured, medium brown: moderately coloured and dark brown: significantly or highly coloured water.

two lakes studied in the present work (von Einem and Granéli 2010). It was found that at least half of the lakes showed a significant increase in colour after AD 1990, while other lakes showed no significant changes, or even a decrease in colour (Fig. 2). Lakes within the same region usually exhibit similar physical, chemical and biological characteristics, and the results suggests there is a possible common regional driving mechanism to the observed increases in DOC concentrations of the majority of the lakes in this area. But these results also suggest that there may be differences in the driving mechanisms of recent brownification, and that variations in the characteristics of the catchment area may lead to different DOC concentrations in individual lakes.

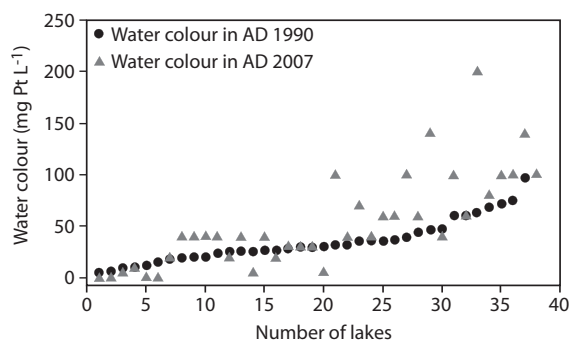


Figure 2. Data from 38 lakes sampled in July 2007 (von Einem and Granéli 2010) in the southern Swedish uplands, showing the change in water water colour since AD1990.

## 2. Summary of Papers

### 2.1 Paper I

*Bragée P, Choudhary P, Routh J, Boyle JF, Hammarlund D (in press) Lake ecosystem responses to catchment disturbance and airborne pollution: an 800-year perspective in southern Sweden. Journal of Paleolimnology*

Variations in nutrient cycling and deposition of lithogenic elements were studied in response to anthropogenic impact and catchment disturbance in two small lakes in the south Swedish uplands, Åbodasjön and Lindhultsgöl, during the past eight centuries. Variations in lake sediment carbon, elemental nitrogen and isotopic ratios (TOC, TN,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) were studied together with *n*-alkane organic geochemistry, to characterize nutrient cycling dynamics, and to distinguish terrestrial and aquatic sources of lacustrine organic matter. Changes in fluvial and airborne delivery of inorganic elements (K, Ti, Rb, Zr, P, Pb and Zn) to the lakes were determined using X-ray fluorescence. The results revealed that population growth and related increases in land-use pressure had a major impact on catchment erosion and thus the input of terrestrial organic matter to the lakes from the 1500s to the end of the 1800s. Evidence was also found of a brief period of catchment disturbance about AD 1200-1300, followed by recovery, probably due to the Black Death pandemic. In about AD 1900 synchronous shifts in most of the proxy records suggest a marked change in external forcing mechanisms common to both lakes, related to a major decrease in population density and the introduction of modern land use following the industrial revolution. The results also demonstrated differences in regional and local forcing of lake ecosystems, but indicated broadly similar sensitivities of the lake ecosystems to human impact, with site-specific responses to local disturbances during the past century due to different nutrient conditions and catchment area properties.

### 2.2 Paper II

*Mazier F, Broström A, Bragée P, Fredh D, Stenberg L, Thiere G, Sugita S, Hammarlund D. (in review) Two*

*hundred years of land-use change in the South Swedish Uplands: comparison of historical map-based estimates with pollen-based reconstruction using the Landscape Reconstruction Algorithm. Review of Palaeobotany and Palynology*

The local variation in land use in the South Swedish Uplands over the past 200 years, based on pollen records from three lake-sediment successions, was studied to assess ecosystem variability and responses to past anthropogenic disturbances. Temporal changes in the proportional cover of 14 plant taxa were quantified as percentages using the Landscape Reconstruction Algorithm (LRA). The LRA-based estimates of the extent of four categories of land use (cropland, meadows/grassland, wetland and outland/woodland) were compared to corresponding estimates based on historical maps and aerial photographs from AD 1769-1823, AD 1837-1895, AD 1946 and AD 2005. The LRA-reconstructed vegetation composition is generally in good agreement with estimates based on the historical records. The LRA approach was used to reconstruct the 200-year history of local land-use dynamics at 20-year intervals around two small lakes, Åbodasjön and Lindhultsgöl. The results showed differences in historical land use between the sites, and that local catchment characteristics, such as soil conditions and wetland cover, also appeared to be important for the development of human impact. Hence, the application of the LRA approach with high-resolution pollen records can provide detailed information on land-use dynamics on decadal to millennial timescales, providing the potential to evaluate the impact of land use on terrestrial and aquatic ecosystems, and land-use dynamics should, therefore, be taken into account when nature conservation strategies are being developed.

### 2.3 Paper III

*Bragée P, Mazier F, Rosén P, Fredh D, Broström A, Granéli W, Hammarlund D. Forcing mechanisms behind variations in total organic carbon (TOC) in lake water during the past eight centuries – Palaeolimnological evidence from southern Sweden. Submitted to Biogeosciences*

The aim of this study was to identify the major



forcing mechanisms behind observed increases in lake-water dissolved organic carbon concentration (DOC) in 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 total organic carbon (TOC) concentration in lake water since AD 1200 inferred from proxy records from two small lakes, Åbodasjön and Lindhultsgöl. Decadal-scale variations in TOC concentration in lake water were reconstructed based on visible-near infrared spectroscopy (VNIRS) of sediment successions. Changes in pH were inferred from diatom analysis. Pollen-based reconstructions of local land use, quantified using the Landscape Reconstruction Algorithm (LRA) and geochemical records, provided information on

catchment-scale environmental changes, and comparisons were made with records of climate and population density. The results revealed generally high TOC concentrations in lake water prior to AD 1900, with second-order variations associated mainly to changes in the intensity of agricultural land use. Significant changes were found in the past century, and unusually low TOC concentrations were recorded in AD 1930-1990, followed by a recent increase. The variation in sulphur deposition, with an increase in the early 1900s leading to a peak around AD 1980, followed by a decrease, was most likely the main driver of these dynamics. Declining atmospheric sulphur emission is the most probable driver of the increase in TOC concentration in lake water during the past thirty years. This suggests that

Table 2. Authors contributions to Papers I-IV

	<b>Paper I</b>	<b>Paper II</b>	<b>Paper III</b>	<b>Paper IV</b>
Fieldwork	P. Bragée F. Mazier A. Broström D. Hammarlund	P. Bragée F. Mazier A. Broström D. Hammarlund	P. Bragée F. Mazier A. Broström D. Hammarlund	P. Bragée F. Mazier A. Broström D. Hammarlund
Core correlation and sample preparation	P. Bragée	P. Bragée	P. Bragée	P. Bragée
Age-depth model	P. Bragée D. Hammarlund	P. Bragée D. Fredh D. Hammarlund	P. Bragée D. Hammarlund	P. Bragée D. Fredh D. Hammarlund
XRF analysis	P. Bragée	P. Bragée	P. Bragée	P. Bragée
C and N elemental and stable isotope analysis	P. Bragée		P. Bragée	
<i>n</i> -alkane analysis	P. Choudhary J. Routh			
VNIRS analysis			P. Rosén	
Diatom analysis			P. Bragée	
Pollen analysis		F. Mazier D. Fredh	F. Mazier D. Fredh	F. Mazier D. Fredh
LRA modeling		F. Mazier	D. Fredh F. Mazier	D. Fredh
Historical maps and aerial photographs		A. Broström L. Stenberg D. Fredh G. Thiere		
Data interpretation	P. Bragée P. Choudhary J. Routh J.F. Boyle D. Hammarlund	F. Mazier A. Broström P. Bragée D. Fredh S. Sugita D. Hammarlund	P. Bragée F. Mazier D. Fredh P. Rosén W. Granéli D. Hammarlund A. Broström	D. Fredh A. Broström F. Mazier P. Lagerås M. Rundgren P. Bragée D. Hammarlund

the lakes studied are recovering towards their natural high-TOC pre-depositional states. Given that the effects of sulphate deposition are declining, other forcing mechanisms related to land use and land management practices, and climate change will possibly become the main drivers of future changes in TOC concentration in the two studied lakes.

## 2.4 Paper IV

*Fredh D, Mazier F, Bragée P, Lagerås P, Rundgren M, Hammarlund D, Broström A. The effect of local land-use change on floristic diversity around two lakes in southern Sweden, AD 1000-2008. Submitted to The Holocene*

The relationship between land use and floristic diversity was analysed around two lakes in southern Sweden, Åbodasjön and Lindhultsgöl, over the past thousand years. Pollen analysis and the Landscape Reconstruction Algorithm approach including the Local Vegetation Estimates (LOVE) model were used to quantify land cover on local scales with high resolution (20 to 50 years). Floristic richness was estimated using palynological richness, and LOVE-based evenness was introduced as a proxy for floristic evenness on a local scale based on the LOVE output. The results revealed a dynamic land-use pattern, with agricultural expansion during the 1200s, partly abandoned landscape around AD 1400, re-establishment during the 1400-1500s and a transition from traditional to modern land use during the 1900s. The results suggest that the more variable landscape and dynamic land use during the 1200s to 1800s characteristic of the traditional landscape were of substantial importance in achieving high floristic diversity. The landscape provided different production services, such as grazing areas for cattle, and regulating services, such as pollination of crops and invasion resistance. During approximately the past 100 years, areas with high floristic and faunal diversity such as meadows and pastures have decreased in favour of crop cultivation and timber production, i.e. potential areas for providing cultural and regulating services. More habitats are also related to coniferous woodlands, and fewer habitats are related to deciduous trees and open land taxa, which may not be sustainable in preserving floristic diversity in the

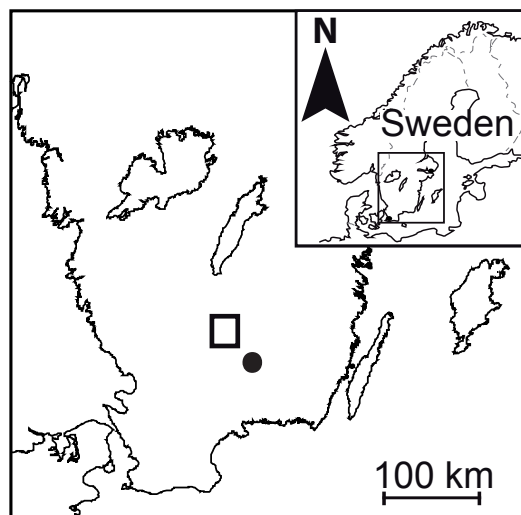


Figure 3. Map of Scandinavia and southern Sweden showing the study area (indicated by the square) and the closest city, Växjö (•).

future. Palynological richness sometimes remains at higher levels during periods of regression, at least during roughly the first 40 years, which suggests that many plants can survive during periods of succession and reforestation. The variability in past agricultural landscape provides information about types of land use that promote floristic diversity, which is potentially useful for nature conservation and the implementation of the framework of ecosystem services.

## 3. Study Area and Lakes

### 3.1 Present conditions

The location of the area studied is illustrated in Figure 3. The crystalline bedrock in the area studied is dominated by granite and gneiss (Wikman 2000), and is covered by sandy glacial till of various thicknesses, with occasional glaciofluvial deposits and scattered peat deposits (Daniel 2009). 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 values from the largest city in the region, Växjö, for AD 1961-1990 (Alexandersson et al. 1991). The regional vegetation belongs to the boreo-nemoral vegetation zone, and is characterized by woodland containing a mixture of coniferous and deciduous

trees (Sjörs 1963; Gustafsson 1996).

The two lakes, Åbodasjön (221 m asl) and Lindhultsgöl (212 m asl), are situated in the upland area of southern Sweden, in a rural setting in the province of Småland. Åbodasjön is an oligotrophic mesohumic lake fed by two inlet streams, from the south and north-east, and has an outlet in the south-west. The village of Åboda (40 residents in 2004) is situated west of the lake, and the catchment area is currently dominated by managed coniferous woodland with semi-open areas of deciduous trees and cropland near the lake margins. Lindhultsgöl is an oligotrophic polyhumic lake with at least two artificial ditches draining into it from nearby wetland and woodland, and there is an outlet consisting of an artificial ditch in the south. The catchment area consists mostly of managed coniferous woodland and wetland with shrubs and pine trees. The morphometric and hydrological characteristics are given in Table 3, and detailed maps of the two lakes are illustrated in Figure 4.

