The role of sediments in the carbon cycle of boreal lakes

HANNAH ELISA CHMIEL
Abstract

Inland waters are active sites of carbon (C) processing and emitters of carbon dioxide (CO₂) and methane (CH₄) to the atmosphere. In the boreal zone, where surface waters receive large quantities of organic carbon (OC) from surrounding forests and wetlands, lakes and streams act as strong sources of these greenhouse gases. Lake sediments provide the only long-term sink of C in boreal inland waters, through burial of OC. However, mineralization of OC counteracts the efficiency of lake sediments in removing C from the short-term C cycle. In this context, this thesis provides a better insight into the dual role of boreal lake sediments as C source and C sink.

The presented work is based on empirical assessments of OC burial and OC mineralization rates in boreal lakes. The temporal variability of OC burial and the stability of the buried OC was assessed on both centennial and millennial timescales. The quantitative importance of sediment OC burial and mineralization in comparison both to other C fluxes within the lake, and to C fluxes within the tributary stream network, was quantified. By simulating the effect of climate change on water temperature, we also gauged the potential future efficiency of lake sediments in storing C.

The results demonstrate that OC mineralization in sediments dominates three-fold over OC burial when observed at a whole-basin and annual scale. The contribution of sediment OC mineralization to annual C emission from the assessed study lake was, however, found to be small (16%), when compared to OC mineralization in the water column (37%) and catchment import of C (47%). Furthermore, C emission from headwater streams was found to dominate greatly over the lake C emission, mainly triggered by the higher gas transfer velocity of streams compared to lakes.

On a long-term (Holocene) scale, the continuous OC burial flux results in a large amount of C stored in sediments. The temporal variability of this OC accumulation was found to vary across lakes, with, however, time-dependent patterns: On a millennial scale, smaller lakes exhibited a higher variability than larger lakes of the study area. For the last century, similar variability and a trend to increased OC accumulation was found for most study lakes, irrespective of their size. Analysis of lignin phenols in the accumulated OC did not indicate post-depositional degradation, independent of the age of the sediment OC, implying that sediments are a very stable sink for land-derived OC in boreal lakes.

Simulation of warming water temperatures in boreal lakes resulted in declines of the OC burial efficiency BE (OCBE; OC burial/OCdeposition) up to 16%, depending, however, on basin morphometry. Predicted declines in OCBE were higher for the more shallow lake compared to the deeper lake.

In conclusion, this thesis illustrates that sediments play, despite a small quantitative impact on aquatic C cycling, an important role as a very stable C sink in boreal lakes. However, the efficiency of this C sink is likely to be reduced in the future.

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To my family
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.


IV  Chmiel, H. E., Natchimuthu, S., Bastviken, D., Ferland, M.-E. & Sobek, S. Decreased efficiency of sediment carbon burial in boreal lakes at warming lake water temperatures. *Manuscript*

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Additional Papers

In addition to the thesis chapters, I have contributed to the following papers:


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Abbreviations

Ad/Al  Acid-to-aldehyde ratio
CMAR  Carbon mass accumulation rate
C/V   Cinnamyl phenol-to-vanillyl phenol ratio
DIC   Dissolved inorganic carbon
DO    Dissolved oxygen
DOC   Dissolved organic carbon
GPP   Gross primary production
IRGA  Infrared Gas Analyzer
LPVI  Lignin Phenol Vegetation Index
NEP   Net ecosystem production
OC    Organic carbon
OCBE  Organic carbon burial efficiency
$pCO_2$ Partial pressure of carbon dioxide
POC   Particulate organic carbon
Q     Discharge
R     Respiration
S/V   Syringyl phenol-to-vanillyl phenol ratio
Introduction

Boreal lakes and carbon cycling

The boreal zone covers about 13% of the continental area on earth (Schultz, 2013) and has an important function in the climate system acting as a terrestrial carbon (C) sink (Pan et al. 2011). Boreal forests remove carbon dioxide (CO$_2$) from the atmosphere and transform it into biomass. The turnover time of this biomass, i.e., the time until organic carbon (OC) is transformed back to CO$_2$ or methane (CH$_4$), depends on various factors, such as respiration in plants or soils, or the release during wildfires, but also on the interplay between the terrestrial and the aquatic C cycle (Cole et al. 2007; Tranvik et al. 2009).

Boreal lakes and streams receive, in addition to their internal (autochthonous) OC production, large quantities of terrestrial (allochthonous) OC from the surrounding catchment (Birge and Juday 1927). The largest share (90-95%) of this allochthonous OC is present in form of dissolved organic carbon (DOC), while particulate organic carbon (POC) forms the smaller fraction (Wetzel, 2001). Allochthonous DOC is mostly derived through leaching of OC from organic-rich soil horizons of forest and wetlands, and is responsible for the brownish colour of boreal surface waters (Rasmussen et al. 1989).

When allochthonous OC enters boreal lakes, it fuels the metabolism of heterotrophic microorganisms in the water column (i.e., bacterioplankton), which results in the production of CO$_2$ (Tranvik 1988). Aquatic primary producers, such as phytoplankton, partially transform the CO$_2$ into biomass again, however, the dark colour of allochthonous DOC strongly inhibits photosynthesis in boreal lakes (Karlsson et al. 2009). Therefore, the majority of boreal lakes are net heterotrophic ecosystems, in which respiration is higher than primary production (Del Giorgio and Peters 1994; Jansson et al. 2000). Besides microbial degradation, OC can be mineralized photochemically by sunlight, either directly to CO$_2$ or by cleavage of structurally complex organic macromolecules to readily bioavailable molecules that stimulate bacterial production (Granéli et al. 1996; Bertilsson and Tranvik 1998). Microbial and photochemical degradation of OC is one of the reasons why most lakes of the boreal zone are supersaturated with CO$_2$ and emit CO$_2$ to the atmosphere (Sobek et al. 2003, 2005). Another reason is dissolved CO$_2$ entering lakes via surface water or ground water inflow (Stets et al. 2009). Also for boreal
streams, high CO₂ emissions were reported and largely attributed to the injection of CO₂ derived from soil respiration via groundwater inflow to the stream (Öquist et al. 2009; Wallin et al. 2013). Hence, the release of CO₂ from the entire aquatic continuum, i.e., both lakes and streams, is an important component of the boreal aquatic C cycle (Crawford et al. 2014) and counteracts the terrestrial C sink.

