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SPATIOTEMPORALITY OF CARBON FLUXES ALONG A BOREAL LAND-STREAM-LAKE CONTINUUM

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ACADEMIC DISSERTATION

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Spatiotemporality of carbon fluxes along a boreal land-stream-lake continuum

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Boreal freshwater ecosystems play an important role in landscape carbon (C) cycle. Streams connecting lakes form an extensive network, where terrestrially fixed C is transported, processed, stored, and released to the atmosphere before reaching the oceans. The terrestrial influence is most significant in headwater streams and lakes. Despite the close connection of terrestrial, lotic, and lentic ecosystems, the ecosystems are mainly studied separately, and the possible interactions between the ecosystems are lost. Also, the C dynamic models are often based on sparse measurements, or singular processes are investigated as snapshot studies. Thus, to reveal the C flux dynamics in the continuum, comprehensive studies based on detailed temporal data series over long periods are needed.

In this thesis, dissolved organic carbon (DOC) concentrations and lateral transport in runoff from an upland catchment site was studied over 15 years (1998-2012). The annual and seasonal dynamics of carbon dioxide (CO2) and methane (CH4) concentrations, lateral fluxes, whole-lake storages and atmospheric release were explored through the aquatic continuum over three years study period (2011-2013). Besides, special attention was paid to CO2 and DOC concentrations and lateral fluxes during spring freshet periods by using automatic high-frequency measurements in the lake and its draining streams.

In general, the continuum showed remarkable spatial and temporal variation in C concentrations and fluxes. The C fluxes in both terrestrial and aquatic ecosystems were seasonally controlled mainly by precipitation and local hydrological conditions. Also, fluxes, concentrations and whole-lake storage of CH4 were regulated by temperature and DOC runoff from upland catchment was regulated by previous year’s net ecosystem exchange and litter production. This study highlighted the importance of spring and autumn for lateral C transport and atmospheric release.

The allochthonous C gas input of terrestrial origin plays an essential role in the temporal C dynamics of the lake. In spring, the laterally transported C gases accumulated under the ice cover during the last weeks of the ice cover period. This connection was confirmed with synchronous changes in concentrations and whole-lake storages in the lake and C gas transport peak in the streams. External input increased the whole-lake storages of C gases as well as the CO2 concentration in the upper water layer of the stratified lake. The atmospheric release at the ice-out was long-lasting, and fluxes were high in comparison to earlier studies. The external input covered up to 24 % of CO2 and 42 % of CH4 released during the first week after ice-out. Due to the transience of the C gas transport and atmospheric release, the lateral impact is easily missed with sparse sampling.

The lateral transport of DOC from the upland catchment on mineral soil was small in comparison to the other ecosystem C fluxes, 0.32 % of the net ecosystem exchange (NEE). Considering the whole catchment, the atmospheric emission from the lake accounted for 9.3 % of the catchment NEE. However, these results shed light
on the increasing importance of freshwaters in transporting and releasing C in the changing future climate. An increasing trend in DOC concentration was found in runoff water from the terrestrial upland catchment, which indicates higher terrestrial C load into freshwaters in the future, too. Warmer winters may result in changes in the seasonal pattern; the differences in snow accumulation did not influence the daily amount of C transported, but the C inputs into the lake took place earlier during the winter months instead of spring. When addressing the impacts of climate change on a catchment scale, it is crucial to consider aquatic and terrestrial ecosystems together to get precise estimates of C sinks and sources.

Keywords: Carbon dioxide, Methane, Dissolved organic carbon, Lateral transport, Spring freshet
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This thesis is based on the following publications:


In the text, the publications are referred to by their roman numerals. This thesis also includes unpublished results.

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Author’s contribution:

H. Miettinen is fully responsible for the summary of this doctoral thesis.

I  HM participated in the field measurements, laboratory analyses, and the writing process.

II  HM was responsible for the field measurements and laboratory analysis. She carried out data analysis and made the figures in the article. MR and HM wrote the first version of the manuscript, which then was commented by other co-authors.

III-IV HM participated in the planning of the study together with AO and JP, was responsible for the field measurements, data analysis and writing of the first version of the manuscript, which then was commented by the other co-authors.
1. Introduction

Freshwater ecosystems, such as streams, rivers, and lakes play an important role in the carbon (C) cycle and the atmospheric release of terrestrial C from the catchment to the global scale (Cole et al., 2007; Battin et al., 2009; Aufdenkampe et al., 2011). Lakes and ponds cover only a small fraction of the total land-area; roughly 2.8 % (Downing et al., 2006). However, their importance is more pronounced in the boreal zone, where the water bodies can cover up to 20 % of the land area (Raatikainen and Kuusisto, 1990). The smallest lakes are the most abundant (Verpoorter et al., 2014), and with the connecting streams and rivers, they form a network which plays a vital role in the regional C cycle.

Aquatic ecosystems are hydrologically connected to terrestrial ecosystems, and the surrounding catchments influence the properties of the lake, e.g., water chemistry and C dynamics. Through photosynthesis, catchment biota fixes the atmospheric C, and C is stored in the living biomass and soil (Fig. 1). Thus, the existing plant communities influence the C quantity in soils, and then, different processes in soils, such as microbial respiration and organic matter (OM) mineralization determine the amount and quality of C in soil water. Terrestrially fixed C is transported from the catchments to the lakes in surface flow or groundwater inputs. It is processed therein, deposited into the sediments, or released into the atmosphere (Battin et al., 2009). Part of the C moves forward in the aquatic continuum. In water, C occurs in particulate (organic and inorganic form; POC, PIC), dissolved (organic and inorganic form; DOC, DIC) or gaseous form (carbon dioxide, methane; CO₂, CH₄, and small amounts of biogenic volatile organic compounds; BVOC). CO₂ is the result of aerobic respiration and soil demineralization processes, while CH₄ is mainly produced in anoxic archael methanogenesis in ecosystems with a high water table (McKenzie et al., 1998). Environmental factors such as temperature, precipitation, and photosynthetically active radiation (PAR) regulate the biotic and abiotic processes and thus, influence the C quantity and quality in water.

On a global scale, annually 2 Pg C is transported through the stream-lake-continua; 0.2 Pg C is stored in sediments, at least 0.8 Pg C is released back to the atmosphere, and 0.9 Pg C ends up in the oceans (Cole et al., 2007). In the boreal region, this C replacement from terrestrial to aquatic ecosystems can decrease the net ecosystem exchange (NEE) of forests from 6 % to 50 % in upland forest and C-rich peatland catchments, respectively (Jonsson et al., 2007; Dinsmore et al., 2010; Huotari et al., 2011; Rasilo, 2013), which addresses the importance of lakes in relation to terrestrial ecosystems. Further, up to 80 % of these allochthonous C inputs — which
enter the aquatic ecosystems mainly in organic form — are released in gaseous form back to the atmosphere from lake-atmosphere interfaces (Algesten et al., 2003).

Since organic C composes the most significant part of the total C transported, it is the most studied among the C species. Lately, an increasing trend of the concentrations of total and dissolved organic C in northern aquatic surfaces has been found (Worrall et al., 2004; Sarkkola et al., 2009). The increase could result from land-use changes (Kortelainen and Saukkonen, 1998; Nieminen, 2004) or changes in atmospheric acid deposition and the consequent release of organic acids, which further influence soil OM solubility (Monteith et al., 2007). However, less attention has been paid to the lateral transport of C gases; CO2 and CH4. The CH4 forms only a negligible part of the total lateral C fluxes (<5 % of total C transport; Leach et al., 2016), but it is important as a greenhouse gas since its 100-year global warming potential (GWP) is 28 times that of CO2 (IPCC, 2014). These C gases are known to have a direct but short-lasting influence on aquatic C dynamics (e.g., Rasilo et al., 2011).

Figure 1. Carbon (C) fluxes in the land-stream-lake continuum. The net carbon dioxide (CO2) uptake by forests is stored in biomass. Streams import terrestrially fixed C into the lakes and forward in the continuum; particulate organic (POC) and inorganic (PIC) carbon, dissolved organic (DOC) and inorganic carbon (DIC), CO2 and methane (CH4). Primary production (PP) in lakes fixes C dissolved in water, and CO2 is released in respiration (R) and mineralization of C. Part of the C is buried in the soil sediments and released in gaseous form. There are also atmospheric release of CO2 and CH4 from all the terrestrial and aquatic surfaces.

