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## Implications of land use on the carbon cycle - Impacts of long-term human activities on terrestrial organic matter input to aquatic ecosystems in southern Sweden

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# Implications of land use on the carbon cycle

Impacts of long-term human activities on terrestrial organic matter input to aquatic ecosystems in southern Sweden

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BINGJIE YANG

QUATERNARY SCIENCES | DEPARTMENT OF GEOLOGY | LUND UNIVERSITY 2020







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# Implications of land use on the carbon cycle

Impacts of long-term human activities on terrestrial organic matter input to aquatic ecosystems in southern Sweden

Bingjie Yang



**LUND**  
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Department of Geology

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Back cover: The cabin where the author stayed during the fieldwork in Skogaryd.

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Implications of land use on the carbon cycle - Impacts of long-term human activities on terrestrial organic matter input to aquatic ecosystems in southern Sweden		
Abstract:  <p>Human activities have a huge impact on the carbon cycle by modifying the landscape. Vegetation cover changes attributed to forest clearance and the expansion of agricultural land increases soil erosion, organic matter and other nutrient delivery from terrestrial to aquatic ecosystems. Long-term records of environmental variation at centennial to millennial time scales are essential for the assessment of ecosystem dynamics in response to early and recent anthropogenic disturbances. Such records can be obtained from well-dated lake sediments, which consist of chronologically deposited materials originating from both within the water body (e.g. organic matter from macrophytes and planktonic algae) and the catchment area (e.g. soil organic matter, plant detritus, pollen and minerogenic material). Multi-proxy analysis of continuous sediment sequences allows us to reconstruct the palaeoenvironment and subsequently investigate aquatic ecosystem response to human activities.</p> <p>In this thesis, we carried out multi-proxy analyses to four sediment sequences from different environmental settings in southern Sweden to explore the variability in organic matter transport between terrestrial and aquatic environments in response to long-term land-use changes. The pollen-based Landscape Reconstruction Algorithm was applied to quantitatively reconstruct the catchment-scale landscape dynamics. Lignin phenols (biomarker) were used to trace the terrestrial organic matter preserved in the sediments. Bulk geochemistry including TOC, C/N ratio and BSi was applied for a general assessment of the proportions of terrestrial and aquatic organic matter deposition and estimation of aquatic production. Ti within the XRF data set was used to assess soil erosion.</p> <p>The study on two sediment sequences from a small forest lake (Lake Skottenesjön) in southwestern Sweden shows that the terrestrial organic matter delivery is sensitive to local land-use variations in the catchment in the past 1000 years. Elevated soil erosion and increased terrestrial organic matter deposition were recorded during intensive wood harvest in the 18th and 19th centuries. No significant change in terrestrial organic matter delivery was observed during the farmland expansion between the 12th and mid-14th century. Export of terrestrial organic and minerogenic matter to the lake was much higher during the period of modern forestry in the 20th century than the period of minor forest disturbance in the 11th century.</p> <p>A similar multi-proxy study was conducted on the sediment sequences from a large lake (Lake Storsjön) and a fjord-like inlet of Baltic Sea (Gåsfjärden) on the east coast of Sweden. The two sites are within the same catchment and connected by a river system. The results show that the composition of lignin-derived organic matter deposited in Gåsfjärden is less sensitive to the variation of vegetation cover than in Storsjön, which is likely due to the sortation and alteration on the terrestrial organic matter during the transportation from the inland to the coast. The concentration of lignin is much lower in Gåsfjärden than Storsjön as the organic matter deposition is dominated by aquatic material in Gåsfjärden. Furthermore, Gåsfjärden receives less degraded terrestrial organic matter, likely because the organic matter liable to degradation has been lost during transportation to the sea.</p> <p>This study highlights the potential of the combined use of lignin phenols and pollen-based quantitative land-cover reconstructions for investigating long-term changes in terrestrial organic matter input to aquatic ecosystems. The findings of the study provide a better understanding of human impacts on organic carbon cycling from a long-term perspective, which is fundamental for the development of environmental management strategies.</p>		
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博学之，审问之，慎思之，明辨之，笃行之  
——孔子

*“Study extensively, Enquire accurately, Reflect carefully,  
Discriminate clearly, Practice earnestly”*

—— Confucius



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# List of papers

This thesis is based on the following three papers:

## Paper I

Yang, B., Nielsen, A.B., Ljung, K., Fahlgren, E., Hormes, A., Hammarlund, D., 2020. Quantitative landscape reconstruction and erosion history during the past 1,100 years in the Skogaryd Research Catchment, southern Sweden. *Vegetation History and Archaeobotany*, 29, 657-670, 10.1007/s00334-020-00770-6. *Available in Open Access*.

## Paper II

Yang, B., Ljung, K., Nielsen, A.B., Fahlgren, A., Hammarlund, D. Impacts of long-term land-use dynamics on terrestrial organic matter input to lakes based on lignin phenols in sediment records from a small Swedish forest lake. *In revision in Science of the Total Environment*.

## Paper III

Yang, B., Ljung, K., Ning, W., Filipsson H.L., Nielsen, A.B. Variations of terrestrial organic matter deposition in a coastal area of the Baltic Sea in response to human impacts over the last 500 years. *Manuscript*.

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# 1 Introduction

Human activities have impacts on the carbon cycle in various ways. In addition to the combustion of fossil fuels and biomass, changes in land use have impacts on the carbon cycle and potentially large impacts on the climate. The change in land cover associated with human activities is one of the major factors controlling the terrestrial organic carbon exports to aquatic ecosystems (e.g. Anderson et al., 2013; Abbas et al., 2020, Lehnhart-Barnett and Waldron, 2020). Soil plays a critical role in the global carbon cycle, as it is the largest storage of organic carbon (over  $1.55 \times 10^{12}$  Mg C) in terrestrial ecosystems (Batjes, 1996; Lal, 2004; Abbas et al., 2020). Conversion of natural vegetation cover into agricultural land results in an elevation in soil erosion that has severe impacts on losses of terrestrial organic carbon (Lal, 2003). The enhanced soil erosion leads to increased input of the terrestrial organic matter, such as plant litter and soil carbon, to aquatic ecosystems (Lehnhart-Barnett and Waldron, 2020). Apart from the land-derived organic matter, increased catchment erosion also contributes to elevated delivery of nutrients to water bodies (Quinton et al., 2010), which can affect aquatic primary production and consequently alter the autochthonous organic matter deposition (e.g. Neumann et al., 2002; Routh et al., 2004; Köster et al., 2005; Li et al., 2008; Das et al., 2009; Haas et al., 2019). The decomposition of elevated organic matter in the bottom water without proper ventilation can result in the development of hypoxia ( $O_2 < 2$  mg/l) in the deep basins (Carstensen et al., 2014), which subsequently has impacts on the aquatic ecosystems.

Increased concentrations of organic carbon in surface waters of lakes, which could be related to human activities, have been observed over the past three decades across parts of Europe and North America (Worrall et al., 2004; Evans et al., 2005; Monteith et al., 2007; Rosén et al., 2011). However, these records of changes in the organic carbon concentrations in aquatic systems observed in monitoring programs only cover a few decades. Long-term records of environmental variation at centennial to millennial time scales are essential for the assessment of the carbon cycle and ecosystem dynamics in response to early and recent anthropogenic disturbances (Willis and Birks, 2006; Meyer-Jacob, 2015). Such records can be obtained from well-dated sediments, which consist of chronologically deposited materials originating from both within the aquatic ecosystem and the catchment area (e.g. Koinig et al., 2003; Klamt et al., 2017; Hyodo et al., 2017; Haas et al.,

2019). Multi-proxy analysis of continuous sediments allows us to explore the aquatic ecosystem responses to anthropogenic disturbances on the catchment (e.g. Meyers, 2003; Brag  e et al., 2013; Dreibrodt and Wiethold, 2015; Ning et al., 2018; Haas et al., 2019).

In this study, the variability of organic matter transport between terrestrial and aquatic environments in response to long-term land-use changes was studied using multi-proxy analysis of four sediment sequences from different environmental settings in southern Sweden. Catchment-scale landscape dynamics were reconstructed quantitatively using the pollen-based Landscape Reconstruction Algorithm (LRA). Lignin phenols were used as biomarkers to trace the terrestrial organic matter preserved in the sediments. Bulk geochemistry including TOC, C/N ratio and BSi was applied for a general assessment of the proportions of terrestrial and aquatic organic matter deposition and estimation of aquatic production. Soil erosion was assessed by XRF analysis.

The aims of this study were:

- 1) To quantify land-use changes at centennial to millennial time scales based on pollen analysis and LRA, and reconstruct the erosion history using geochemical analyses;
- 2) To investigate the variation in terrestrial organic matter export to the aquatic system in response to land-use changes (forestry, agriculture, drainage patterns) by comparing the content of lignin-derived organic matter in the sediments with the landscape reconstruction;
- 3) To assess the difference in the character of terrestrial organic matter deposited in aquatic systems with different environmental characteristics (small lake vs. large lake, inland lake vs. coastal area).

## 2 Background

### 2.1 Land-use history in southern Sweden

Agriculture was introduced to Sweden in around 4000 BC, and cereal growing and animal husbandry were established in southern Sweden, especially in the lowlands, within a few thousand years (Berglund 1985). Even before that, in the late Mesolithic, hunting camps started to be built up on the south-east coast of Sweden which possibly led to some small clearings (Berglund, 1992). During the Neolithic and early Bronze

Age, settlements were mainly located along the coast, and the agrarian economy was introduced and developed with expansion of arable land, wood pastures and meadows (Berglund, 1992). In an agrarian economy, human impacts on the landscape are characterized by deforestation and the creation of secondary woodlands and grassland (Rackham, 1986; Berglund, 1992). Compared with the low-land, the human impact came later in the uplands of southern Sweden, and the land-use there was dominated by grazing (Berglund, 2003). During the late Bronze Age (9th – 6th century BC), there was a large increase in agricultural landscape opening from former forests and woodlands in southernmost Sweden followed by the transition of land-use development, from shifting cultivation to farming based on permanent fields, around the first millennium BC (Berglund, 1991; Gaillard et al., 1994; Berglund et al., 2008; Widgren and Pedersen, 2011). The land was shifted to an anthropogenic mosaic of fields, meadows, pastures and managed woodlands (Widgren and Pedersen, 2011). The agricultural and grazing activities did not change much throughout the Late Bronze Age, Pre-Roman Iron Age and Roman Iron Age (Gaillard et al., 1991). This period of land-use ended around AD 500 with the widespread abandonment of pastures, arable fields and regrowth of woodland (aspen, birch and hazel) (Gaillard et al., 1991; Lagerås et al., 1995, 1996). The agricultural decline may be connected with either a general decline in population associated with the Migration Period crisis during the 6th century or the shift in arable cultivation on the deeper and heavier soils related to the improvement in ploughing techniques in the Iron Age (Godwin, 1975; Gaillard, 1991).

During the early Medieval (900-1200 AD), land use expanded in southern Sweden, which was related to social expansion (Berglund, 1991; Lindbladh et al., 2000). This expansion was interrupted by the Black Death in AD 1348-1351, which is seen as the beginning of the Late Medieval agrarian crisis, and approximately half of the farmland was abandoned (Lindbladh and Bradshaw, 1998; Hultberg et al., 2010; Myrdal, 2012; Lagerås et al., 2016). The population increased and arable fields and pastures were reestablished during the 16th century (Myrdal and Morell, 2011; Lagerås, 2013; Fredh et al., 2019). During the agricultural revolution (AD 1700-1900), much higher productivity in agriculture was achieved because of better management of manure, crop rotation, irrigation and marling (Myrdal and Morell, 2011). The expansion of agricultural land reached a maximum in the 19th century (Fredh, 2019). Traditionally, the agricultural land was divided into in-

fields and outland. Infields were located near the villages and consisted of arable fields and hay meadows on fertile soils, and outlands on poor soils were used for grazing and collecting fodder and fuel (Eriksson et al., 2002; Fredh, 2019). This farming system existed until mid-19th to early 20th century when it was abandoned due to the increasing use of artificial fertilizers, production of ley and grain fields and the development of forestry later on (Eriksson et al., 2002).

From the early 20th century, conifers, especially spruce were extensively planted in southern Sweden with the development of the managed forest (Björkman, 2001; Lindbladh et al., 2014; Mazier et al., 2015; Åkesson et al., 2015). The small-scale agriculture was converted into modern crop cultivation and commercial forestry (Antonsson and Jansson 2011).

## 2.2 Pollen-based land-cover reconstruction

Pollen preserved in sediments has been widely used to investigate past vegetation changes since it can be identified based on relatively distinct shapes and surface structures (Seppä, 2007). In early studies, pollen analysis of vegetation changes was based on original pollen percentages. However, different plant taxa have variable pollen production and dispersal, so pollen proportions cannot fully represent the vegetation cover. Thus, in the mid-20th century, researchers started to revise the pollen records, and try to formulate the pollen-vegetation relationship to convert pollen percentages into quantitative estimates of vegetation composition (Erdtman, 1943; Fagerlind, 1952). A theoretical framework called R-value was developed by Davis (1963) based on the comparison of pollen percentages in modern lake sediment with different taxa percentages in the surrounding forest in Vermont, US. R-value was further developed by the inclusion of a regional background term by Andersen (1970, 1974), who also presented several correction factors for key tree species. Andersen's model was later expanded into the Extended R-value (ERV) model, which takes the non-linear relationship between vegetation and pollen percentage as well as the properties of pollen dispersal and deposition into consideration, and thus makes a significant improvement on the application of pollen analysis to past vegetation and land-use changes (Parsons and Prentice, 1981; Prentice, 1985).