At the same time as CO₂ is emitted from lakes and streams to the atmosphere, OC is stored in lake sediments over long time scales.

The role of lake sediments for carbon cycling

In lakes, organic and inorganic particles of different origin settle down to the lake bottom, where they form sediments. These sediments play a dual role in the aquatic C cycle since they act both as a source and sink of C. The deposition of organic matter at the lake bottom results, on the one hand, in burial of OC, which removes C from the active short-term C cycle. Hence, sediments represent a long-term C sink. On the other hand, ongoing microbial degradation in the sediments prior to burial results in mineralization of OC to CO₂ and CH₄, which are recycled again in the lake or emitted to the atmosphere (Figure 1).

In the boreal zone, it has been shown that lake sediments store more C than the surrounding forest soils and biomass (Kortelainen et al. 2004). The transport of POC to boreal lakes certainly contributes to sediment OC burial, and it seems that in-lake flocculation of DOC to POC is an additional important source of sediment OC in boreal lakes (von Wachenfeldt and Tranvik 2008).

Figure 1. Carbon fluxes and processes in the aquatic C cycle of lakes.
The production of CO$_2$ and CH$_4$ in sediments depends on different factors that act on OC mineralization: The source of the organic material (i.e., autochthonous or allochthonous origin) influences mineralization rates (Sobek et al. 2009) as autochthonous OC is more easily degraded by microorganisms than allochthonous OC (Burdige 2007). Furthermore, temperature strongly controls OC mineralization rates in sediments, with warmer temperatures stimulating higher mineralization rates (Bergström et al. 2010; Gudasz et al. 2010). Additionally, mineralization rates and the production of CH$_4$ depend on the presence and absence of oxygen: Under anoxic conditions, degradation rates typically slow down, lowering the production rates of CO$_2$ (Zehnder and Svensson 1986) while methanogenesis occurs. In lakes, CH$_4$ typically has a small contribution in C units, however, CH$_4$ is a 28-fold more powerful greenhouse gas than CO$_2$ on a 100 year scale (Intergovernmental Panel on Climate Change, IPCC, 2013) and therefore important to consider with respect to climate.

To quantify this dual function of lake sediments as C source and sink, the OC burial efficiency (OCBE, %) is calculated. It is defined as the ratio of OC burial:OC deposition onto the sediment surface, and hence represents the fraction of OC that remains permanently in the sediments after deposition. Since the OCBE of sediments is intimately linked to OC mineralization (OC that is not mineralized is buried), it depends on the same factors, i.e., temperature, organic matter sources and the exposure time to oxygen, but also on the sediment accumulation rate (Burdige 2007; Sobek et al. 2009).

**Knowns and Unknowns**

The role of lake sediments for aquatic C cycling has been investigated in a multitude of studies (e.g., Mulholland and Elwood 1982; Molot and Dillon 1996; Kortelainen et al. 2004, 2013; Algesten et al. 2005; Sobek et al. 2009; Bergström et al. 2010; Gudasz et al. 2010; Ferland et al. 2012; Fenner and Freeman 2013). Some of these studies have focused on regulatory factors of OC mineralization and preservation in boreal lakes. For instance, the temperature dependence of OC mineralization rates in lake sediments was investigated in a literature survey and exemplified for boreal lakes of different trophic state (Gudasz et al. 2010). Also, the exposure time of oxygen and anoxic conditions in lakes have been highlighted as key factors for OC preservation in boreal lake sediments (Sobek et al. 2009; Fenner and Freeman 2013).

Other studies have explored quantitative aspects of C fluxes at the lake bottom, in order to gauge the importance of sediments for aquatic C cycling. For example, a large-scale study on Finnish lakes concluded that OC mineralization in sediments was an important driver for CO$_2$ emission (Kortelainen et al. 2006). However, an investigation of sediment OC mineralization
in boreal lakes during summer, demonstrates that this is not always the case (Algesten et al. 2005). Also, studies on the OCBE have come to divergent results. In a survey of sediments, which were investigated at the deepest point in lakes, it was found that boreal lake sediments had a comparatively high OCBE (range, 45-67%; Sobek et al. 2009), while a study on the OCBE in boreal lake sediments in Québec revealed a much wider range (4-62%), by accounting for the spatial variability in sediment deposition across lake basins (Ferland et al. 2014). Hence, dissimilar findings of studies that investigate sediment C fluxes in lakes may reflect the difference in spatial and temporal scale on which these studies were carried out (Hobbs et al. 2013). For better comparison between studies, and a comprehensive assessment of the role of lake sediments in the aquatic C cycle, it is therefore necessary to integrate spatial and temporal patterns in sediment-, lake-, and catchment-scale C fluxes.

Furthermore, it is important to look at different time scales, when assessing OC accumulation in lake sediments. Studies that address the temporal variability in OC accumulation mostly focus on trends in the recent past, i.e., the last century (Anderson et al. 2013; Dietz et al. 2015). The variability in OC accumulation on long-term scales, i.e., over millennia of the Holocene is, however, often neglected. Instead, when addressing Holocene-scale OC accumulation it is mostly referred to it as mean values, and less is known about the variability (Anderson et al. 2009; Kastowski et al. 2011; Kortelainen et al. 2013). Long-term variations in OC burial, however, might provide important insights into the functioning and evolution of lake sediments as a C sink, in response to past environmental change.

Also, the source of the buried OC is important, particularly in terms of how the lake sediment C sink is accounted. If the buried OC originates from land (i.e., is allochthonous) it may simply be viewed as soil C that was transported to the lake bottom, and may therefore not be accounted as a new sink. If the buried OC was produced within the lake, i.e., is autochthonous, it would represent a new C sink. There are contrasting views on the source of buried in OC in boreal lakes (Dean and Gorham 1998; Gudasz et al. 2012). More specific knowledge on the different source types of terrestrial OC in sediments and how their contribution changed over time is needed. In addition, there are indications that lake sediment OC can continue to degrade for hundreds of years after deposition, albeit, at very low rates (Sobek et al. 2014). For boreal lakes, however, the long-term stability of the buried sediment OC is presently not well understood.