1.1. Spatial variation of C dynamics

The catchment characteristics and ecosystem productivity are connected to lakes’ C gas concentrations and release of gases to the atmosphere (e.g., Sobek et al., 2003; Maberly et al., 2012), whereas local and current hydrological conditions control the C
mobilization between streams and catchment (e.g., Ledesma et al., 2018). These factors together influence the quantity and quality of allochthonous inputs into the lake, e.g., inorganic and organic C and nutrients. Most of the C exchange from terrestrial to aquatic ecosystems takes place in narrow areas at the interface of aquatic and terrestrial ecosystems, i.e., in the riparian zone. These areas are usually rich in OM and referred to as biogeochemical hot spots due to vigorous transformation, transportation, retention and production of OM (Bishop et al., 1994; Wetzel, 2001; Larmola, 2005). Local hydrological conditions, e.g., seasonal water table fluctuations, are of utmost importance for connectivity between terrestrial and aquatic ecosystems (Lyon et al., 2011; Leith et al., 2015; Ledesma et al., 2018). Due to external inputs from adjacent soil layers, boreal streams (Hope et al., 2001; Dinsmore et al., 2013; Wallin et al., 2013) and lakes (Cole et al., 1994; Sobek et al., 2003) are usually supersaturated with CO₂. Lakes are also sources of CH₄ into the atmosphere (e.g., Juutinen et al., 2009) and more recent studies show that streams (Campeau et al., 2014; Crawford et al., 2014a) and peatland ditches also (Minkkinen et al., 2018) act as sources of CH₄. The riparian soil influence is most considerable in smaller headwater streams and lakes (Kling et al., 2000; Teodoru et al., 2009).

1.2. Temporal variation of C dynamics

The interactions and C fluxes in the land-stream-lake continuum are highly dynamic and controlled by environmental variables; i.e., seasonal and annual changes in air temperature and precipitation and subsequent changes in hydrology (Einola et al., 2011; Ojala et al., 2011; López Bellido et al., 2012). Among other effects, the large seasonal variation in the boreal region results in a long snow accumulation period and subsequent melting period. Spring freshet, which is the most important high-flow event, dominates the annual and seasonal hydrological regime, and thus has a great influence on C transport and atmospheric fluxes. More than half of the annual organic and inorganic C transport can occur during the spring freshet (Laudon et al., 2004; Dinsmore et al., 2011; Dyson et al., 2011). Also, large atmospheric emissions from stream surfaces have been found during high flow events (Natchimuthu et al., 2017). In terms of total annual C transport, summer and winter are less critical, but the transport can occasionally be enhanced by short-lasting extreme precipitation events, most notably in headwater streams (Dinsmore and Billett, 2008; Rasilo et al., 2011).

The ice-cover period, lasting for several months, has clear consequences for C dynamics in lakes. Low water temperatures, low PAR, weak stratification and low external inputs influence OM mineralization, respiration and gas production as well as C accumulation (e.g., Baehr and DeGrandpre, 2002; Karlsson et al., 2008; Huotari et al., 2009) in conditions, where the ice cover acts as a physical barrier of the C exchange between aquatic surfaces and atmosphere. At the ice-out in spring, there are large, but short-lasting peaks of C gas emissions resulted from hypolimnetic C gas accumulation in the lake (e.g., Michmerhuizen et al., 1996; Striegl et al., 2001; Karlsson et al., 2013). Intra-lake metabolism controls the release of gases in the summer. Another important seasonal flux peak takes place during the autumn turnover when gases produced and accumulated in the bottom layer are released. This
release can even exceed the spring release (Riera et al., 1999; Huotari et al., 2011). However, knowledge of detailed temporal flux and concentration dynamics and timing in relation to external inputs is still lacking.

Predicted changes in precipitation and temperature patterns in the boreal zone will strongly influence C dynamics. Increases in winter and autumn precipitation in the near-future (IPCC, 2013) will increase the water transport from mineral soils and peatlands (Carey et al., 2010) as well as induce changes in the annual and seasonal pattern and alter the intensity of discharge regime (Korhonen and Kuusisto, 2010). It is known that in comparison to larger streams, headwater streams are especially sensitive to changes in precipitation and hydrology (Baker et al., 2004). Higher annual and seasonal temperatures may alter the processes related to C production and mineralization, e.g., higher temperatures may accelerate the decomposition of soil OM (Piao et al., 2008; Vesala et al., 2010). Given this background, the C dynamics in the stream-lake continuum, which is tightly connected to terrestrial C sources and water movements in the catchment, may experience drastic changes in the coming years. For evaluating the intensity and importance of these changes, it is fundamental to reveal the temporal and spatial variation of C dynamics with long-lasting high-frequency measurements. Until now, many studies on C dynamics in streams or lakes are based on short time-periods or are snapshot studies, which give important information about the processes behind, but provide only rough estimations about the seasonal or spatial variability in the system. In this context, to study the dependence of C dynamics on environmental variables, extended measurement periods with high-frequency data are needed.

1.3. Objectives of the study

This study aimed to quantify the lateral DOC, CO₂ and CH₄ fluxes and atmospheric C gas release and to form a comprehensive understanding of C flux dynamics in a boreal land-stream-lake continuum. The overall hypothesis was that C dynamics in aquatic systems are highly variable at both the seasonal and annual timeframes and are strongly influenced by terrestrial lateral C inputs, which are regulated by the local hydrology.

This study about the C dynamics in the land-stream-lake continuum located in Southern Finland consists of a 15-year long time-series (1998–2012; I) from two small upland catchments and a 4-year time-period (2011–2014; II-IV) over an aquatic headwater stream-lake continuum. The extensive field measurement data set covered remarkable annual and seasonal variation in environmental conditions. Particular attention was paid to the spring snowmelt period and subsequent flooding (i.e., spring freshet), a period, which has specific importance during the hydrological year, and it is sensitive to the changing environmental conditions (IV).
The principal aims of this study were:

- To reveal the seasonal and spatial pattern of C gas fluxes (CO₂, CH₄) in an aquatic continuum and the regulating factors behind them (II, III)
- To reveal the lateral signal of terrestrial C in the lake (III, IV)
- To estimate the annual atmospheric gas release and lateral transport in the aquatic system; from the lake and the main streams (II, III)
- To identify the most important drivers behind long-term DOC downstream transport (I)
- To estimate and evaluate the importance of C fluxes in relation to the catchment (I, III)
- To investigate the possible changes due to changing climate in lateral CO₂ and DOC transport during the ice cover period and spring freshet (IV)
2. Material and methods

2.1. Study sites

*Lake Kuivajärvi*

The study lake, Lake Kuivajärvi, is situated in the boreal zone in Southern Finland (61°51’N, 24°17’E, 180 m a.s.l) next to the Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II Station; Hari & Kulmala 2005; Fig. 2). The 30-year mean temperature and precipitation in the area are 3.3 °C and 711 mm, respectively (Pirinen et al., 2012). The lake is small with an area and a mean depth of 63.8 ha and 6.4 m, respectively, oblong in shape (length 2.6 km), and unregulated. It is a typical boreal dimictic lake, and the ice cover period usually lasts from late November to early May. It is mesotrophic and humic and contains a high quantity of DOC (three-year mean in surface water 13.0 mg L⁻¹). The measurements in the lake were performed at a permanent measurement platform with ongoing measurements since 2009. The platform is situated in the deepest part of the lake, where the depth is 13 m.

Figure 2. Location of Lake Kuivajärvi and upland catchment sites in Finland, bathymetric map of Lake Kuivajärvi and its draining inlet streams (S1-S15, blue, green and red color indicate small, mediate and large sized streams, respectively) and the lake outlet and schematic presentation of upland catchment sites at SMEAR II Station, which is located next to the study lake.
The lake catchment area is 914 ha (of which lakes cover 71 ha) and it is mainly covered by managed Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* (L). Karst) forests and a small amount of peatlands and agricultural land (Table 1). The upland primary soil type is haplic podzol. Besides mineral soil, the riparian zone also consists of organic soil.