### 3.2 Local history

The lakes are situated in Slätthög Parish (Fig. 4), an elongated area (22 km in length) covering about 138 km<sup>2</sup>, established in around AD 1000. Archaeological remains in the area reveal human presence at least during the last 6000 years. The first local population data are available in church records from AD 1571, and show that the parish had 301 inhabitants (<3 inhabitants km<sup>-2</sup>; Andersson Palm 2000). During the 1700s, the population started to increase rapidly, and reached a peak of 2485 in AD 1865 (about 20 inhabitants km<sup>-2</sup>), followed by a decrease in rural population in response to industrialisation in the late 1800s.

The general land-use history of the southern Swedish uplands is known through palaeo-ecological studies, archaeological evidence and historical records. The area has been subject to a number of agricultural expansions and regressions during the past 6000 years (Lagerås 1996; Berglund et al. 2002). The early expansions were dominated by grazing animals and single farms with small-scale agriculture (Berglund et al. 2002). This was followed by a general agricultural development in the uplands of southern Sweden in Mediaeval times (about AD

*Table 3. Morphometric and hydrological characteristics of the two lakes studied, sampled in July 2007 (von Einem and Granéli 2010)*

	Åbodasjön	Lindhultsgöl
Altitude (m)	221	212
Lake surface area (km <sup>2</sup> )	0.5	0.07
Maximum depth (m)	9	5
Catchment area (km <sup>2</sup> )	9.5	0.6
Residence time (years)	0.5	-
pH	7.0	6.4
Alkalinity	0.56	0.83
Chlorophyll a conc.(µg L <sup>-1</sup> )	7.7	14.9
DOC conc.(mg L <sup>-1</sup> )	11.0	23.8
Water colour (mg Pt L <sup>-1</sup> )	40	960
Liming started	1984	1993

900-1200) towards permanent farming with permanent cropland and meadows, used for hay production, located close to the settlements. The land outside this centre, the outland, was the common land used for cattle grazing and the collection of firewood and timber. A second expansion took place in the 1500s, and records showed that 49 farms were established in the parish in the mid-1500s, and that farms were situated within 1-2 km of the two lakes studied in this work. Agriculture developed during the agricultural revolution (about AD 1700-1900) leading to improvements in yield from small-scale agriculture through better management of manure, crop rotation and marling (Emanuelsson 2009; Myrdal and Morell 2011). The maximum degree of agricultural land use was seen in the late 1800s, and during the past century small-scale agriculture has been replaced by modern land use dominated by commercial forestry and crop cultivation (Antonsson and Jansson 2011).

Historically, important local industries in the study area have included iron production based on bog ore (AD 1530-1610), potash manufacturing based mainly on beech wood (AD 1753-1800), the production of tar and charcoal based mainly on pine wood (AD 1600-1850), and nitrate production, based on manure, for gunpowder (AD 1780-1817). Gamleboda mill, situated at the outlet of Åbodasjön from AD 1756 and onwards, has



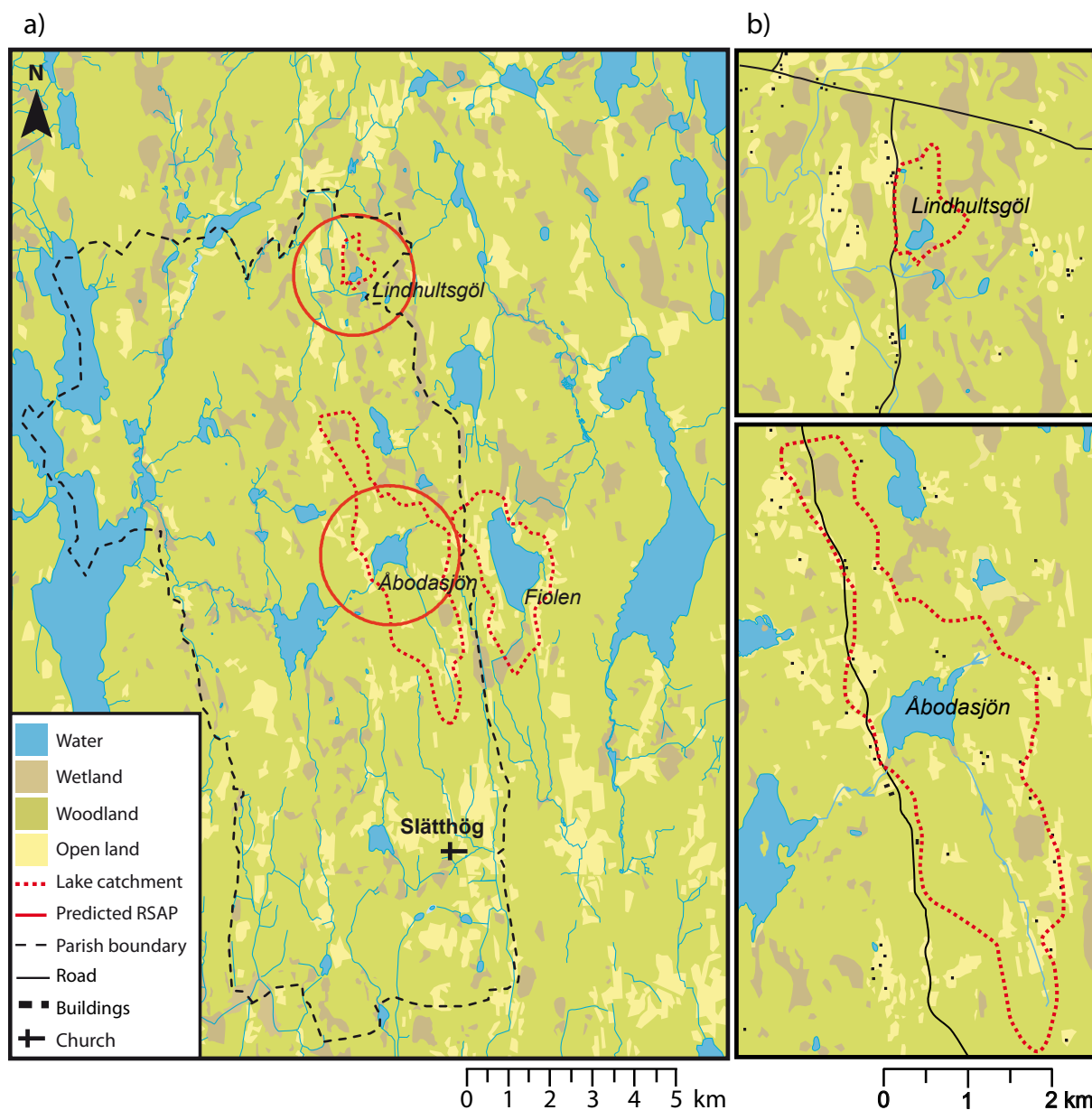


Figure 4. (a) Map showing the location of the lakes Åbodasjön, Lindhultsgöl and Fiolen. Modern land-use properties, catchment areas for each lake (red dashed lines), relevant source area of pollen (RSAP) (red circles), and the parish boundary (black dashed line) are shown. (b) Maps of the studied lakes showing modern land-use properties, catchment areas for each lake and important infrastructure.

regulated the water level of the lake. Several sawmills were established in the study area in the 1950s.

### 3.3 Site-specific trends in lake-water DOC and colour

In 2007, Åbodasjön was moderately coloured (40 mg Pt L<sup>-1</sup>) with a DOC concentration of 11 mg L<sup>-1</sup>. Lindhultsgöl was highly coloured (960 mg Pt L<sup>-1</sup>) and had a DOC concentration of 23.8 mg L<sup>-1</sup>

(Table 1). Yearly monitoring data from the inlet of Åbodasjön, based on an average of several samples (County Administrative Board of Kronoberg, unpublished data), also showed an increasing trend in water colour since the 1990s with an R<sup>2</sup> value of 0.5188 (Fig. 5). Based on the monitoring data available, it can be concluded that Åbodasjön has been subject to brownification during the past three decades. Unfortunately, no monitoring data are available for Lindhultsgöl.

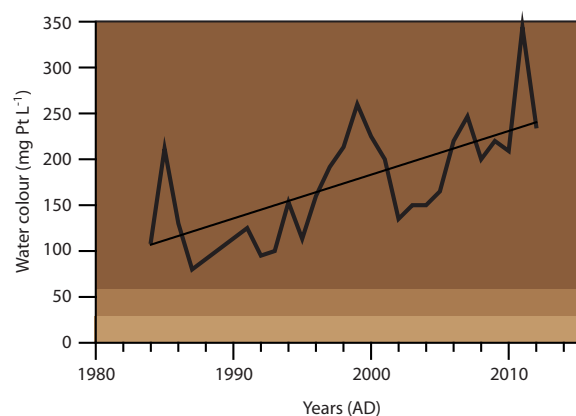


Figure 5. Water colour in Åbodasjön, based on yearly mean values from monitoring data plotted against time, fitted with linear regression ( $R^2$ : 0.5188). The brown shaded areas represent different colour regimes, according to Figure 1.

## 4. Methods - Reconstructing the Past

### 4.1 Stratigraphic approach

#### 4.1.1 Sediment coring and subsampling

Sediment cores were collected at the central parts of the lakes in spring 2008, at a water depth of 8.6 m in Åbodasjön and 5.2 m in Lindhultsgöl. Surface sediment sequences, 32 cm in Åbodasjön and 30 cm in Lindhultsgöl, were obtained at both sites using a gravity corer (HTH-Kajak corer: Renberg and Hansson 2008) from a platform, and divided in the field into 5-mm contiguous intervals. Overlapping sediment cores of 4.5 m in Åbodasjön and 2.6 m in Lindhultsgöl, were collected using 1-m-long Russian peat corers. All cores were transferred to supportive plastic liners, wrapped in plastic and transported to Lund, where the uppermost 1-m from each lake was divided into 5-mm contiguous sections in the laboratory. All sediment samples were kept in cold storage (4°C) prior to analyses. Correlations between the surface sediments and Russian core segments were based on mineral magnetic properties (Thompson 1980) measured at the Palaeomagnetic and Mineral Magnetic Laboratory at Lund University, and X-ray fluorescence measurements of element compositions (Boyle 2000).

### 4.1.2 Dating and chronologies

Accurate dating of sediment cores and the construction of robust chronologies with high temporal resolution is necessary for the comparison of data collected at the two lakes and to ensure reliable interpretations of events. For this purpose, a combination of different dating methods was used.

For dating of the most recently deposited sediments (e.g. <150 y), a lead radioisotope ( $^{210}\text{Pb}$ ) was used to date sediment samples from the gravity cores (Appleby 2002). Sediment samples were analysed to determine the activities of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  using gamma spectrometry at the Gamma Dating Center, Institute of Geography at the University of Copenhagen, Denmark. The total signal of  $^{210}\text{Pb}$  in the sediments consists of supported  $^{210}\text{Pb}$  from autogenic material and unsupported  $^{210}\text{Pb}$  originating from atmospheric deposition. The unsupported  $^{210}\text{Pb}$  activity is the fraction used for dating, and was calculated using the  $^{226}\text{Ra}$  concentration, based on the assumption that supported  $^{210}\text{Pb}$  is in equilibrium with its parent nuclide  $^{226}\text{Ra}$ . The  $^{210}\text{Pb}$  decays with a half-life of 22 years, and the remaining amount of unsupported  $^{210}\text{Pb}$  activity left at a certain depth reveals the age of the sediment. To calculate sediment ages, the constant rate of supply (CRS) model (Appleby 2002) was applied to the profiles. Variations in the radioisotope  $^{137}\text{Cs}$  was used as time markers based on peak concentrations associated with stratospheric testing of atomic weapons beginning in AD 1952, and the Chernobyl nuclear power plant accident in AD 1986.