Based on the theoretical framework developed before, Sugita (2007a, b) developed the Landscape Reconstruction Algorithm (LRA). LRA is a multistep model

for quantifying vegetation composition based on pollen assemblages with fewer biases on pollen productivity and dispersal (Sugita, 2007a, b). LRA includes two models, REVEALS (Regional Estimates of VEgetation Abundance from Large Sites) and LOVE (Local Vegetation Estimates). REVEALS is used to estimate regional vegetation composition within an area of 50-100 km radius using pollen records from large lakes (100-500 ha) or several small lakes (Sugita, 2007a). LOVE is used to estimate local vegetation composition using pollen records from small lakes and the regional vegetation composition estimated by REVEALS (Sugita, 2007b). Over the last decade, LRA has been validated in southern Sweden (Cui et al., 2013, 2014; Hultberg et al., 2015; Mazier et al., 2015; Åkesson et al., 2015; Fredh et al., 2017, 2019), Denmark (Nielsen and Odgaard, 2010; Overballe-Petersen et al., 2013), Norway (Mehl et al., 2015; Hjelle et al., 2017), Switzerland (Soepboer et al., 2010) and Czech (Abraham et al., 2017).

### 2.3 Significance of organic matter in sediments

Organic carbon in sediments is an important part of the global carbon cycle. Previous studies have shown that on a global scale,  $\sim 5.1$  Pg C per year is exported from the terrestrial environment to inland waters (Drake et al., 2018), 0.2 to 1.6 Pg C per year is buried in inland water sediments (Mendonça et al., 2017 and references therein), 0.95 Pg C per year is further delivered to the marine ecosystem (Regnier et al., 2013) and  $\sim 0.2$  Pg C per year is buried in marine sediments (IPCC, 2013). The majority of organic carbon accumulation in the global ocean happens in coastal areas. Besides the burial of organic carbon produced by in-situ aquatic primary production, coastal areas are also a significant sink for terrestrial organic carbon (Hedges and Keil, 1995; Bianchi et al., 1999).

Lakes play an important role in the terrestrial carbon budget. The terrestrial carbon delivered to lakes can be released to the atmosphere as  $\text{CO}_2$ , be deposited in lake sediments, or be further transported to the sea (Algesten et al., 2004; Cole et al., 2007; Mendonça et al., 2017). Lakes are very efficient carbon sinks, and the C burial flux in lakes is generally higher than in oceans, which is partly due to the fact that lakes receive a higher proportion of terrestrial organic matter (Sobek et al., 2009). Although organic carbon burial in the lake and marine sediments is often small compared to other fluxes in the carbon cycle, it leads to net  $\text{CO}_2$  removal from the atmosphere and serves as long-term carbon

storage (Burdige, 2007; Sobek et al., 2009; Mendonça et al., 2017).

Sedimentary organic matter consists of organic compounds with a wide range of sources, and they are different from each other in terms of chemical and physical properties (Mayer, 1994; Hedges and Oades, 1997; Hedges et al., 1997; Goñi et al., 1998). In general, there are two main groups of organic matters in the lake and marine sediments, and both of them hold information about the origin, the preservation state and the transportation mode to the depositional basin. First, autochthonous organic particles are produced by the aquatic community (Meyers, 1997). Detritus produced by phytoplankton through photosynthesis is the dominant source of primary organic matter in sediments in most aquatic systems (Meyer, 1997). Second, allochthonous organic matter, which is brought to lakes by surface runoff and fluvial transport, mainly includes detritus from land plants and soil-derived organic matters (Meyers, 1997). Allochthonous organic matter can also be from the spores, pollen, leaves and branches of land plants, soot from forest-fires and fine soil from distant sources transported by wind (Meyers and Ishiwatari, 1993; Meyers, 1997). The composition of organic matter is often altered through degradation during transport. However, degradation mostly happens in the uppermost layers of the sediments, so organic matter composition does not change much after burial in the deeper layers (Hodell and Schelske, 1998; Routh et al., 2004). Hence, the organic matter preserved in aquatic sediments keeps considerable information about its origin, delivery and preservation that can be extracted by the combined use of geochemical proxies, such as total organic carbon content (TOC), total organic carbon to nitrogen (C/N) ratios, stable carbon isotopes ( $\delta^{13}\text{C}$ ) and biomarkers such as lignin phenols (Meyers, 1997).

### 2.4 Biomarkers: lignin phenols

Organic biomarkers are molecules synthesized by distinctive biotic sources. Biomarkers are seen as molecular fossils since they keep the information of their origin after burial in sediments (Meyers and Teranes, 2001). Lignin is a type of biopolymer which is almost exclusively synthesized by terrestrial vascular plants (Hedges and Mann, 1979; Ertel and Hedges, 1984). It is the most abundant plant product and makes a large contribution to bulk organic matter in sediments (Brown, 1969; Ertel and Hedges, 1985; Goñi and Hedges, 1992; Jex et al., 2014). Lignin is highly resistant to microbial degradation and can still contain information of its or-



igin after deposition in sediments (Hedges et al., 1982; Goñi and Hedges, 1992; Jex et al., 2014). Sedimentary lignin can be broken down into eleven dominant phenols through alkaline CuO oxidation in the lab, which are usually classified into four functional groups (vanillyl, syringyl, cinnamyl and *p*-hydroxyl) (Fig. 1; Hedges and Ertel, 1982). Vanillyl phenols (V) which include vanillic acid, vanillin and acetovanillone are degradation products of both woody and non-woody tissues of angiosperms and gymnosperms; Syringyl phenols (S) which include syringic acid, syringaldehyde and acetosyringone are exclusively found in angiosperms (Hedges and Mann, 1979); Cinnamyl phenols (C) which include *p*-coumaric acid and ferulic acid are only found in non-woody plant tissues (Hedges and Mann, 1979). The *p*-hydroxyl phenols (P) are not exclusively derived from terrestrial vascular plants, but can also be generated by plankton and bacteria (Hedges et al., 1988a).

The relative abundance of syringyl, vanillyl and cinnamyl phenols can reflect original plant types and tissues of the lignin (Hedges and Mann, 1979). Syringyl to vanillyl ratios (S/V) can be used to assess the relative abundance of angiosperm and gymnosperm production,

with high S/V ratios in the sediments reflecting domination of angiosperm-derived organic matter (Hedges and Mann, 1979). Cinnamyl to vanillyl (C/V) ratios can be used to indicate the relative composition of woody to non-woody tissues origin (Hedges and Mann, 1979). The ratio between *p*-hydroxyacetophenone and total *p*-hydroxyl phenols (PON/P) can be used to indicate the relative abundance of lignin-containing plants and lignin-free plants, as PON is derived from lignin while *p*-hydroxybenzaldehyde and *p*-hydroxybenzoic acid can be produced by lignin-free organisms, such as phytoplankton and bacteria (Hedges et al., 1988a).

Different lignin phenols respond differently to degradation, with cinnamyl phenols being more liable to the degradation than syringyl and vanillyl phenols (Hedges et al., 1988b; Opsahl and Benner, 1995). This differential degradation response leads to lower C/V ratio when the terrestrial organic matter is highly degraded (Hedges et al., 1988; Opsahl and Benner, 1995). Also, enhanced degradation by microbial activity leads to increase in the acids compared to the aldehyde, so the acid to aldehyde ratio of vanillyl phenols (Ad/Al)<sub>V</sub> is also a proxy for the degradation state of the terres-

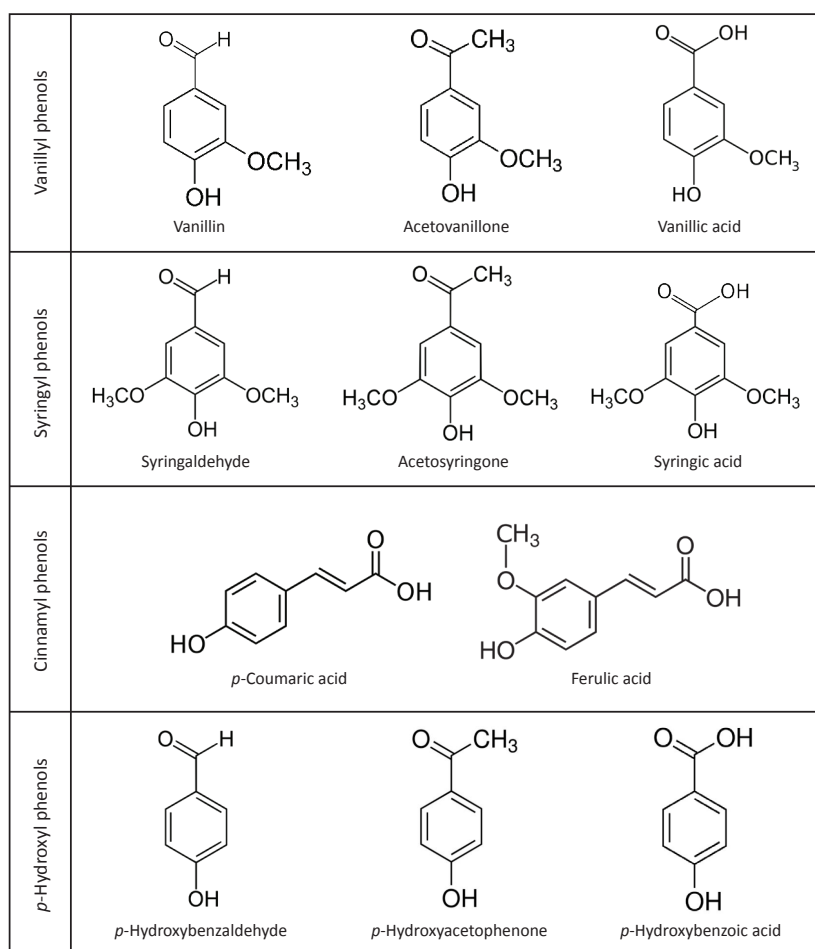


Figure 1 Structure of lignin phenols.

trial organic matter (Ertel and Hedges, 1984; Opsahl and Benner, 1995). The P phenols are more resistant to degradation than the other three groups, so the P/(S+V) can be used as another indicator of degradation (Dittmar and Lara, 2001). Lignin phenols have been used to identify the terrestrial organic matter sources in coastal marine environments (e.g. Miltner and Emeis, 2001; Visser et al., 2004; Sun et al., 2011; Cui et al., 2017), large lake catchments (e.g. Hedges et al. 1982; Hu et al. 1999), and a few small lakes (e.g. Glaser and Zech, 2005; Pempkowiak et al., 2006; Kuliński et al., 2007; Chmiel et al., 2015; Hyodo et al., 2017).

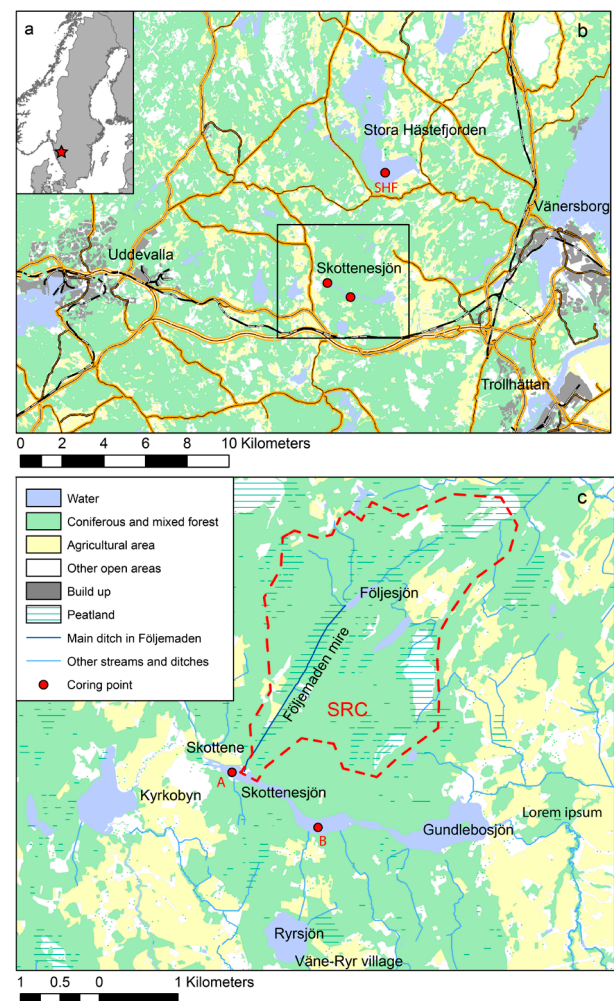
## 2.5 Study description

### Western study region

Lake Skottenesjön (58°21'N, 12°08'E; 60 m above sea level) is an elongated lake located downstream of the Skogaryd Research Catchment (SRC; Fig. 2). SRC is situated in the province of Västra Götaland, about 90 km north of Gothenburg, southwestern Sweden. The SRC was established to characterize and quantify greenhouse gas balances in forested regions including carbon and nitrogen exchange between land-atmosphere, land-water and water-atmosphere (<https://www.fieldsites.se/en-GB/research-stations/skogaryd>). Skottenesjön has a surface area of 72 ha (Natchimuthu et al. 2016) and the main inflow from Lake Gundlebosjön. The maximum water depth is close to the main inflow in the east, and the water depth shallows towards the outflow in the west. Except for the main inflow, a few smaller streams and ditches, including the one that passes through the wetland Följemaden, drain into Lake Skottenesjön.

Skogaryd is located in the hemi-boreal region with a humid and warm climate (SITES, 2015). The long-term (during 1961-1990) mean annual temperature was 6.2°C, and mean annual precipitation was 709 mm according to the Vänersborg weather station which is situated 12 km east of Skogaryd. This area is a mixed coniferous forest with Norway spruce (*Picea abies*) and Scots Pine (*Pinus sylvestris*) as main tree species. The understory vegetation is dominated by *Vaccinium myrtillus*, *Luzula pilosa*, and *Oxalis acetosella*, and the bottom layer is dominated by *Pleurozium schreberi*, *Dicranum majus*, *Hylocomium splendens*, *Plagiomnium affine*, *Polytrichum formosum* and *Sciuro-hypnum oedipodium* (Ernfors et al., 2011).