Fifteen streams are draining to the lake. The main inlet, Saarijärvenpuro (width 3.2 m), is located in the north end of the lake. It is flowing all year round and draining a small (15.3 ha) upper lake. The rest of the inlets (hereafter referred to as secondary inlets), are located evenly around the lake, and mostly ephemeral, flowing only during notable hydrological events. The outlet stream Huikonjoki (width 8 m) is situated in the south end of the lake, which is shallow and covered by dense stands of *Menyanthes trifoliata* L. and other aquatic plants like *Carex rostrata* Stokes and *Equisetum spp*.

### Table 1. The lake catchment characteristics.

<table>
<thead>
<tr>
<th>Area</th>
<th>Area (ha)</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lake</td>
<td>71</td>
<td>8</td>
</tr>
<tr>
<td>Forest</td>
<td>721</td>
<td>78</td>
</tr>
<tr>
<td>Peatland</td>
<td>97</td>
<td>11</td>
</tr>
<tr>
<td>Natural</td>
<td>25</td>
<td>3</td>
</tr>
<tr>
<td>Drained</td>
<td>72</td>
<td>8</td>
</tr>
<tr>
<td>Agricultural land</td>
<td>7</td>
<td>1</td>
</tr>
<tr>
<td>Urban areas</td>
<td>18</td>
<td>2</td>
</tr>
</tbody>
</table>

**Upland catchment sites**

The upland catchment study was conducted in two small hydrological catchments right next to the catchment of Lake Kuivajärvi, in SMEAR II Station (I). The area of the catchments is 889 m² and 301 m² in catchment 1 and 2, respectively, and they are separate hydrological units. The catchments are natural and formed on the granite bedrock with shallow (0.50–0.70 m) haplic podzol-type soil above. The catchments are mainly covered by Scots pine forest with understorey vegetation of *Vaccinium myrtillus* L. and *Vaccinium vitis-idaea* L. The area was clear-cut, treated with prescribed burning and sown with Scots pine seeds in 1962.

### 2.2. Field measurements

**Manual gas sampling from water**

The CO₂ and CH₄ concentrations in lake and stream water were measured using the headspace equilibrium technique (McAuliffe, 1971). The sampling was performed
from January 2011 until the end of May 2014. In the lake, the sampling frequency was once a week and fortnightly in open water and ice cover periods, respectively. During the ice thaw, i.e., two to five weeks before the ice-out, the manual sampling in the lake had to be postponed. In the main inlet and outlet streams, the sampling was carried out once a fortnight, except during the spring freshet, when the sampling frequency was one to three times per week. In the spring of 2013 and 2014, all 15 inlets and the outlet of Lake Kuivajärvi were sampled once a week.

In the lake, water samples were taken with a Limnos® water sampler (volume 2.0 dm³, length 30 cm) from 8 different depths from surface to the bottom (depths of 0.2, 1.0, 3.0, 5.0, 7.0, 9.0, 11.0 and 12.0 m). Two replicate gas samples were drawn into 60 ml plastic syringes equipped with a 3-way stopcock valve. In the streams, the replicate gas samples were taken directly into the syringes from freely moving surface water. All the measurements in the streams were performed as close to the lake as possible.

The gas samples were processed in the laboratory of Hyytiälä Forestry Field Station next to the lake immediately after sampling. In syringes, 30 ml of water was replaced with 30 ml of N₂ gas (AGA Instrument Nitrogen 5.0). Gas and water phases were equilibrated, placing them first in 20 °C water bath for 30 minutes and then shaking vigorously for 3 minutes. The gas-phase was pushed into pre-evacuated airtight 12 ml Exetainer® vials (Labro Ltd., Lampeter, Ceredigion, UK) and stored at 4 °C until the analysis. Gas samples were analysed with a gas chromatograph (Agilent 7890, Agilent Technologies, Palo Alto, CA, USA) equipped with a flame-ionization detector (FID; 300 °C, for CH₄ and CO₂) and thermal conductivity detector (TCD; 250 °C, for CO₂). The gas concentrations were calculated using Henry’s Law adjusted temperature correction for 20 °C.

**Automatic CO₂ concentration measurement in water**

The continuous measurement system of dissolved CO₂ in lake water was installed at the depths of 1.5, 2.5, and 7.0 m. It is a closed system for each depth individually, formed with a semipermeable silicon rubber tube in measuring depth allowing the gas exchange between the air phase in the tube and the lake water. The silicon rubber tubes are connected with gas-impermeable tubes with CO₂ silicon-based nondispersive infrared flow-through probes (CARBO-CAP GMP343, Vaisala Oyj, Helsinki, Finland). The air inside the tubes is continuously circulated with a pump (Gardener Denver Thomas GmbH SMG-4, Puchheim, Germany). In the platform, the probes are located in a temperature-controlled box. For more information on the measurement setup, see Hari et al. (2008) and Provenzale et al. (2018). The concentrations were calculated using the temperature dependence of CO₂ solubility in water according to Hari et al. (2008):

\[
C_{CO_2} = X_{CO_2} P_{H} K_h
\]  

Where the concentration of CO₂ measured with the sensor \(C_{CO_2}\) is calculated with atmospheric pressure \(P, \text{ atm}\) and Henry’s law constant with appropriate temperature in measuring depth \(K_h\). The gaps due to malfunctioning or power cutoffs in the data set were filled using values from regressions between manual and
automatic CO₂ measurements. In this study, data over six months, covering periods of ice-cover, ice thaw, and ice out were compared between the years 2013 and 2014.

Additional measurements

Temperature in the water column was measured with continuously logging thermistor string of Pt100 resistance thermometers placed at 16 different depths (0.2, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, 6.0, 7.0, 8.0, 10.0, and 12.0 m) at the platform. We also measured dissolved O₂ concentrations (mg L⁻¹) with an optical temperature-compensated dissolved oxygen meter (YSI ProODO, Yellow Springs Instruments, Yellow Springs, OH, USA). The measurements were taken at 0.5 m intervals from the surface layer to 9.0 m, and then to 12.0 m at 1.0 m intervals. Chlorophyll a (Chl a) in the illuminated euphotic layer was measured and determined with a standard method (SFS-EN ISO 6878) using spectrophotometer (Shimadzu ultraviolet (UV)-1800 spectrophotometer, Shimadzu Corp., Kyoto, Japan).

Discharge measurements

The discharge in the main inlet and outlet streams was measured with an acoustic flow meter (SonTek FlowTracker Handheld ADV®, SonTek, San Diego, CA, USA) weekly or biweekly during spring and summer in 2012 and 2013 so that the sampling covered the times of high and low flow periods in the streams. The water level in both streams was measured continuously at 30 min intervals by measuring the hydrostatic pressure with Levelogger Edge data loggers (Solinst Canada Ltd., Georgetown, Ontario, Canada) placed on the bottom of both streams. The pressure readings were compensated with air pressure above the water surface measured with a Barologger Gold data logger (Solinst Canada Ltd., Canada). The continuous discharge was then estimated from the relation between the results of discharge and water level. Besides the discharge estimated in the main inlet and outlet for the study period 2011–2013 (III), we measured discharge in the secondary inlets during two spring freshets in 2013 and 2014 (IV). We measured the discharge in every inlet with the acoustic flow meter in three representative points and used the mean for further estimations. For estimates of the daily discharge, the gaps between the sampling days were filled with linear interpolation.

The runoff in the upland catchments was measured from two weirs cast on the bedrock for both catchments separately (I). Water from the catchments were flowing through the runoff tubes and the flow was continuously measured with flow meters (Schlumberger, Schlumberger Ltd., Houston, TX), which were connected to the runoff tubes.

