WATERBORNE CARBON IN NORTHERN STREAMS

Controls on dissolved carbon transport across sub-arctic Scandinavia

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Abstract

Waterborne carbon (C) forms an active and significant part of the global C cycle, which is important in the Arctic where greater temperature increases and variability are anticipated relative to the rest of the globe with potential implications for the C cycle. Understanding and quantification of the current processes governing the movement of C by connecting terrestrial and marine systems is necessary to better estimate future changes of waterborne C. This thesis investigates how the sub-arctic landscape influences the waterborne carbon export by combining data-driven and modeling methods across spatial and temporal scales. First, a study of the state of total organic carbon monitoring in northern Scandinavia was carried out using national-scale monitoring data and detailed data from scientific literature. This study, which highlights the consistency in land cover and hydroclimatic controls on waterborne C across northern Scandinavia, was combined with three more detailed studies leveraging field measurements and modeling. These focused on the Abisko region to provide insight to processes and mechanisms across scales. The thesis highlights that the governing transport mechanisms of dissolved organic and inorganic carbon (DOC and DIC respectively) are fundamentally different due to differences in release rates associated with the nature of their terrestrial sources (geogenic and organic matter respectively). As such, the DIC mass flux exhibits a high flow-dependence whereas DOC is relatively flow-independent. Furthermore, these investigations identified significant relationships between waterborne C and biogeophysical as well as hydroclimatic variables across large to small spatial scales. This thesis demonstrates that both surface and sub-surface hydrological processes (such as flow pathway distributions) in combination with distributions of C sources and associated release rates are prerequisite for understanding waterborne C dynamics in northern streams.

Keywords: dissolved carbon, DOC, DIC, TOC, sub-arctic, hydrology, Abisko

Svensk sammanfattning

Vattenburet kol (C) utgör en aktiv och betydande del av den globala C-cykeln. Detta är speciellt angeläget i Arktis där större temperaturökningar och -variationer förekommer jämfört med resten av världen vilket har tydliga potentiella konsekvenser för C-cykeln. Det är nödvändigt att skapa förståelse och kvantifieringar av rådande processer som styr transporten av kol i det akvatiska systemet eftersom det kopplar samman det terrestra och marina systemen och för att bättre uppskatta framtida förändringar i dessa miljöer på höga breddgrader. Denna avhandling undersöker hur det arktiska och sub-arktiska landskapet påverkar exporten av vattenburet kol genom att kombinera datadrivna- och modelleringsmetoder över olika tids- och rumsliga skalor. Först, genom att skapa en kontext för vår aktuella kunskap med avseende på vattenburet C i den nordliga regionen undersöktes övervakningssituationen för total organisk kol (TOC) i boreala och tundralandskap i norra Norden. Alltså, en studie utfördes genom att sammanställa nationell övervakningsdata tillsammans med mer detaljerad data från avrinningsområden från forskningslokaler. Genom att utröna den relativa betydelsen av olika vegetationstyper för TOC över hela norra Norden erhålls en bas att bygga processbaserad förståelse på. Denna storskaliga studie, som belyser betydelsen av vegetationen och den hydroklimatiska kontrollen på vattenburet C i norra Norden, kombinerades i avhandlingen med tre mer detaljerade studier med fältmätningar och modellering som grund. De detaljerade studierna fokuserade på Abisko-regionen i nordligaste Sverige för att få förståelse för de processer och mekanismer som råder på dessa nordliga breddgrader. Studierna omfattar olika rumsliga skalor; från källflöden till hela avrinningsområden. Genom detta tillvägagångssätt belyser avhandlingen att de dominerande transportmekanismerna för löst organiskt kol (DOC) och löst oorganiskt kol (DIC) är fundamentalt olika på grund av deras olika upplösningshastigheter, vilket i sig har att göra med deras terrestra ursprungsmaterial (berggrunden respektive organiskt material). Av den orsaken uppvisar massflödet av DIC hög korrelation med vattenflödet medan DOC är relativt flödesoberoende. Vidare, dessa studier identifierade signifikanta förhållanden mellan vattenburet C och biogeofysiska och hydroklimatiska variabler över olika tids- och rumsliga skalor. Denna avhandling visar att både hydrologiska processer över och under markytan (så som fördelning av flödesvägar) i kombination med fördelning av kolets ursprungsmaterial i landskapet och sammanhörande upplösningshastigheter är grundförutsättningar för att förstå det vattenburna kolets dynamik i nordliga vattendrag.

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Thesis content

This doctoral thesis consists of a summary and four papers. The papers are referred to as papers I to IV in the summary text.

I

Jantze, E. J., Dahlke, H.E., Jaramillo, F. and Lyon, S.W., The state of dissolved carbon export across boreal and tundra environments in Scandinavia. In review at Science of the Total Environment.

II

Jantze, E.J., Lyon, S.W. and Destouni, G., 2013, Subsurface release and transport of dissolved carbon in a discontinuous permafrost region, Hydrology and Earth Systems Sciences, 17, 3827-3839. doi:10.5194/hess-17-3827-2013.

III

Giesler, R., Lyon, S.W., Mörth, C-M., Karlsson, J., Karlsson, E.M., Jantze, E.J., Destouni, G. and Humborg, C, 2014, Catchment-scale dissolved carbon concentrations and export estimates across six subarctic streams in northern Sweden, Biogeosciences, 11, 525-537, doi:10.5194/bg-11-525-2014.

IV

Jantze, E. J., Laudon, H., Dahlke, H.E. and Lyon, S.W., 2015, Spatial variability of dissolved organic and inorganic carbon in sub-arctic headwater streams. Accepted in Arctic, Antarctic, and Alpine Research.