The SRC is located in Väne-Ryr Parish historically

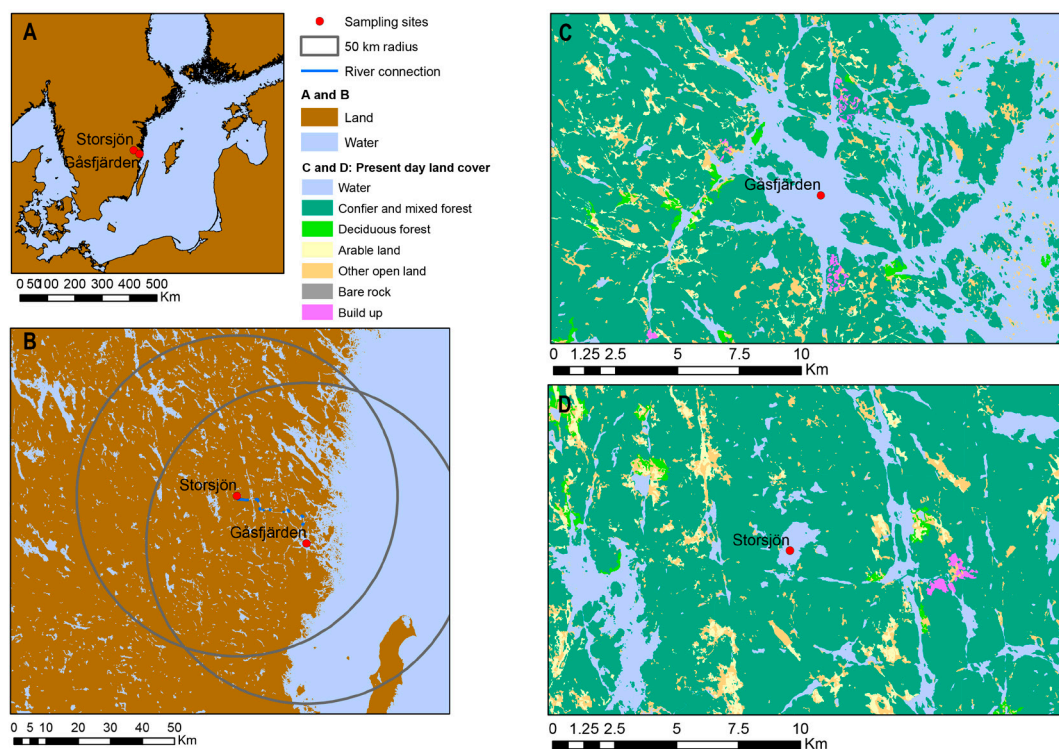


**Figure 2** Map of the western study region.

(Hill, 1999). There is sparse archaeological evidence of prehistoric settlement in the area, but the first documented history of human activity at Skogaryd was in the mid-1600s, and this area has experienced significant changes in land use since then. Records from AD 1571 to 1966 shows a steady increase in the population between ad 1571 and 1880 and a decline afterwards (Palm 2000). The study area was owned by the local farmers prior to AD 1692, and belonged to the crown between ad 1692 and 1738 (Hill 1999). In the early 1700s, the first steel factory, Kollerö Bruk, in western Sweden was established in the Skogaryd area (Hill, 1999). The area surrounding the peatland Följemaden was covered by forest in the 1700s and early 1800s. In 1872, the peatland was drained by a ditch called Krondiket from Lake Följesjön down to Lake Skottenesjön.

### Eastern study region

Gåsfjärden (57°34' N, 16°34' E; Fig 3) is located on the southeast coast of Sweden. It is a fjord-like inlet connected to Baltic Sea through an approximately 500 m



**Figure 3** Map of the eastern study region.

wide and less than 20 m deep channel. Gåsfjärden has a surface area of 22 km<sup>2</sup> and an average water depth of 10 m. The hydrological catchment is around 1500 km<sup>2</sup>. Water exchange between Gåsfjärden and the open sea is restricted by numerous small islands.

Lake Storsjön (57°42'N, 16°14'E; Fig 3) has a mean water depth of 5 m and a surface area of 2.1 km<sup>2</sup>. It is situated around 25 km NW of Gåsfjärden and drains into the river Botorpsströmmen which is one of the rivers draining into Gåsfjärden.

The region where the two study sites are located is covered by mixed coniferous forest with small areas of agricultural land. There are more deciduous forests dominated by oak along the coast than the further inland. There was a copper mine on the shore of Gåsfjärden between the 1630s and AD 1920 (Söderhjelm and Sundblad 1996). Additionally, a sawmill industry in Blankaholm by Gåsfjärden started in AD 1886 and expanded during the 1910s and the 1920s (Ning et al, 2018).

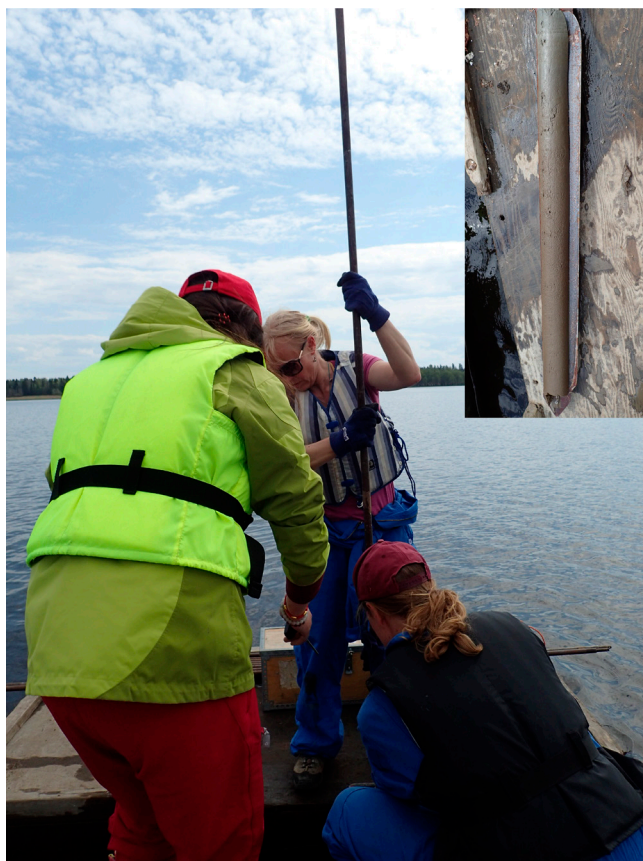
## 3 Material and methods

### 3.1 Sampling

Two fieldworks were carried out at Skogaryd (Fig. 4). The first one was performed on January 27-29, 2016 on Lake Skottenesjön. During the fieldwork, sediment cores were retrieved from two points. Site A (58°21'18"N, 12°07'43"E) is situated in the western basin of the lake and close to the outlet of the ditch Krondiket which transports runoff from the Skogaryd Research Catchment into the lake. Site B (58°20'58"N, 12°08'52"E) is located in the eastern basin of the lake, close to the main inflow from the adjacent Lake Gundlebosjön. At site A, a sediment sequence spanning 4.6 m was retrieved at a water depth of 1.9 m. At site B, a sediment sequence spanning 3 m was retrieved at a water depth of 5.1 m. The loose surface sediments were retrieved from both sites using a HTH-type gravity corer with an inner diameter of 66 mm (Renberg and Hansson, 2008) and subsampled in 1-cm intervals in the field. The deeper parts of the two sediment sequences were retrieved using a Russian corer (chamber length 1 m; 75 mm in diameter). The cores were collected with overlaps of roughly 60 cm between successive 1 m sections. All the sediment sequences were wrapped in plastic film, placed in supportive plastic liners and transported to the laboratory at the Department of Geology, Lund University where they were stored in the cold room (5 °C) until further analysis.

The second fieldwork was performed on Lake Stora Hästefjorden on May 10, 2016. Lake Stora Häste-





**Figure 4** Sediment sampling in Lake Stora Hästefjärden and a sediment core retrieved by a Russian corer.

fjärden (58°24'21''N, 12°10'33''E) is located 5 km to the north of Lake Skottenesjön. A 1.8 m long sediment sequence was obtained with a Russian corer at a water depth of 7.5 m and the loose surface sediment was collected with an HTH-type gravity corer (Renberg and Hansson, 2008) and subsampled in every 1 cm in the field.

In Lake Storsjön, a 4.5 m long sediment sequence was retrieved using a Russian corer and a HON-Kajak corer (Renberg, 1991; Ning et al., 2018).

In Gåsfjärden, the sediment sequences were collected at a water depth of 31 m using a Gemini corer (Ning et al., 2018).

### 3.2 $^{210}\text{Pb}$ and $^{14}\text{C}$ dating

The loose sediments of the uppermost part of the sediment sequences collected by gravity corer were freeze-dried and analysed for  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  by gamma spectrometry at the Environmental Radiometric Facility at University College London, using a coaxial well-type low background intrinsic germanium detector (ORTEC® HPGe GWL series). The chronologies

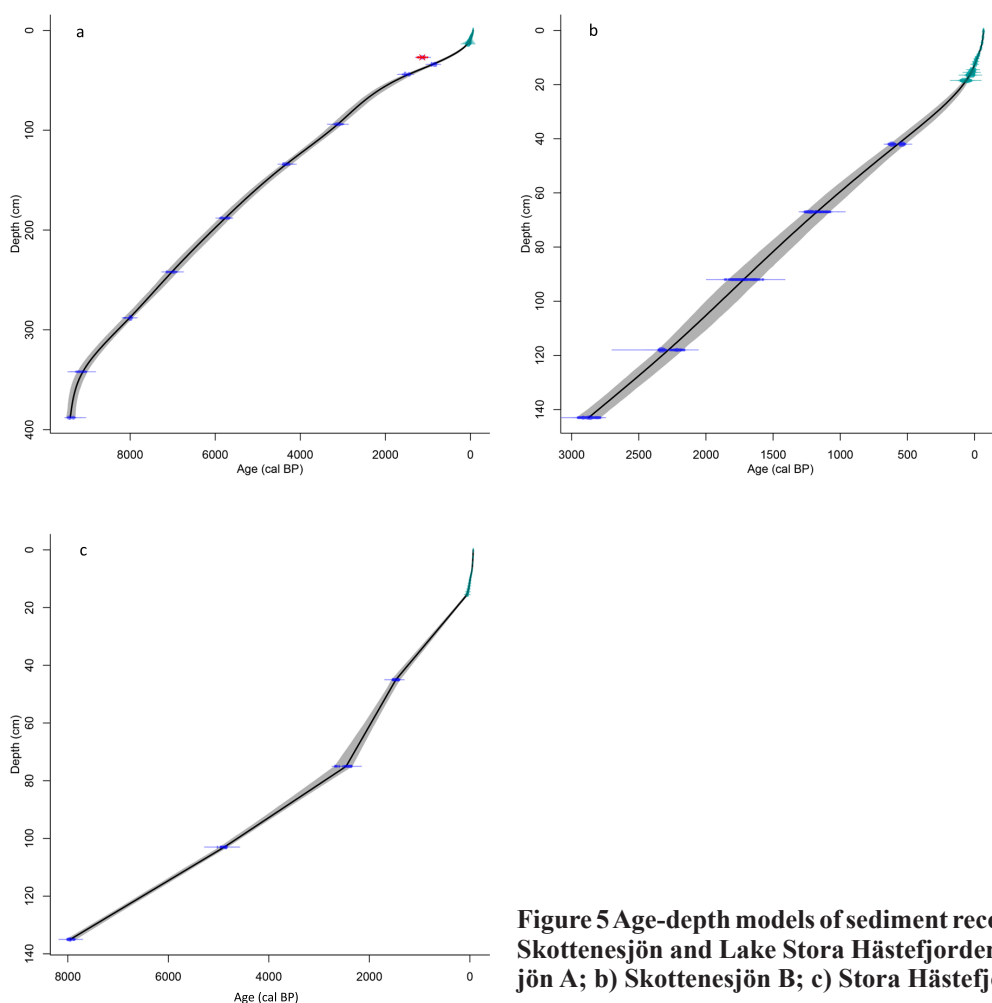
**Table 1** Accelerator-mass-spectrometry (AMS)  $^{14}\text{C}$  dates from Lake Skottenejön and Lake Stora Hästefjärden

Depth (cm)	Dated Material	Weight (mg C)	Lab no. <sup>a</sup>	Radiocarbon age (BP)	Calibrated age <sup>b</sup> (cal. BP $\pm 2\sigma$ ranges)
Skottenesjön A					
26-27	Macrofossils	1.8	LuS 15003	1200 $\pm$ 40	1051-1189
33-35	Macrofossils	1.3	LuS 15004	915 $\pm$ 35	791-928
43-45	Twig, <i>Betula</i> seed and catkin	0.6	LuS 12229	1620 $\pm$ 40	1410-1573 <sup>c</sup>
93-95	Leaf and twig	0.8	LuS 12230	2950 $\pm$ 40	2975-3214 <sup>c</sup>
133-135	Big twig	1.8	LuS 12231	3895 $\pm$ 35	4235-4422 <sup>c</sup>
187-189	Twig and <i>Betula</i> seed	1.4	LuS 12232	5010 $\pm$ 50	5647-5898 <sup>c</sup>
241-243	Bark, twig, catkin	0.5	LuS 12233	6120 $\pm$ 45	6896-7158 <sup>c</sup>
287-289	Leaf	0.7	LuS 12234	7185 $\pm$ 45	7936-8062 <sup>c</sup>
341-343	Twig, leaf	0.7	LuS 12235	8200 $\pm$ 50	9017-9296 <sup>c</sup>
387-389	Twig	1.6	LuS 12236	8365 $\pm$ 45	9283-9482 <sup>c</sup>
Skottenesjön B					
41-43	Macrofossils	0.5	LuS 13513	550 $\pm$ 40	513-565
66-68	Macrofossils	0.7	LuS 13514	1240 $\pm$ 40	1070-1270
97-93	Macrofossils	1.1	LuS 13515	1795 $\pm$ 60	1593-1836
117-118	Macrofossils	0.7	LuS 13516	2290 $\pm$ 45	2292-2360
142-143	Macrofossils	1.4	LuS 13517	2775 $\pm$ 40	2791-2959
Stora Hästefjärden					
44-46	Small Macrofossils	0.4	LuS 13509	1585 $\pm$ 40	1392-1554 <sup>c</sup>
74-76	Small Macrofossils	0.5	LuS 13510	2390 $\pm$ 45	2337-2516 <sup>c</sup>
102-104	Small Macrofossils	0.6	LuS 13511	4305 $\pm$ 45	4823-4976 <sup>c</sup>
133-136	Small Macrofossils	0.4	LuS 13512	7130 $\pm$ 50	7912-8025 <sup>c</sup>

<sup>a</sup> LuS = Lund University Radiocarbon Dating Laboratory.