Dissolved organic carbon

The DOC concentration was analysed from water samples taken from the lake (depths of 0.2 m and 12.0 m) and the precipitation and runoff from the upland catchment site once a month. During the spring events in 2013 and 2014, samples from the 15 inlets and the outlet of Lake Kuivajärvi were taken once a week. The water samples were filtered through a 0.45 μm membrane filter with a vacuum filtering system (Millipore, Millipore Corporation, Billerica, MA) within the same day and then stored in the dark at -18 °C. Later, the samples were analyzed with a C analyzer in Hyytiälä Forestry
Field Station until the end of the year 2012 (Shimadzu TOC 5000A, Shimadzu Corporation, Kyoto, Japan) and since then, in the laboratory of the Department of Forest Sciences at the University of Helsinki (TOC-Vcpb, Shimadzu Corporation, Kyoto, Japan).

On the sampling days, pH and specific conductance were also measured from the water samples using a combined pH and conductivity meter (ACCUMET 20, Thermo Fischer Scientific, Waltham, MA), and the water temperatures in the streams were measured in situ.

Net ecosystem exchange and litterfall in the upland catchment site

The NEE in the upland catchments was measured with the eddy covariance (EC) technique (Aubinet et al., 2012). The measurement system was located at 23.3 m height and consisted of an ultrasonic anemometer (Solent Research 101R2, Gill Instruments Ltd., Lymington, Hampshire, England) for measuring three wind speed components and temperature, and a closed-path infrared gas analyser (LI-6262, LI-COR Biosciences, Lincoln, NE) for measuring CO2 and H2O concentrations.

Litterfall from the tree canopies was collected monthly from 21 litter traps. The samples were oven-dried at 60°C for 24 h, and the mass was measured immediately after that. The C concentrations were analysed from the material ground with a ball mill using elemental analyzer (Vario MAX CN elemental analyzer, Elementar Analysensysteme GmbH, Hanau, Germany).

2.3. Calculations

Whole-lake C gas storage

To calculate the whole-lake storage of C gases for the years 2011 to 2013, we used the depth-zone specific volume data (obtained from the Finnish Environment Institute) with the discrete gas concentration data according to Striegl and Michmerhuizen (1998).

Lateral fluxes

For filling the gaps between the measurements in the export data sets, the DOC, CO2, and CH4 concentrations were estimated by using linear interpolation (I-IV). Then, to calculate the daily and annual lateral fluxes in the streams, the daily discharge was multiplied by the corresponding daily C concentration. Annual C fluxes were obtained by integrating the daily results over the year (II, III).

Atmospheric gas flux calculations

The atmospheric gas fluxes from the aquatic continuum were estimated from the concentration difference between the water surface and atmosphere. The different physical characteristics between the streams and the lake forced us to use different methods between the sites. In the lake, the atmospheric flux was estimated according to the equation:
\[ Flux_{\text{gas}} = \alpha k_{\text{gas}} (C_{\text{gas}} - C_{\text{eq}}) \]  

(2)

Where \( Flux_{\text{gas}} \) (mol m\(^{-2}\) d\(^{-1}\)) is the flux of studied gas (CO\(_2\) or CH\(_4\)), \( k_{\text{gas}} \) is the gas transfer coefficient (m d\(^{-1}\)), \( C_{\text{gas}} \) is the concentration (mol m\(^{-3}\)) of given gas and \( C_{\text{eq}} \) is the concentration (mol m\(^{-3}\)) in equilibrium with the atmosphere (Cole and Caraco, 1998). Due to the low pH in the lake, the chemical enhancement factor, \( \alpha \), is here assumed to be 1 (Cole and Caraco, 1998). In the lake, the gas transfer coefficients were based on chamber flux measurements made simultaneously with surface water concentrations measurements (Cf. Cole et al., 2010) between August and November in 2011 \((n = 14)\), and the mean was used. The gas transfer coefficients calculated by this method are in good agreement with the gas transfer coefficient measured with the EC system in the same period (Heiskanen et al., 2014).

To estimate the gas transfer coefficient in streams, we used the empirically determined equation of Raymond et al. (2012):

\[ k_{600} = 929 (V S)^{0.75} Q^{0.011} \]  

(3)

Where \( V \) is the stream velocity (m s\(^{-1}\)), \( S \) is the stream slope (unitless), and \( Q \) is the discharge (m\(^3\) s\(^{-1}\)). The gas transfer coefficient was calculated using the equation of Riera et al. (1999):

\[ k_{\text{gas}} = k_{600} (S C_{\text{gas}}/600)^n \]  

(4)

Where \( S C_{\text{gas}} \) is the Schmidt number for the given gas adjusted to the in situ temperature (Jähne et al., 1987). For \( n \) we used a value of -0.5 according to the experimental measurements of Clark et al. (1995).

### 2.4. Statistical analyses

The seasonal data was divided into four seasons using the thermal criterion based on daily mean air temperature (winter <0 °C; spring 0–10 °C; summer >10 °C; autumn 10–0 °C), thus the length of the seasons varied from year to year (III). As the data sets did not fulfil the normality assumptions, the temporal and spatial differences in gas concentration and fluxes and temporal trends in data sets were analysed with non-parametric tests (Kruskal-Wallis median test, Mann-Kendall trend test) (II, III, IV). The correlations between hydrological and biological parameters and C concentrations and fluxes were determined with Spearman correlations, or when normally distributed, with Pearson correlations (III, IV).

The factors behind the interannual variation in long-term time series of DOC fluxes and concentrations in the upland catchment were studied by using linear mixed-effects models (I). The tested fixed factors were annual precipitation and catchment area, and random factors were NEE of the current year, NEE of the previous year, litterfall of the previous year, snow water storage in March, soil water content, the temperature sum of the previous year, soil water content of the previous...
July and the H+ ion concentration in the runoff water. First, the annual precipitation and catchment area were tested in a simple model:

\[ Y = b_0 + b_1 X_1 + b_2 X_2 \]  

(5)

Where \( Y \) is the annual DOC flux/concentration, \( b_0 \) is the intercept of the model, \( b_1 \) is the regression coefficient for annual precipitation \( (X_1) \), and \( b_2 \) is the regression coefficient for catchment area \( (X_2) \). Later, other variables were added, and the models with the best combination of explaining factors were looked for, and finally, the models with the best-adjusted \( R^2 \) and the lowest Akaike’s Information Criterion (AIC) value were selected for sensitivity analysis of the variables. Then, in the sensitivity analysis, ±10% change was added in the annual mean values of the variables. With these simulated values the robustness of the linear mixed-model results was estimated.
3. Results

3.1. Spatial and temporal patterns in the aquatic continuum (II, III, IV)

Weather conditions

Annual mean air temperature in the years 2011 and 2013 was 5.4 and 5.1 °C, respectively (III). They were warmer years than 2012 when the annual mean air temperature was 3.3 °C. The coldest year 2012 was also the rainiest, and annual precipitation was 917 mm, while it was 767 mm in 2011 and less in the dry year 2013, 616 mm.

There were no significant differences in length of snow cover or ice-cover periods in the lake in years 2011 to 2013. Winter 2014, however, was warmer, and this reduced snow accumulation and depth, as well as ice-cover length and thickness in the lake in comparison to the earlier years (IV).

Hydrology

The discharge in the main inlet (Q\textsubscript{in}) and the outlet (Q\textsubscript{out}) co-varied with the environmental conditions and thus, had a seasonal pattern (III). The highest seasonal discharge was observed in spring simultaneously with the spring freshet (2011–2013; Q\textsubscript{in}, 0.35 m\textsuperscript{3} s\textsuperscript{-1}; Q\textsubscript{out} 0.73 m\textsuperscript{3} s\textsuperscript{-1}), which contributed one-third of the annual values. Another seasonal peak was observed in autumn when the precipitation was also high, even if the seasonal mean did not reach the spring values (2011–2013; Q\textsubscript{in}, 0.18 m\textsuperscript{3} s\textsuperscript{-1}; Q\textsubscript{out} 0.51 m\textsuperscript{3} s\textsuperscript{-1}). In general, during winter and summer seasons the discharge was low in both streams (e.g., in summer Q\textsubscript{in}, 0.09 m\textsuperscript{3} s\textsuperscript{-1}; Q\textsubscript{out} 0.16 m\textsuperscript{3} s\textsuperscript{-1}). However, there were substantial inter-annual differences in winter discharge, mostly influenced by the total precipitation in late autumn and early winter.