Apparently, important gaps remain in our understanding of lake sediments as sources and sinks of C.
Aims of the Thesis

This thesis aims at gaining more insights into the role of lake sediments in the C cycle of boreal lakes. Thereby, the main focus is on assessing C fluxes on integrated spatial and temporal scales, in order to put sediment C fluxes in perspective to lake and catchment-scale C cycling. In addition, the thesis addresses the role of lake sediments as past and future C sink.

More specifically, the different thesis chapters focus on:

1) the temporal variability of OC accumulation in boreal lake sediments over the past 10,000 years, and the stability of the sediment C sink (Paper I),

2) the contemporary role of lake sediments as C source and sink in the annual C balance of boreal lakes (Papers II and IV),

3) the role of C loss and transport in the headwaters of a small boreal lake in relation to lake-internal C cycling (Papers II and III),

4) the future role of lake sediments as C sink in a warmer climate (Paper IV).
Methods

Study sites

The study lakes and streams (Table 1) are located in the boreal zone of central Sweden (Paper I-IV) and southwestern Sweden (Paper IV). They are small sized systems (<2 km²) with medium to high DOC contents (range, 10-28 mg L⁻¹), brownish water colour, and a glacial origin some 8,000-10,000 years ago (Lundqvist 1986).

Table 1. Investigated lakes and streams in Papers I-IV.

<table>
<thead>
<tr>
<th>Paper</th>
<th>Study site</th>
<th>Surface Area km²</th>
<th>Location</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lake</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td>Dagarn</td>
<td>1.72</td>
<td>59°54'</td>
</tr>
<tr>
<td>I</td>
<td>Övre Skärsjön</td>
<td>1.65</td>
<td>59°51'</td>
</tr>
<tr>
<td>I</td>
<td>Oppsveten</td>
<td>0.65</td>
<td>60°01'</td>
</tr>
<tr>
<td>I</td>
<td>Lilla Sångaren</td>
<td>0.24</td>
<td>59°54'</td>
</tr>
<tr>
<td>I</td>
<td>Grästjärn</td>
<td>0.09</td>
<td>59°53'</td>
</tr>
<tr>
<td>I I I I IV</td>
<td>Gäddtjärn</td>
<td>0.07</td>
<td>59°51'</td>
</tr>
<tr>
<td>I I I I IV</td>
<td>Erssjön</td>
<td>0.06</td>
<td>58°22'</td>
</tr>
<tr>
<td>I</td>
<td>Svarttjärn</td>
<td>&lt;0.01</td>
<td>59°53'</td>
</tr>
<tr>
<td>III</td>
<td>Prästjärn</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Kringeltjärn</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Svintjärn</td>
<td>&lt;0.01</td>
<td>59°52'</td>
</tr>
<tr>
<td>Stream</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>III</td>
<td>Gäddtjärn inlet 1</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Gäddtjärn inlet 2</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Gäddtjärn outlet</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Wetland outlet</td>
<td>&lt;0.01</td>
<td>59°51'</td>
</tr>
<tr>
<td>III</td>
<td>Svintjärn outlet</td>
<td>&lt;0.01</td>
<td>59°52'</td>
</tr>
</tbody>
</table>
Analysis of carbon mass accumulation rates

General approach

Carbon mass accumulation rates (CMARs; g C m\(^{-2}\) yr\(^{-1}\)) in sediments were calculated from the dry bulk density, the carbon content, and from sedimentation rates in vertical sediment core profiles according to the equation:

\[
CMAR = CC \times \rho_{bulk} \times SR
\]

where \(CC\) is the C content in mass %, \(\rho_{bulk}\) is the dry bulk density in g cm\(^{-3}\), and \(SR\) is sedimentation rate in cm yr\(^{-1}\).

Sampling, preparation, and analyses

To investigate CMARs over the Holocene (Paper I) sediment long-cores (1m) were sampled with a Livingstone corer from deep areas of the lakes with maximum sediment thickness. The cores were taken in overlapping sections and split and sliced into 1-5 cm thick subsamples. For determinations of CMARs rates over the past century (Papers I, II, and IV) sediment cores were sampled with a gravity corer (UWITEK) from the deepest point of the lakes. These cores (20 cm) were sliced into 0.5-1.0 cm increments. All subsamples were freeze-dried and homogenized for further analyses.

C contents, dry bulk density, and sedimentation rates

Carbon and nitrogen contents were measured on subsamples of both long- and short-cores, using an elemental analyzer (ECS 4010 Elemental Combustion System, CHNS-O).

To derive the dry bulk density of sediment in long-core samples (Paper I) we applied a core scanning technique before core splitting. This technique operates by measurements of gamma ray attenuation, which can be translated into the wet bulk density of the material, from which the dry bulk density was calculated. For measurements a GEOTEK Multi Sensor Core Logger (MSCL) at the Department of Geological Sciences at Stockholm University was used. The dry bulk density of sediment in short cores was determined manually on 1 cm\(^3\) subsamples.

Sedimentation rates in long-core samples were derived through radiocarbon \(^{14}\)C dating of macrofossils or bulk sediment samples taken at different core depths. The \(^{14}\)C ages were measured on chemically pre-treated samples by accelerated mass spectrometry (AMS) at the Ångstöm Laboratory at Uppsala University. Calibrations of \(^{14}\)C ages were performed using the IntCal09 calibration curve (Reimer et al. 2009). Sedimentation rates in short-
core samples were determined by lead ($^{210}$Pb) dating. The unsupported $^{210}$Pb activity was measured on chemically pre-treated subsamples using gamma spectrometry. Sedimentation rates were calculated by assuming a constant rate of supply of unsupported $^{210}$Pb to the sediment (Appleby and Oldfield 1978).