<sup>b</sup>  $^{14}\text{C}$  dates were calibrated using the IntCal13 radiocarbon calibration dataset (Reimer et al., 2013).

<sup>c</sup> Yang et al. (2020)



**Figure 5** Age-depth models of sediment records from Lake Skottenesjön and Lake Stora Hästefjärden. a) Skottenesjön A; b) Skottenesjön B; c) Stora Hästefjärden.

were constructed based on the constant rate of supply model (CRS) and corrected based on  $^{137}\text{Cs}$  data (Appleby, 2001). The lower sediment sections beyond the reach of  $^{210}\text{Pb}$  dating were dated by radiocarbon dating. Sediment samples from selected intervals of the sediment sequences were wet-sieved to obtain terrestrial macroscopic plant remains that were later dated at the Radiocarbon Dating Laboratory at Lund University (Table 1).

The radiocarbon dates were calibrated using the IntCal13 radiocarbon calibration dataset (Reimer et al. 2013). The age-depth models including both radiocarbon dates and  $^{210}\text{Pb}$  ages were constructed using the R-code CLAM (Blaauw 2010; Fig. 5).

### 3.3 Lignin phenols analysis

The alkaline cupric oxide (CuO) oxidation method developed by Hedges and Ertel (1982) with modifications following Sun et al. (2015) was applied to break down lignin and extract lignin phenols from sediments. Freeze-dried sediment samples were ho-

mogenized and treated with ~500 mg CuO, ~100mg  $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$  (oxygen scavenger) and 4 ml 2 N NaOH bubbled with  $\text{N}_2$  gas overnight in Teflon vessels. The Teflon vessels were purged with Ar gas for 30 min to prevent the introduction of  $\text{O}_2$  before being sealed and coated with a Parr® metal-jacketed vessel. The samples were heated while they were rotated in an oven at 155 °C for 3 hours. After cooling, known amounts of ethyl vanillin and trans-cinnamic acid were added to each sample as internal standards. The contents in the Teflon vessels were transferred and rinsed with 1 N NaOH into a centrifuge tube and centrifuged at 3000 rpm for 10 min. The supernatant was acidified to pH 1 with 6 N HCl and extracted with ethyl acetate three times. The extracts were passed through a  $\text{Na}_2\text{SO}_4$  column to remove water, and dried under steady  $\text{N}_2$  flows.

The products were redissolved in pyridine and derivatized with bis-(trimethylsilyl)-trifluoroacetamide-trimethylchlorosilane (BSTFA-TMCS) for analysis and quantification using a gas chromatography-mass spectrometry (Shimadzu QP2010 GC-MS). Chromatographic separation was achieved using a capillary

**Table 2 Abbreviations, retention times and characteristic mass fragments for CuO oxidation products**

Compound	Abbreviation	Retention time (min)	Major ions (m/z) <sup>a</sup>
<i>p</i> -hydroxybenzaldehyde	PAL	9.403	<b>151</b> , 179, 194
<i>p</i> -hydroxyacetophenone	PON	11.919	<b>193</b> , 194, 208
vanillin	VAL	13.628	<b>194</b> , 209, 224
<i>trans</i> -cinnamic acid <sup>b</sup>	CiAD	13.868	131, 161, <b>205</b>
ethyl-vanillin <sup>b</sup>	EVAL	15.340	<b>167</b> , 195, 238
acetovanillone	VON	15.982	208, <b>223</b> , 238
<i>p</i> -hydroxybenzoic acid	PAD	16.292	223, <b>267</b> , 282
syringaldehyde	SAL	18.186	<b>224</b> , 239, 254
vanillic acid	VAD	19.972	<b>267</b> , 282, 297
acetosyringone	SON	20.027	<b>238</b> , 253, 268
syringic acid	SAD	23.415	297, 312, <b>327</b>
<i>p</i> -coumaric acid	CAD	24.231	219, <b>293</b> , 308
ferulic acid	FAD	27.860	249, 323, <b>338</b>

<sup>a</sup> Bolded ions were used for SIM quantification.

<sup>b</sup> Internal standards

column (DB-5, 30 m × 0.25 mm i.d., 0.25 µm film thickness), and the program was set with an initial temperature of 100 °C, a 4°C/min temperature ramp and a final temperature of 300 °C.

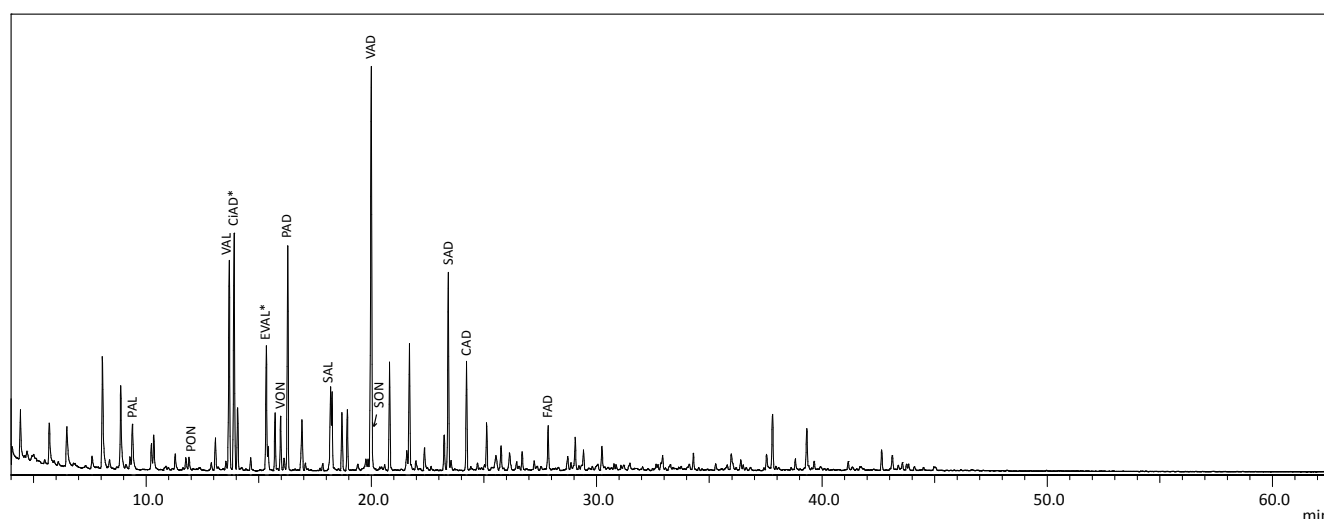
The analysis was conducted with total ion monitoring (TIM, range m/z 50-650; Table 2) and single ion monitoring (SIM, one characteristic ion for quantification with two reference ions). The eleven target compounds were identified based on their mass spectra and comparison of their retention times to a standard mix containing all target phenols. Vanillic acid and acetosyringone co-eluted and were not possible to separate chromatographically (Fig. 6). Instead, these two compounds were separated and quantified based on the ion m/z 267 for vanillic acid and m/z 238 for acetosyringone. Lignin phenols in the sediment samples from Lake Skottenesjön were quantified using *trans*-cinnamic acid as a single point internal standard. Lignin

phenols in the sediment samples from Lake Storsjön and Gåsfjärden were quantified using *trans*-cinnamic acid as an internal standard and a four-point calibration curve.

### 3.4 Pollen analysis and landscape reconstruction

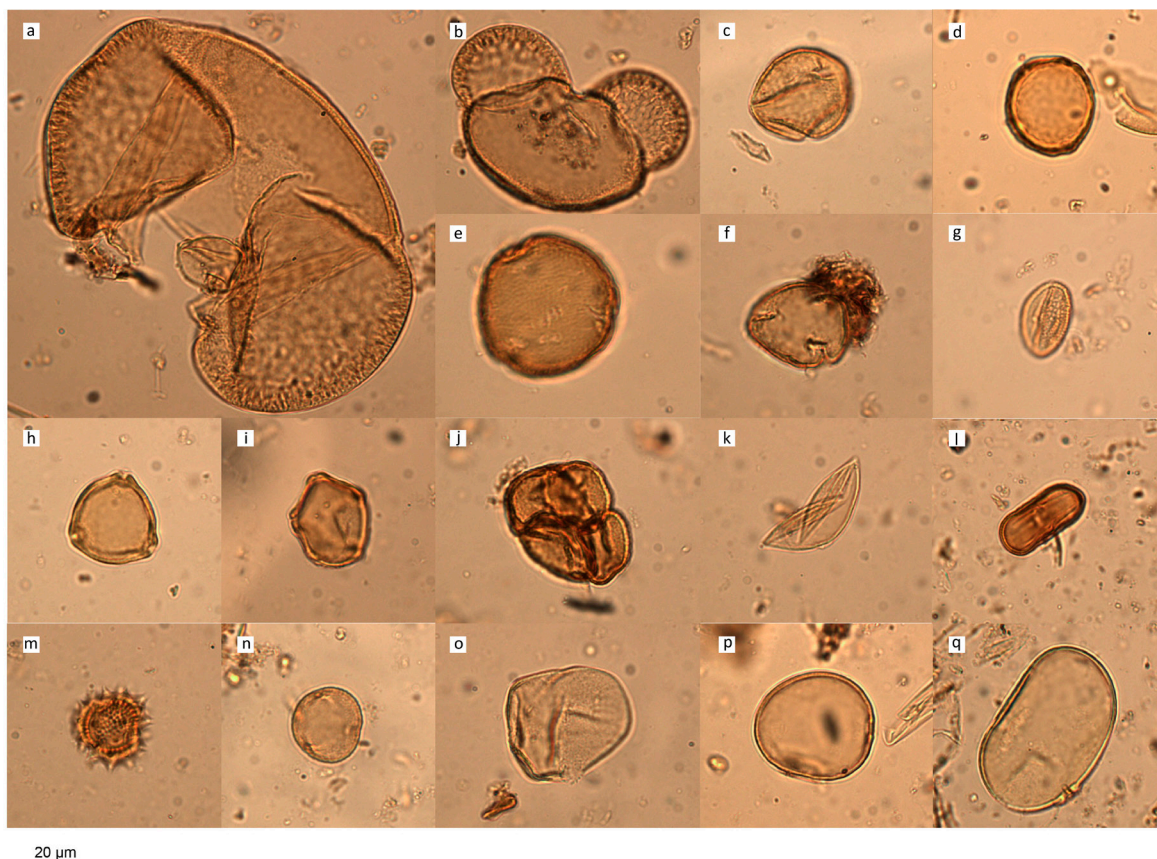
The sediment samples for pollen analysis were treated following the standard acetolysis method (Berglund and Ralska-Jasiewiczowa, 1986). At least 1000 pollen grains in each sample, which is required for LRA, were counted and identified under the Olympus BX41 microscope at ×400 magnification at the Department of Geology, Lund University (Fig. 7).

REVEALS and LOVE in the LRA model (Sugita, 2007 a,b) were applied to the pollen counts at an interval of approximately 50 years to obtain the land cover reconstruction. In the western study region, pollen counts



**Figure 6 Chromatogram of a sediment sample from Lake Skottenesjön with identification of the lignin phenols. \* Internal standards. Abbreviations are explained in Table 2.**





**Figure 7** Examples of pollen grains in sediment samples of Lake Skottenesjön. a) *Picea*; b) *Pinus*; c) *Quercus*; d) *Ulmus*; e) *Fagus*; f) *Fraxinus*; g) *Salix*; h) *Betula*; i) *Alnus*; j) *Calluna*; k) *Juniperus*; l) *Apiaceae*; m) *Asteraceae*; n) *Rumex*; o) *Cyperaceae*; p) *Hordeum*; q) *Secale cereale*.

from Lake Stora Hästefjorden were used in REVEALS to estimate the regional vegetation composition (Yang et al., 2020). The pollen counts from Lake Skottenesjön, together with the regional vegetation estimates from Lake Stora Hästefjorden, were used in LOVE to estimate the local vegetation composition of the catchment of Lake Skottenesjön (Yang et al., 2020; Fig. 8). In the eastern study region, pollen counts from Lake Storsjön and Gåsfjärden were used in REVEALS to estimate the regional vegetation composition. The relative pollen productivity (RPP) estimates and fall speeds of pollen used in the REVEALS and LOVE are shown in Table 3. The area of the water basins was assumed constant throughout the study period, atmospheric conditions were assumed to be neutral and the wind speed was set to 3 m/s (Yang et al., 2020). The program REVEALS.v5.0 was applied for REVEALS, and LOVE.v5.1 was applied for LOVE (Sugita, unpublished).