Annually, the Q\textsubscript{in} and Q\textsubscript{out} were higher in the rainy year 2012, (daily mean, 0.17 and 0.44 m\textsuperscript{3} s\textsuperscript{-1}, respectively) than in the drier years 2011 (0.12 and 0.28 m\textsuperscript{3} s\textsuperscript{-1}, respectively) and 2013 (0.11 and 0.28 m\textsuperscript{3} s\textsuperscript{-1}, respectively). The Q\textsubscript{out} exceeded the Q\textsubscript{in} most often, and the difference was most substantial during high flow events like spring freshet, e.g., in years 2011–2013, the Q\textsubscript{in} was 46–57 % of that of the Q\textsubscript{out}. During the freshet period, the importance of total discharge in secondary inlets (n = 14) remained low in comparison to Q\textsubscript{in}, covering 21 % of the Q\textsubscript{out} in 2013 (IV). The contribution by Q\textsubscript{in} and secondary inlets indicates that the lake receives additional
water inputs as direct surface flow from the riparian zone or through groundwater inputs, and these are especially important during the high flow events.

The snow accumulation and melt influenced the discharge in all the inlets and the outlet during spring freshet periods in 2013 and 2014 (IV). The total discharge was higher in 2013 than in 2014. In 2013, after the temperature rose above 0 °C and subsequent snowmelt, the discharge increased quickly within five days into its maximum (1.57 m$^3$ s$^{-1}$ and 1.82 m$^3$ s$^{-1}$ in all the inlets and the outlet, respectively). In the year 2014, there was no apparent increase in discharge like in 2013. In 2014, the snow melted partly already between mid-December and mid-January and resulted in a high flow event in streams (peak discharge 0.46 m$^3$ s$^{-1}$ and 1.01 m$^3$ s$^{-1}$ at all the inlets and the outlet, respectively). Later, the precipitation induced three small events in March–April, but no clear spring freshet, as in the years 2011 to 2013, was detected. The proportion of secondary inlets of the total input was 29 and 17 % in 2013 and 2014, respectively.

Concentrations and lateral fluxes of CO$_2$

Concentrations of CO$_2$ in the main inlet and outlet as well as in the lake had a seasonal pattern (III). In general, the concentrations were highest in winter and spring, and lowest in summer and autumn. Despite the differences in hydrology between the years, only small differences in the annual pattern were observed. Spatially, the concentrations were highest in the main inlet, followed by the concentrations in the outlet and lowest in the lake. Besides, in the main inlet, there was more variation in concentrations than in the other sites, indicating more noticeable terrestrial influence. During the ice cover period in the lake, i.e., winter and spring seasons, the concentrations measured in the outlet were of similar magnitude with the lake concentrations. The open-water period was characterized by efficient gas exchange between lake surface and atmosphere, and the lake CO$_2$ concentrations decreased in comparison to the main inlet and outlet. During the spring snowmelt in 2013 and 2014, the spatial variation in CO$_2$ concentrations between the 15 inlet streams and the outlet was high. However, despite the hydrological differences between the years, there were no significant differences in the CO$_2$ concentrations in the outlet or smaller sized inlets. Higher concentrations of CO$_2$ in 2013 than in 2014 were only found in some of larger inlets with high discharge, and the difference between the years was small.

A positive relationship between discharge and CO$_2$ concentrations was found in the main inlet (III). Interestingly, the concentrations in winter showed a different pattern since they were high despite the low discharge. In comparison with the main inlet, a positive relationship between discharge and concentrations was only found in the spring season in the outlet. We also found a connection with the stream size and concentrations (IV). The positive relationship between concentrations and discharge, similar to the main inlet, was confirmed in the secondary inlets with copious discharge, while in the inlets with lower discharge the relationship was negative, i.e., the concentrations decreased by increasing discharge. No relationship between CO$_2$ concentration and precipitation was found.
The main inlet and outlet had a similar seasonal load pattern of dissolved CO$_2$ (Fig. 3a; III). The highest transport coincided with seasons with high flow, i.e., spring and autumn, whereas the transport was low during seasons of low flow, i.e., winter and summer. The transport was highest during the spring freshet when almost half of the annual CO$_2$ transport took place. In years 2011 to 2013, the hydrological spring events in the streams took place earlier than the ice-out in the lake and already 80–85 % of the CO$_2$ had passed the main inlet and 64–74 % the outlet at the ice-out and concurrent lake turn-over. High precipitation increased the transport occasionally in the summer. In the autumn the precipitation-induced transport peak was higher and lasted longer than in summer, although it remained at a lower level than in the spring. The variability of the daily transport was closely connected to current hydrology, and there was no visible influence of lake turn-over in the outlet stream. The discharge also dominated the annual CO$_2$ transport, which was highest during the rainy year, and lowest during the dry year.

![Graphs showing lateral fluxes and lake storages of CO$_2$-C](image)

**Figure 3.** Daily lateral fluxes of CO$_2$-C in the main inlet (dark solid line) and the outlet (light solid line; a) and whole-lake storages of CO$_2$-C (b) in the year 2011. Vertical dashed lines represent the spring and autumn turnover in the lake. The bold horizontal line in the x-axis displays the ice cover period.

**Concentrations and lateral fluxes of DOC during spring in 2013 and 2014**

Despite the significant differences in snow accumulation and melting between the years 2013 and 2014, there were no substantial differences in mean concentrations of DOC in the inlet streams between the years (IV). The mean concentrations in the outlet were higher in 2013 than in 2014. The total transport of DOC was slightly higher in the year 2013 with higher discharge than in 2014, mostly derived from the
differences in snow accumulation, while the precipitation between the years was the same. Compared to the transport of CO₂, DOC dominated the total transport in both years; 88 % of C was in organic form and 12 % in gaseous form. The secondary inlets contributed 34 and 18 % of DOC in 2013 and 2014, respectively, indicating higher importance of secondary inlets in terms of C transport during significant freshet events like that in 2013.

As the timing of the highest C transport was connected to hydrological high flow events, it differed between hydrologically different years. In 2013, approximately half of the transport into the lake was observed during freshet, whereas in 2014, 2/3 of the total transport took place earlier during ice cover period. In the outlet, the transport had a similar pattern, with approximately half of the total transport during freshet in 2013 and 82 % during ice cover period in 2014. However, in the outlet in 2013, the C transport during ice cover period was nearly the same as during freshet.

Lake CO₂ dynamics

In the years 2011 to 2013, the freeze-over time of the lake was relatively constant and freezing took place at the end of November. The CO₂ accumulated during the ice cover period and the accumulation was most intense in the hypolimnion, but also detectable in the upper layers of the lake (II). There was a clear linear accumulation of CO₂ estimated from the whole-lake storages with a similar growth rate between the years. In the same years, the ice-out in the lake took place around the beginning of May and varied ten days between the years (III). Annually, the highest concentrations in the lake surface and the highest whole-lake storage were measured at the ice-out and subsequent lake turn-over. In comparison to the linear CO₂ accumulation during the ice cover period, there was a 30 % increase in the whole-lake storage at the ice-out in 2011 (Fig. 3b) and 2012. However, such a surplus was not observed in 2013.