**Lignin phenol analysis**

The molecular composition of lignin phenols was analyzed in long-core sediment samples (Paper I). Lignin is the major component in the cell wall of vascular plants, and its phenolic composition indicates the source of OC, i.e., the plant type, and the degree to which the organic material is degraded. A set of lignin phenol parameters used to identify OC sources and degradation state is presented in Table 2.

**Table 2. Definitions of Lignin phenol parameters used to assess OC sources and degradation state in sediment samples .**

<table>
<thead>
<tr>
<th>Lignin parameter</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Source parameters</strong></td>
<td></td>
</tr>
<tr>
<td>$X_{\text{lignin}}$</td>
<td>sum of all vanillyl, syringyl, and cinnamyl phenols (mmol C/mol OC in sample)</td>
</tr>
<tr>
<td>$\Lambda_8$</td>
<td>sum of all vanillyl, syringyl, and cinnamyl phenols (mg phenol/100 mg OC in sample)</td>
</tr>
<tr>
<td>PON/P</td>
<td>$p$-hydroxyacetophenone/$p$-hydroxyl phenols (molar ratio)</td>
</tr>
<tr>
<td>S/V</td>
<td>syringyl phenols /vanillyl phenols (molar ratio)</td>
</tr>
<tr>
<td>C/V</td>
<td>cinnamyl phenols /vanillyl phenols (molar ratio)</td>
</tr>
<tr>
<td>LPVI</td>
<td>$= [S(S+1)/(V+1)+1] x [C(C+1)/(V+1)+1]$ whereby S, C, and V are in % of $\Lambda_8$</td>
</tr>
<tr>
<td><strong>Degradation state parameters</strong></td>
<td></td>
</tr>
<tr>
<td>$P/(V+S)$</td>
<td>$p$-hydroxyl phenols/sum of vanillyl and syringyl phenols (molar ratio)</td>
</tr>
<tr>
<td>Ad/Al)p</td>
<td>$p$-hydrobenzoic acid/$p$-hydrobenzaldehyde (molar ratio)</td>
</tr>
<tr>
<td>(Ad/Al)v</td>
<td>vanillic acid/vanillin (molar ratio)</td>
</tr>
<tr>
<td>(Ad/Al)s</td>
<td>syringic acid/syringaldehyde (molar ratio)</td>
</tr>
</tbody>
</table>
Lignin phenols were chemically extracted from sediment samples following the cupric oxide (CuO) oxidation method (Hedges and Erte 1982). The sample extracts were measured on an ultra performance liquid chromatography (UPLC) system (Waters Acquity UPLC), using a modified method after Lobbes et al. (1999).

**Sediment and water incubation experiments**

OC mineralization rates in water and sediment samples (Papers II and IV) were determined by laboratory incubation experiments following the experimental setup by Gudasz et al. (2010). Oxic mineralization rates were quantified as the change in DIC concentration in water and in water overlying sediment samples over time. DIC concentrations were measured on a Total Carbon (TC) Analyzer (Sievers 900). For the determination of anoxic mineralization rates the change in CH$_4$ concentration was additionally quantified, using a gas chromatograph (7890A GC system, Agilent Technologies) for quantifications. Both water and sediment samples were kept in temperature-controlled water chambers during the incubation periods to assure stable conditions and to assess OC mineralization rates at different temperatures.

**Sediment traps**

To quantify the sinking flux of OC in the water column (Paper II) sediment traps were deployed at the deep center of one study lake. The traps consisted of cylindrical, polycarbonate tubes, which were open at the top and closed at the bottom. The tubes were placed in tube holders that were deployed at 1 m below the water surface and at 1 m above the lake bottom. The tube holders were attached to a rope that was anchored in the sediment and held upright with a buoy. The traps material was collected about monthly during the ice-free study period and once after ice-melt. The trap samples were freeze-dried, homogenized and analyzed for C and N contents on an elemental analyzer (ECS 4010 Elemental Combustion System, CHNS-O).

**Sub-bottom profiling**

The morphometry of two study lakes (Papers II and IV) was mapped with a sub bottom profiler as described in Ferland et al. (2012). Briefly, the instrument simultaneously measures the interfaces of water-sediment and sediment bedrock. The difference between these two layers equals the sediment thickness.
For measurements, a triple beam sub-bottom profiler (BSS+3, Specialty Devices Inc.), was used that was suspended next to the boat in the surface water. Measurements were performed in about 5 m distances on transects across the study lakes and the data points were spatially interpolated.

Monitoring of lakes and streams

Water column monitoring and the diel DO technique
Temperature, dissolved oxygen (DO), pH, and conductivity were automatically and continuously monitored in the water column of two study lakes at the deepest point (Papers II and IV). Temperature sensors were deployed at 0.5-1.0 m increments throughout the water column, while DO, pH, and conductivity were measured in surface and bottom waters. Logging was performed over at least one annual cycle including the ice-cover period. Surface water DO concentration was logged at 20 min intervals, and the diurnal patterns of variability in DO were used to model gross primary production (GPP), net ecosystem production (NEP), and respiration (R) following Staehr et al. (2010).

Stream monitoring of discharge and CO₂
To calculate the load of C to and from one study lake (Paper III), discharge (Q), and pCO₂ were automatically monitored in the inlet- and in the outlet-stream. Q was measured using the salt dilution method (Day 1975) and correlated to water level that was automatically monitored with a pressure sensor. For pCO₂ a non-dispersive infrared sensor was used, which was covered with a polytetrafluoroethylene membrane that is impermeable to water but permeable to CO₂. Logging was performed during the ice-free season of one annual cycle. Automated measurements were complemented by repeated manual measurements of pCO₂, DOC, DIC, and POC at various locations in the headwaters of the study lake.

Lake warming simulations
In Paper IV, lake warming was simulated for two lakes to test the effect of higher water temperatures and altered stratification patterns on the OCBE of sediments. Using water column monitoring data of temperature and oxygen, we first assessed present-day conditions in the study lakes and quantified the basin-wide, annual OCBE. Lake warming simulation were performed according to regional climate change scenarios (Kjellström et al. 2014). These
scenarios predict changes in seasonal air temperatures for different regions in Sweden.