Co-authorship

The co-authorship of the papers reflects the collaborative nature of the underlying research. For Papers I, II and IV, I was the main responsible for all the analysis and had the responsibility of organizing and writing the papers. For Papers I and IV I had the main responsibility for designing the studies. In Paper III, I acquired, compiled and processed parts of the data that significantly contributed to the study.

Abbreviations and symbols

C   Carbon
CRU   Climate research unit
DIC   Dissolved inorganic carbon
DOC   Dissolved organic carbon
EEA   European Environmental Agency
k   Release rate constant
MAAT Mean annual air temperature
MLR   Multiple linear regression
NIVA   The Norwegian Institute for Water Research
NLS   National Land Survey of Finland
NMA   The Norwegian Mapping Authority
P   Precipitation
PCA   Principle component analysis
PET   Potential evapotranspiration
Q   Discharge
q   Specific discharge
r   DOC/DIC dissolution rate
SGU   Swedish Geological Survey
SLU   The Swedish University of Agricultural Sciences
SMHI   The Swedish Meteorological and Hydrological Institute
SYKE The Finnish Environment Institute
T   Temperature
TOC   Total organic carbon

Errata

Page 3834 in Paper II: Furthermore, the DIC concentration *increasing* trend is consistent with increasing average

Introduction

The Arctic has undergone significant biophysical and climatic changes during past decades (ACIA, 2005; Serreze and Barry, 2011). It is recognized that the Arctic's climate has experienced greater temperature increases and variability compared to the rest of the globe. Over the past 80 years, arctic air temperatures over land have increased with significant impact of the cycling of water and carbon (C). Shifts in the Arctic climate have led to changes in the storage and fluxes of surface waters, in amounts and intensities of precipitation and in overall aquatic ecosystem health and functioning (ACIA, 2005) with consequence to C fluxes. This places the Arctic in a precarious position with regards to climatic changes. For example, tundra and permafrost environments, which cover much of the Arctic and are typically found at high latitudes and northern landscapes, are considered as future potential tipping elements (Lenton et al., 2008) whereby they could enter a new state of ecosystem functioning and C cycling under changes in climate. Further, shifts in the functioning of these landscapes brought about by climatic change could feedback to an amplification of global warming through the release of C from thawing permafrost soils. This is significant since Arctic ecosystems and permafrost soils have, despite the low levels of primary productivity, accumulated and stored organic matter in the subsurface because the cold climate inhibits decomposition. The total C stock in arctic soils is estimated to be about 1300Pg of which 800Pg is found in perennially frozen ground and 500Pg is found in non-permafrost or seasonally thawed soils (Hugelius et al., 2014). Soils in the Arctic have functioned primarily as C sinks during the Holocene; however, the decomposition rate of organic matter in arctic and sub-arctic soils has started to increase due to ongoing climatic warming. These systems thus could shift such that they no longer serve as net carbon sinks but potentially become large net carbon sources (Kling et al., 1991; ACIA, 2005; Fung et al., 2005; Schuur et al., 2008; Koven et al., 2011). The critical point at which these systems shift from fundamental sinks to sources of C is not clear (Lenton et al., 2008) but could cause the most significant feedback for the climate at high latitudes (Schuur et al., 2008). What is clear is that we lack a fundamental understanding of the cycling and movement of C through northern landscapes that forms the prerequisite for estimating future changes.

This lack of understanding is largely due to the complexity of interactions and mechanism governing current and future carbon cycling and transport in Arctic and northern environments. Many other factors besides increased decomposition of organic matter influence the cycling of C in high latitudes. For example, warmer air temperatures in combination with increased CO_2 levels in the atmosphere have promoted vegetation growth in high latitudes. Thus, the Arctic has been "greening" over the past decades, including increases in shrub and forest cover (Sturm el al., 2001; Frost and Epstein 2014; ACIA, 2005). This increased greening has consequences for processes both at local and regional scales in ecosystems, the water balance and active layer/permafrost dynamics (all of which have impact on C cycling and transport). For example, expansion of woody vegetation can alter the biotic and abiotic controls over C cycling, such as the decomposing rates of plant litter (DeMarco et al. 2014), albedo and biomass C storage (de Wit et al., 2014). Changes in land cover and climate can subsequently lead to changes in hydrological flow pathways, which in turn affect dissolved nutrient fluxes (Turner and Rabalais, 2003) and C fluxes (Lyon et al., 2010). The spatiotemporal extent of vegetation change, however, is not consistent throughout the Arctic, which complicates a direct estimation of its impact. Rather, Arctic greening varies widely depending on landscape and region (Frost and Epstein 2014). This heterogeneity in greening impact (and in other rapidly changing Arctic characteristics) necessitates investigations across scales to isolate mechanisms connecting terrestrial C stores to aquatic systems leading to variability in waterborne C.

It is precisely the waterborne transport of C that integrates terrestrial and aquatic ecosystems. For example, terrestrial soils are the primary source of dissolved organic C (DOC) in aquatic systems (Guo and Macdonald, 2006). However, the main sources of stream water DOC are different for boreal and tundra landscapes due to variations in hydrology and heterogeneities in land cover, whose coupled control on dynamics of waterborne C is still poorly understood. Recent ecosystem assessments have begun to acknowledge the role that lateral movement of water through terrestrial systems plays for DOC and dissolved inorganic (DIC) cycling (Cole et al., 2007). Cole et al. (2007) further acknowledged the loss of C during transport from land to ocean, through sedimentation and outgassing, and lack of process representation in global climate models. Considering the transport of DOC from the terrestrial to marine environment, there are increasing DOC concentrations in surface waters across northern Scandinavia (e.g., Hongve et al., 2004) and similar trends having been observed for DIC (e.g. Paper II of this thesis). For the case of DIC, the C originates from weathering and dissolution of unconsolidated sediment or bedrock system. This is a significantly different source loading zone and process compared to DOC in the landscape. Weathering is a relatively slow process (compared to the decomposition and dissolution rates for organic carbon) and often more homogenous across scales, which impacts the dynamics of DIC in stream water (Paper II).