We monitored the under-ice CO₂ concentrations with automatic measurements during the typical winter 2013 and warm winter 2014 (Fig. 4; IV). At the beginning of the ice cover period in 2013, the CO₂ concentrations were similar at all depths, but later during the ice-cover period concentrations at 1.5 and 2.5 m depths remained constant, while concentrations at 7.0 m increased. At the onset of the freshet, when the lake was stratified, concentrations in the upper layer increased within four days. The concentrations at 2.5 m increased smoothly after ten days from the onset of the freshet, simultaneously with the lake surface water mixing. The lake turned over entirely in the next day after the ice-out and the CO₂ accumulated in the water column was rapidly released to the atmosphere before the lake started to stratify again. In 2014, the warm period during winter months induced a small discharge peak, which was in synchrony with the increase in CO₂ concentrations at 1.5 m depth in the lake, but it did not influence the concentrations at deeper depths. The warm spring influenced the water stratification, and the mixing started already under the ice cover, although it did not affect the deeper (>6 m) layers before the last days of the ice cover period. The turn-over was completed one day before the ice-out. The concentrations in the upper layer were significantly lower in 2014 than in 2013. The concentrations at 7.0 m depth were lower during the ice cover period in 2014, but were at similar levels during the ice thaw period between the years.
In the summer, the surface layer concentrations slowly increased towards the autumn, but the whole-lake storages remained stable during the open-water period. There was a weak negative relationship between the surface water CO₂ and Chl a, indicating possible biological control (II). In the autumn, the gas ventilation was efficient, and the storage was smallest just before the freeze over. We observed increased CO₂ concentrations in the bottom layer simultaneously with decreasing O₂ concentrations, which indicate in-lake mineralization in the hypolimnion (II). These high CO₂ concentrations decreased before the freeze-over, during the autumn turn-over.

Figure 4. CO₂ concentrations (mg L⁻¹) at 1.5, 2.5, and 7.0 m depths and the CO₂ input (kg d⁻¹) into the lake via inlets in 2013 (a) and 2014 (b). For each depth, dotted lines represent the gap-filled periods. The moment of ice-out and spring turn-over is marked with the black arrow.

Concentrations and fluxes of CH₄

In general, the CH₄ concentrations were lower in the lake than in the streams (III). Different from CO₂, the concentrations of CH₄ did not have a similar seasonal pattern in the study sites, although the pattern was found within the study sites in the main inlet and the lake. In the main inlet, the highest peaks were measured in the spring. At the spring freshet, the lake influenced the outlet, and the concentrations were of similar size. In summer and towards autumn, the outlet concentrations increased, while in the main inlet and lake, the concentrations were lower without differences between the summer and autumn. The highest concentrations in the outlet were measured in the autumn. The connection between hydrology and transport was not as clear with CH₄ as it was with CO₂. Seasonally, in the main inlet stream, there was mostly a positive relationship between concentrations and discharge, and it was
strongest during the spring snowmelt, but in the outlet, this relationship was negative.

Similar to CO₂, the annual output of CH₄ was higher than the input (Fig. 5a; III). Annually, the CH₄ transport through the lake was highest in the rainy and cold year of 2012. In comparison, the transport from the lake was highest in 2011, and thus it did not correlate with the highest discharge but coincided with the highest annual air temperature and highest summertime accumulation of CH₄ in the lake. In both streams, the loads were smallest in the dry year of 2013.

![Figure 5. Daily lateral fluxes of CH₄-C in the main inlet (dark solid line) and the outlet (light solid line; a) and whole-lake storages of CH₄-C (b) in the year 2011. Vertical dashed lines represent the spring and autumn turnover in the lake. The bold horizontal line in the x-axis displays the ice cover period.](image)

Seasonally, the CH₄ transport had no similar pattern between the streams. In winter, the transport was low in both streams. The importance of spring differed between the streams: most of the transport in the main inlet took place in the spring, 52–65 % of the annual transport (III). In the outlet, instead, only 15–21 % of the annual transport took place in the spring. The summer transport was higher from the lake than into the lake and was connected to precipitation events, which in the outlet increased the transport, but was modest or undetectable in the main inlet. Autumn had the highest importance in the outlet, while in the main inlet the increase in transport in the autumn was only modest. The highest transport in the outlet, up to 51 % of annual transport took place in the autumn. There were also high transport episodes during autumn, which occurred at the time of the autumn turn-over in the
lake and were linked to high concentrations but did not coincide with the hydrological events.

**Lake CH$_4$ dynamics**

There was never a sign of wintertime anoxia or hypoxia in the hypolimnion and thus CH$_4$, different from CO$_2$, did not accumulate under the permanent ice cover (Fig. 5b; II). The concentrations, as well as the whole-lake CH$_4$ storage under the ice cover, were very small. However, after the complete ice-out, the whole-lake storage suddenly increased 40- to 200-fold compared to the estimate based on the last under-ice measurements (III). Interestingly, this increase took place in 2011 after five days from the ice-out, while during the other years the whole-lake storage was most substantial on the very first day after the ice-out. The CH$_4$ concentrations in the water column were rapidly exhausted and vanished within a week (II).

Annually, the storages and concentrations in the upper layers were highest at the ice-out. The whole-lake storage remained low until late summer when the CH$_4$ started to accumulate in the hypolimnion. This timing was most apparent during the hot summer of 2011, but also clearly visible in 2013 (III). In the cold year of 2012, the whole-lake storage was low compared to other years. The whole-lake CH$_4$ storage vanished by mid-October every year during the autumn turn-over.

**Atmospheric gas fluxes**

The annual mean atmospheric CO$_2$-C release per unit of surface area was highest in the main inlet and lowest in the lake (Fig. 6a–c; III). In all study sites, the precipitation and consequently, increased discharge and C transport affected the CO$_2$-C release, and it was highest in the rainy year and lowest in the dry year. The surface water concentrations were supersaturated in relation to the atmosphere during the whole study period, and all the study sites were continuous sources of CO$_2$-C to the atmosphere.

In the lake, the first month after the ice-out was the most important period of CO$_2$-C emissions from the lake when 25 to 31 % of the annual emissions took place (II). Due to high daily mean fluxes, the autumn season was also an important period of releasing CO$_2$, although high peaks during the autumn turn-over were not detected.

Similar to CO$_2$ fluxes, the annual mean CH$_4$-C release was highest in the main inlet and lowest in the lake (Fig. 6d–f; III). Even the lowest lake surface concentrations were still tens of times above the atmospheric equilibrium, and thus, all sites acted as a continuous source of CH$_4$-C to the atmosphere (II). Besides the annual precipitation sum, also air temperatures affected the atmospheric fluxes of CH$_4$-C. The influence of precipitation and temperature was seen in the outlet and in the lake, where the CH$_4$-C release was highest in 2011 coinciding with the highest annual air temperature, lateral transport, and largest storage, whereas in the main inlet, the discharge controlled the atmospheric release. Dry, warm year decreased the release from the lake and the main inlet, while in the outlet the release remained similar to the year 2012.
The most crucial period for CH₄ release was summer in the outlet and the lake, while the spring was the most important in the main inlet. However, despite the substantial differences in accumulation of CH₄ during the open water period, the atmospheric release in autumn turn-over was similar between the years (II).

### 3.2. Long-term DOC flux in the upland catchment (I)

**Concentrations and lateral flux of DOC**

The results from the two upland catchments were similar, and thus, in this thesis, only the results from catchment 1 are presented. Monthly mean DOC concentration in the runoff during the 15-year study period from the upland catchment was 3.84 mg L⁻¹. Seasonally, the highest DOC concentrations were observed at the beginning of the hydrological spring event, and then they decreased towards summer. An increase in DOC concentrations in runoff was observed over time.

The monthly DOC fluxes showed clear seasonal variation following the downstream runoff. The fluxes were highest in April (peak values from 0.020 to 0.025 g C m⁻²) and peaked another time in late November. The annual DOC fluxes correlated positively with the annual runoff. However, there was no correlation between the annual DOC flux and annual NEE or annual litterfall. There was a high interannual
variation between the years due to high variation in annual runoff; the dry year minimum of 0.20 g C m\(^{-2}\) yr\(^{-1}\) and wet year maximum of 1.89 g C m\(^{-2}\) yr\(^{-1}\).

The models testing the factors behind the interannual variation of annual DOC fluxes revealed especially sensitivity to precipitation. Also, all the models testing annual DOC fluxes and concentrations were sensitive to changes in NEE. In addition to these two factors, models also showed the importance of litterfall from the previous year and snow water storage in March.