**Photochemical modeling**

In Paper II, the DOC photomineralization in lake water was simulated. Briefly, the DOC photomineralization was simulated as daily DIC production following the method and calculations described in detail in Koehler et al. (2014). For simulations the chromophoric dissolved organic matter (CDOM) absorption coefficient (m⁻¹) was determined for lake water from absorbance measurements. The apparent quantum yield of DIC photoproduction \( \Phi \) (mol C mol photons⁻¹) of the study was determined earlier in Koehler et al. (2014) and used here for simulations.

**Gas flux determination**

**General approach**

The diffusive flux \( F \) of CO₂ and CH₄ from water to the atmosphere can be determined by concentration measurements of the respective gas in water and in air and by the determination of the gas exchange coefficient \( k \) according to:

\[
F = k(C_{aq} - C_{eq})
\]

where \( F \) is the diffusive flux of CO₂ or CH₄ in mg C m⁻² d⁻¹, \( k \) is the gas exchange coefficient, also termed as the piston velocity, in m d⁻¹, \( C_{aq} \) is the concentration of the respective gas in water, and \( C_{eq} \) is the theoretical concentration of the respective gas in water if it was in equilibration with air. For lakes, \( k \) was derived using wind speed data and the empirical relationship after Cole and Caraco (1998). For streams, \( k \) was derived by tracer injections as described below.

**Headspace equilibration method**

The concentration of CO₂ in lake and stream water (Papers II and III) was determined by measurements of the partial pressure of CO₂ (\( p_{CO₂} \)) using an infrared gas analyzer (IRGA, EGM-3) and the headspace equilibration method according to Sobek et al. (2003). Briefly, water was sampled in polyethylene syringes and equilibrated with ambient air by shaking. After shaking the equilibrated air was analyzed on an IRGA. The \( p_{CO₂} \) of ambient air
was also measured during sampling campaigns to correct for the $p\text{CO}_2$ in equilibrated air samples. The $p\text{CO}_2$ and $\text{CO}_2$ concentration in water was calculated according to Weiss (1947) and using Henry’s constant.

Floating chamber method

$\text{CO}_2$ and $\text{CH}_4$ fluxes from lakes (Papers II and IV) were measured by floating chambers, which were placed in transects across the lake surface to cover different depth zones. Briefly, the flux of the respective gas was determined as the change in gas concentration in the chamber air over time. $\text{CO}_2$ chambers were equipped with $\text{CO}_2$ mini-loggers (CO2 Engine® ELG, SenseAir AB), and the flux was determined as the rate of change in $\text{CO}_2$ concentration over 30 minute measuring periods. $\text{CH}_4$ chambers were placed on the lakes for 24 h periods and the change in $\text{CH}_4$ concentration was determined from initial and final air samples that were extracted from the chambers by syringes and measured on a gas chromatograph (GC-FID; Shimadzu GC-8, PoropackN column).

Determination of $k$ in streams by propane injections

For determinations of $k$ in streams (Paper III) we used propane ($\text{C}_3\text{H}_8$) as a volatile tracer gas. Briefly, gas injections were performed at three stream reaches at different discharge ($Q$) conditions for which the reach travel time ($\tau$) was determined by the salt dilution method according to Day (1975). $\text{C}_3\text{H}_4$ was injected to the streams through an air curtain upstream of the sampling points and 10-15 min prior to sampling to achieve steady state conditions. Stream water samples were taken from upper and lower reach ends in polypropylene syringes and according to the travel time $\tau$. The headspace equilibration method described above, was applied to transfer $\text{C}_3\text{H}_8$ from water samples into air samples, which were measured on a gas chromatograph (7890A GC system, Agilent Technologies). The gas transfer coefficient $k$ for $\text{C}_3\text{H}_8$ was calculated according to Genereux and Hemond (1990) modified by Wallin et al. (2011), and the $k_{\text{CO}_2}$ was calculated following Jones and Mulholand (1998) and Wanninkhof et al. (1990).
Results and Discussion

Role of sediments for C cycling in a lake and catchment perspective

Burial and mineralization of OC in lake sediments
In Papers II and IV we investigated two small lakes with respect to OC burial and OC mineralization in sediments. The quantification of burial and mineralization in these lakes was performed on a whole-basin scale and over an entire year, in order to account for spatio-temporal dynamics of the C fluxes.

The results of these studies illustrate, that OC mineralization in lake sediments dominates over OC burial, if accounting for the spatio-temporal variability of C fluxes over the year. OC burial in the study lakes (0.3 and 0.5 t C yr\(^{-1}\), respectively) was found to be about one third of sediment OC mineralization (1.0 and 1.2 t C yr\(^{-1}\), respectively), which implies that about 25% of the OC that reaches the lake bottom is buried in the sediments. The results were remarkably similar for these two lakes and suggest that sediments of small boreal lakes are a stronger source than sink of C. However, it is important to state that even the small of OC burial flux in these lakes represents a permanent removal of C from the short-term C cycle.

Sediment C fluxes vs. emission C flux from lakes
In addition to sediment C fluxes, the total annual emission of CO\(_2\) and CH\(_4\) was quantified for the two study lakes (Papers II and IV). Annual CO\(_2\) emission equaled 6.4 and 6.9 t C yr\(^{-1}\), and the CH\(_4\) emission was estimated at 0.08 and 0.03 t C yr\(^{-1}\), respectively. Hence, the total C emission was 21- and 13-times higher than the respective annual OC burial flux of each lake, which agrees with values (range, 4-86; mean, 30), reported in a large scale study of boreal and arctic Finnish lakes (Kortelainen et al. 2013).