Carbon sources are only part of the story. The flow of water within the terrestrial system is the other integral component of dissolved C transport. The flow of water through the Arctic landscape, however, is often complicated by the presence of frozen zones in the subsurface. The subsurface pathways of water

flow in many Arctic environments are determined by permafrost and ground ice distributions in the landscape (Carey and Woo, 2001; Framton et al., 2011; Sjöberg et al., 2013; Bosson et al., 2012). In a warming climate, deepening of water flow pathways in the arctic landscape can be brought about as an effect of permafrost degradation (Frey and McClelland, 2009; Rowland et al., 2010). As such, the terrestrial freshwater cycle in the Arctic is intimately coupled to the cryosphere and with the existence (or absence) of permafrost. Also, seasonally frozen ground conditions in winter can create an abundance of relatively deep flow pathways affecting mineral weathering (Figure 1; MacLean et al., 1999) and DIC production (Lyon et al., 2010). Counter to this, during the spring freshet and summer seasons, there may instead be a dominance of near-surface flow pathways through organic-rich soil horizons (Figure 1; Carey, 2003; Carey and Quinton, 2004) leading to increased concentrations of DOC in aquatic systems. Once into the aquatic system, several other processes can affect DOC and DIC transport including sorption in mineral soils (Carey, 2003, Ågren et al. 2007), in stream processes such as microbial degradation, transformation and mineralization (Kalbitz et al., 2003), degassing and sedimentation (Cole et al., 2007). There remains limited mechanistic understanding of the coupling between hydrological and biogeochemical processes that is relevant across northern Scandinavia in specific (and northern latitudes in general). This is, in part, owing to the remoteness of these landscapes that leads to a lack of empirical studies and data across scales to support development (and testing) of process representations.

A more complete mechanistic understanding and quantification framework is needed to characterize current conditions and estimate potential future changes to the hydrological transport of dissolved carbon in Arctic landscape. This thesis develops such a framework and exemplifies its application to a sub-arctic landscape in northern Sweden. The main goal of this thesis is to understand how the sub-arctic landscape influences the waterborne carbon export by combining data-driven and modeling methods on various spatial and temporal scales. The main objectives for this thesis and the papers in which they are addressed (Table 1) include:

Objective A: To identify potential links between waterborne C in streams and landscape characteristics for northern environments.

Objective B: To investigate the spatial variability of waterborne C in streams across Scandinavia.

Objective C: To resolve catchment-scale mechanisms that control waterborne C concentration and load dynamics in streams.

Table 1: Coverage of the thesis objectives by the included thesis papers.

Objective / Paper		I	II	III	IV
A	Potential links between waterborne C and landscape characteristics.	■			■
B	Spatial variability of waterborne C across Scandinavia.	■		■	■
C	Catchment scale mechanisms controlling the dynamics of dissolved C		■	■	

Study sites and data

The research in this thesis starts off with a synthesis study of the state of total organic carbon (TOC) monitoring in the boreal and tundra environments in Norway, Sweden and Finland using national monitoring data and scientific literature. Three more detailed papers using field measurements and modeling focused on the Abisko region in northernmost Sweden follow this broad-scale investigation. Together, this field-model coupled design allowed for development of process and mechanism insights across scales. The scales and study sites considered are presented in the following sections and summarized in Table 2.

Paper I

Waterborne C in boreal and tundra environments of Scandinavia were investigated in Paper I using catchment-scale inventories (Table 2). The studied catchments covered approximately half of Sweden's area and almost the entirety of Norway and Finland. The land cover types in the region (to a large extent) are comprised of forest, shrubs, wetlands, agriculture and tundra. The main data considered (here TOC concentrations) were obtained from both national monitoring programs and research catchment sites located in Sweden (Krycklan, Stordalen and Tarfala) and Norway (Langtjern and Storgama). These research catchments were selected to complement scale and provide more homogenous land covers across the range of national monitoring catchments for comparison. The studied catchments (a total of 59 national monitoring catchments and 7 research catchments) span large temperature and precipitation gradients from north-to-south due to latitude and east-to-west due to altitude. The mean annual air temperatures for all catchments ranged between -5.8 and 6.1°C and the mean annual precipitation for catchments ranged between 463 and 3285mm yr^{-1}, with the highest precipitation in Norway where the mountains give an orographic lift. For this study, TOC concentrations in streams at the outlets of the catchments and topographic data were accessed from publically available national monitoring programs and mapping authorities, respectively, in each country. For the research catchments, the TOC