### 3.5 Bulk elemental and $\delta^{13}\text{C}$ analysis

The sediment samples for bulk elemental analysis were freeze-dried and homogenized. We assumed that no carbonate was present in the lake sediments since the lakes in this study and their catchments are situated on

**Table 3** Relative pollen productivity estimates (RPP) relative to Poaceae and fall speeds of the taxa used in the LRA (Yang et al., 2020)

Pollen taxa	RPP	Fall speed (m/s)
<i>Pinus</i>	5.663 <sup>a</sup>	0.031 <sup>b</sup>
<i>Picea</i>	1.757 <sup>a</sup>	0.056 <sup>b</sup>
<i>Quercus</i>	7.533 <sup>a</sup>	0.035 <sup>b</sup>
<i>Fagus</i>	6.667 <sup>a</sup>	0.057 <sup>c</sup>
<i>Ulmus</i>	1.267 <sup>a</sup>	0.032 <sup>c</sup>
<i>Tilia</i>	0.8 <sup>a</sup>	0.032 <sup>c</sup>
<i>Fraxinus</i>	0.667 <sup>a</sup>	0.022 <sup>b</sup>
<i>Alnus</i>	4.2 <sup>a</sup>	0.021 <sup>b</sup>
<i>Betula</i>	8.867 <sup>a</sup>	0.024 <sup>b</sup>
<i>Corylus</i>	1.4 <sup>a</sup>	0.025 <sup>c</sup>
<i>Carpinus betulus</i>	2.553 <sup>a</sup>	0.042 <sup>b</sup>
<i>Salix</i>	1.267 <sup>a</sup>	0.022 <sup>c</sup>
<i>Juniperus</i>	2.067 <sup>a</sup>	0.016 <sup>b</sup>
<i>Calluna</i>	1.102 <sup>d</sup>	0.038 <sup>e</sup>
Poaceae	1	0.035 <sup>a</sup>
<i>Filipendula</i>	2.48 <sup>e</sup>	0.006 <sup>e</sup>
<i>Rumex acetosella</i>	1.559 <sup>d</sup>	0.018 <sup>e</sup>
<i>Plantago lanceolata</i>	0.897 <sup>d</sup>	0.029 <sup>e</sup>
<i>Plantago media/major</i>	1.27 <sup>f</sup>	0.024 <sup>f</sup>
<i>Artemisia</i>	3.48 <sup>g</sup>	0.025 <sup>h</sup>
Chenopodiaceae	4.28 <sup>i</sup>	0.019 <sup>i</sup>
Compositae Cichorioideae	0.244 <sup>c</sup>	0.051 <sup>e</sup>
Apiaceae	0.26 <sup>j</sup>	0.042 <sup>c</sup>
Cyperaceae	1.002 <sup>a</sup>	0.035 <sup>a</sup>
<i>Secale cereale</i>	3.017 <sup>a</sup>	0.06 <sup>c</sup>
Cereal-t	0.747 <sup>d</sup>	0.06 <sup>c</sup>

<sup>a</sup> Sugita et al. 1999; <sup>b</sup> Eisenhut 1961; <sup>c</sup> Gregory 1973;

<sup>d</sup> Nielsen 2004; <sup>e</sup> Broström et al. 2004; <sup>f</sup> Mazier et al. 2008;

<sup>h</sup> Mazier et al. 2012; <sup>i</sup> Abraham and Kozáková 2012; <sup>j</sup> Hjelle 1998



crystalline bedrock with acidic soils. Thus, lake sediment samples were wrapped in the tin capsules without decalcification. The marine sediment samples from Gåsfjärden were decalcified by adding an excessive amount of 1 N HCl in the silver capsules which were later wrapped in the tin capsules. The samples were then analysed using an elemental analyser (COSTECH ECS4010) at the Department of Geology, Lund University and an elemental analyser coupled to an Isotope Ratio Mass Spectrometer (Sercon Ltd HS2022) at the Department of Earth Sciences, Gothenburg University. The atomic C/N ratios were calculated using the weight percentages of total organic carbon (TOC) and total nitrogen (TN) multiplied by 1.167 (Meyers and Teranes, 2001).

### 3.6 Biogenic silica (BSi) analysis

The sequential alkaline digestion method was applied to measure the BSi content at the Department of Geology, Lund University (Conley and Shelske 2001). Approximately 30 mg freeze-dried sediment samples were homogenized and digested with 40 ml 0.1 N Na<sub>2</sub>CO<sub>3</sub> at 85 °C for 5 hours. One ml aliquot was withdrawn from each sample after 3, 4, 5 hours, respectively. Aliquots were transferred to 9 ml 0.021 N HCl for neutralization. Concentrations of dissolved silica in the samples were measured via the molybdate-blue methodology using a discrete chemical analyser (SmartChem 200) (Grasshoff, 1983). The BSi contents were estimated using the intercept of a linear regression through three subsampled Si content and extraction time (Conley 1998; Sauer et al. 2006). Mean values of three subsampled Si content were used to estimate BSi contents for

the samples with the linear regression having a slope around 0.

### 3.7 X-Ray Fluorescence (XRF)

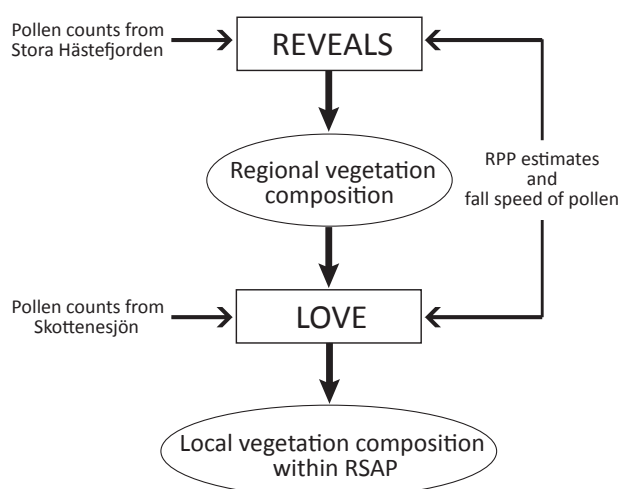
The sediment core sections for X-ray fluorescence (XRF) analysis were surface-cleaned and scanned with an Itrax Core Scanner at the Natural History Museum of Denmark, Copenhagen. The XRF scanner was set with a count time of 35 s and a step size of 1 mm at 50 kV and 30 mA. The results are presented as counts per second (cps) (Croudace et al. 2006; Davies et al. 2015).

## 4 Summary of papers

### Paper I

The population in southern Sweden experienced several phases of growth and decline during the past 1000 years. Population growth led to the expansion of agricultural land and increased catchment erosion (Williams 2000; Boyle 2001; Koinig et al. 2003; Brag  e et al. 2013). Increased detrital input caused eutrophication and elevation of aquatic primary productivity and in lake waters (Neumann et al. 2002; Routh et al. 2004; K  ster et al. 2005; Li et al. 2008). The lake ecosystems affected by human activities may return to natural conditions during the periods of population decrease. However, such recovery can take decades or even centuries (Haas et al. 2019). Long-term records of land use and aquatic environmental changes at centennial to millennial time scales can provide a better understanding of ecosystem dynamics in response to early and recent anthropogenic disturbances.

In this study, the effects of forestry and agricultural land-use on catchment erosion and delivery of organic and minerogenic matter to the lake during the past 1100 years were investigated based on a sediment sequence from a small forest lake in southwestern Sweden. Catchment-scale vegetation cover was quantified at 50-year resolution using pollen analysis and the Landscape Reconstruction Algorithm (LRA) (Sugita, 2007 a, b). Variations in terrestrial organic matter input to lake sediments were assessed by total organic carbon (TOC) content and carbon to nitrogen (C/N) ratios. Changes in the minerogenic matter were analysed using X-ray fluorescence (XRF) scanning. The results show that Skogaryd was not intensively used for agriculture throughout the past 1,100 years, but its land-use



**Figure 8** Flow chart of Landscape Reconstruction Algorithm (LRA) used to quantify the land cover in the western study region. RSAP: relative source area of pollen. RPP: relative pollen productivity estimates.

**Table 4 Author contributions**

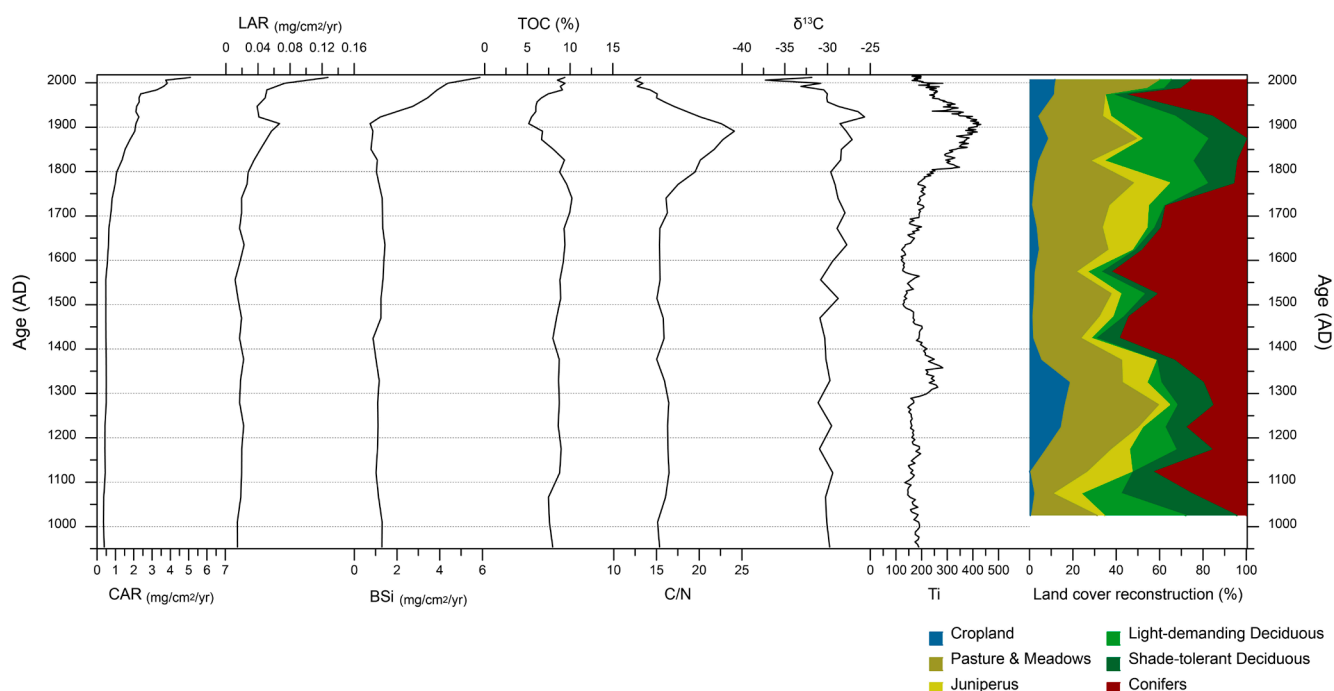
	<i>Paper I</i>	<i>Paper II</i>	<i>Paper III</i>
<i>Study design</i>	B. Yang D. Hammarlund K. Ljung A. B. Nielsen	B. Yang K. Ljung A. B. Nielsen	B. Yang K. Ljung A. B. Nielsen
<i>Sampling</i>	B. Yang E. Fahlgren D. Hammarlund K. Ljung A. B. Nielsen	B. Yang D. Hammarlund K. Ljung A. B. Nielsen	H. L. Filipsson J. Nyberg
<i>Chronological analysis</i>	B. Yang	B. Yang	Published
<i>XRF analysis</i>	M. L. Siggaard-Andersen	M. L. Siggaard-Andersen	-
<i>TOC, TN analysis</i>	E. Fahlgren	E. Fahlgren	B. Yang K. Ljung
<i>BSi analysis</i>	-	B. Yang	-
<i>Pollen analysis</i>	B. Yang A. B. Nielsen	-	A. B. Nielsen W. Ning C. Åkesson
<i>Lignin phenols analysis</i>	-	B. Yang K. Ljung	B. Yang K. Ljung
<i>LRA</i>	B. Yang A. B. Nielsen	-	B. Yang A. B. Nielsen
<i>Data interpretation &amp; discussion</i>	B. Yang S. Björck D. Hammarlund K. Ljung A. B. Nielsen	B. Yang D. Hammarlund K. Ljung A. B. Nielsen	B. Yang H. L. Filipsson K. Ljung A. B. Nielsen

variations were very sensitive to societal changes. Between ca. AD 950 and 1350, local land-use was characterized by small-scale agricultural activities related to the Medieval expansion, and enhanced soil erosion was recorded by increased Ti deposition (Fig. 9). Around AD 1350 much of the farmland was abandoned, most likely in response to outbreaks of plague. The abandonment of farmland led to reduced soil erosion and reestablishment of coniferous woodland. From the 16th century, land-use expanded and gradually intensified, concurrent with a population increase documented in the study area between ca. AD 1600 and 1850. Intensive wood-harvesting from ca. AD 1700 resulted in soil erosion and increased terrestrial organic and minerogenic matter export to the lake (Fig. 9). These processes peaked with the artificial drainage of a nearby wetland for agricultural purposes in the late 19th century. During the 20th century, modern forestry management started with the plantation of conifers, and the input of organic and minerogenic matter to the lake reduced within a few decades (Fig. 9). The outcomes of this study are helpful for a better understanding of the impacts of long-term land-use on elemental cycles.

## Paper II

Lakes play an important role in the global carbon cycle. The land-derived organic matter delivered to lakes is partly transferred into atmospheric CO<sub>2</sub>, buried in sediments or further transported to marine ecosystems (Algesten et al., 2004). Organic carbon burial in lake sediments is heavily affected by the terrestrial organic matter input. However, few studies have focused on long-term changes in terrestrial organic matter input to lakes in response to land-use changes.

In this study, we assessed variations in sedimentary terrestrial organic matter over the last 1000 years based on lignin biomarker records from two sediment cores from Lake Skottenesjön, southwestern Sweden. In combination with pollen-based quantitative land cover reconstruction, we investigated the impacts of centennial-scale changes in land use on terrestrial organic matter input to lake sediments. The results show that human activities in the catchment have made significant impacts on terrestrial organic export by modifying the vegetation cover. When farmland expanded between the 12th and the mid-14th century, no significant change in terrestrial organic matter input was observed



**Figure 9** Mass accumulation rates of carbon (CAR) and lignin (LAR), biogenic silica flux (BSi), total organic carbon content (TOC), carbon/nitrogen ratio (C/N), stable carbon isotope ( $\delta^{13}\text{C}$ ), X-ray fluorescence Ti of sediment from Skottenesjön A, together with land-cover reconstruction based on the pollen from Skottenesjön A. LAR is the sum of the mass accumulation rates of vanillyl, syringyl and cinnamyl phenols. Ti is expressed as elemental intensity in counts per second (cps). C/N, TOC, Ti and land-cover reconstruction are based on Yang et al. (2020).