Our results also demonstrate that sediment OC mineralization, i.e., the production of CO\(_2\) and CH\(_4\) in sediments, can in total account for a significant share of the annual C emission (16 and 17\%, respectively). In this perspective, sediments are considered to be the major source of the CH\(_4\) flux to the atmosphere, as the production of CH\(_4\) in lakes is mostly restricted to the anoxic environments found in the sediments (Bastviken 2009). However, the
sediment CO₂ production could not account for the 6 times larger amount of CO₂ emitted to the atmosphere on an annual scale. This finding was surprising since previous studies have pointed out sediments as a major source of CO₂ emission from boreal lakes (Kortelainen et al. 2006), and given the shallow basin morphometry of the study lakes with a high share of sediments located in warm and oxygen-rich epilimnetic waters, which stimulate sediment OC mineralization (Sobek et al. 2009; Gudasz et al. 2010).

**Figure 3.** Comparison of OC burial and mineralization in lake sediment to total C emission for two small boreal lakes (Papers II and IV). Numbers express C flux in t C yr⁻¹.

**Role of sediments in an annual lake C budget**

In Paper II, we investigated the full annual C budget of a small boreal lake (Figure 2) in order to put OC burial and mineralization in sediments into perspective to other C fluxes in the lake. In doing so, the importance of different CO₂ sources for the annual lake CO₂ emission could be assessed.

We found that OC mineralization in the water column of the lake (2.4 t C yr⁻¹) was more than twice the annual sediment OC mineralization and accounted for about 40% of the annual CO₂ emission. About 16% of the OC mineralization in water was ascribed to photochemical OC mineralization, which amounted to about 0.4 t C yr⁻¹, and hence was alone larger than annual OC burial in the lake. Most OC mineralization in water was however attributed to the net heterotrophic character of the lake, with low primary production but high respiration of OC. The dominance of water column DOC mineralization over sediment OC mineralization in this lake can be explained by similar mineralization rates in 1 m² of sediment (mean, 69 mg C
m\(^{-2}\) d\(^{-1}\) at 15°C) as in 1 m\(^3\) of water (48 mg C m\(^{-3}\) d\(^{-1}\), respectively) in combination with the bathymetric properties of the lake with 3.8 m\(^3\) of water per 1 m\(^2\) sediment area.

The contribution of the different OC mineralization processes to the gain in lake CO\(_2\), however, varied over the annual cycle. OC mineralization in water dominated over OC mineralization in sediments during the ice-free season, whereas during months of ice cover, sediment and water contributed about equally to the overall OC mineralization in the lakes.

OC mineralization in sediment and in water accounted together for about 50% of the annual CO\(_2\) emission from the study lake. To identify and relate the remaining CO\(_2\) sources in the lake C budget, we investigated the supply of C to the lake from the surrounding catchment (Papers II and III). The import and export of C via fluvial stream transport (13.5 and 14.5 t C yr\(^{-1}\), respectively) was found to dominate quantitatively over all other C fluxes in the lake. Furthermore, the inflow of shallow groundwater supplied a substantial amount of C to the lake (5.1 t C yr\(^{-1}\)), which illustrates the strong influence of the catchment for the C balance in the lake.

**Figure 2.** A simplified annual C budget of Lake Gäddtjärn (Paper II) showing the means, expressed in t C yr\(^{-1}\), of OC burial, sediment OC mineralization (SM), net water OC mineralization (WM), photochemical OC mineralization (PM), C emissions, C import from the catchment via surface water and groundwater, and C export from the lake via the outlet stream.
Comparison of lake- and catchment-scale C fluxes

In Paper III, the headwater systems and the outlet stream of the study lake in Paper II were explored with respect to fluvial C transport downstream and CO$_2$ emission to the atmosphere. This study was carried out in order to compare C losses from the aquatic network upstream of the lake with C losses of the lake itself.

C loss by CO$_2$ emission from headwater systems was found to be higher than the total C loss of the study lake via fluvial downstream export and atmospheric emission (Figure 3). The largest source of aquatic CO$_2$ emission from the entire catchment to the atmosphere was the headwater streams, despite their small areal coverage of the catchment (<0.1%). The high CO$_2$ emission from streams was mostly explained by the large difference in the gas transfer velocity $k$, which was on average 30 times higher in streams than in the lakes.

Hence, in a catchment perspective lake C fluxes played a secondary role, when comparing the overall C loss from the system, which illustrates the importance of addressing C fluxes on integrated scales. Including upstream aquatic emissions further illustrates that only a very small fraction of the total C that enters boreal surface waters will be buried in the sediments.

Figure 3. C losses from the catchment of Lake Gäddtjärn (Paper III) during the open-water sampling period. Error bars represent the minimum and maximum estimate obtained from cumulative standard errors of discharge and concentration measurements for DOC and DIC, and cumulative standard errors of tracer injection replicates (streams), wind speed model-derived $k_{CO2}$ (lakes) and concentration measurements for CO$_2$ evasion.
Temporal variability of the lake sediment C sink

Variability in OC accumulation over the Holocene

In Paper I, we analyzed vertical sediment profiles of seven Swedish boreal lakes for centennial- and millennial-scale OC accumulation rates, i.e., CMARs, to indicate changes in OC accumulation over time. Both centennial and millennial CMARs exhibited variability over time, i.e., over sediment core depth (Figure 4), which was mostly attributed to the variability in sedimentation rate. However, while the millennial variability in CMARs seemed to be related to lake size, with smaller lakes showing higher variability (regression of lake area against standard deviations of CMARs; $R^2=0.63$, $p<0.01$, n=7), there was no such pattern obvious for CMARs on a centennial time scale. Furthermore, there was no common temporal trend observed for millennial CMARs across lakes, however, in six out of seven lakes centennial-scale CMARs increased towards recent time.

The larger variability in millennial CMARs in the smallest lakes was explained by the more dominant impact of local-scale changes, while the larger lakes exhibit more resilience towards changes in the local environment. Changes in OC accumulation in the larger lakes might therefore reflect rather regional-scale changes, such as climatic shifts over the Holocene (Seppä et al. 2005).