For older sedimentary sequences, the most common method of choice is radiocarbon ( $^{14}\text{C}$ ) dating (Björck and Wohlfarth 2002). Radiocarbon is produced in the atmosphere by cosmic rays, and is taken up by all living organic matter on earth. When an organism dies,  $^{14}\text{C}$  is no longer absorbed and the remaining radiocarbon decays with a half-life of 5730 years. Selected macrofossil remains and bulk samples from both lakes were subjected to accelerator mass spectrometry  $^{14}\text{C}$  dating at the Radiocarbon Dating Laboratory, Lund University. Bulk samples can be subjected to “old carbon

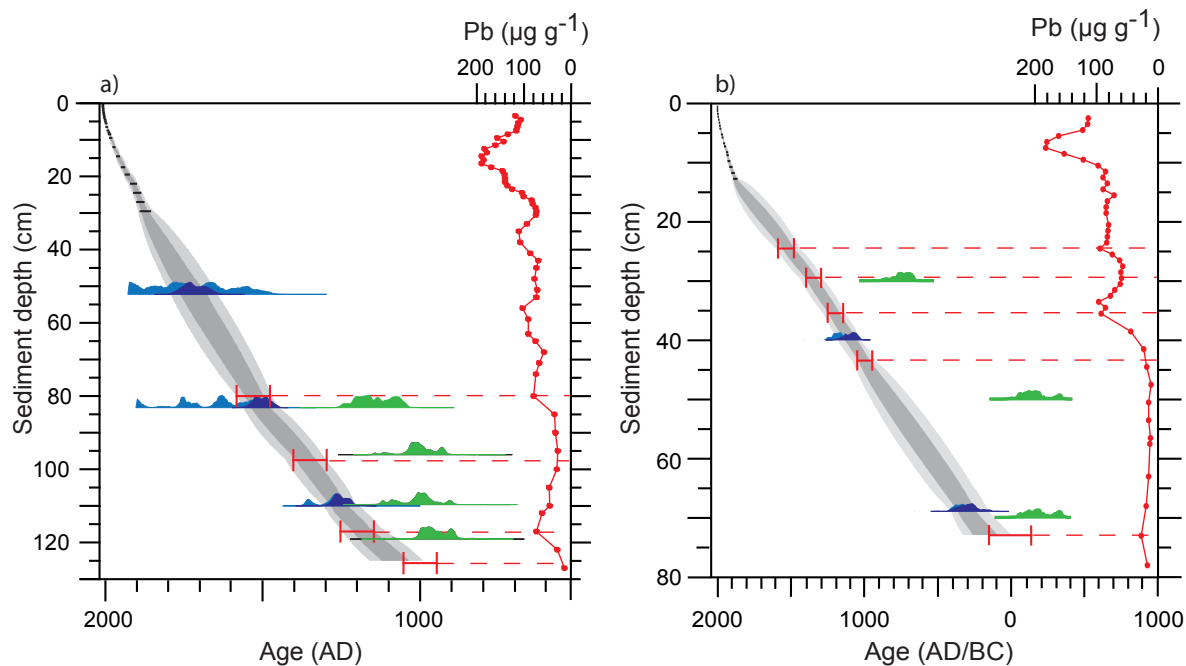


Figure 6. Age-depth models based on  $^{210}\text{Pb}$  dates (horizontal black bars), calibrated radiocarbon dates obtained from terrestrial macrofossils (blue) and bulk samples (green), and Pb pollution marker horizons (horizontal red bars), together with the records on total Pb concentration (red) for: a) Åbodasjön and b) Lindhultsgöl. Depths of Pb pollution marker horizons are indicated by the red dashed lines. The modelled  $\pm 1\sigma$  and  $\pm 2\sigma$  confidence intervals are indicated by dark and pale grey shading, respectively.

effects”, i.e. the incorporation of older carbon due to reworking of the sediment, resulting in higher ages (Barnekow et al. 1998; Reuss et al. 2010). Therefore, only dated macrofossils were used to determine the chronology. The radiocarbon age of each sample is not equivalent to calendar years, due to natural variations in  $^{14}\text{C}$  production. All  $^{14}\text{C}$  dates were converted into calendar years using the IntCal09 radiocarbon calibration dataset (Reimer et al. 2009) and the calibration program OxCal4.1 (Bronk Ramsey 2009).

The concentration of Pb in sediment can be used as a time marker, based on the regionally coherent pattern of Pb deposition resulting from airborne pollution over Sweden during recent millennia (Brännvall et al. 2001; Renberg et al. 2001). The large-scale trends include a substantial increase in Pb deposition around AD 1000, mainly related to mining and metal production in Europe, which reached a peak in about AD 1200. This was followed by a minimum around AD 1350 in response to plagues and recession, and another peak around AD 1530, reflecting increased silver ore processing in continental Europe (Brännvall et al. 2001). These four marker horizons were all clearly

seen in the Åbodasjön and Lindhultsgöl Pb records. Subsequent fluctuations at relatively high levels from about AD 1530 to 1900, followed by rising values to maximal Pb concentrations in the 1970s, due to the extensive use of leaded petrol, were also recorded. The 1970s Pb peaks were not incorporated into the age models as robust  $^{210}\text{Pb}$  data are available in this part of the sediment records. The Lindhultsgöl record also includes a minor peak in Pb deeper in the sediments, which is believed to represent the Greek and Roman civilizations around BC/AD 0 (Brännvall et al. 2001). The concentrations of Pb and other elements were obtained using X-ray fluorescence (XRF) analysis (Boyle 2000). Sediment samples were analysed using an S2 Ranger XRF spectrometer at the Department of Geography, University of Liverpool.

The chronology for Åbodasjön was based on 25  $^{210}\text{Pb}$  dates, three  $^{14}\text{C}$  macrofossil dates and four Pb pollution age markers. The chronology for Lindhultsgöl was based on 19  $^{210}\text{Pb}$  dates, two  $^{14}\text{C}$  macrofossil dates and five Pb pollution age markers (Paper I). To establish chronologies for the two lakes, stratigraphic age-depth models were constructed using the P\_Sequence deposition

model in OxCal4.1 (Bronk Ramsey 2008), where depths between samples were introduced to constrain the calibration probability intervals. All ages were expressed as calendar years AD, with a  $2\sigma$  confidence interval (95.4% probability). Åbodasjön chronology covers the period from AD 1200 to 2008, and the chronology for Lindhultsgöl covers approximately 2000 years. Figure 6 shows the chronologies for Åbodasjön and Lindhultsgöl. The chronology of the nearby lake Fiolen is described in detail by Fredh et al. (2012).

## 4.2 Environmental reconstructions

### 4.2.1 Lake-water chemistry

Two different methods, both based on the same approach using surface-sediment training sets for biological or geochemical indicators, can be used to reconstruct past changes in environmental variables in lake water, such as TOC and pH. Samples are collected from a series of lakes to analyse present-day environmental variables of interest, and these are then compared to the indicators preserved in the surface sediments using statistical techniques. If there is a high degree of certainty, quantitative transfer functions can be developed and used to infer past specific environmental variables.

In this study, VNIRS was used to infer the TOC concentrations of the lake water (Paper III). The use of VNIRS in palaeolimnology was introduced in the 1990s (Korsman et al. 1999) for quantitative inferences of, for example, TOC concentrations, pH and phosphorus concentrations in lake water. The method has since been developed using training sets for northern and southern Sweden (Rosén, 2005; Cunningham et al., 2011), and northern Canada (Rouillard et al. 2011). VNIRS measures the absorbance of wavelengths of 400-2500 nm, reflecting the molecular vibrations of organic compounds in the sediment. Past changes in TOC concentration in lake water were reconstructed using a calibration model based on VNIRS of surface sediments from 140 Swedish lakes covering a gradient from 0.7 to 24 mg L<sup>-1</sup> (Cunningham et al. 2011). The inferred TOC concentrations in Lindhultsgöl exceeded the range covered by the calibration set, and an additional inference model

from Canada, based on 160 lakes with a DOC range of 0.6-39.6 mg L<sup>-1</sup> was used (Rouillard et al. 2011). The performance of the combined Swedish and Canadian calibration set is similar to that of the Swedish calibration set, showing an R<sup>2</sup> value of 0.6 between measured and predicted TOC concentration in lake water and a root mean square error of prediction of 4.1 mg L<sup>-1</sup> (10.5% of the gradient).

The pH in the lakes was inferred from sedimentary diatom assemblages using calibration models (Paper III). Diatoms are unicellular, eukaryotic organisms well preserved in lake sediments (Smol and Cumming 2008). Different taxa have different environmental optima, and analysis of assemblages of fossil species can therefore be used to reconstruct environmental variables (Battarbee et al. 1999). Diatom assemblages were separated from the sediment in the laboratory using the water-bath technique described by Renberg (1990b) with digestion of H<sub>2</sub>O<sub>2</sub> (Battarbee et al. 2001). To estimate diatom concentrations, a known quantity of divinylbenzene microspheres was added to the digested and cleaned samples (Battarbee and Kneen 1982; Wolfe 1997). The samples were evaporated onto cover slips, and at least 400 diatom valves per sample were counted and identified using a light microscope, largely using available reference material, for instance, Krammer and Lange-Bertalot (1986-1991). The pH was inferred using the quantified diatom data in the European Diatom Database combined pH training set online (<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 in the lake water was analysed using a locally weighted average model and inverse deshrinking. In total, 18 samples were included for each lake and the results are presented in Figure 7.

### 4.2.2 Nutrient cycling and the origin of organic matter

Organic matter in the lake basin originates from the terrestrial surroundings and aquatic sources, in dissolved and particulate form. Sedimentary organic geochemical records can provide fundamental information on the origin and fate of the organic

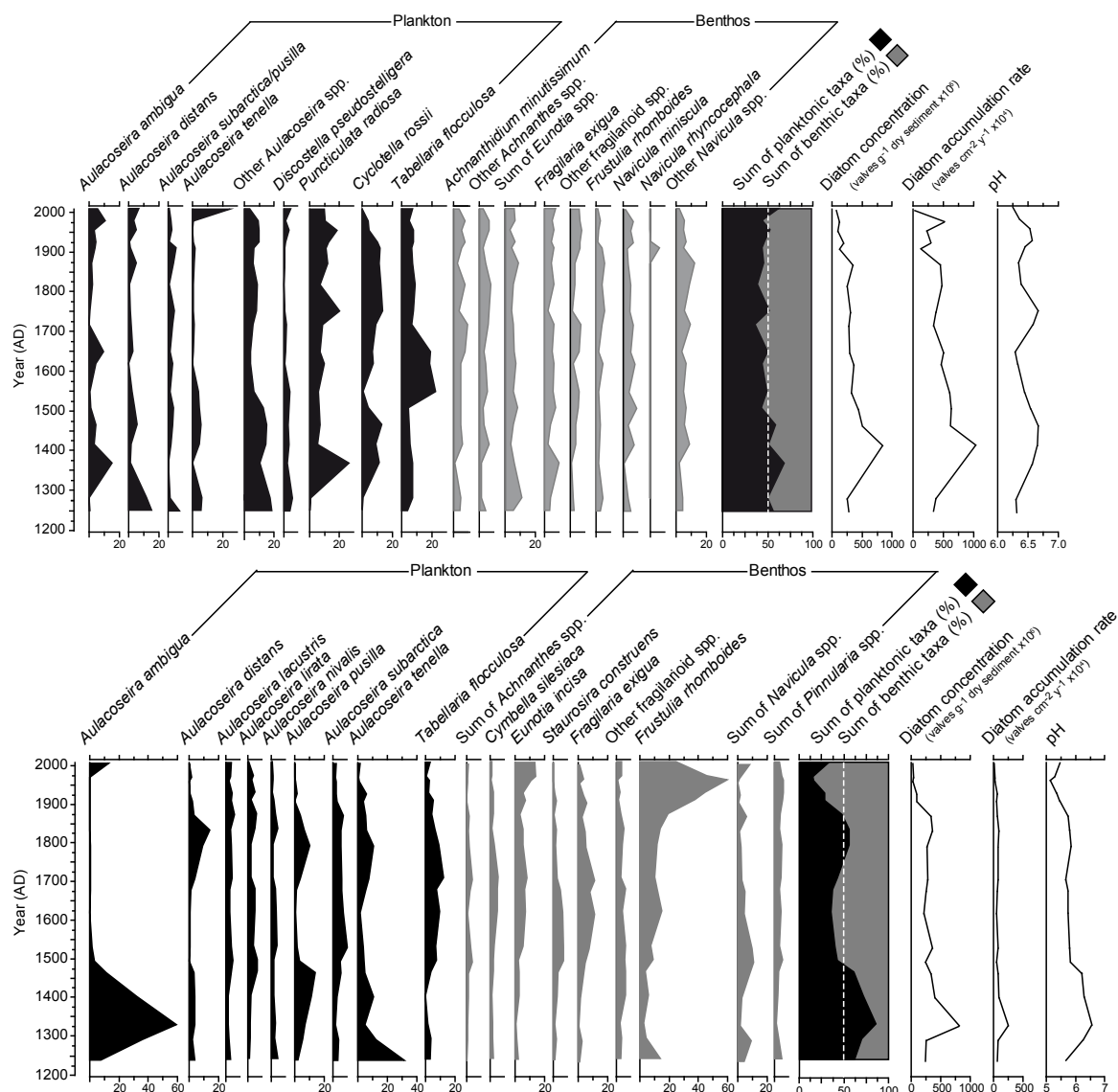


Figure 7. Diatom stratigraphy of the major (>5%) taxa, in: Åbodasjön (upper panel) and Lindhultsgöl (lower panel), expressed as relative abundance (%), proportion of planktonic and benthic taxa (%), diatom concentration, diatom accumulation rate and inferred pH.