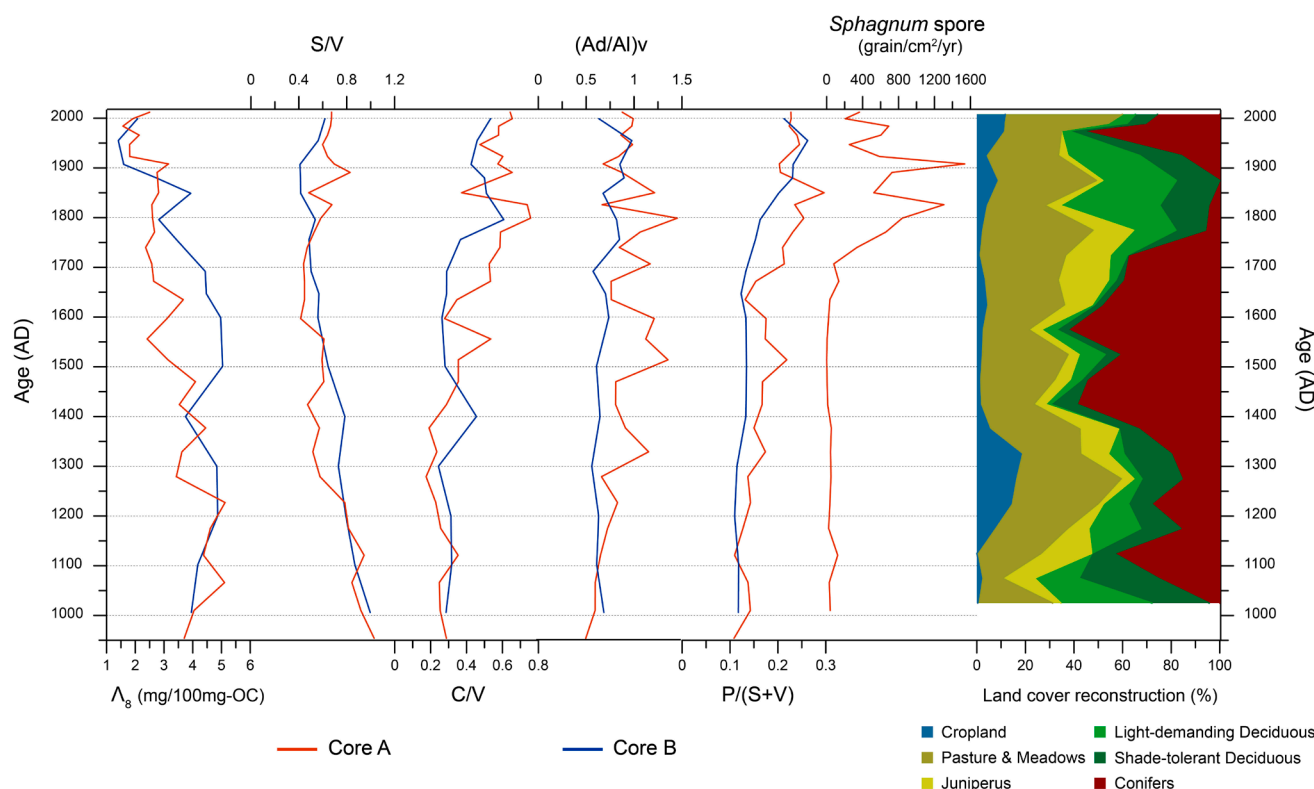
(Fig. 9). Intensified use of the forest in the 18th and 19th centuries resulted in enhanced soil erosion, and increased terrestrial organic matter input to the lake. Much higher export of terrestrial organic and mineralogenic matter to the lake was recorded during the period of modern forestry in the 20th century as compared to previous periods of minor forest disturbance, such as 11th century (Fig. 9). Hence, the changes in land use inferred from pollen considerably modified the composition of terrestrial organic matter deposited in the lake sediments. The comparison of lignin phenols contents from two sediment cores in the same lake shows that the terrestrial organic matter delivery is sensitive to the local land-use changes and spatial variability within the catchment (Fig. 10). The lignin parameters can reflect the vegetation types in the catchment. For example, the ratio of syringyl to vanillyl phenols of lignin shows a good correlation with the coverage of deciduous trees. This study demonstrates that a combined use of lignin phenols analysis and pollen-based quantitative land cover reconstruction is useful for investigation long-term changes in terrestrial organic matter delivery to lake ecosystems.

### Paper III

Coastal areas are where the majority of organic carbon is accumulated in the global ocean, and is also a

significant sink for terrestrial organic carbon (Hedges and Keil, 1995; Bianchi et al., 1999). The terrestrial organic matter buried in coastal marine basins consists of a mixture of long-transported material from further inland and material from the local area closer to the coastline. The terrestrial organic matter undergoes alteration through both chemical and physical processes during transportation (Bauer et al., 2013). Comparison of the character of the organic matter in sediment cores from inland water bodies and coastal marine basins connected through a river system can provide insights into the sources and fate of terrestrial organic matter as it is transported from the inland to coastal basins.

In this study, the lignin phenols in the sediments of Gåsfjärden, a fjord-like inlet of Baltic Sea, and Lake Storsjön, an upstream lake within the same watershed were analysed and compared with pollen-based land-cover reconstructions over the last 500 years. The results show that the deposition of terrestrial organic matter in the two water basin has been affected by the land-use changes (Fig. 11 & 12). Both basins received higher amounts of lignin-derived organic matter during the periods with intensified agricultural land use. However, substantial differences with respect to the concentration, composition and degradation state of the sedimentary lignin-derived organic matter were observed between the two sites. The concentration of lignin-de-



**Figure 10** Lignin data from records A (red) and B (blue) together with *Sphagnum* spore accumulation rate and pollen-based land-cover reconstruction from Skottenesjön A.  $\Lambda_g$ : sum of vanillyl, syringyl and cinnamyl phenols normalized to 100 mg organic carbon; S/V: ratios of syringyl to vanillyl phenols; C/V: ratios of cinnamyl to vanillyl phenols; (Ad/Al)<sub>v</sub>: ratios of vanillic acid to vanillin; P/(S+V): ratios of *p*-hydroxyl to sum of vanillyl and syringyl phenols. Land-cover reconstruction is based on Yang et al. (2020).

rived organic matter is much lower in Gåsfjärden than Storsjön as the organic matter deposition is dominated by aquatic material in Gåsfjärden. Although the vegetation history is similar within the whole watershed where the two sites are located, the composition of lignin deposited in Gåsfjärden is less sensitive to the variations in vegetation cover than in Storsjön. Furthermore, Gåsfjärden receives less degraded terrestrial organic matter, likely because the organic matter liable to degradation has been lost during transportation to the sea. The results demonstrate that the terrestrial organic matter deposition in the coastal-marine basin and the lake upstream from the coast within the same watershed exhibit different characters in terms of concentration, composition and degradation state despite the similar vegetation history.

## 5 Discussion

### 5.1 Land-use history and its impacts on terrestrial organic matter export and aquatic ecosystems

From the 12th to the middle of the 14th century, relatively small-scale land use characterized by an ex-

pansion of cropland is revealed by the pollen-based vegetation cover estimates of Lake Skottenesjön located in southwestern Sweden (Fig 9). This change can be linked to the Medieval expansion with population growth in southern Sweden (Larsson, 1975; Berglund, 1991; Lindbladh and Bradshaw, 1998; Lindbladh, et al. 2000; Myrdal, 2011; Lagerås, 2013; Fredh, et al. 2019). The development of farming and grazing at the expense of forest cover likely contributed to an elevation of soil erosion indicated by an increase in the deposition of Ti between ca. AD 1200 and ca. AD 1350 (Yang et al., 2020). Enhanced erosion due to the conversion of land cover from forest to farmland has been shown to lead to increased terrestrial organic matter input to nearby lakes (Kaushal and Binford 1999; Meyers and Lallier-Vergés 1999; Routh et al. 2004; Das et al. 2009; Li et al. 2014). However, no significant changes were observed in the organic carbon mass accumulation rate (CAR) and lignin mass accumulation rate (LAR) during this early period of agricultural expansion at Skogaryd (Fig. 9). The lack of response in LAR could be explained by the relatively small-scale farming which was not intensive enough to supply significant amounts of terrestrial organic matter to the lake. Furthermore, the soils were not highly disturbed by farming, as the

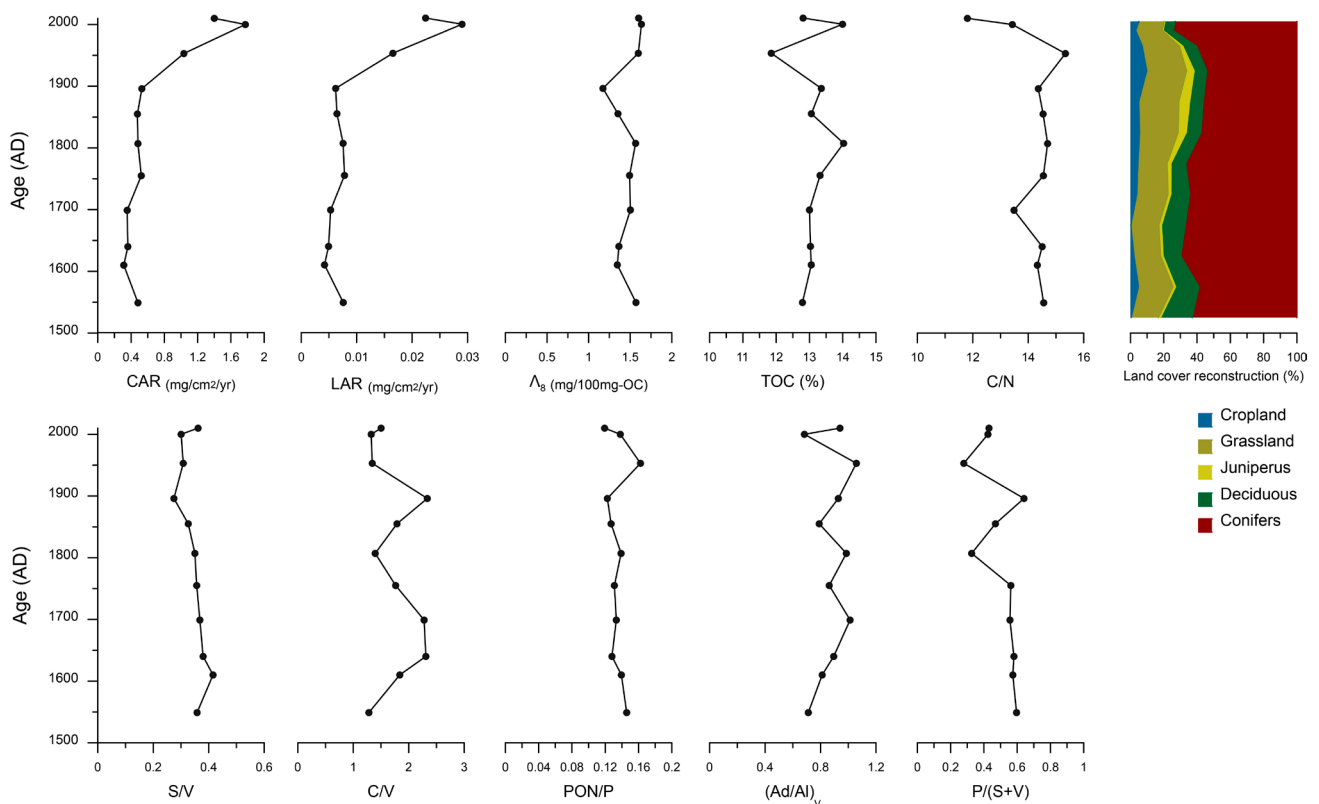


medieval agricultural practices were less advanced than in modern times (Bringéus, 2003). Additionally, the constantly low values of BSi flux suggest that the variation in the nutrients input to the lake did not affect the diatom production.

The Medieval expansion was interrupted by the outbreaks of the plague epidemic which is also known as “the black death”. The epidemic started in AD 1350 in most parts of Sweden, followed by several localized plague outbreaks in the second half of 14th century and at the beginning of the 15th (Benedictow, 2004; Myrdal, 2012; Lagerås, 2013). A decline in the grazing and arable fields and reestablishment of coniferous woodland in the catchment of Lake Skottenesjön at the onset of the Late Medieval crisis is indicated by the pollen-based vegetation cover estimates (Fig. 9; Yang et al., 2020). The abandonment of agricultural land was likely the result of a population decline in the western study region, where Lake Skottenesjön is located, in the wake of the plague outbreaks (Yang et al., 2020). The decrease in the deposition of Ti during this period suggest a decrease in catchment erosion in response to the reforestation. The agricultural activities

in the catchment of Lake Skottenesjön did not recover until ca. AD 1600 (Fig. 9). However, it is suggested that the population increased and agriculture expanded in southern Sweden during the 16th century (Lagerås, 2013). The delay of the land use expansion at our study sites was likely due to the fact that Skogaryd is located in a rather marginal region in term of agriculture, which might not be the prior choice for settlement and farming at beginning of the population increase. In the eastern study region where Lake Storsjön is located, on the other hand, a small-scale farming expansion with an increase in cropland cover before ca. AD 1600 is observed from the vegetation cover estimates based on the pollen record of Lake Storsjön (Ning et al., 2018). Relatively high LAR in the sediment of Lake Storsjön during this period reflects increased export of terrestrial organic matter (Fig. 11).