Table 2: Aspects accounted for in the methodology in Papers I-IV

Aspects	Paper I	Paper II	Paper III	Paper IV
Time period(s) used for the calculations of mean dissolved C and climate data	Years with available data 1967-2013	1986-2010	Years with available data 1982-2010	Synoptic study 10 days in August 2012
Catchments investigated (number of catchments and area)	Regional scale (13 Norwegian, 31 Swedish and 23 Finnish) 0.1-49217km^2, median: 295km2.	Abiskojokka 566km^2	Homojokka, Stream 2, Pessijokka, Stream 4, Miellajokka and Abiskojokka 5.2-566km^2	Sub-catchments of Abiskojokka (32) 0.10-576km^2
Waterborne carbon variables	TOC	DOC, DIC	DOC, DIC	DOC, DIC, CO_2
Hydroclimatic and water chemistry variables considered	T, P, PET	Q, r	Flow pathway length and flow pathway length/ gradient	Q, q, EC, water travel time, base cations, Si^{4+} and alkalinity
Landscape characteristics considered	Catchment area, land cover	Not considered	Catchment area, elevation, slope, aspect	Catchment area, land cover, lithology
Data (source)	Köppen's climate classifications (Peel et al., 2007), TOC (NIVA, SLU, SYKE, Laudon et al., 2013; de Wit et al., 2007; Olefeldt et al., 2013) Land cover (Corine Landcover 2006, EEA) DEM (NMA, SLU, NLS) T, P, PET (CRU, SMHI)	DOC, DIC, alkalinity (SLU, Lyon et al. (2010), Q (SMHI) DEM (SLU)	Q, T, P (SMHI) DEM (SLU)	DEM (SLU) Q, T, P (SMHI Land cover and geology (SGU)
Statistics	Simple and multiple linear regression, PCA	Simple linear regression, generalized least squares approach (Box et al., 2008)	Time series regression, simple linear regression, mean absolute percentage error, mean absolute deviation and mean square deviation	Simple and multiple linear regression, Spearman's and Kendall's rank correlation coefficients

concentration data came from the scientific literature (Olefeldt et al., 2013, Laudon et al., 2013; and de Wit et al., 2007) or, in the case of Tarfala, from first-hand sampling and analysis. Land coverages were taken from the Corine raster map (Corine Landcover 2006) that was accessed from European Environmental Agency (EEA) for all catchments (Table 2). For several of the research catchments, these coarse data were augmented with reported land coverage percentages.

Papers II-IV

To further study dissolved C in tundra environments, detailed observations and modeling simulations were carried out in the Abisko region (Figure 2; 68°21'36" N, 18°46'48" E). The Abisko region is situated in a mountainous sub-arctic environment with glacially eroded valleys, 200km north of the Arctic Circle in the discontinuous permafrost zone. The permafrost decreases in coverage along a gradient from west to east (Brown et al., 1998; Johansson et al., 2006). Mean annual air temperature was -0.8°C in the reference time period 1961–1990 but it has since 2000 significantly exceeded 0°C, which is a threshold for many ecological and cryospheric processes (Callaghan et al., 2010). Mean annual precipitation for the same time period was 304mm yr^{-1}, but there is a strong altitudinal and eastward increasing gradient in precipitation. Paper II and Paper IV deal with Abiskojokka catchment (Table 2) that has an elevation range of 350m to 1600m a.s.l. causing strong precipitation gradients within the catchment. Paper III deals with dissolved C at the outlet of Abiskojokka and in streams draining 5 additional catchments (Table 2) located between the towns of Abisko in the west and Kiruna in the east, across the aforementioned permafrost and precipitation gradients. All 6 catchments are north-facing and drain into to the Torne River system. The vegetation both in Abiskojokka catchment and across the region is dominated by birch forest (*Betula pubescens*

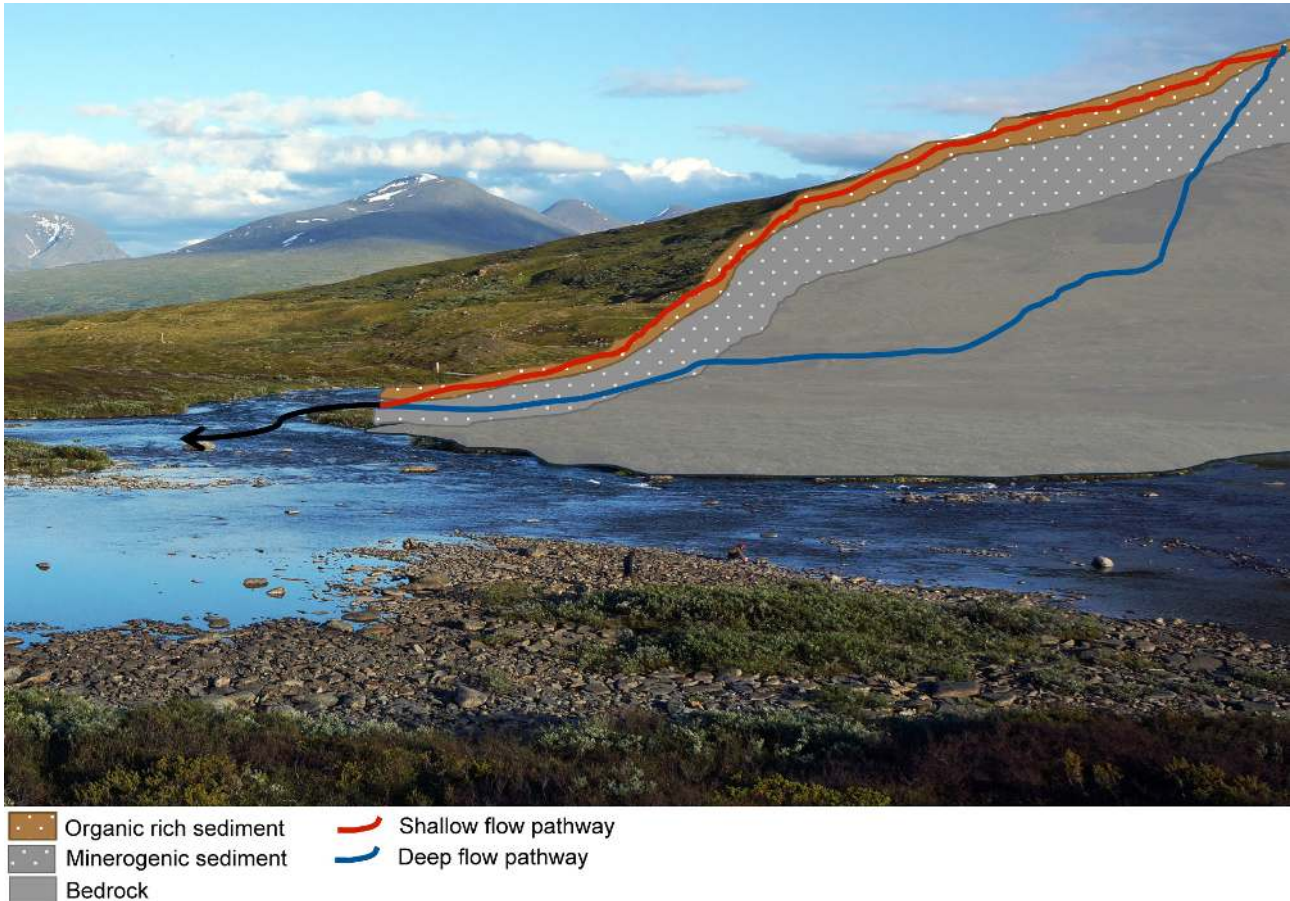| Organic rich sediment | Shallow flow pathway |
| Minerogenic sediment | Deep flow pathway |
| Bedrock | |