The trend of increasing CMARs over the last century has been observed by several other studies investigating OC accumulation over time (Kastowski et al. 2011; Anderson et al. 2013). These studies concluded that recent changes in CMARs are caused by changes in land use and associated soil erosion. For the investigated lakes of this study this explanation is, however, unlikely since no intensified agriculture took place in their catchments over the past century. Instead, the effect of re-forestation during the twentieth century, after a long history of mining activities in central Sweden (Eriksson 1960), might be reflected here in elevated CMARs. Also, increased leaching of OC from soils as a consequence of recovery from acidification might be responsible for a larger quantity of OC supply to lakes in the more recent past (Monteith et al. 2007; Bragée et al. 2015).
Moreover, it can be discussed whether the temporal variability in OC accumulation reflects a changing strengths of the sediment C sink over time, or if periods of higher OC accumulation indicate periods of higher OC input and therefore also higher C emission if assuming that the OC burial efficiency remained stable.

Sources and stability of terrestrial OC in boreal lake sediments

The vertical sediment profiles in Paper I were investigated for terrestrial sources and the degradation state of OC by analyzing the molecular composition of lignin phenols in the sediments.

The analysis revealed that wood-containing material from gymnosperms was a constant and dominating source of OC to the lakes over the Holocene. This was indicated by different lignin phenol parameters, such as low S/V and C/V values throughout the entire sediment core profiles. Moreover, the lignin phenol indices Ad/Al and P/(V+S), which are indicative of the degradation state of the OC, reflected that no consistent degradation of the organic material was detectable even though the organic matter has resided in the sediment for thousands of years. Together with the CMARs, these results indicate that OC is buried in sediments within the first century of deposition and stabilized on a permanent basis.
OC burial efficiency of lake sediments: now and in the future

In Papers II and IV, the basin-wide OC burial efficiency (OCBE) of sediment was quantified for two small lakes with differing basin bathymetry. Additionally, in Paper IV, the effect of lake warming on the OCBE was simulated for both lakes following different climate change scenarios.

The whole-basin OCBE was found to be low under present-day conditions, and of similar magnitude in both study lakes (28 and 25%, respectively). Different lake warming simulations caused consistent decreases in the OCBE of on average 5-16% and 3-11% in the two study lakes respectively (Figure 4). The more shallow lake exhibited in general higher declines in the OCBE, related to more sediment area being affected to changes in epilimnetic water temperatures. The simulated extent of hypolimnetic anoxia during summer stratification counteracted the effect of increased OC mineralization in epilimnetic sediments only to a minor degree.

Hence, this study illustrates that the single effect of warmer water temperatures in lakes may reduce the efficiency of sediments in storing OC in the future, however the magnitude in OCBE reduction will, among other factors, depend on morphometric properties of lakes.

Figure 4. Decline of the OC burial efficiency (OCBE) in Lake Erssjön and Lake Gåddtjärn in response to lake warming, calculated following climate change scenarios for each lake region. Temperature on the x-axis is predicted increase in mean annual temperature. PD=present-day conditions, L=low-, M=medium-, H=high-case scenario. Future projections assume no change in OC deposition onto the sediment surface. Error bars indicate minimum and maximum estimates of 95% confidence intervals.
Conclusions and Perspective

This thesis provides an insight into the role of lake sediments in the C cycle of boreal lakes on integrated spatial and temporal scales. By exploring sediment processes and C fluxes within lakes and across the aquatic network, and over long and short time periods, we put sediments into a larger perspective and elucidate their function for C cycling from the past to the future. The major conclusions of this thesis are that:

1) OC burial in sediments represents one of the smallest aquatic C fluxes at both the lake and the catchment scale, but is the only flux that continuously withdraws C from the active cycling loop.

2) Sediment OC mineralization is not the dominating CO$_2$ source for CO$_2$ emission, even in a shallow lake, given the strong heterotrophic character of the water column and the dominating influence from the lake catchment.

3) At a catchment scale, C emission in headwaters dominates greatly over the entire C loss from a small boreal lake via emission and fluvial export.

4) OC accumulation in lake sediments varied differently in boreal lakes on a millennial time scale, however, similar patterns in OC accumulation are visible in most investigated lakes on a centennial scale.

5) Allochthonous OC is buried and stabilized in sediments within the first century of deposition.

6) The OC burial efficiency of sediments is low if integrated on a whole-basin scale and is likely to decrease in warming lakes in response to future climate change.

Future studies that address the role of sediments for C cycling in boreal lakes may focus even more on the spatio-temporal dynamics of CH$_4$ in lake C budgets given its strong greenhouse gas effect, despite the comparatively small C flux. Moreover, studies that investigate OC storage could operate more on a catchment-scale and investigate the role of headwaters as first OC storage sites. Finally, studies could explore the temporal variability of the OC burial efficiency in sediments over the past, by combining investigations of OC burial rates with paleolimnological proxies that are used to reconstruct the past environment of lakes. This would further help to better project and assess the role of lake sediments as C source and C sink in the future.
Sammanfattning på Svenska

Det finns ungefär 120 miljoner sjöar på jorden. Trots att de sammanlagt täcker bara drygt 3% av den del av kontinenterna som inte är täckt av inlandsisar, har de en viktig funktion i det globala kolkretsloppet. Grundämnet kol finns i allt organiskt material, t ex växtdelar och humus från skog och mark. Mycket av detta organiska material sköljs ner till sjöar och vattendrag, där det antingen sjunker ner och lagras som sediment på sjöbotten, eller bryts ner av mikroorganismer. Denna nedbrytning av organiskt material leder slutligen till att växthusgaserna koldioxid och metan bildas. En del av den koldioxid och metan som bildas i sjöarna när atmosfären vilket medför att sjöar påverkar klimatet. Å andra sidan kan det organiskt materialet i sedimenten bevaras där under mycket lång tid – sjösedimenten har kontinuerligt bygotts upp sedan sjöarna bildades. I svenska sjöar har detta pågått sedan landskapet blev isfritt efter den senaste istiden, dvs ungefär tio tusen år, på andra håll betydligt längre – i Tanganyikasjön är till exempel de äldsta sedimenten ungefär 10 miljoner år gamla. Genom att sjöar avger växthusgaser till atmosfären samtidigt som de utgör ett förvar av kol i sedimenten, utgör de samtidigt både en kolkälla och en kolhot. Summan av de dessa båda processer, dvs. inlandsvattnens kolutsläpp till atmosfären och deras kolinlagring i sedimenten, är en omsättning av kol lika stor som landväxternas samlade netttoupptag av koldioxid. Det är uppenbart att inlandsvatten spelar en viktig roll i hela landskapsets och planetens omsättning av kol.