From ca. AD 1700 and onward, land uses intensified in both the western and eastern study regions, which are indicated by the pollen-based vegetation cover estimates of Lake Skottenesjön and Lake Storsjön (Ning et al., 2018; Yang et al., 2020). In the Skogaryd catchment, the conifer-dominated forest was gradual-



**Figure 11** Mass accumulation rates of carbon (CAR), lignin (LAR), total organic carbon content (TOC), Carbon/nitrogen ratios (C/N), lignin data and land-cover reconstruction based on the pollen of the sediment record from Lake Storsjön. A<sub>8</sub>: sum of vanillyl, syringyl and cinnamyl phenols normalized to 100 mg organic carbon; PON/P: ratios of *p*-hydroxyacetophenone to total *p*-hydroxyl phenols; S/V: ratios of syringyl to vanillyl phenols; C/V: ratios of cinnamyl to vanillyl phenols; (Ad/Al)<sub>v</sub>: ratios of vanillic acid to vanillin; P/(S+V): ratios of *p*-hydroxyl to sum of vanillyl and syringyl phenols. The pollen-based land-cover reconstruction is based on Ning et al. (2018).

ly cleared from the early 18th century due to the high demand for charcoal and wood fuel for a steel factory (Yang et al., 2020). The large increase in the deposition of Ti reflects substantially increased erosion due to the intensified logging in the catchment (Yang et al., 2020). Soils may lose the capacity of organic carbon storage when the forest is cleared, which can result in an elevation of organic carbon export (Eglin et al., 2010; Sulman et al., 2020). The significant increase in LAR of the lake sediment of Lake Skottenesjön between ca. AD 1700 and 1900 reflect a remarkable increase in terrestrial organic matter deposition (Fig. 9). The increase is also observed in LAR in the sediment of Lake Storsjön from ca. AD 1700 (Fig. 11). The CAR and LAR are strongly correlated in each of the lakes ( $r^2=0.8976$ ,  $p<0.01$  for Skottenesjön and  $r^2=0.9946$ ,  $p<0.01$  for Lake Storsjön), suggesting that the organic carbon accumulation in the two lakes was highly controlled by terrestrial sources.

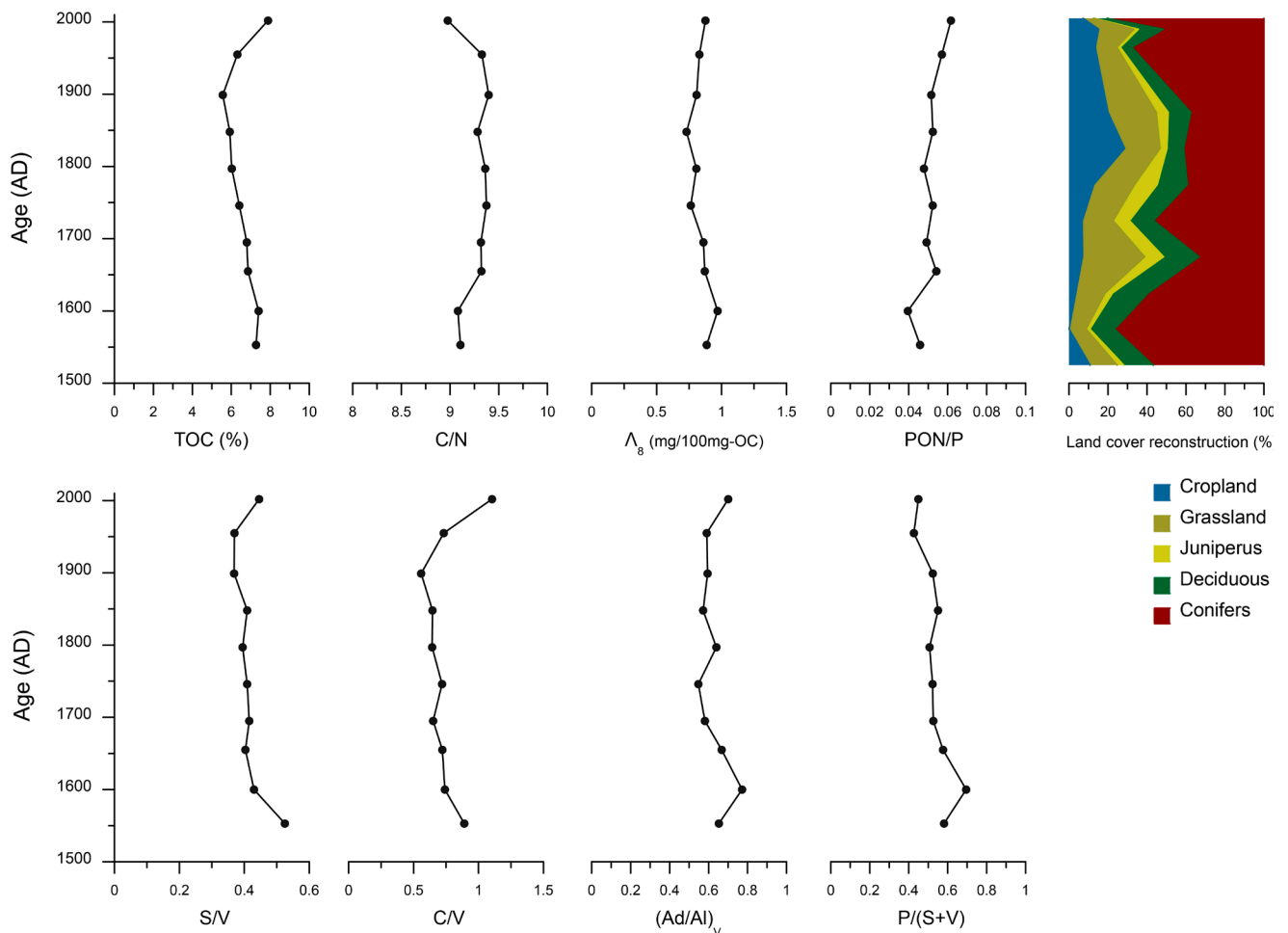
In the 19th century, parts of the catchment in Skogaryd were drained through a system of ditches to transform wetlands to farmland and increase forest productivity, and a major ditch was conducted to drain the wetland Följemaden into the western basin of Lake Skottenesjön in the 1870s (Fig. 2; Hill, 1999). The ditching led to a massive increase in the export of terrestrial organic matter as indicated by the LAR. In addition to the terrestrial organic matter, the enhanced soil erosion likely led to an increase in nutrients input to the lake, which may have promoted the aquatic primary productivity. The BSi flux was rather low and stable between ca. AD 1700 and 1900, suggesting little change in diatom production (Fig. 9). However, the  $\delta^{13}\text{C}$  shows higher values from AD 1700, which probably related to the enhanced aquatic primary productivity (Li et al., 2008). The  $\delta^{13}\text{C}$  reached the maximum and the BSi increased rapidly after the ditching, reflecting a variation in the nutrient supply that contributed to a further elevation of the aquatic primary production.

Between ca. AD 1700 and 1900, the land use in the eastern study region was characterized by the expansion of agriculture instead of wood harvesting (Ning et al., 2018). The deposition of terrestrial organic matter in Lake Storsjön indicated by LAR increased in the 18th century, which is likely associated with the enhanced soil erosion due to increased openness in the landscape (Fig. 11). The agricultural activities in the region reached the maximum in the early 20th century as indicated by the vegetation cover estimates (Ning et al., 2018). The soil erosion intensified and led to a

substantial increase in the terrestrial organic matter input to Lake Storsjön, which is evidenced by the large increases in LAR and C/N ratio (Fig. 9).

The land use in large parts of the western study region was converted to the conifer-dominated managed forest from ca. AD 1900 (Yang et al., 2020), which follows the general land-use history in Sweden (e.g. Björkman, 2001; Mazier et al., 2015; Åkesson et al., 2015; Ning et al., 2018). The land use in the eastern study region was converted to managed coniferous forest from ca. AD 1950 (Ning et al., 2018). When the landscapes were reforested, the geochemical proxies of the lake sediments from the two lakes indicate reduced soil erosion. In Lake Skottenesjön, the decrease in Ti from ca. 1900 suggests a decline in the minerogenic matter delivery from the catchment in response to the reforestation (Fig. 9). In Lake Storsjön, the large increase in TOC probably resulted from less dilution from minerogenic matter (Fig. 11). However, the level of terrestrial organic matter exported to the aquatic ecosystem did not return to the condition before intense human impacts. In Lake Storsjön, the terrestrial organic matter export from the catchment, indicated by the LAR, continued to increase until around fifty years after the establishment of the managed forest. The LAR decreased from ca. AD 2000, but the values were much higher than they were before the 18th century (Fig. 11). Although the terrestrial organic matter export to Lake Skottenesjön tended to decrease shortly after the reforestation on large parts of the catchment, the accumulation of lignin-derived organic matter indicated by LAR in the sediment was also much higher than it was when the catchment was mostly covered by natural forest with minor human disturbance (Fig. 9).

The substantial drops in C/N ratios of the sediments from Lake Skottenesjön and Lake Storsjön in the late 19th century were likely attributed to significant input of inorganic nitrogen from artificial fertilizers. The extensive use of artificial fertilizers led to a significant increase in the aquatic primary production indicated by BSi flux in sediments from the two lakes. Hence, both allochthonous and autochthonous organic matter deposition contributed to the relatively high total organic carbon accumulation in the sediments. Additionally, the  $\delta^{13}\text{C}$  in the sediment of Lake Skottenesjön decreased by 5‰ from ca. AD 1950 to 2000 (Fig. 9). A similar change has been observed in the sediment from White Lake in northwestern New Jersey, USA (Li et al., 2008). The organic respiration in soil increased as the landscape was reforested and the soil carbon started



**Figure 12** Total organic carbon content (TOC), carbon/nitrogen ratios (C/N), lignin data and land-cover reconstruction based on the pollen of the sediment record from Gåsfjärden.  $\Lambda_g$ : sum of vanillyl, syringyl and cinnamyl phenols normalized to 100 mg organic carbon; PON/P: ratios of *p*-hydroxyacetophenone to total *p*-hydroxyl phenols; S/V: ratios of syringyl to vanillyl phenols; C/V: ratios of cinnamyl to vanillyl phenols; (Ad/Al)<sub>v</sub>: ratios of vanillic acid to vanillin; P/(S+V): ratios of *p*-hydroxyl to sum of vanillyl and syringyl phenols.

to accumulated (Richter et al., 1999). The increasing organic respiration in the soil led to a decrease in the  $\delta^{13}\text{C}$  of the dissolved carbon source in the groundwater, which subsequently cause the shift in the  $\delta^{13}\text{C}$  of the organic matter in the lake (Richter et al., 1999; Li et al., 2008). In the late 20th century, local clear-cuttings in the managed forest are revealed in the vegetation cover estimate of Lake Skottenesjön (Yang et al., 2020). As the consequence of intensive deforestation within a quite short period, a sharp increase is observed in the terrestrial organic matter export to the lake, and the increase was nearly twice the amount of lignin-derived organic matter increase during the wood-harvesting period between AD 1700 and 1900 (Fig. 9).

## 5.2 Lignin signatures in aquatic systems with different characteristics

The degradation state of lignin can be traced by a series of lignin parameters including (Ad/Al)<sub>v</sub> and P/

(S+V) ratios (Ertel and Hedges, 1984; Opsahl and Benner, 1995; Dittmar and Lara, 2001). Typically, fresh plant material has (Ad/Al)<sub>v</sub> less than 0.3 (Opsahl and Benner, 1995), and the sedimentary plant fragments have (Ad/Al)<sub>v</sub> above 0.3 (Hedges et al., 1988b). The average of (Ad/Al)<sub>v</sub> in the sediments from Lake Skottenesjön and Lake Storsjön is above 0.6, indicating that the lignin in our lake sediments is highly degraded (Fig. 10 & 11). The early diagenesis of lignin during the deposition in the lakes could potentially cause bias on the indication of the origin of lignin. However, studies suggest that degradation of the lignin fraction of terrestrial organic matter within the water column is insignificant (Farella et al., 2001). Besides, diagenesis ceases very soon after the burial of terrestrial organic matter in the sediments as conditions become anoxic a few millimetres below the sediment surface (Maerki et al., 2006; Sobek et al., 2009; Chmiel et al., 2015). Additionally, the (Ad/Al)<sub>v</sub> and P/(S+V) ratios do not show an increasing downward trend in any of the sediment



profiles, suggesting no or only minor post-depositional degradation. Therefore, lignin phenols signatures in the sediment records studied here are considered to retain information on the source and degradation state of terrestrial organic matter before reaching the lakes. In addition to the degree of degradation,  $(Ad/Al)_V$  ratios are also influenced by the relative abundance of soil organic matter versus plant fragments in sediments (Tareq et al., 2011). The relatively high  $(Ad/Al)_V$  ratios in the sediments of the studied lakes from ca. AD 1700 onwards probably indicate increased soil organic matter input that was likely caused an increased human disturbance in the forest and a general opening of the landscape (Fig. 10 & 11). The P phenols are not exclusively produced by vascular plants, as *p*-hydroxybenzaldehyde and *p*-hydroxybenzoic acid can be derived from protein-rich organisms, such as plankton (Hedges et al., 1988a). Hence, the variation of  $P/(S+V)$  ratio can be driven by changes in the input of organic matter originating from phytoplankton. The lack of relationship between  $P/(S+V)$  and  $(Ad/Al)_V$  ratios and the negative relationship between  $P/(S+V)$  and  $PON/P$  ratios in the sediment of Lake Storsjön suggest that the  $P/(S+V)$  ratios here are affected by the input of aquatic organic matter, and do not necessarily provide information about the organic matter degradation (Fig. 11). Also, *Sphagnum* mosses lack V and S phenols but are rich in P phenols, and hence could exhibit high  $P/(S+V)$  ratios (Gross, 1979; Williams et al., 1998; Zacccone et al., 2008). The elevated  $P/(S+V)$  ratios in the sediment from the western basin of Lake Skottenesjön during the last three centuries may be partly derived from enhanced input of organic matter originating from *Sphagnum* mosses in response to the drainage and disturbance of peatlands including Följemaden, which is consistent with a substantial increase in the deposition of *Sphagnum* spores (Fig. 10).

In the sediments of Gåsfjärden, there is a similar trend between the  $(Ad/Al)_V$  and  $P/(S+V)$  ratios (Fig. 12), suggesting that the variations in both proxies can provide information about lignin degradation. Although both proxies exhibit relatively high values at the bottom part of the sediment profile, the lack of downward decreasing trend in  $\Lambda_8$  and C/V ratios suggests the absence of the downcore degradation. The  $(Ad/Al)_V$  ratio in the sediment from Gåsfjärden has an average of 0.6, which is less than in the sediments from Lake Skottenesjön and Lake Storsjön. The degrees of degradation of lignin indicated by  $(Ad/Al)_V$  ratios were thus lower in the sediments of Gåsfjärden than Storsjön, reflecting that the terrestrial organic matter buried in Gåsfjärden

was less degraded. This difference may reflect processes that affect the terrestrial organic matter during transportation to the coast. Both physical and chemical processes during transportation alter the terrestrial organic matter, so some of the particulate terrestrial organic matters exported from inland are lost on the way to the coastal area (Bauer et al., 2013). Our data would then indicate that the material liable to degradation is preferentially removed, which could result in a situation where more resistant organic matter is reaching the coastal region that will appear to be less degraded.