Figure 1: A photo from a tributary stream to Abiskojokka showing conceptual example of different flow pathways of water and the subsurface sediment and bedrock. The colored lines exemplifies different water flowpaths; the red line shows shallow (quick) pathways that prevails in wet conditions, e.g. during spring flood. The blue line marks deep flow pathways that prevail during stable low flow conditions, e.g. during winter months.

tortuosa) in the sub-alpine zone, willows (*Salix sp.*) in the low alpine zone and dwarf birch (*Betula nana*) and heath in the middle to high alpine zone (Rafstedt et al., 1985). Bedrock outcrops dominate the landscape with non-continuous to thin quaternary deposits cover. The mineral sediments are covered by a thin O-horizon that ranges between 0 and 50cm; soils are cryoturbated at higher elevations (Becher et al., 2013).

Methodology

The hydrological catchment has been used as the fundamental spatial unit throughout this thesis because it provides a topographically-limited boundary that makes it possible to analyze and compare hydrological and solute fluxes. All investigations in this thesis were carried out in the boreal and tundra environments of Scandinavia. Spatial variability and dynamics of waterborne C were studied in relation to hydrology and biophysical catchment characteristics throughout this thesis. Spatially, the studies comprise a wide range from headwater systems and streams (Papers II-IV) to large river basins (Paper I). Specifically, the investigations were carried out within the Abiskojokka catchment (Paper II; Paper IV), the

Abisko region (Paper III) and the boreal and tundra zone of Scandinavia (Paper I; Figure 2). Temporally they range from synoptic sampling to analysis of decadal long-term monitoring data (Table 2).