matter preserved in lake sediments. In this study, various organic proxies were applied, including bulk organic carbon (TC) and nitrogen (TN) content, atomic C/N ratios (Papers I and III), stable carbon and nitrogen isotope ratios ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) and hydrocarbon concentrations (*n*-alkanes) (Paper I). Atomic C/N ratios of lignin- and cellulose-rich terrestrial organic matter are typically >20, whereas aquatic organic matter is characterized by atomic C/N ratios <10 (Meyers and Lallier-Vergès 1999). Values of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  for organic matter are influenced by a number of factors, and they have been used in combination with TC and TN composition of lake sediments to assess changes in

carbon and nitrogen sources and nutrient balance, often in response to anthropogenic activities in watersheds (Meyers and Teranes 2001; Talbot 2001). Hydrocarbon molecules (*n*-alkanes) are widely used to assess sources of organic matter because of their long residence times and resistance to degradation. Molecular distributions of *n*-alkanes can be used to differentiate biological sources. Most aquatic algae and photosynthetic bacteria are dominated by *n*-C<sub>15</sub>, C<sub>17</sub> and C<sub>19</sub> alkanes, while vascular plants contain large proportions of *n*-C<sub>27</sub>, C<sub>29</sub>, and C<sub>31</sub> alkanes in their epicuticular wax coatings (Cranwell et al. 1987; Rieley et al. 1991; Tenzer et al. 1999).



Bulk organic TC and TN contents were analysed during combustion using a Costech ECS 4010 elemental analyser at the Department of Geology, Lund University. Values of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  were analysed using a VG-isotope Micromass dual-inlet mass spectrometer equipped with a EuroVector elemental analyser and a continuous flow inlet, at the Department of Geography and Geology, University of Copenhagen. The data are reported in  $\delta$ -notation;  $\delta = [\text{R}_{\text{sample}}/\text{R}_{\text{std}} - 1] \times 1000$ , where R denotes the  $^{13}\text{C}/^{12}\text{C}$  and  $^{15}\text{N}/^{14}\text{N}$  ratios in the samples, and VPDB and AIR standards, respectively.

To determine the *n*-alkanes, sediment samples were extracted with a mixture of  $\text{CH}_2\text{Cl}_2$  and  $\text{CH}_3\text{OH}$  (9:1 v/v). The extracts were reduced using a Büchi rotovapor and injected in splitless mode into an Agilent 6890 gas chromatograph equipped with a HP5-MS column, at the Department of Geological Sciences, Stockholm University. The chromatograph was interfaced with an Agilent 5973 mass spectrometer. The *n*-alkane concentrations were normalized with respect to sediment carbon contents.

#### 4.2.3 Catchment-scale land use

Palaeoecological studies based on fossil pollen assemblages found in sediments have been an important tool, for almost a century, in reconstructing past vegetation and anthropogenic activities, such as tree succession and woodland clearance for crop cultivation (Bennett and Willis 2001). It has previously been difficult to estimate past vegetation based on fossil pollen, due to known biases in the way in which vegetation is represented in the fossil pollen record, for example, differences in pollen productivity and dispersal between taxa (Broström et al. 1998; Sugita 2007 a; b). In the present work, fossil pollen records from Åbodasjön, Lindhultsgöl and Fiolen were used to reconstruct and quantify past local land use and associated vegetation using the LRA (Sugita 2007a; b), which compensates for these differences (Papers II, III and IV).

For pollen analysis, sediment samples were treated with the acetolysis method and mounted on slides for identification using a light microscope

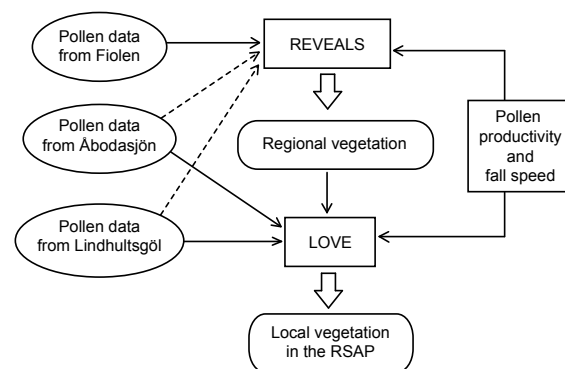


Figure 8. Flow chart illustrating how the Landscape Reconstruction Algorithm was used in this study to quantify past regional and local land use.

(Berglund and Ralska-Jasiewiczowa 1986). At least 1000 pollen grains corresponding to the selected taxa were identified and counted in each 20-year time window, using identification keys (Moore et al. 1991; Punt et al 1976-2009, Beug 2004) and the reference collection at the Department of Geology, Lund University.

For a reliable reconstruction of regional and local vegetation and land use there are two steps using the LRA approach illustrated in Figure 8. The first step in the LRA is to reconstruct the regional plant abundance and composition within a 50-100 km radius using the REVEALS (Regional Estimates of Vegetation Abundance from Large Sites) model. The second step is to apply the LOVE (Local Vegetation Estimates) model for the reconstruction of local vegetation and land use using the background pollen loading based on the regional plant abundance estimates obtained from the REVEALS model. The REVEALS model was applied to pollen counts from Fiolen and Åbodasjön, and the LOVE model was subsequently applied to pollen counts from Lindhultsgöl. Similarly, the REVEALS model was applied to pollen counts from Fiolen and Lindhultsgöl, and the LOVE model was subsequently applied to pollen counts from Åbodasjön. The modelled radii of the relevant source area of pollen (RSAP) for the two lakes, Åbodasjön and Lindhultsgöl, were 1740 and 1440 m from their respective centres (Fig. 4). It is important to note that the model covers a larger area (RSAP) than the actual catchment area, and in this study, changes in land use within the RSAP are assumed to reflect catchment variations in general.

In total, 26 taxa were used in the model, and were sorted and calculated to represent five different types of land use: 1) coniferous woodland, 2) deciduous woodland, 3) grassland, 4) cropland and 5) wetland, based on assumptions regarding the habitats of different taxa (Paper II).

In addition to pollen-based reconstructions of land use, historical maps provide important information on changes in site-specific land use. In Sweden, detailed maps are available from AD 1749, when cadastral maps were produced to reduce the fragmentation of land patches owned by individual farmers, in order to increase agricultural efficiency and productivity (Örback 1998). From AD 1939 to 1976, aerial photograph-based economic maps replaced traditional mapping methods for governmental administration, including forest and agriculture management. Cadastral maps are available for Åbodasjön and Lindhultsgöl from AD

1769-1826 and AD 1837-1895, economic maps for AD 1946, and rectified aerial photographs (orthophotos) for AD 2005. The categories of land use obtained from the historical maps and aerial photographs were quantified for comparison and evaluation of the LRA-based reconstructions of local land use (Paper II). The cadastral and economic maps were rectified using control points (i.e. crossroads, lake borders and administrative borders) on a modern georeferenced base map. Categories of land use were classified and their areas calculated from the maps using ArcGIS 9.2. This enabled comparisons between four different time windows over the past 200 years, and quantification of the changes in land use within the area studied (Fig. 9). The LRA model LOVE was applied on a longer timescale covering the past 800 and 1000 years in the studies described in Papers III and IV, respectively.

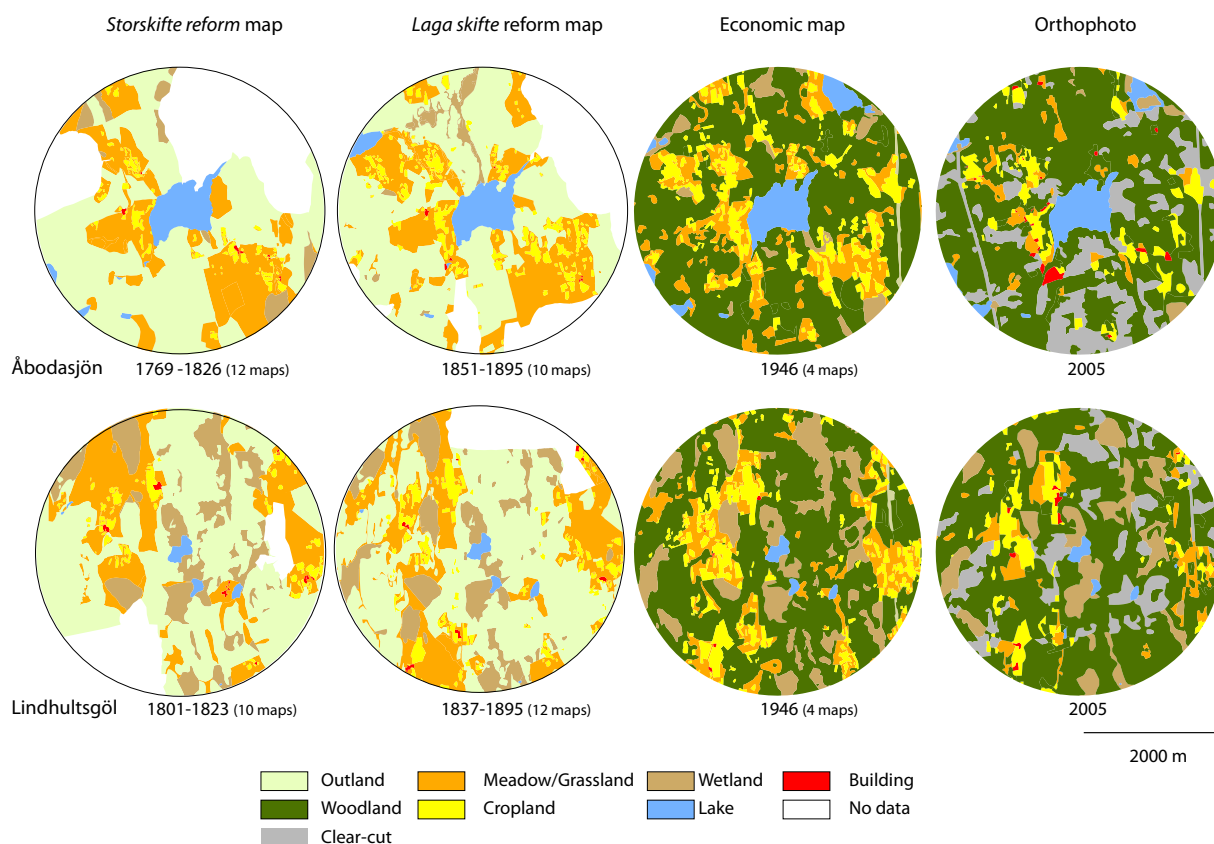


Figure 9. Documented categories of land use derived from historical maps and available orthophotos within 2-km radii around Åbodasjön and Lindhultsgöl.

#### 4.2.4 Catchment-scale erosion

Catchment erosion, which affects the terrestrial influx of inorganic and organic matter to the lakes, is regulated by precipitation, vegetation and type of land use, as well as human activities. Enhanced catchment erosion may be reflected by elevated concentrations of lithogenic elements potassium (K), titanium (Ti), rubidium (Rb), and zirconium (Zr) in the sediment profiles (Engstrom and Wright 1984; Boyle 2001). The total concentrations of 35 different inorganic elements, including the lithogenic elements, were determined using XRF analysis (Boyle 2000) (Papers I, III and IV). Elemental Rb and Zr/Ti ratios were also used as indicators of variations in mineral grain size in the lake sediments, as Zr is associated with silt particles, and Ti and Rb are often found in the fine silt and clay fractions (Koinig et al. 2003; Taboada et al. 2005).