The  $\Lambda_8$ , which is the sum of V, S and C phenols normalized to 100 mg organic carbon, is an indicator of the relative abundance of terrestrial plant-derived organic matter in the TOC content of lake sediments (Hedges and Mann, 1979).  $\Lambda_8$  of the sediment cannot reflect the changes in the amount of lignin-derived organic matter delivery, as it is always affected by the deposition of the organic matter originating from non-vascular plants. In Lake Skottenesjön, the relatively high proportion of lignin phenols in the total organic carbon deposition indicated by  $\Lambda_8$  of core A during the medieval expansion period was likely the result of the export of lignin-rich terrestrial organic matter during the transition from lignin-rich forest soils to lignin-poor farmland soils (Fig. 10). From ca. AD 1700 and onwards,  $\Lambda_8$  in core A shows relatively low and stable values while LAR increases substantially. The low  $\Lambda_8$  was concurrent with the increases in both *Sphagnum* spores deposition and  $P/(S+V)$  ratio, indicating that  $\Lambda_8$  was affected by the dilution effect of the *Sphagnum*-derived organic matter. In the 19th century, the  $\Lambda_8$  in the sediment from Lake Storsjön decreased while farmland continued expanding (Fig. 11). The expansion of farming and grazing likely contributed to increased nutrient input to the lake, which subsequently caused enhanced aquatic primary production. The decreasing proportion of lignin-derived organic matter indicated by  $\Lambda_8$  was probably partly attributed to an increase in the autochthonous input relative to the allochthonous input. This interpretation is further supported by a decrease in the  $PON/P$  ratios during the same period (Fig. 11), likely indicating an increase in the relative abundance of lignin-free organic matter originating from phytoplankton in the sediment.

The  $\Lambda_8$  in the sediment of Lake Storsjön (1.5 mg/100 mg-OC) is much lower than it is in the two sediment sequences from Lake Skottenesjön (2.5 mg/100 mg-OC in core A, 3.4 mg/100 mg-OC in core B) on average between ca. AD 1500 and 2010. The land use in

the catchment of Skottenesjön was characterized by wood-harvesting, while the catchment of Lake Storsjön was mainly used for agriculture. The higher content of lignin in the sediment of Lake Skottenesjön than the sediment of Lake Storsjön may suggest that compared to the farming and grazing, intensive logging could result in higher amounts of terrestrial organic matter export to lakes. Besides, it is suggested that the lake area and catchment area have effects on the terrestrial organic carbon export to lakes (Sepp et al., 2019; Toming et al., 2020). Therefore, the difference of the lignin content between our two lakes could also be associated with the difference in the physical properties of the catchment and morphometric characteristics of the two lakes.

In the sediments of Gåsfjärden,  $\Lambda_8$  had an average of 0.8 mg/100 mg-OC (Fig. 12), which is much lower than in the sediments from Lake Skottenesjön and Lake Storsjön. It has been reported that organic carbon is decreasing as it is transported from the inland to coastal area through river systems, and the causes include biotic and abiotic degradation, sedimentation, scavenging, and salinity-induced flocculation of both particulate and dissolved organic matter along the transport path (Bauer et al., 2013). This may explain why Gåsfjärden received lower amounts of terrestrial organic matter from the catchment than Storsjön. The sediment of Gåsfjärden had C/N ratios less than 10, which suggest that the organic matter was dominated by the aquatic production (Fig. 12; Meyers, 1994). Thus, the lower proportion of lignin in the total organic matter in the sediment of Gåsfjärden compared to Storsjön was also likely attributed to a relatively larger amount of aquatic organic matter input to the sediments. In addition, higher landscape openness in the coastal region near Gåsfjärden throughout the study period is indicated by the pollen-based land cover estimates (Fig. 12). Gåsfjärden might have received greater input of organic matter originating from leaves and grasses that are relatively richer in carbon but lower in lignin compared to woody tissues, which subsequently led to lower lignin content in the sediment (Routh et al., 2014).

The lignin phenol parameters S/V and C/V ratios are commonly used to estimate relative contributions of plant types (angiosperm vs gymnosperm) and tissue types (non-woody vs woody tissue) to the terrestrial organic matter component of sediments (Hedges and Mann, 1979). Several studies have shown positive correlations between S/V ratios and proportions of angiosperm plants in watersheds (Kuliński et al., 2007;

Teisserenc et al., 2010; Hyodo et al., 2017). In core A from Lake Skottenesjön, the S/V ratio is positively correlated with the cover of deciduous trees in the catchment ( $r^2=0.3525$ ,  $p<0.01$ ), suggesting that the sediment organic matter composition is affected by catchment vegetation dynamics (Fig. 10). However, the good correlations between the land-cover estimate and S/V ratios were not seen in core B or the sediments from Storsjön and Gåsfjärden. The better correlation in Lake Skottenesjön with its relative small basin is likely a result of a more direct supply of terrestrial organic matter (Kuliński et al., 2007). Lake Skottenesjön has a smaller catchment compared to Lake Storsjön. The small lake with small catchment receives terrestrial material from immediate surroundings, which makes the lignin composition in the sediment sensitive to the local environmental conditions (Kuliński et al., 2007). The lack of relationship between the land cover estimate and S/V ratios in core B in Lake Skottenesjön is likely due to the different lignin source areas of the two basins. The eastern basin receives materials from the catchment area of the inflowing stream on the east, while the western basin receives materials from the SRC on the north in addition to some materials from the eastern basin (Fig. 2). The stronger correlation between land cover estimates and S/V ratios in core A probably reflects that the lignin and pollen source areas for the western basin match each other well.

None of the C/V ratios in our sediments from three sites show any good relationship with the coverage of grassland indicated by the reconstructed vegetation cover. It is suggested that the C/V ratio is a useful indicator of palaeovegetation and climate change over the last hundreds of thousands of years (e.g. Castañeda et al., 2009; Tareq et al., 2011; Routh et al., 2014). Shifts of vegetation driven by climate change in the late Quaternary were far greater compared to the vegetation changes over the last 1000 years at our study sites. The lack of clear response of C/V ratios in our study compared to the studies on larger temporal and spatial scales was probably due to the extent of the shifts of vegetation between grassland and forest not being large enough. Also, C phenols can be generated by both leaves and grasses. A replacement of conifers by early successional deciduous trees, such as *Betula*, could increase the C/V ratios. The relatively high C/V ratios in both cores in Lake Skottenesjön after ca. AD 1700 were likely attributed to expansions of both grassland cover and early successional forest in response to intensified wood harvest (Fig. 10). Additionally, C phenols are more liable to degradation than S and V phenols. C/V

ratios might have lost some of the information about the origin during the long transport in a large catchment before reaching aquatic basins.

In Gåsfjärden, the S/V and C/V ratios in the sediment exhibit much less variability than in the sediments from the two lakes (Fig. 12). More variable vegetation cover around Gåsfjärden than around Storsjön during the last 500 years was revealed by the pollen-based land-cover estimates. Thus, the difference in variability of the S/V and C/V ratios between Gåsfjärden and the two lakes cannot be explained by differences in vegetation. The terrestrial organic matter sourced from the inland system has likely been sorted and altered through the long transportation, and subsequently the fraction reaching the coastal area and deposited in the sediments of the coastal marine basin is rather homogenized. Hence, the variability of terrestrial organic matter deposition in the coastal marine water basin seemed less sensitive to the changes in vegetation composition in the catchment compared to inland lakes.

## 6 Summary and conclusions

- The Skogaryd area was sensitive to variations in the extent and intensity of human activities. Between ca. AD 950 and 1350, the local land use was characterized by small-scale agricultural activities associated with the Medieval expansion. From ca. AD 1350, much of the farmland was abandoned and coniferous woodland cover expanded as a consequence of population decline due to plague outbreaks. In the 1600s, agricultural land-use expanded again in response to the population recovery. Between ca. AD 1700 and 1850, abandonment of agriculture and intensified wood-harvesting are observed, which can be attributed to the high demand for fuel and labour for a local steel factory set up in the early 1700s and active until the late 1800s. Artificial drainage was established to create more arable land to sustain the population in the late 19th century when the population in the area reached a maximum. From the 20th century, forestry management started with the plantations of *Picea* and *Pinus*.
- Land-use types in the catchment have different effects on soil erosion and terrestrial matter export. The agriculture in the Medieval expansion caused an increase in minerogenic material input to the

lake, but little change in terrestrial organic matter export, which was probably because medieval agricultural practices were less advanced than in modern times with less intense tillage. Artificial drainage in modern times caused a great amount of terrestrial organic matter input to the lake and subsequently elevated the organic carbon accumulation in the sediment. Compared to agricultural activities, intensive wood harvest led to more enhanced soil erosion and a substantial increase in the supply of terrestrial organic matter to the lake. Soil erosion reduced within decades after reforestation. However, compared to the period with minor human disturbance in the forest, a higher amount of terrestrial organic and minerogenic matter was delivered to the lake during the period of modern forestry.

- The comparison of lignin phenols content from two sediment cores in the same lake shows that the terrestrial organic matter delivery is sensitive to spatial variability within the catchment.
- The composition of lignin-derived organic matter in the sediment of lakes varied in response to vegetation cover changes in the watershed. However, the composition of lignin-derived organic matter deposited in Gåsfjärden was not sensitive to the variation of vegetation cover, which is likely due to sortation and alteration of the terrestrial organic matter during the transportation from the inland to the coast. Thus, caution is needed when using lignin phenols in marine sediments as proxies to reflect the vegetation composition in the catchment.
- This study highlights the potential of the combined use of lignin phenols and pollen-based quantitative land-cover reconstructions for the investigation of long-term changes in terrestrial organic matter export to aquatic ecosystems.
- The outcomes of this study provide a better understanding of the impacts of land-use changes on carbon cycles from a long-term perspective, which are fundamental for the development of strategies for environmental management.

# Svensk sammanfattning

Mänskliga aktiviteter har stor inverkan på kolcykeln genom att ändra landskapet. Förändringar av vegetationstäcket till följd av skogsröjning och expansion av jordbruksmark ökar markerosion och transport av organiskt material och andra näringsämnen från land till akvatiska ekosystem.

Rekonstruktioner av tidigare miljövariationer på hundra- till tusenårsskalor är väsentliga för bedömningar av ekosystemens reaktioner på mänskliga störningar av miljön. Sådana rekonstruktioner kan erhållas från väl daterade sjösediment, som består av kronologiskt avsatta material som härstammar både från vattenmiljön (t.ex. organiskt material från makrofyter och planktonalger) och avrinningsområdet (t.ex. organiskt jordmaterial, växtrester, pollen och minerogent material). Mätningar av biologiska och kemiska variationer i kontinuerliga sedimentsekvenser gör det möjligt för oss att rekonstruera och undersöka ekosystemens respons på mänskliga aktiviteter över hundratals till tusentals år.

I den här avhandlingen presenteras kemiska och biologiska analyser av fyra sedimentsekvenser från olika miljöer i södra Sverige för att utforska variationen i transport av organiskt material mellan mark- och vattenmiljöer som resultat av långvariga förändringar av markanvändningen. Den pollenbaserade "Landscape Reconstruction Algorithm" (LRA) tillämpades för att kvantitativt rekonstruera vegetationsdynamiken i landskapet. Ligninfenoler (biomarkörer) användes för att spåra det terrestriska organiska materialet som bevarats i sedimenten. Bulkgeokemi, inklusive totalhalt av organiskt kol och kväve, samt biologiskt producerat kisel, användes för bedömning av den akvatiska primärproduktionen och andelen material med terrestriskt ursprung i sedimenten.

Resultaten från två sedimentssekvenser från en liten skogsjö (Skottenesjön) i sydvästra Sverige visar att den terrestriska organiska exporten till sjön är känslig för lokala markanvändningsvariationer i avrinningsområdet under de senaste 1000 åren. Förhöjd markerosion och ökad export av organiskt material till sjösedimenten registrerades under en period med intensiv avverkning på 1700- och 1800-talet. Ingen signifikant förändring av transporten av organisk material observerades under jordbruksmarkens expansion mellan 1100- och mitten av 1300-talet. Exporten av markbundna organi-

ska och minerogena ämnen till sjön var mycket högre under modernt skogsbruk på 1900-talet än i perioder med mindre skogsstörningar under 1100-talet.

En liknande studie genomfördes av sedimentsekvenser från en stor sjö (Storsjön) och en fjärd i Östersjön (Gåsfjärden) på Sveriges östkust. De två platserna ligger inom samma avrinningsområde och är förbundna med ett system av vattendrag. Resultaten visar att halten av ligninfenoler som deponerats i Gåsfjärden är mindre känslig för variationer i vegetationstäcket än i Storsjön. Koncentrationen av ligninhaltigt organiskt material är mycket lägre i Gåsfjärden än Storsjön, eftersom den organiska materialavsättningen i Gåsfjärden domineras av akvatisk biomassa. Dessutom är det terrestriska organiska materialet som är bevarat i Gåsfjärden mindre nedbrutet än i Storsjön, troligtvis för att det mer lättnedbrytbara organiska materialet har gått förlorat under transporten till havet.

Denna studie lyfter fram potentialen med att använda ligninfenoler i kombination med pollenbaserade kvantitativa vegetationsrekonstruktioner för undersökning av de processer som styr förändringar i transporten av organiskt material från den terrestra miljön till akvatiska ekosystem. Resultaten av studien ger en bättre förståelse för långsiktiga mänskliga effekter på den organiska kolcykeln, vilket är grundläggande för utvecklingen av strategier för att hantera framtida klimat- och miljöförändringar.



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