Paper I: Region-scale overview

To identify potential controls on total organic carbon (TOC) concentration in stream water as well as the TOC variability regionally, Paper I investigated the relationship between TOC concentration and land cover across northern Scandinavia. To do this we summarized TOC concentrations for 66 streams (12 Norwegian, 31 Swedish and 23 Finnish) of which 59 were national monitoring locations and 7 were research catchment sites (Figure 2) using current national stream monitoring data and research catchment data reported in the scientific literature. To enable comparison and limit the spatial extent of the study, the studied catchments were confined to the boreal and tundra region of Scandinavia, thus a region with comparable climate. With this synthesized TOC and land cover data set we further made a gap analysis to find potential monitoring bias and to estimate the representativeness of the monitored sites in relation to the land coverage. This was done by mapping the spatial variability of TOC concentrations in monitored streams relative to land
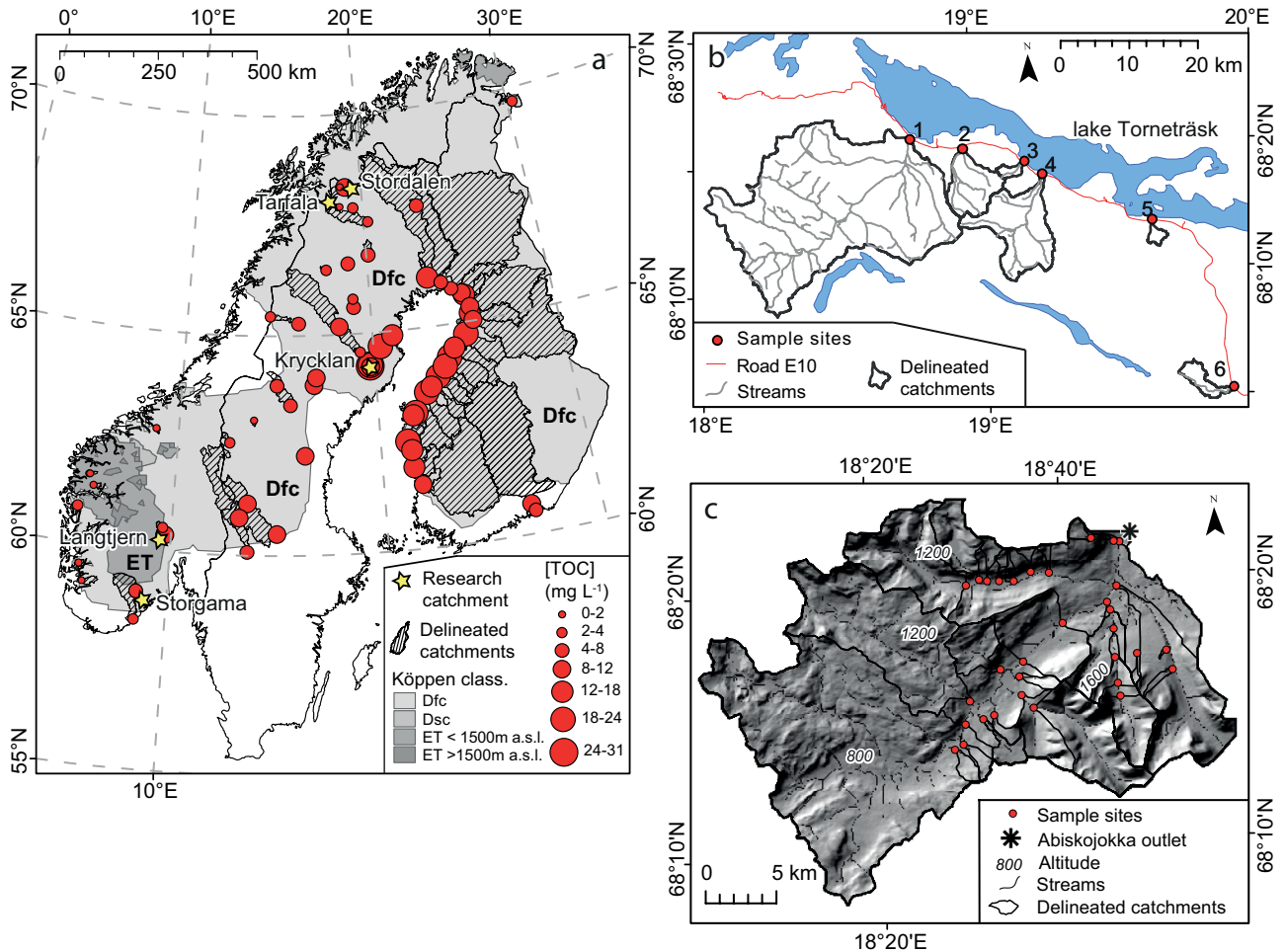
Figure 2: Maps showing studied catchments in this thesis. a) Catchments considered in Paper I and the annual average TOC concentrations (red circles) at the outlet within the boreal and tundra zone of Norway, Sweden and Finland according to the Köppen classifications. The research catchment sites are indicated with stars. b) Catchments 1-6 considered in Paper III; Stream 1 (Homojokka), Stream 2, Stream 3 (Pessijokka), Stream 4, Stream 5 (Miellajokka) and Stream 6 (Abiskojokka) are all parts of the Torne river basin. Sample sites are indicated with red circle. c) Abiskojokka catchment and sub-catchments. For Paper II, the monitoring site of Abiskojokka outlet is indicated with black star. For Paper IV, the sample sites are indicated with red circles.

coverage distributions. For each site, time series of average monthly and annual TOC concentrations and climate data were assembled for the common overlapping time period 1967-2013 (Table 2). A raster land cover map (Table 2) was further used to calculate land cover percentage for each catchment area draining through each monitoring location. The relationships among TOC, climate variables and land cover were studied on both monthly and annual basis to investigate possible links (see Table 2 for a list of the statistical analysis considered). We further investigated the monthly TOC concentration dynamics in relation to percentage forest cover of the national monitoring catchments. This was done by ranking the monthly TOC concentration dataset in relation to increasing forest cover (from 0 to 100%) of the catchment area and extracting the upper and lower quintiles (80-100% and 0-20% respectively) of monthly TOC concentrations.

Paper II: Developing a transport framework

A framework to characterize the relevant conditions for hydrological transport of dissolved carbon was developed to resolve mechanisms behind dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) transport landscape to stream (see Box 1). This framework was based on a Lagrangian approach considering hydrological pathways and travel times of water. Tracking the movement of water makes it possible to estimate fluxes of solute through the landscape and its underlying geological formations. Together, this framework coupling water movement and solute transport makes it possible to quantify the release, transport and export of dissolved carbon (given the appropriate observations to calibrate against). Abiskojokka catchment was used as a specific case to exemplify and test this approach in a terrestrial sub-arctic landscape. Time series of annual flow-weighted DOC and DIC concentrations, annual DOC and DIC mass fluxes as well as annual total discharge for the Abiskojokka catchment were calculated for the time period 1986-2010 to test the mechanistic model using observed data. To compare the model and observed data we used the fluctuations around the average of solute fluxes for the model and the long-term data from Abiskojokka catchment. For the observed data we further investigated the

Box 1: A mechanistic framework for modeling of solute release and transport

Parameter	Measure	Dimensions
c	Annual flow-weighted mean concentration	$[M\ L^{-3}]$
s	Mass flux $s = c\ q_s$	$[M\ L^{-2}\ T^{-1}]$
S	Mass flow rate $S = cQ$	$[M\ T^{-1}]$
q_s	Specific discharge	$[L\ T^{-1}]$
Q	Total annual discharge	$[L^3\ T^{-1}]$
r	DOC/DIC dissolution rate	$[M\ T^{-1}\ L^{-3}]$
k	Release rate constant, with $1/k$ quantifying a characteristic time for the entire c_0^* to dissolve.	$[T^{-1}]$
τ	Advective (water) travel time through the stream tube	$[T]$
θ	Average volumetric water content	$[-]\ \dfrac{[L^3\text{water}]}{[L^3\text{bulk}]}$