*NEE and litterfall*

The upland catchment site acted as a C sink over the study period; the mean annual NEE was -234 g C m\(^{-2}\) yr\(^{-1}\). The monthly mean NEE showed a clear trend over the 15 years. The mean annual litterfall rate was 149 g C m\(^{-2}\) yr\(^{-1}\), but it did not show any decreasing or increasing trend over the 15 years.
4. Discussion

4.1. Spatio-temporal variation of C dynamics in the land-stream-lake continuum

Seasonality of lateral and vertical C gas concentrations and fluxes

There was a clear seasonal pattern of CO₂ concentrations and fluxes in the land-stream-lake continuum. The pattern appeared steady and predictable and was connected to prevailing hydrological conditions. This observation is in line with earlier results from boreal streams (e.g., Hope et al., 2004; Öquist et al., 2009). However, the seasonality of CH₄ concentrations and fluxes was less pronounced, since there was no seasonal pattern across the continuum, although there was a seasonal pattern in the main inlet and the lake. Earlier studies made in peatland catchments have not reported a seasonal pattern of CH₄ in streams (e.g., Hope et al., 2004; Crawford et al., 2013; Leach et al., 2016). The distinct pattern between the main inlet and outlet streams can reflect the adjacent soil type, which around the inlet consists of mineral soil, but close to the outlet the soil is mainly composed of peat. Different soil types alter the production and oxidation of CH₄ in streams and adjacent soil layers (Crawford et al., 2014b; Rasilo et al., 2017), and these processes are known to be spatially highly variable (Campeau and del Giorgio, 2014). The soil type also influences the pathways for C mobilization (Laudon et al., 2007; Dinsmore et al., 2013), and thus the event response of terrestrial C transport between the sites can vary. Thus, the differences in the seasonal pattern between the streams could be a result of spatial soil variability.

The results from the years 2011 to 2013 confirm the earlier results regarding the importance of spring for C transport (Laudon et al., 2004; Dinsmore et al., 2011; Dyson et al., 2011) and atmospheric release from aquatic surfaces (Michmerhuizen et al., 1996; Striegl et al., 2001). The highest atmospheric release in the lake and streams took place in spring. The seasonal emissions at the ice-out and subsequent thermal stratification release in the lake were 40% and 35% of the annual emissions of CO₂ and CH₄, respectively. In autumn, the rain increased the hydrological inputs in streams and thus, the transport of CO₂, and autumn appeared second to spring. The importance of autumn was especially clear in the output of CH₄ since 38% of the annual CH₄ transport from the lake took place in autumn. Autumn was less important in the main inlet, where the riverine transport of CH₄ was less than 20% of the annual transport. High atmospheric release and transport of CH₄ have been found before in
a peatland catchment and related to late-summer productivity (Dinsmore et al., 2010), which again addresses the soil type derived differences along the continuum.

**Sources of C in the discharge and runoff**

The stream concentrations of CO$_2$ and CH$_4$ revealed that the streams were supersaturated with these gases throughout the study period and had large spatial variation. As the gas concentrations in shallow boreal streams with moderate discharge equilibrate quickly with the atmosphere (Teodoru et al., 2009; Wallin et al., 2013), the supersaturation of C gases in streams is a sign of replenishment from adjacent soils (Rasilo et al., 2017). Thus, the concentrations of C gases, as well as concentrations of DOC in streams, reflect the biological and hydrological conditions in the catchment, especially in the riparian zone. Seasonal changes in the water table regulates the connection between the stream and the groundwater sources, which especially in the riparian zone contain large quantities of CO$_2$ (Rasilo et al., 2011; Leith et al., 2015), CH$_4$ (Hope et al., 2004) and DOC (Lyon et al., 2011). The different pattern of concentration of C species could result from a different location and different export rates in the soil profile. When water infiltrates through the mineral soil, most of the DOC in water precipitates in the illuvial layer of the podzolic soil. Thus, the deeper groundwater is usually more diluted with DOC compared to the surface layers. The highest CO$_2$ export in riparian soil has been observed at 30–50 cm depth, while continuous, but smaller export takes place in layers deeper than 65 cm (Leith et al., 2015). Also, the CH$_4$ increases in the soil profile towards the deeper anoxic layers (Hope et al., 2004).

Furthermore, prevailing hydrological conditions in the catchment alter the soil water movements and C mobilization from different depths through different infiltration rate in dry and wet soils. The event response in wet soils is immediate, while in dry soils, the time lag between the event and water flow could be up to 36 days (Ilvesniemi et al., 2010). However, the soils in the upland catchment typically reach the maximum water capacity in late autumn or early winter (Ilvesniemi et al., 2010), which predict the fast response of events both in autumn and spring. Besides, the environmental conditions of previous seasons and years on C concentrations in aquatic surfaces have been addressed in numerous studies before (Ågren et al., 2010; Einola et al., 2011; Tiwari et al., 2018).

**External input into the lake**

In years 2011 to 2013, the highest lateral transport took place in spring, when a remarkable quantity of C gases (reported from years 2011–2013) and DOC (reported from 2013) were transported. There was a time lag between the peak of the riverine C input into the lake and the atmospheric C release from the lake at the ice-out, i.e., the peak input took place ca. 10 days before and the hydrological spring event was almost over at the time of the ice-out. This mismatch in timing of riverine transport and ice-out indicate relatively large C inputs under the ice cover. The riverine input accounted for 15–24 % of the atmospheric release of CO$_2$ and 18–42 % that of CH$_4$ during the first week after the ice-out and thus, has an essential role in lake C dynamics. The involved external C inputs are mainly expected in organic form in late autumn (Karlsson et al., 2013), and they thus influence the under-ice CO$_2$ dynamics in lakes.
through mineralization of DOC (Striegl et al., 2001). Winter season has typically low flow conditions and thus, negligible external C inputs (Ågren et al., 2007). Even though less reported, there are recent studies on high inputs of allochthonous CO₂ (Pasche et al., 2019) and DOC (Cortés et al., 2017) in late winter in lakes. This thesis confirms that their importance can temporarily be remarkable in lake C dynamics and form an essential part of the atmospheric C gas release.

At the ice-out, there was a large increase in the whole-lake storage of both C gases in comparison to estimates based on the last under-ice measurements 2 to 5 weeks before. With CH₄, this increase was unexpected, since, during ice cover period, we detected neither high concentrations nor gas accumulation. No anoxia in the hypolimnion was detected either, which typically is required for high CH₄ accumulation (Juutinen et al., 2009). The growth of whole-lake CO₂ storage was linear under the ice cover, but similarly to CH₄, there was an unexpected increase at the end of the ice cover period. Thus, the plausible explanation for increases is the C-rich hydrological peak, which determines the under-ice storage of both gases. The CH₄ input through the main inlet before the ice-out was remarkable and equalled the whole-lake storage of CH₄ at the time of the ice-out either completely (2011, 2012) or partly (2013). The increase in the whole-lake storage of CO₂ also agrees with the latter, although the external input was less pronounced.

For further proof, the synchrony between lateral C inputs in all the 15 inlets around the lake and automatically measured lake CO₂ dynamics in the stratified lake during the ice cover period confirm the terrestrial lateral signal in the lake. Depending on the volume and intensity of the hydrological events in streams in spring and water storage conditions in the catchment, the terrestrial inputs entered the lake as surface inputs and increased the CO₂ concentrations at surface layers in the lake with corresponding volume and intensity. For verifying the origin of CO₂ and distinguishing it from the in-lake movements and transport from hypolimnion, at the time of the increase, the lake was thermally stratified. Thus, the cold stream water input was forced to flow to the uppermost layer under the ice cover with the same temperature gradient (Bengtsson, 1996). After the onset of water column mixing, the terrestrial signal was lost.