### 5. Results and Discussion

#### 5.1 VNIRS-inferred TOC concentrations, colour and long-term trends in Åbodasjön and Lindhultsgöl

The VNIRS-inferred TOC reconstructions for the two lakes studied show substantial increases in TOC concentration during the 1990s. The TOC concentration in Åbodasjön increased by 2 mg L<sup>-1</sup> between AD 1990 and 1995, after which it levelled out. The increase in TOC in Lindhultsgöl was considerably higher, 10 mg L<sup>-1</sup>. The increase started at around AD 1980, and was accentuated after AD 1990, followed by a continuous increase to the present day (Fig. 10). The highest annual increases in DOC concentration have previously been observed in waters with the highest average DOC concentrations (Worrall and Burt 2004a), as found for the 12 lakes monitored in southern Sweden with the highest DOC concentrations (Fig. 1). The findings for Lindhultsgöl are thus in line with this observation. The reconstructed present-day TOC concentrations (AD 2007), of 9.9 mg L<sup>-1</sup> for Åbodasjön and 21.8 mg L<sup>-1</sup> for Lindhultsgöl, were slightly lower than the present-day measured DOC concentrations which were 11 mg L<sup>-1</sup> and 23.8 mg

L<sup>-1</sup>, respectively (Table 3). However, the agreement is good, bearing the mind the root mean square error of prediction for the VNIRS model of 4.1 mg L<sup>-1</sup>. Furthermore, the TOC concentration was calculated as a yearly mean, while the DOC concentration was derived from a single measurement, hence lacking a considerable amount of temporal variability in the data.

During the past 800 years, the VNIRS-reconstructed TOC records for both lakes generally show concentrations similar to or higher than the present day, apart from some low and exceptionally low values during parts of the previous century (Fig. 10), which is in accordance with previous long-term studies of lake-water TOC records in southern Sweden (Cunningham et al. 2011; Rosén et al. 2011). Generally, the TOC records for Åbodasjön show greater variability than those for Lindhultsgöl, and the TOC concentration in Lindhultsgöl exhibited higher and more stable values prior to AD 1900.

The discrepancy between TOC concentration and colour in Åbodasjön during recent decades suggests a change in the composition of organic carbon (i.e. humic substances) and/or a change in iron concentration. This implies that it is also reasonable to assume that past variations in TOC may not follow changes in colour. It is not possible to differentiate between differences in the composition of organic carbon using the VNIRS method, and the proportion of coloured humic substances in the TOC pool in the lakes is therefore unknown in this study. However, fossil diatoms may provide information on past light penetration, which affects water colour, as decreased light penetration reduces benthic growth, leading to a higher diatom planktonic/benthic (P/B) ratio (Rosén et al. 2009; Hansson 1992). Additional data, such as those from reconstructions of land use, may also provide important information on the availability of soil DOC and humic substances as this is regulated by the amount of organic-rich soil (Wilson and Xenopoulos 2008).

The diatom record from Åbodasjön generally exhibited a rather stable proportion of planktonic and benthic taxa over the past 800 years, except for the period between about AD 1200 and 1450, which was characterized by a higher P/B ratio and



an abundance of planktonic taxa (Figs. 7 and 10), indicating more coloured water. This is in agreement with high water TOC concentrations, and the highest abundance of wetland, as wetland is usually associated with organic-rich and coloured high-molecular-weight organic matter (Wilson and Xenopoulos 2008). However, this also coincided with early agricultural expansion, and planktonic taxa are typically favoured by high pH commonly caused by anthropogenic alkalisation resulting from the release of bases and nutrients as a result of burning in the landscape (Renberg et al. 1993; Boyle 2007). Accordingly, increasing diatom-inferred pH are recorded during this period (Fig. 7 & 10). There was no general agreement between the P/B ratio and TOC trend in the Åbodasjön records, implying a weak relationship between colour and TOC concentration on a long temporal scale in this particular lake. This was further confirmed by the lack of change in the P/B ratio during the substantial increase in TOC concentration in the 1800s, which peaked at AD 1860-1910. During this period, land use was associated with an increase in open land with a dominance of grasslands used for grazing and cultivation (Fig. 10). Agricultural land and the transformation to agricultural systems typically results in the export of structurally less complex dissolved organic matter characterized by colourless organic matter with a lower molecular weight (Dalzell et al. 2011; Kalbitz et al. 2003; Wilson and Xenopoulos 2008), and would explain the absence of any significant changes in lake-water colour during this period.

Two periods of elevated P/B ratios were observed at Lindhultsgöl between AD 1200 and 1500 and in the 1800s (Fig. 7 and 10). The first period was associated with a high TOC concentration and an increase in wetland and conifer woodland, typically associated with high-DOC soils and organic matter export (Sobek et al. 2007, Xenopoulos et al. 2003). However, the gradual decrease in TOC concentration during the second period, in the 1800s, despite increased wetland cover and a higher P/B ratio, indicates limited light penetration. It may be hypothesized that the more coloured conditions were driven by a change in the composition of organic carbon associated with the

observed increase in wetland during this period. Starting in the late 1800s, a considerable increase in benthic taxa (80%) was seen, indicating increased light penetration, accompanied by a significantly decreasing and low TOC concentration. Decreasing diatom-inferred pH during the same period indicates more acidic conditions, probably leading to a reduction in soil DOC export. However, considering the generally high TOC concentration of  $>12 \text{ mg L}^{-1}$ , indicative of significantly coloured water, together with the abundance of wetland, it is unlikely that the water was very clear at Lindhultsgöl during the period of low P/B ratios. The P/B ratio was strongly influenced by the dominance of *Frustulia rhomboides* (Fig. 7), an epipelagic benthic taxon generally abundant in lakes with a high DOC concentration (Spaulding and Edlund 2008), and the interpretation of the P/B ratio is therefore uncertain.

The observed brownification at Åbodasjön in the 1990s was also accompanied by a slight increase in P/B ratio together with a substantial increase in *Aulacoseira tenella* (Fig. 7), which is mainly found in highly coloured waters (Huttunen and Turkia 1994). At Lindhultsgöl this time period was also associated with an increase in P/B ratio, implying that the water in Lindhultsgöl has become more coloured in recent decades, together with the increase in TOC concentration.

An increase in iron concentration may also be a potential driver of brownification in Swedish waters (Kritzberg and Ekström 2012). Both lakes show a considerable increase in sedimentary iron concentration during the previous century (Fig. 10), but lack any similarity to VNIRS-inferred TOC or P/B ratio trends. Iron in sediments usually originates from the terrestrial surroundings, and may be associated with the lithogenic or the organic fraction of the sediments. Sedimentary records of iron are usually difficult to interpret, due to chemical precipitation or the dissolution of iron in the sediments or at the sediment surface-water interface. Therefore, it is not possible to establish the role of iron in determining the colour of these particular lakes. Further studies based on monitoring data would improve our understanding of this relation.

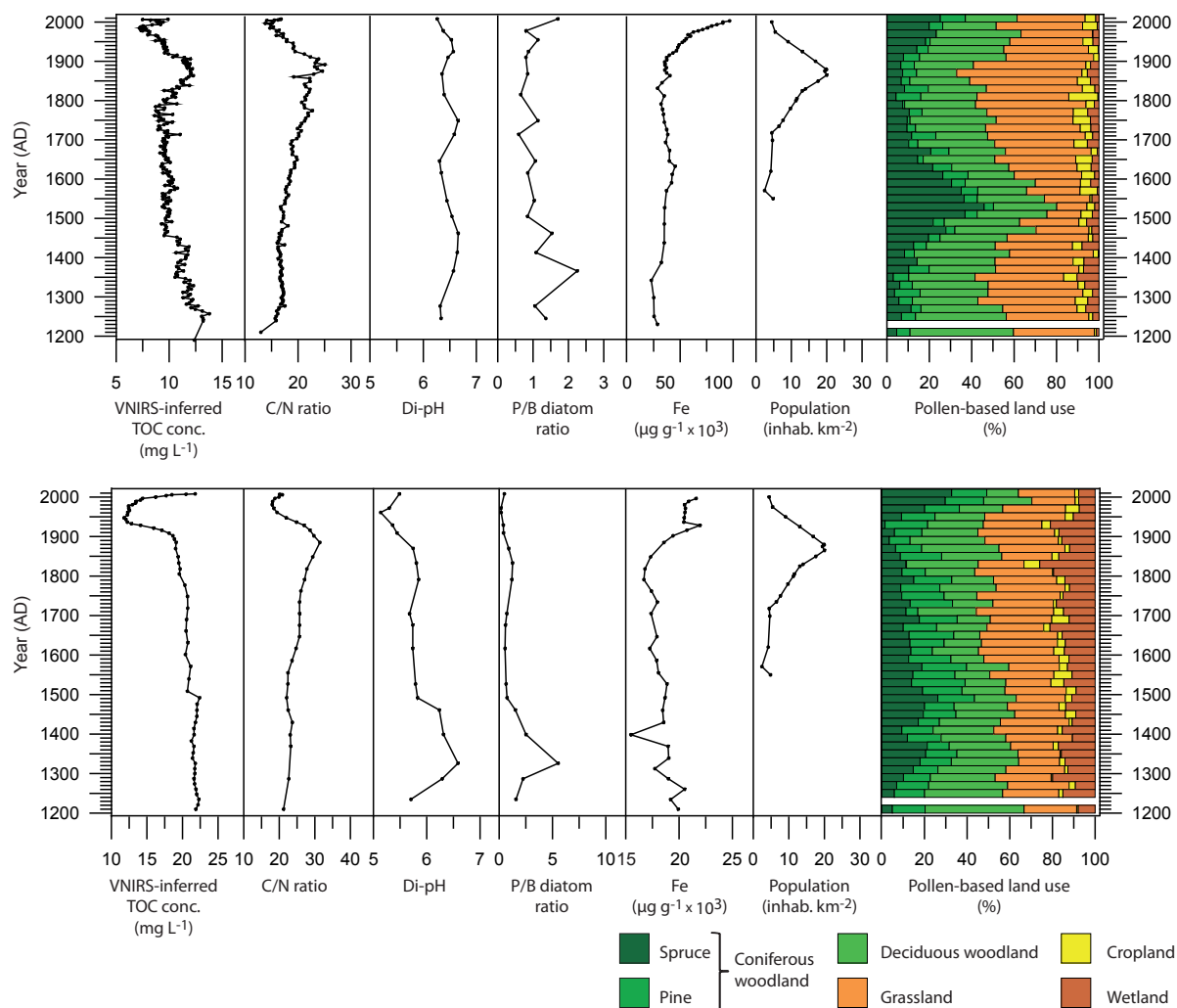


Figure 10. Records of VNIRS-inferred total organic carbon (TOC) concentration, atomic carbon:nitrogen (C/N) ratio, diatom-inferred pH (Di-pH), diatom planktonic:benthic (P/B) ratio, iron concentration (Fe) in lake-water, population density and pollen-based land use plotted against time for Åbodasjön (upper panel) and Lindhultsgöl (lower panel)

## 5.2 Identifying anthropogenic and climate impact on DOC

### 5.2.1 Land use

Land cover determines soil carbon stocks, and the presence of highly organic soils associated with boreal coniferous woodland and wetland is positively correlated to DOC concentrations in lakes in various regions (Sobek et al. 2007; Xenopoulos et al. 2003). The difference in land cover around the two lakes, with a much higher proportion of wetland in the Lindhultsgöl catchment area, occupying 10-20% of the RSAP during the past 800 years, resulted in a generally

higher TOC concentration in Lindhultsgöl than in Åbodasjön. This indicates the importance of wetland in long-term changes in TOC, and wetland loss has been shown to decrease the export of DOC to waters in the USA (Raymond et al. 2004). Elevated TOC concentration was recorded in Åbodasjön during the period of the highest wetland abundance, from AD 1200 to 1450. The loss of wetland around Lindhultsgöl from AD 1940 to the present day was found to be associated with very low TOC concentrations. However, the TOC concentration decreased shortly before the decrease in wetland. Long-term changes in coniferous woodland did not seem to affect the TOC concentration in either of the lakes, on the contrary, periods of distinct increases in coniferous woodland

surrounding Åbodasjön, beginning in the 1400s and in the 1900s, were associated with low and decreasing TOC content.

Changes in land use have significant effects on soil carbon stocks, thus influencing DOC export. Losses of soil carbon occur when grassland or native forest is converted to cropland, but increases when cropland is converted to grassland or forest with a natural succession (Ostle et al. 2009, Guo and Gifford 2002). Changes in DOC in response to land management have also been observed. DOC export has increased in the UK as a result of artificial drainage and heather burning of peatland (Worrall et al. 2003; Yallop et al. 2010). Agricultural land has commonly been associated with elevated DOC concentrations in lakes (Correll et al. 2001; McTiernan et al. 2001) as modern management practices lead to increased organic production and export as a result of organic and inorganic fertilization (McTiernan et al. 2001), and increased soil erosion (Correll et al. 2001). This effect has been observed even with small proportions of agriculture land (12%) within a boreal catchment area (Mattsson et al. 2005). The effects of forestry on DOC export appear to be most significant following felling (Neal et al. 2003).