Starting from Lyon et al. (2010), the concentration, c [$M\ L^{-3}$-water], and mass flux, s [$M\ L^{-2}\ T^{-1}$], of DOC or DIC in an arbitrary stream tube from the land surface through the soil-groundwater system to the stream can be expressed as:

$$c = r\frac{\tau}{\theta} \qquad s = r\ \frac{q_s\tau}{\theta} \qquad\qquad\qquad\qquad \text{(Eq. 1)}$$

where r is the average release (dissolution) rate [$M\ T^{-1}\ L^{-3}$] of DOC or DIC in the stream tube, τ is the advective (water) travel time [T] through the stream tube (essentially the time for 1 pore volume of water to flow through the stream tube), θ is the average volumetric water content (-) [L^3-water L^{-3}-bulk] (equals the porosity under fully saturated, groundwater conditions), and q_S is the local specific groundwater discharge (Darcy flux) [$L\ T^{-1}$] flowing into the stream through the interface with the soil-groundwater system.

Neglecting local, molecular diffusion and considering the simplest case of constant release (dissolution) rate (zeroth-order kinetics) from the solid into the mobile water phase, r can be further resolved and expressed as (Eriksson and Destouni, 1997):

$$r = \frac{dc}{dt} = -kc_0^* \qquad\qquad\qquad\qquad \text{(Eq. 2)}$$

where t is running time [T], c^* is the average bulk concentration [$M\ L^{-3}$-bulk] of organic carbon for DOC and inorganic carbon for DIC in the solid, soil or aquifer, phase within the stream tube, c_0^* is the initial c^* at time $t = 0$, and k is a release rate constant [T^{-1}] (with $1/k$ quantifying a characteristic time for the entire initial bulk concentration c_0^* to dissolve). Inserting equation (2) into equation (1) (through r) and solving for initial conditions $c^*(0) =$ and $c(0) = 0$ throughout the entire stream tube, and boundary condition $c(t,0) = 0$ (zero concentration in incoming water to the stream tube) yields the following set of solutions for solute concentration c and mass flux s in the water flowing from the soil-groundwater stream tube into the stream at time t based on (Eriksson and Destouni, 1997).

For travel time $\tau \le 1/k$ (implying that c_0^* dissolves entirely (at time $t = 1/k$) first after the first pore volume of water has flown through the stream tube (at time $t = \tau$)) and different running time t, the resulting solution expressions for concentration c and mass flux s are:

for $t \le \tau$ (i.e., before 1 whole pore volume of water has flown through the stream tube)

$$\frac{\theta c}{c_0^*} = kt; \qquad \frac{\theta s}{q_s c_0^*} = kt \qquad\qquad\qquad\qquad \text{(Eq. 3a)}$$

for $\tau < t \le 1/k$ (i.e., after 1 pore volume has flown through the stream tube, but before c_0^* has been entirely dissolved)

$$\frac{\theta c}{c_0^*} = k\tau; \qquad \frac{\theta s}{q_s c_0^*} = k\tau \qquad\qquad\qquad\qquad \text{(Eq. 3b)}$$

for $1/k < t \le 1/k + \tau$ (i.e., after c_0^* has been entirely dissolved, but before 1 pore volume has flown through the stream tube thereafter)

$$\frac{\theta c}{c_0^*} = k(\tau - 1) + 1; \qquad \frac{\theta s}{q_s c_0^*} = k(\tau - 1) + 1 \qquad\qquad \text{(Eq. 3c)}$$

For travel time $\tau > 1/k$ (implying that c_0^* dissolves entirely (at time $t = 1/k$) before the first pore volume of water has flown through the stream tube (at time $t = \tau$) and different running time t, the resulting solution expressions for concentration c and mass flux s are: for $t \le 1/k$ (i.e., before c_0^* has been entirely dissolved)

$$\frac{\theta c}{c_0^*} = kt; \qquad \frac{\theta s}{q_s c_0^*} = kt \qquad\qquad\qquad\qquad \text{(Eq. 4a)}$$

for $1/k < t \le \tau$ (i.e., after c_0^* has been entirely dissolved, but before 1 pore volume has flown through the stream tube)

$$\frac{\theta c}{c_0^*} = 1; \qquad \frac{\theta s}{q_s c_0^*} = 1 \qquad\qquad\qquad\qquad \text{(Eq. 4b)}$$

for $\tau < t \le \tau + 1/k$ (i.e., after 1 pore volume has flown through, but before the last solute mass fraction dissolved from c_0^* has also been transported through thereafter)

$$\frac{\theta c}{c_0^*} = k(\tau - 1) + 1; \qquad \frac{\theta s}{q_s c_0^*} = k(\tau - 1) + 1 \qquad\qquad \text{(Eq. 4c)}$$

The solutions (3)-(4) apply for solute release and transport through a single stream tube, characterized by a single value of τ. Averaging over the whole ensemble of different stream tubes, with associated different τ, to the nearest stream in a catchment, yields average flux concentration $\overline{c_f}$ and mass flux into the stream network as (Lyon et al., 2010):

$$\overline{c_f} = \frac{1}{\overline{q_s}} \int_0^\infty s g(\tau) d\tau; \qquad \overline{s} = \overline{q_s}\, \overline{c_f} \qquad\qquad\qquad \text{(Eq. 5)}$$

where $\overline{q_s}$ is the average water flux into the stream, and $g(\tau)$ is the probability density function (pdf) that quantifies the spatial distribution of advective solute travel times τ through the catchment to the stream.