Speciellt i den nordliga barrskogsregionen, som sträcker sig genom stora delar av Sverige, är inlandsvatten viktiga. Jämfört med andra regioner på jorden finns här osedvanligt många sjöar, och dessutom exceptionellt mycket organiskt material i skogmarkens och myrarnas humuslager. I den nordliga barrskogen har det exempelvis visats att mer kol lagras i sjösediment än i växter och mark i den omkringliggande skogen. Vidare svarar all miljontals sjöar som ligger insprängda i barrskogslandskapet för en viktig del av koldioxidutsläppet från jordens inlandsvatten.

I denna avhandling har jag undersökt sedimenten i olika svenska skogs-sjöar för att utröna följande frågor:

1) Hur stor andel av det kol i organiskt material som sjunker ner till sjöbotten inlagras där, och hur stor andel bryts ner av mikroorganismer till koldioxid och metan?
2) Hur mycket av en sjöns utsläpp av växthusgaser till atmosfären härstammar från sediementen? Vilka andra processer bidrar till en sjöns utsläpp av växthusgaser?

3) Hur mycket har inlagringen av kol i sjösedimenten varierat sedan den senaste istiden? Hur stabil är kolsänkan i sjösediment?

4) Hur kommer ett varmare klimat påverka sjösedimentens effektivitet som kolsänka?

För att kunna besvara dessa frågor studerade vi ett antal olika skogssjöar i centrala och sydvästra Sverige. Samtliga sjöar är typiska för det svenska barrskogsområdet och kännetecknas av att vattnet är brun av humusämnen från omgivningarna.

I två av sjöarna placerade vi sensorer i vattnet som gav oss kontinuerliga mätvärden under ett års tid som är viktiga i samband med kolomsättningen, bland annat temperatur och syrgas löst vattnet. Koldioxid och metan mättes även direkt i sjöarna, och vi beräknade hur mycket av dessa växthusgaser avges till atmosfären. Vid en av sjöarna följde vi även halten av koldioxid med liknande sensorer, placerade i bäckar och i grundvatten. På så vis kunde vi undersöka betydelsen av tillförseln av koldioxid, jämfört med hur mycket som produceras i sjön.

Vi undersökte sedimenten med den särskild provtagare som tar upp ett osoört prov på hela sedimentet i ett långt plaströr. I dessa prover mätte vi kol och andra organiska ämnen, och bestämna sedimentens ålder bland annat med hjälp av kol-14 metoden. Vi kunde sedan beräkna vilken takt materialet har lagrats på botten, och hur takten varierat över tiden. Vi gjorde också experiment med både sediment och sjövatten för att se hur mycket koldioxid och metan som bildas. På så sätt kunde vi jämföra sjöarnas produktion av växthusgaser med deras inlagring av kol i sediment – det vill säga jämföra sjöarnas roll som kolkälla och som kolsänka.

Slutligen använde vi regionala klimatmodeller för att simulera hur en varmare lufttemperatur påverkar vattnets temperatur och syrehalt, för att därifrån göra en prognos över hur inlagring av kol i sjösediment kommer att förändras i framtiden.

Denna avhandling visar att sjösedimenten i små skogssjöar avger ungefär tre gånger så mycket kol i form av koldioxid och metan till atmosfären än de lagrar in i sedimenten. Trots att kolinlagringen är förhållandevis liten så pågår den kontinuerligt, och utgör därmed en viktig kolsänka över längre tidsperioder. Även om sjösedimentens växthusgasproduktion var större än deras kolinlagring, bidrog sedimenten bara med en liten andel (~15%) av sjöns utsläpp av koldioxid till atmosfären. Andra processer, såsom nedbrytning av löst humus i vattnet och inflödet av löst koldioxid via bäckar och grundvatten hade större påverkan på sjöarnas utsläpp av koldioxid. Metan utgjorde bara en liten del av sjöarnas kolomsättning, men eftersom den är en mycket
kraftigare växthusgas än koldioxid är den ändå viktig för sjöarnas effekt på klimatet.

Inlagringen av kol i skogssjöars sediment varierade över tid. Sedan den senaste istiden (upp till 9000 år sedan) var variationen större i små sjöar jämfört med större sjöar. Under de senaste 100 åren uppvisade de flesta sjöarna liknande trender. Detta tyder på en ökad kolinlagring, möjligen som en följd av ökad urlakning av humus från skogsmark. Analys av sammansättningen av molekyler på olika djup i sedimenten visade inga tydliga tecken på nedbrytning, oberoende av hur mer i sedimentet proverna togs – det vill säga hur gammalt det är. Detta pekar på att skogssjöars sediment utgör en stabil kolsänka. För att förutspå effekterna av ett framtida varmare klimat på sedimentens kolinlagring simulerade vi temperatur och syresättning i två små skogssjöar utifrån klimatmodellernas prognoser. Dessa simuleringar visade att kolinlagringen i skogssjöars sediment sannolikt kommer att bli mindre effektiv i framtiden.

Sammantaget visar denna avhandling att sedimenten spelar en förhållandevis liten roll i skogssjöarnas omsättning av kol, och bidrar med bara en liten andel till sjöarnas utsläpp av kol till atmosfären. Å andra sidan är de en stabil kolsänka, och är därmed av stor betydelse för barrskogslandskapets långsiktiga kolbalans. Ett varmare klimat kommer däremot sannolikt göra sediments kolinlagring mindre effektiv.
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References


A doctoral dissertation from the Faculty of Science and Technology, Uppsala University, is usually a summary of a number of papers. A few copies of the complete dissertation are kept at major Swedish research libraries, while the summary alone is distributed internationally through the series Digital Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology. (Prior to January, 2005, the series was published under the title “Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology”.)