During the past century, land use and land management practices have undergone major changes, and the long-term (decadal to centennial) impact on DOC in lake water has been less studied. In the present work, the combination of high-resolution (20-y) pollen analysis with detailed absolute chronologies and quantitative estimates of past vegetation allowed the impact of changes in land use on TOC to be estimated. Based on the reconstructions of land use, the continuous presence of agricultural indicators (Papers III and IV) together with evidence of catchment disturbance (Paper I) suggests that humans have been present within the RSAP throughout the past millennium. Furthermore, evidence was found of two periods of agricultural expansion with different intensities in both lake sediment records; one during AD 1260-1400 followed by partly abandoned landscape (Papers I, III and IV), and the other during AD 1550-1880, with increasing agrarian intensity from the end of the 1700s (Fig. 11). The long-term reconstructions revealed responses of TOC in the

lakes to changes in the intensity of agricultural land use. At Åbodasjön, a gradual decrease in TOC concentration was recorded concurrently with the first period of agrarian expansion (Fig. 11). Land-use reconstruction showed an increase in grassland and cropland, indicating a more open to semi-open landscape, probably associated with lower biomass production, which could have resulted in a reduction in DOC export (Rosén et al., 2011). During the second period of agrarian expansion an inverse response of TOC concentration was seen in Åbodasjön, showing a significant increase from AD 1800 peaking at AD 1850-1900, simultaneously with an increase in population density. Extensive catchment disturbance was indicated by the high C/N ratio, and open land, predominantly grassland, increased to more than 60% of the RSAP (Fig. 10). The increase in open land categories and associated biomass loss was probably counteracted by modern agricultural practices, such as increased organic production and erosion rates, leading to elevated DOC concentrations in lake water (McTiernan et al. 2001; Correll et al. 2001). This suggests that not only changes in land use, but also changes in land management practices (traditional vs. modern), influence long-term TOC concentration in lake water.

Anthropogenic disturbance and agrarian intensity were considerably lower around Lindhultsgöl than around Åbodasjön. The more rural setting and the high proportion of wetland made the catchment area less suitable for crop cultivation, and human impact was probably limited to animal grazing and small-scale forestry. However, between AD 1780-1880, agrarian intensity increased at Lindhultsgöl, resulting in the expansion of grassland and deciduous woodland at the expense of coniferous woodland, which were reflected by a gradual decrease in TOC concentration, possibly similar to the first agrarian expansion around Åbodasjön (AD 1260-1350). This may indicate the sensitivity of a relatively pristine environment to initial catchment disturbance, resulting in a reduction in biomass production and significant losses of soil organic carbon stocks upon conversion from native woodland to cropland (Ostle et al. 2009), rather than a general response to agrarian activity, as

suggested by the different responses of TOC to the two periods of agrarian expansion at Åbodasjön.

The transition to the previous century involved a range of new possible regional and catchment-scale forcing mechanisms affecting TOC concentration in the lakes (Fig. 11). Significant shifts in most of the sediment proxy records for both lakes, including a temporal coherent decrease in TOC concentration, indicate a transition towards contrasting geochemical states in the two lakes at AD 1900 (Papers I and III). The onset of industrialism at this time led to a decline in the rural population as urbanization began, indicated by reductions in open land use and related increases in woodland, especially the coniferous woodland surrounding both lakes. The decrease in agrarian intensity around Åbodasjön around AD 1900 led to a decrease in the transport of terrestrial organic matter from the catchment area, as reflected by a decrease in C/N ratios and lake-water TOC concentration, suggesting recovery from increased land-use intensity around AD 1880-1900. A similar coherent decline in C/N ratio was also seen in Lindhultsgöl, however, quantitative changes in the total area of woodland were only minor (although coniferous woodland was increasing), suggesting other processes with a stronger influence on the decreasing TOC concentration in this lake.

The increase in coniferous woodland around Åbodasjön from about AD 1900, and around Lindhultsgöl from AD 1940 (Fig. 12), represents the onset of commercial forestry in the catchment area, associated with management practices such as clear-cut felling, ditching and fertilizing to improve forest productivity (Antikainen et al. 2008). Clear-cut felling is known to significantly affect DOC levels in stream water in boreal forests (Lepistö et al. 2008; Laudon et al. 2009), and artificial drainage and ditching of woodland and wetland has also been coupled to changes in DOC export. However, the effects of ditching and drainage of woodland, wetland and cropland are complex. Some have reported increases in surface-water DOC (Kalén 2007; Ecke 2009; Dalzell et al. 2011) and others decreases (Åström et al. 2001; McTiernan et al. 2001; Rantakari et al. 2010).

Both catchment areas have been subject to clear-cut felling, resulting in distinct increases in clear-

cuts between AD 1946 and 2005 (Fig. 9), together with different degrees of artificial drainage and ditching (Fig. 11), both of which may have influenced DOC export to the lakes. However, the temporal and spatial uncertainties of these operations make it difficult to evaluate the effects on TOC concentrations in either of the lakes. Furthermore, the response of DOC to forestry management practices may be variable or exhibit a short-term effect (Neal et al. 2003; Laudon et al. 2009), and it may be difficult to detect long-term changes in lake-water DOC concentration. However, a major ditching operation along the southern inlet to Åbodasjön in the 1920s led to significant increases in lithogenic element concentrations (Paper I), and may have led to an increase in the supply of soil-derived DOC from surrounding crop cultivation, resulting in the observed stabilization of inferred TOC concentration. Crop cultivation along the inlet was abandoned in the 1950s, reflected by decreases in lithogenic material and TOC concentration in the lake water. Apart from this well-documented ditching operation, there is no compelling evidence of the effects of ditching or forestry on TOC concentrations in the two lakes studied, and the major variations in TOC were, therefore, probably driven by other processes.

### 5.2.2 Sulphur deposition

Elevated sulphur deposition in the form of  $\text{SO}_4^{2-}$  has been shown to reduce the solubility and export of soil DOC as a result of changes in pH and ionic strength. Conversely, a reduction in sulphur deposition leads to increased solubility and leaching of DOC into lakes and running water (Evans et al. 2006; Monteith et al. 2007). Monteith et al. (2007) studied 522 lakes and streams in North America and Northern Europe and concluded that increasing trends in DOC concentration between AD 1990 and 2004 were caused by changes in sulphur deposition. Several studies support this hypothesis, suggesting that the observed increase in DOC is the result of a common regional forcing mechanism. However, increasing DOC trends have also been observed in areas not subject to atmospheric sulphur deposition and acidification (Freeman et al. 2001;

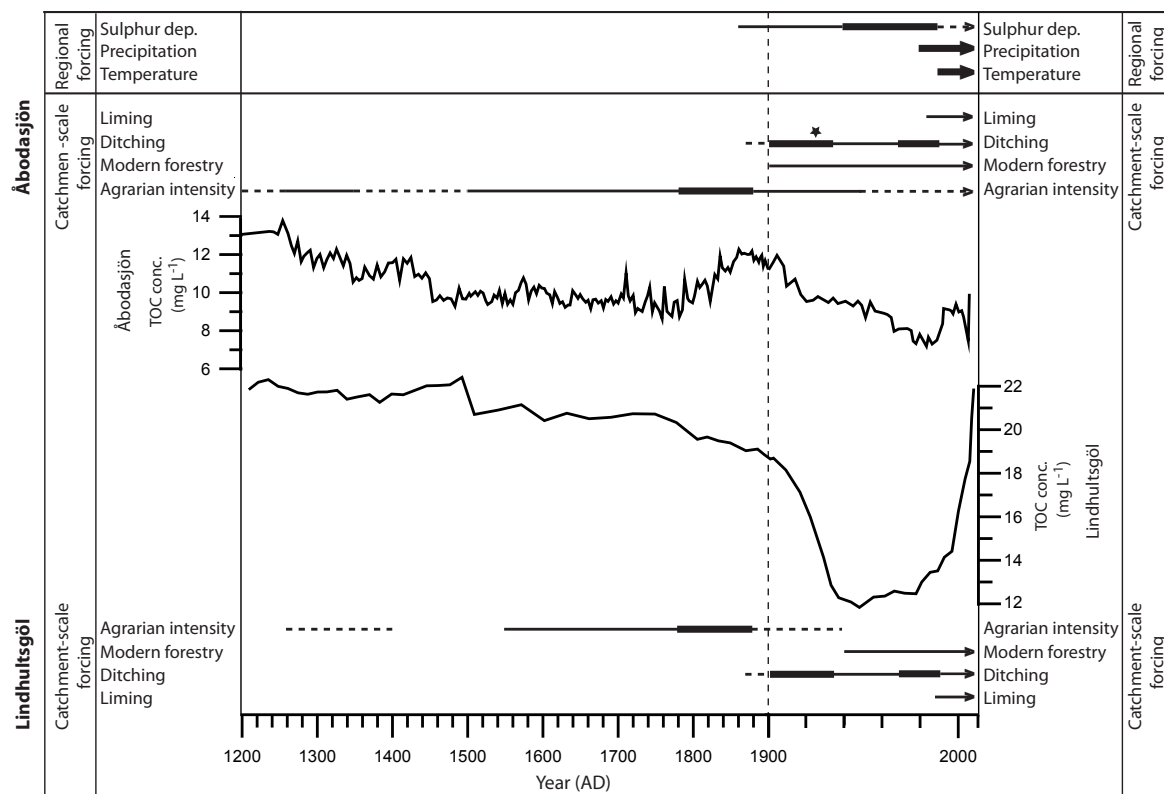


Figure 11. Records of VNIRS-inferred total organic carbon (TOC) concentration in Åbodasjön (upper graph) and Lindhultsgöl (lower graph), with possible regional and catchment-scale drivers of TOC changes. Regional drivers include sulphur deposition, precipitation and temperature (Fig. 12). Local drivers include site-specific liming history, regional trends in ditching (Hånell 2009) and changes in land use inferred from pollen data (Fig. 10) and historical accounts (agrarian intensity and modern forestry). Horizontal lines represent periods of activity, heavy lines represent periods of increased or high intensity, and dashed lines represent periods of decreased or low intensity. Arrows indicate ongoing processes. The asterisk indicates major drainage work undertaken at the southern inlet of Åbodasjön in AD 1922. The vertical dashed line indicates AD 1900. Note the different scale for the period AD 1900–2010

Evans et al. 2005). Monteith et al. (2007) found that declining concentrations of sea salt ( $\text{Cl}^-$ ), which has similar effects on soil solubility to sulphur deposition, were responsible for increases in DOC concentrations at some coastal sites where sulphur deposition was low.

The effect of a decrease in sulphur deposition on DOC leaching is commonly related to catchment area sensitivity to acidification (Monteith et al. 2007). Also soil properties are important and DOC trends in organic soil horizons usually match increases in surface waters resulting from changes in acidification (Oulehle and Hruška 2009; Evans et al. 2012). Less consistent results have been reported for mineral soil horizons exhibiting decreasing DOC concentrations at some sites in Sweden (Löfgren and Zetterberg 2011) and Norway (Wu et al. 2010). Löfgren and Zetterberg (2011) suggested that processes in the riparian zones and peatland,

rather than in dry uphill soils, govern the variation in stream-water DOC, and that organic-rich soils with connectivity to the lakes were responsible for the changes. Other experiments have indicated that soil-water colour increases as a result of changes in the composition of dissolved organic matter due to reduced acid deposition (Ekström et al. 2011, SanClements et al. 2012), leading to significant coherent changes in lake-water colour.

Sulphur emissions started to increase at the onset of industrialisation at the end of the 1800s (Fig.12), which led to the acidification of soil and surface water over large parts of Europe due to the acid deposition. Sulphur emissions increased significantly in the 1940s, peaking around AD 1980–1995 (Schöpp et al. 2003). This was followed by progressively decreasing deposition and widespread recovery of lakes and streams from acidification 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 concentration seen in the two lakes studied in this work. Moreover, sulphur deposition prior to the 1990s is generally inversely correlated to the inferred variations in TOC in the two lakes, strongly suggesting that sulphur deposition is the main forcing mechanism of TOC in both lakes during the past century.