**The export through the outlet**

Surprisingly high CH₄ concentrations and export from the lake in the autumn in comparison to the export and concentrations in the main inlet and open lake indicate that the source of CH₄ can be local and close to the outlet. The high C export from the lake refers to inputs from the surrounding peaty soils during high flow events and/or from the lake bottom from the shallow part, where the C tends to accumulate. Higher DOC export from the lakes in comparison to the inputs have been observed before, especially in lakes with similar kinds of dense macrophyte beds (Goodman 2011). The export from shallow lake bottom is a plausible explanation for high export from the lake, as also the CH₄ probably originated from the part of the lake with dense stands of aquatic macrophytes, where the CH₄ production can be high (Kankaala et al., 2005).
The volume of the output water constantly exceeded the volume of the main inlet and in spring, the volume of all inlets together. This indicates additional water inputs, e.g., groundwater flows. They can form a small but continuous source of CO₂ (Leith et al., 2015) and can be responsible for up to 64 % of the total external DIC inputs into the boreal lake (Einarsdottir et al., 2017). However, the groundwater flow paths through the spatial gradient differ from the event-related surface inputs and are affected e.g., by the hydraulic conductivity and porosity of the soil (Alley et al., 2002). Thus, the groundwater inputs from riparian soils, rich in CO₂ (e.g., Rasilo et al., 2011) are a plausible explanation for increases of CO₂ in deeper portions of the water column during the ice cover period, although in this study, their detection from the CO₂ gradients in the lake was not possible.

**Importance of lateral and vertical transport in relation to the catchment**

The estimates of terrestrial fixed C ventilated through the lake show that the lake is an important conduit for terrestrial C to the atmosphere. The mean annual atmospheric C gas release over the three years study period from the lake was 9.3 % of the 15-year annual mean NEE value measured from the upland catchment area and upscaled over the lake catchment area. This result is in accordance with an earlier study from a boreal lake catchment, where the corresponding proportion was estimated to be approximately 10 % (Huotari et al., 2011).

The DOC runoff from the upland catchment was 0.75 g C m⁻² yr⁻¹ (15-year mean; 0.32 % of annual NEE) and small in comparison to other studies from forested upland catchments (e.g., 4 g C m⁻² yr⁻¹; Buffam et al., 2011). Unfortunately, we did not estimate the DOC export in the aquatic continuum in this study. However, we can assume that the DOC export may be higher in the lake catchment than in the upland catchment for several reasons, e.g., higher discharge, which is the primary driver of DOC fluxes in lotic ecosystems (Mattsson et al., 2015) and higher peatland coverage, which is known to increase both DOC concentrations and export from the catchment (Laudon et al., 2004; Ågren et al., 2007; Rantakari et al., 2010). Also, most of the peatlands in the lake catchment were drained. However, the drainage of peatlands does not necessarily influence the lateral DOC export, since the increased runoff compensates the decreased DOC concentrations after the drainage, and as a result, the lateral export remains similar in size to undrained peatlands (Rantakari et al., 2010).

Annual aquatic C gas transport in relation to the lake catchment was 1.70 g C m⁻² yr⁻¹. It was mainly in the form of CO₂, while CH₄ forms a negligible part of the total C export (ca. 0.04 %). The annual C gas export observed is in agreement with earlier results from headwater streams in Northern Sweden (Leith et al., 2015), Southern Sweden (Kokic et al., 2015) and Northern Finland (Juutinen et al., 2013). The large fluxes of all C gases in the outlet, especially in the autumn, indicate that the measuring point may not have been representative of the total catchment export, as the shallow area around the outlet appears to be a hotspot of lateral and atmospheric C fluxes.
4.2. Long-term DOC fluxes in the upland catchment

The DOC concentrations in runoff from the upland catchment increased over time, as found in other recent studies of lake and stream water DOC/TOC concentrations in the boreal zone (e.g., Sarkkola et al., 2009; Peltomaa and Ojala, 2012). The increasing trend in the DOC concentrations can be a result of increasing NEE within the catchment. A large proportion of the C assimilated by trees is allocated belowground, e.g., 18–21 % of assimilated C could be allocated to belowground respiration (Epron et al., 2011). Thus, the increase in NEE, which results in higher belowground C assimilation, may contribute to an increase in DOC in soil water and its export from the catchment area.

We observed an increase in annual soil temperature sum, as well as in the prolonged autumn season with higher temperatures. Higher temperatures can contribute to higher decomposition rate of soil OM and thus, increase the amount of DOC in soil water and then DOC export (Piao et al., 2008; Vesala et al., 2010). Higher soil temperatures can also increase the GPP of vegetation and lead to increased belowground C sink of photoassimilates (Pumpanen et al., 2012) and stimulate the decomposition through a priming effect (Fontaine et al., 2007; Kuzyakov, 2010). However, the litterfall from the tree canopy remained the same over the study period, and thus, the increasing litter production cannot explain the increasing DOC.

The long-term results show that the water balance, i.e., precipitation and runoff, especially during the spring freshet, ultimately determine the interannual changes in DOC export and concentrations, whereas the net primary production and litterfall of the previous year in the catchment sets the limits for the DOC export. This is probably explained by a time delay and C transport processes in the soil. During the first year, the mass loss of the needle litter through decomposition is ca. 30 % (Prescott et al., 2000), and a small proportion of this is converted to DOC (Cotrufo et al., 1994) and dissolved in water.

4.3. Prospects

Changes in lateral transport in spring

As riverine C transport is mainly controlled by hydrology, the predicted future changes in annual precipitation and typical snowmelt patterns can be crucial for C dynamics in the aquatic continuum. The results during the ice-cover period in warm winter and the following spring support this, as the timing of C transport differed significantly from years with typical winter conditions. In years 2011 to 2013, the lateral C transport mainly took place during the last two weeks before the ice-out after winter months with low lateral transport, a typical pattern confirmed also by the 15-year time series in the upland catchment. In 2014, however, the lateral transport occurred earlier during the winter months, when ca. 2/3 of total C transport of the six month study period took place. This change in timing of the transport differed significantly from results represented in Ågren et al. (2007), which showed that only
3–13 % of annual lateral transport of DOC takes place during winter months. Climate models predict increases in winter precipitation and warmer winters (IPCC, 2013), which may shift the lateral C transport towards winter months and thus advance the C inputs into the lakes. Nevertheless, despite the change in timing of transport, our results show that the amount of C transported does not necessarily change if the precipitation sum remains the same. Also, the proportion of transported organic and inorganic C remained unchanged.

However, as the C inputs during the ice cover period affect the C dynamics in a lake and atmospheric releases at the ice-out, the change in timing can probably influence the atmospheric C gas release. The spring emissions of CO₂ from the lake were typically high in comparison to a small headwater lake without external stream inputs (Huotari et al., 2011) and long-lasting in comparison to a larger humic lake, where the emission lasted for one week (Ojala et al., 2011). In contrast, after a warm winter and advanced external inputs, the surface water concentrations remained low at the beginning of open water period. This indicates that external inputs right before the ice-out enhance surface water concentrations in receiving lakes and increase the atmospheric emissions at the ice-out.

**Changes in annual fluxes**

Annually, the lateral transport from upland catchment until the export from the lake, as well as whole-lake storages and atmospheric release in the aquatic continuum were related to total precipitation and thus, discharge. The results in this study show that the lateral transport was higher during wet years, i.e., we observed an increase in C runoff of ca. 800 % from the upland catchment and 150 % more export from the lake during the wet year in comparison to the dry year. Besides, the lake atmospheric C release was 33 % higher during the wet year with higher lateral transport and weaker water column stability than in a dry and warm year. The corresponding values in the streams were 49 % and 80 % in the inlet and outlet streams, respectively. Thus, the predicted increases in precipitation in the boreal zone may result in remarkable increases in the terrestrial C transport from the catchment to the aquatic continuum as well as enhance the atmospheric C release from the aquatic surfaces.
4.4. Conclusions

Revealing the patterns and the C dynamics in less studied ecosystem interfaces and connections is essential for a more in-depth understanding of ecosystem functions and defining the natural C sources and sinks. The findings of this study, which was carried out in a typical boreal forested catchment with an unregulated lake and its stream network, improve the understanding of the land-stream-lake continuum as a whole and show that the C fluxes in the system must be studied concomitantly. By taking into consideration the connections in the hydrological continuum, we can improve the estimates of both lateral C fluxes as well as atmospheric C release from this dynamic system.

The results showed that, in the catchment scale, laterally transported terrestrial carbon is of great importance. The C release of the lake corresponded to almost 10 % of the NEE of the lake catchment. Besides, the aquatic lateral and atmospheric C fluxes will likely increase in the future. Thus, accurate and extensive time series are needed when estimating the future directions and intensity of the increased C fluxes in the land-stream-lake continua.
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