The initial decline in TOC in Åbodasjön around AD 1900 to 1920 was rapid, and a second significant decline in TOC was recorded in the 1950s, simultaneously with the highest increase in sulphur deposition. This was followed by a decrease in pH, indicating increased acidity in response to the strong mineral acids associated with elevated sulphur emissions. Changes in land use concurrent with changes in sulphur deposition may also have contributed to the observed changes in lake-water TOC concentration during the 1900s. The decrease in TOC concentrations around AD 1900 was probably also a result of the conversion from open land to woodland, resulting in more stable soils and less export of DOC to the lake. Also, the suggested increase in soil-derived DOC input to the lake, associated with the agrarian activities along the southern inlet between AD 1920 and the 1950s, may have reduced any sulphur-deposition-driven decrease in TOC concentration in the lake. The decline in sulphur deposition starting in AD 1990 was not accompanied by any changes in pH indicative of recovery from acidification, an effect also observed at other sites (SanClements et al. 2012), and may indicate a time lag or suppressed recovery from acidification due to organic acids replacing mineral acids (Skjelkvåle et al. 2005).

The rapid decline in TOC concentration between AD 1900 and 1930 in Lindhultsgöl corresponded to the onset of sulphur deposition, and resulted in exceptionally low values, below those during the past 800 years. No changes in land use during this period can explain the significant decline in TOC concentration, suggesting that the system of the already acidic organic soils was highly sensitive to sulphur deposition. The lake showed acidification, as reflected by the decreasing inferred pH. Lindhultsgöl had been exhibiting stable, low values of TOC since the 1930s, and the largest

increase in sulphur emission, in AD 1950, did not influence the concentration. This may be explained by the high proportion of wetland and already acidified organic soils in the catchment area. Experiments have shown that decreases in DOC concentration and leaching stabilized at a certain pH, despite continuing acid treatment (Evans et al. 2012). The distinct increases in TOC concentration in Lindhultsgöl in the 1990s were accompanied by pH recovery from the most acidic conditions.

These results clearly indicate that decreasing sulphur deposition was the main driver of the observed increases in TOC concentrations and colour from the 1990s in the two lakes studied, in agreement with the prevailing theory that declining acidification is the main driver of the brownification of water. Site-specific catchment characteristics are of great importance regarding the effects of sulphur deposition on the variation of DOC in lake water, as exemplified by the sediment-based records from these two lakes, which exhibited clear differences in the magnitude of TOC response. The effect of decreasing sulphur deposition on soil solubility is greatest in acid-sensitive regions (Monteith et al. 2007) and organic soil horizons (Oulehle and Hruška 2009; Evans et al. 2012). Thus, the acid-sensitive locations and the abundance of conifer woodland and wetland may play an important role in the observed increases in TOC at the sites studied in this work, in comparison to other southern Swedish lakes, showing small or no changes in DOC concentration during the past three decades. It is also important to note that Åbodasjön and Lindhultsgöl have been treated for acidification by yearly liming of the surface waters and surrounding wetlands from AD 1984 and 1993, respectively.

### 5.2.3 Climate

The effects of climate on DOC concentrations in lake water are driven by a variety of different processes, including temperature-driven soil organic productivity and decomposition, as well as precipitation-driven runoff and groundwater fluctuations, which affect soil DOC export (e.g. Sobek et al. 2007). However, any positive effect of increased temperatures on soil DOC production may be counteracted by accelerated

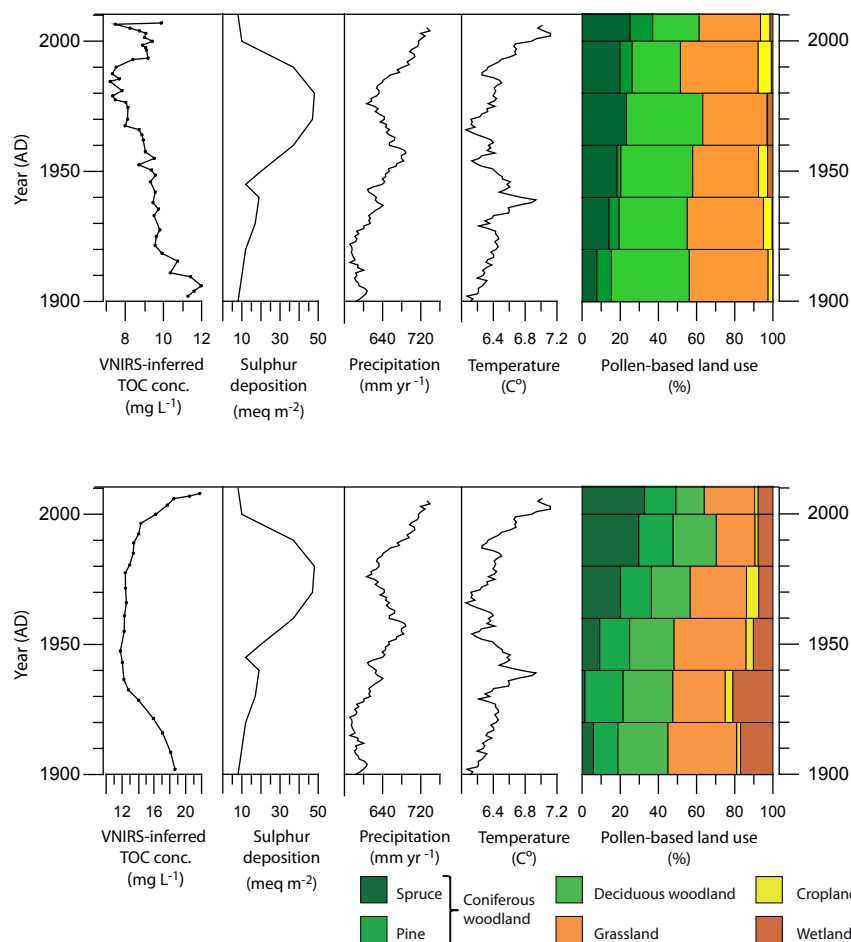


Figure 12. Records of VNIRS-inferred lake-water total organic carbon (TOC) concentration and pollen-based land use since AD 1900 from Åbodasjön (upper panel) and Lindhultsgöl (lower panel) plotted together with sulphur deposition (modified from the Swedish Environmental Research Institute MAGIC-model) and climate data from Växjö (annual precipitation and mean annual temperature expressed as 15-year running averages)

evapotranspiration and respiration of soil organic matter, leading to a loss of leachable DOC through emissions of CO<sub>2</sub> and CH<sub>4</sub> (Fenner et al. 2007). In addition, increasing temperature could also intensify aquatic respiration and the mineralization of organic carbon in lake sediments, increasing emission rates of CO<sub>2</sub> and CH<sub>4</sub> (Gudasz et al. 2010). Together with a longer season of biological consumption due to longer ice-free seasons, this will result in a reduction of DOC concentrations and organic sedimentation in lakes. Thus, the influence of temperature on temporal variations in DOC concentrations at individual sites is likely to be very different.

Results concerning the effects of changes in precipitation are inconsistent. A general relationship between DOC concentration and colour, and precipitation and discharge during the past three decades has been reported in several studies (Hongve et al. 2004; Dillon and Molot 2005; Sarkkola et al. 2009; Zhang et al. 2010). This is in contrast to the negative relationship found between runoff and

DOC concentration in different continents (Sobek et al. 2007). Short-term rainfall events may lead to a dilution effect, lowering the DOC concentration (Clark et al. 2007), or an increase in DOC following dry seasons or periods during which leachable organic matter is stored in the soil (Tipping et al. 1999). This implies that changes in DOC export resulting from long-term changes in precipitation will depend on the organic production, and if there is no increase in productivity, increased precipitation and discharge will lead to a decrease in DOC in the long term.

Changes in water table levels and hydrology may influence DOC production and export, and increased leaching of DOC has been observed as a rise in water table or increased surface water flow due to contact with upper forest floors rich in organic material (Hongve et al. 2004). Lowering of water tables and reductions in stream flow as a result of drier conditions have been shown to decrease lake-water colour and DOC concentrations in Canada, due to the reduced export of DOC

(Schindler et al. 1996). Furthermore, a decrease in discharge will increase the water residence time in lakes, resulting in a greater amount of DOC being removed from the lake through bacterial mineralization, sedimentation and photooxidation. This is especially important in lakes with an intermediate water residence time (a few years), which are more sensitive to changes in water balance with respect to lake-water colour and DOC concentrations than lakes with rapid water exchange, where the removal of DOC is insignificant in comparison to the DOC influx, and than lakes with a long residence time (>5 years), where in-lake processes are less influenced by changes in discharge. In contrast, periods of drought have been associated with an increase in DOC concentration in two UK river catchment areas (Worrall and Burt, 2004b), as drought may lower the water table and increase the production of DOC under aerated conditions. Increased DOC concentrations in Estonian rivers were explained by increasing trends in hydrological drought caused by a man-made drainage system, and extremely low water tables (Pärn and Mander 2012). However, evidence of the role of water table drawdown is contradictory, and aerobic conditions may instead favour the production of CO<sub>2</sub> over DOC (Kalbitz et al. 2000). There may also be a reduction in microbial decomposition and consumption during dry conditions (Scott et al. 1998).

Meteorological data from AD 1860 to the present for the nearby city of Växjö (Swedish Meteorological and Hydrological Institute) (Fig. 12), showed an increase in precipitation since AD 1980, and in temperature since AD 1990. Hence, climate change may have contributed to the observed and reconstructed increases in TOC concentration in the lakes over recent decades. However, no coherent variations in TOC were detected with variations in temperature and precipitation at either of the lakes on the centennial scale (AD 1860-present). Prior to the period covered by monitoring data, information is available on long-term changes in temperature, based mainly on large-scale reconstructions. Regional variability may deviate from the general patterns, making it difficult to establish any long-term effect on variations of TOC concentration in the lakes. During the

previous millennium, there was a period of high temperatures around AD 1000-1100, and minimum temperatures around AD 1600 in the Northern hemisphere, compared to the average temperature for AD 1961-1990 (Moberg et al. 2005). No effects of climate change on long-term TOC concentrations in the two lakes were observed in this study, probably due to the lack of sufficiently detailed regional climate records, but such an effect cannot be ruled out.

### 5.3 Future developments

Apart from the many ecological consequences, increasing DOC concentration and colour also have implications for society. Over half of the drinking water supply in Sweden is derived from surface water, and in recent years water treatment plants have experienced problems associated with an increased amount of DOC, in particular humic substances. If brownification continues, drinking water quality will be threatened, and major investments will be necessary in water treatment. It may even be necessary to change from surface water to ground water to provide potable drinking water. Another consequence of increasing DOC concentrations is that changes have been necessary in national liming programmes as the large amounts of organic acids released with increased DOC during recent decades have now, to some extent, replaced the strong acid associated with sulphur deposition, delaying recovery from acidification (Erlandsson et al. 2010). Tourism is one of the fastest growing industries in Sweden today, and recreational activities such as swimming and fishing will be threatened by browner water in lakes, rivers and coastal areas. Therefore, predictions of future DOC concentrations and the colour of surface waters are of considerable value.

Based on the results presented in this thesis, decreasing sulphur deposition is the most plausible driver of increasing TOC and colour during the past 30 years in the two lakes studied. This suggests recovery from the phase of maximum sulphur deposition, which resulted in exceptionally low TOC concentrations during AD 1930-90, and a return to pre-depositional conditions and higher TOC concentrations. With the return of sulphur



emission levels to background levels (Schöpp et al. 2003), it is likely that TOC concentrations will level off. The timing is dependent on the quantity of organic carbon stored in catchment soils due to previous suppression of DOC leaching, and the composition of DOC. However, TOC concentrations have already reached historically “normal” levels, and other drivers/factors will become progressively more important for TOC concentration in the future.

The long-term records also demonstrate that TOC concentrations in lake water were influenced by changes in wetland abundance, agricultural practices and general land use during the past 800 years. Agricultural and forestry management practices have changed considerably over the past century, potentially increasing DOC production and export associated with fertilization, clear-cut felling and ditching. However, the strong effects of sulphur deposition on inferred variations of TOC in lake water over the past century may have masked any effects of modern land use. The interaction between contemporary processes potentially causing DOC changes during the past century makes it difficult to discern the impact of these changes in land use and land management.

In addition, changes in climate may affect lake-water DOC concentration in a number of ways. Future climate predictions for Northern Europe include higher seasonal amounts and intensity of precipitation, and increasing mean annual air temperatures (Alcamo et al. 2002). It has been hypothesized that these changes will result in continued increases in DOC in boreal areas, mediated by increased terrestrial vegetation (Larsen et al. 2010). Increasing trends in annual precipitation from about AD 1980 and mean annual temperature from about AD 1990 may have contributed to the observed and reconstructed increases in TOC concentrations over recent decades. On the other hand, the long-term records provide no evidence of climate forcing TOC variations.

It is difficult to distinguish the effects of individual mechanisms in our efforts to identify the main driver or drivers of increasing DOC. It is also likely that there are interactions between different drivers, and there will be spatial differences in future DOC variations depending on catchment

characteristics and anthropogenic activities. The long-term TOC records for the two lakes studied showed unusually low TOC concentrations during maximum sulphur deposition, and pre-depositional TOC concentrations have now been reached. Considering the past variability in TOC concentrations in Åbodasjön, with periods of higher than present TOC concentrations, it is likely that such concentrations may be reached again in this particular environment. This demonstrates the importance of applying a long-term perspective in order to differentiate between causal relationships and to predict future trends for environmental planning and management.

## **6. Conclusions**

The conclusions of the work presented here can be summarized as follows.

- The long-term VNIRS reconstructions revealed that TOC concentrations in the two lakes were generally high prior to AD 1900, with similar or higher concentrations than at present. Significant changes took place during the past century, and unusually low TOC concentrations were recorded in AD 1930-1990, followed by a recent increase. There are indications that colour occasionally deviates from long-term TOC variations in the lakes, due to variations in the composition of organic matter, which is generally regulated by land use and land management.

- It was found that the long-term TOC in lake water is mainly influenced by changes in the intensity of agricultural land use and the abundance of wetland. Site-specific catchment characteristics are of great importance in determining variations in lake-water TOC concentrations in response to land-use dynamics, as exemplified by the sediment-based records from the two lakes studied, showing clear historical differences in agricultural activity in the catchment areas.

- Based on the results presented here, it can be concluded that sulphur deposition has been the main driver of TOC trends in both lakes during the past century, and that increases in TOC concentration and colour in the lakes during the past thirty years were driven mainly by an increase in soil solubility and the export of DOC due to

declining atmospheric sulphur emissions.

- The results suggests that the two lakes are recovering towards their natural high-DOC pre-depositional states, and TOC concentrations have now reached historically “normal” levels in the two lakes, and are likely to level out. Given that the effects of sulphate deposition now subside, the future development of DOC concentrations in lakes will probably be driven by previously suppressed or masked effects of climate, land use and land management during the past century.

- This study clearly illustrates the importance of adopting a long-term perspective, on time-scales beyond monitoring series, in our attempts to understand lake-water DOC dynamics, and to help predict the future development of lake-water quality in boreal environments. It can be argued that palaeolimnological studies should be carried out when planning, for example, new water plants and drinking water supplies based on lake surface water, to avoid costly investments required at a later date to deal with changing lake-water quality that could have been foreseen with the aid of historical reconstructions.

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## **Svensk sammanfattning**

De senaste ca trettio åren har man observerat en kraftig ökning av löst organiskt kol i sjöar, vattendrag och kustnära vatten i olika delar av det norra halvklotet, bl.a. i delar av Nordamerika, Storbritannien, Baltikum, centrala Europa och Skandinavien. I Sverige har ökningen främst varit koncentrerad till de södra delarna av landet. Löst organiskt kol i sjöar belägna i den boreala regionen består vanligtvis främst av nedbrutet organiskt material från växter och organismer som löses i vatten och lakas ut ur den omgivande marken. Av det lösta organiska kolet utgörs vanligtvis 50-75 % av färgade humuspartiklar som ger sjöar en karakteristisk brun färg. Ökningen av löst organiskt kol i sjöar har således oftast följts av en kraftig färgning av vattnen, s.k. brunifiering.

Brunifieringen av våra sjöar medför en rad konsekvenser för de akvatiska ekosystemen, vattnet blir mörkare vilket leder till sämre ljusförhållanden och högre vattentemperaturer som påverkar den akvatiska faunan och florans betydande förändringar i den biologiska mångfalden. Sjövattnen har även en avgörande betydelse för Sveriges vattenförsörjning då ca hälften av vårt dricksvatten tas från ytvattentäkter. Ökningen av humus leder till en högre bakteriell tillväxt och till högre halter av miljöfarliga ämnen med risk för vår hälsa. Den ökade humushalten gör det svårare att framställa dricksvatten av god kvalitet och vattenverken kan bli tvingade att investera i ny reningsteknik, eller eventuellt byta vattentäkt med stora ekonomiska konsekvenser. Vattenkvaliteten i sjöar och vattendrag är även viktiga för rekreation i form av fiske och bad, brunifieringen kan medföra färre besökare och intäkter från turism i de värst drabbade områdena.

Man tror att den ökade brunifieringen kan bero

på förändringar i nedfall av surt regn, klimat och markanvändning. Mängden löst organiskt kol som lakas ut från marken till sjövattnet styrs av faktorer som påverkar organisk produktion och nedbrytning, löslighet och transport. Nedfall av surt regn påverkar lösligheten av organiskt kol och försurning av marken leder till ett lägre pH som ger lägre löslighet. De senaste trettio åren har nedfallet av surt regn minskat dramatiskt och bidragit till en ökad pH och återhämtning från sura markförhållanden, vilket kan leda till en högre löslighet av organiskt kol och en ökad urlakning. Om detta är orsaken till den ökande brunifieringen så innebär det att mark- och vattensystemen återhämtar sig från en tidigare miljöpåverkan till mer normala förhållanden.

Ett varmare klimat och mer nederbörd kan i sin tur öka den organiska produktionen och transporten av löst organiskt kol till sjöar. Men i själva verket är det mer komplicerat än så, högre temperatur kan istället leda till en ökad mängd av organiskt kol till atmosfären i form av koldioxid och metan än löst organiskt kol till sjöarna. Ökad nederbörd kan på lång sikt leda till en minskning av löst organiskt kol i sjöar genom utspädning och en urlakning av markerna. Om dagens klimatförändringar är orsaken bakom den ökade brunifieringen kan detta potentiellt leda till fortsatta ökningar med stora konsekvenser på sjöarnas funktion.

Markanvändning har genomgått stora förändringar det senaste århundradet, från traditionellt småskaligt jordbruk till dagens kommersiella moderna skogs- och jordbruk. Förändringar i markanvändning resulterar i förändringar i markens lagrade kol, t.ex. skog till åkermark leder till en förlust i kol. Dränering, avverkning och gödsling av skogs- och jordbruksmark påverkar också urlakning och transport av löst organiskt kol till sjöar. Det är viktigt att förstå hur människan kan ha bidragit till den ökande brunifieringen, för att möjliggöra en hållbar utveckling i skogs- och jordbruk med så lite påverkan som möjligt på halten organiskt kol i våra sjöar.

De potentiella orsakerna till den ökade brunifieringen är processer som pågått under en längre tid än vad som har kunnat studeras då det saknas, eller finns bara begränsad mätdata från tiden före 1960. För att kunna ta reda på om den ökade brunifieringen är en produkt av mänsklig påverkan

eller är en del av en naturlig utveckling behövs ett längre tidsperspektiv studeras. Paleolimnologi och analyser av sjösediment kan ge unik information om den historiska miljöutvecklingen i en sjö och dess omgivning. Sjösediment består av en blandning av material som kommer från sjöns omgivning, atmosfäriska föroreningar och material som bildats i sjön. Genom att analysera sammansättningen av det material som kontinuerligt avlagras på sjöns botten i en kronologisk följd får man information om tiden innan industrialismen och modernt skogs- och jordbruk.

I den här avhandlingen analyseras hur halten löst organiskt kol (TOC) i två sjöar (Åbodasjön och Lindhultsgöl) på Sydsvenska höglandet under 800 år har påverkats av förändringar i nedfall av surt regn, klimat och markanvändning för att försöka förstå mekanismerna bakom dagens ökade brunifiering. Flera olika paleolimnologiska metoder användes såsom radiometrisk datering av sedimenten, närinfraröd spektroskopi (VNIRS) för rekonstruktion av variationer i TOC-halt, analys av diatoméer (kiselalger) för rekonstruktioner av pH, pollenanalys och modeller för att kvantifiera utbredningen av olika typer av markanvändning under olika tidsperioder, samt geokemiska analyser av organiskt och oorganiskt innehåll för information om dess ursprung, näringskretslopp och perioder av markerosion. Detta jämfördes med dokumentation om befolkningsutvecklingen i området, surt nedfall, samt förändringar i temperatur och nederbörd.

Resultaten från undersökningarna visar att sjöarna har varit påverkade av mänsklig aktivitet av olika intensitet i sjöarnas närområde de senaste 1000 åren. Det finns bevis för två perioder av ökad aktivitet och jordbruksutbredning kring Åbodasjön och Lindhultsgöl. En period mellan 1260-1400 följd av delvis övergivna jordbruksmarker som kan härröras till den effekt digerdödens utbrott 1350 tros ha haft på Sveriges befolkning. En återetablering registrerade på 1500-talet, med kontinuerligt ökad utbredning av jordbruksmark i takt med en ökad befolkning till ett maximum på 1880-talet. Därefter avbefolkades bygden och modern markanvändning med ett industrialiserat skogs- och jordbruk introducerades. Idag dominerar bygden av skogsbruk med inriktning främst på gran, och befolkningen har minskat till 1700-talets låga



nivåer.

De historiska rekonstruktionerna visar att TOC-halterna var höga fram till 1900-talet, och variationer kunde kopplas till förändringar i jordbruksutbredning och intensitet samt förekomsten av torvmark som är rika på humuspartiklar. Det är viktigt att poängtera att halten humus i sjövattnet kan variera och resultaten från Åbodasjön tyder på att en period av ökad TOC-halt p.g.a. ökad intensitet i jordbruksutbredning förmodligen inte gav upphov till någon större förändring i vattenfärg då TOC från jordbruk har en sammansättning av främst ofärgade partiklar. De historiska variationerna i TOC-halten kan alltså inte alltid tolkas som förändringar i vattnets färg.

I början av 1900-talet minskade TOC-halten i båda sjöarna, mest kraftigt i Lindhultsgöl, för att nå låga eller ovanligt låga nivåer ur ett historiskt tidsperspektiv under 1930-1990, därefter ökade nivåerna till en mer "normal" nivå i linje med pre-industriella koncentrationer. Dessa förändringar under 1900-talet kan kopplas till surt nedfall som startade kring 1900 för att nå de högsta nivåerna 1980-1995 följt av en kraftig minskning. Det finns inga förändringar i markanvändning eller klimat som kan förklara de ovanligt låga nivåerna av TOC-halten i sjöarna under 1900-talet, inte heller den hastiga ökningen som inträffade på 1990-talet. Minskningarna i surt nedfall och en ökad urlakning av ackumulerat organiskt kol som ej lösts ut i vatten p.g.a. tidigare sura markförhållanden är den troligaste förklaringen till den ökade brunifieringen de senaste trettio åren. Detta innebär att brunifieringen är en återhämtningsprocess och att halterna av TOC kommer att plana ut. Hur länge TOC-halten och färgen kommer att öka är osäkert, då det beror på hur mycket organiskt material som har lagrats i markerna under försurningen, och hur mycket av det som består av humuspartiklar. Det är också osäkert om andra mekanismer såsom klimatförändringar och/eller olika metoder inom modernt skogs- och jordbruk, tidigare varit underordnad den starka effekt surt nedfall hade på lösligheten av TOC, och nu kan komma att påverka den framtida utvecklingen av TOC-halten i sjöar på det sydsvenska höglandet.

Med den här studien vill jag illustrera att ett

långtidsperspektiv, bortom limnologisk mätdata kan hjälpa till att bidra med viktig bakgrundsinformation som kan hjälpa till att förutsäga den troliga framtida utvecklingen av sjöar. Den här studien visar också hur olika typer av vegetation och markanvändning i sjöns omgivning påverkar mottagligheten för en miljörelaterad stress och det är viktigt att ta de lokala förhållandena i beaktning. Man kan argumentera för att paleolimnologiska undersökningar bör utföras vid planering av större investeringar, såsom ny vattentäkt för dricksvattensförsörjning, för att analysera sjöns naturliga variationer kontra känslighet för mänsklig påverkan för att undvika problem associerade med förändringar i kvalitén av sjövattnet som kunde ha förutspåtts.

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