The solutions (3)-(5) provide a generic framework for solute release and transport through a catchment, which can be used to quantify different cases of subsurface DOC and DIC release and subsequent transport into streams.

existence of simple (linear) relationships for flow-weighted DOC and DIC concentrations, annual DOC and DIC mass fluxes and annual total discharge.

Paper III-IV: Detailed investigations across scales

Waterborne C export together with the mechanisms that control the dynamics of dissolved C in stream water can be explored across scales through field investigations, which was done in this thesis via two data-driven studies. Such approaches offer natural cross-scale follow up to the regional inventory of Paper I and the theoretical development of Paper II. As such, Paper III combines one year of field measurements of stream discharge, DOC, DIC, conductivity, alkalinity, silica and base cations for 6 sub-arctic streams (including Abiskojokka) together

with long-term dissolved C and discharge monitoring from the national monitoring programs for 2 of those streams (Figure 2). DIC concentration was estimated from previously established relationships between alkalinity and DIC concentration. The annual loads of DOC and DIC were calculated as the product of daily concentrations and stream flows. Using a topographic map, a basic terrain analysis was created as a proxy for the hydrology including slope, aspect and flow pathway length. Hydrological characteristics were investigated in relation to the one-year estimates of DOC and DIC loads. Furthermore, to put these values into perspective the hydrological characteristics were also compared to the long-term water chemistry. Thus, Paper III includes analysis of both long-term and seasonal temporal scales as well as across spatial scale.

Paper IV aims to identify possible links and the

spatial variability of dissolved C (DOC, DIC and CO_2) and landscape characteristics (vegetation, geology, physical variables) at smaller scales. The study was carried out as a data-driven analysis based on a synoptic (snapshot) field campaign of headwaters and sub-catchments to Abiskojokka catchment (Figure 2). In addition to the stream flow and dissolved C measurements, the study also includes stream water chemistry measurements of base cations (Ca^{2+}, K^+, Mg^{2+} and Na^+), silica, electrical conductivity (EC) and alkalinity. The topographic and biophysical features that were considered as potential controlling variables were: the catchment area, the average catchment elevation, and the percent land cover for various vegetation types, quaternary geology and lithologies within each catchment. A water travel time model (Lyon et al., 2010) was used to provide a proxy for the distributed hydrology in the catchments. We investigated the existence of relationship for C concentration and mass flux rates versus the catchments' biophysical characteristics, stream water flow and chemistry (Table 2). The spatial variability of dissolved C was tested in relation to the catchment characteristics with the aim to explore whether C export from catchments with a dominant landscape characteristic (i.e. relatively more homogeneous) were different compared to catchments in which the landscape relatively more heterogeneous. The spatial variability was estimated by comparing the variance for the measured dissolved C concentrations between catchments with higher-than-median values of a given characteristic and catchments with lower-than-median values of a given characteristic.

Summary of papers with main results

Paper I

Jantze, E. J., Dahlke, H.E., Jaramillo, F., Lyon, S.W., The state of dissolved carbon export across boreal and tundra environments in Scandinavia. In review at Science of the Total Environment.

Paper I presents a summary of TOC concentrations in national monitoring and research catchments across northern Scandinavia. The overview of TOC concentration in relation to land coverage shows that the current national-scale monitoring data have an even distribution in monitoring of small to large catchments and further that they represent a relatively good distribution of land coverage. This is a rather unique feature for a national-scale monitoring effort compared to sub-arctic to arctic environments in, for example, northern Asia and North America (Bring and Destouni, 2009). Specifically, forest coverage, which can be considered important due to its connections to TOC cycling, across the monitored streams has an even distribution from low to high

coverage among the catchments. There is however a slight overrepresentation of forested land (8%) and an underrepresentation of tundra environments (11%) relative to the distribution of these land cover types across tundra and boreal Scandinavia. Looking at annual average TOC concentrations, there was a significantly positive relationship to forest and wetland cover and a significantly negatively relationship to open space coverage (Figure 3).

Regarding at the TOC concentrations, the median and standard deviation of annual average TOC concentration for all catchments considered within the national monitoring programs was 8.0 ± 6.9 mg L^{-1}. This value can be compared to the individual monthly ranges of TOC concentrations across all catchments (Figure 4). To help put these large-scale monitoring sites into perspective, we also consider the seasonal fluctuations of TOC concentrations for smaller research catchments with considerably more homogeneous land covers. For the analysis of monthly TOC dynamics the national monitoring data set was sorted by the forest percentage cover and the upper and lower quintile of these coverages were identified. For the catchments in each of these quintiles, the average monthly TOC concentrations were determined (Figure 4). This monthly TOC analysis shows that catchments in the upper quintile of forest cover measured a wide range of TOC concentrations while the range for catchments in the lower quintile was small. Five of the research catchment sites (namely, Krycklan C2, Krycklan C7, Langtjern, Stordalen and Storgama) had monthly mean TOC concentrations that fell within the upper quintile of TOC concentration (Figure 4). All monthly TOC concentrations for Krycklan C4 were higher than upper quintile and all monthly TOC concentrations for Tarfala were below the lower quintile of the forest cover (Figure 4).

Multiple linear regression (MLR) was used to determine how landscape and climatic variables related to TOC concentrations across the national monitoring sites on a monthly scale. By using this method we were able to partially identify the most important variables for predicting TOC concentrations for the different months of monitoring. The MLR results are relatively consistent in showing that average maximum July temperature was the most important variable for predicting TOC concentration in most of the months of TOC concentration measures (Table 3). This indicates the importance of summer season temperature when biomass productivity is the highest, active layer depth and snowmelt (in the alpine environments). There were also consistent results indicating that catchment area and average total August precipitation were frequently ranked as important variables for predicting monthly TOC concentrations.
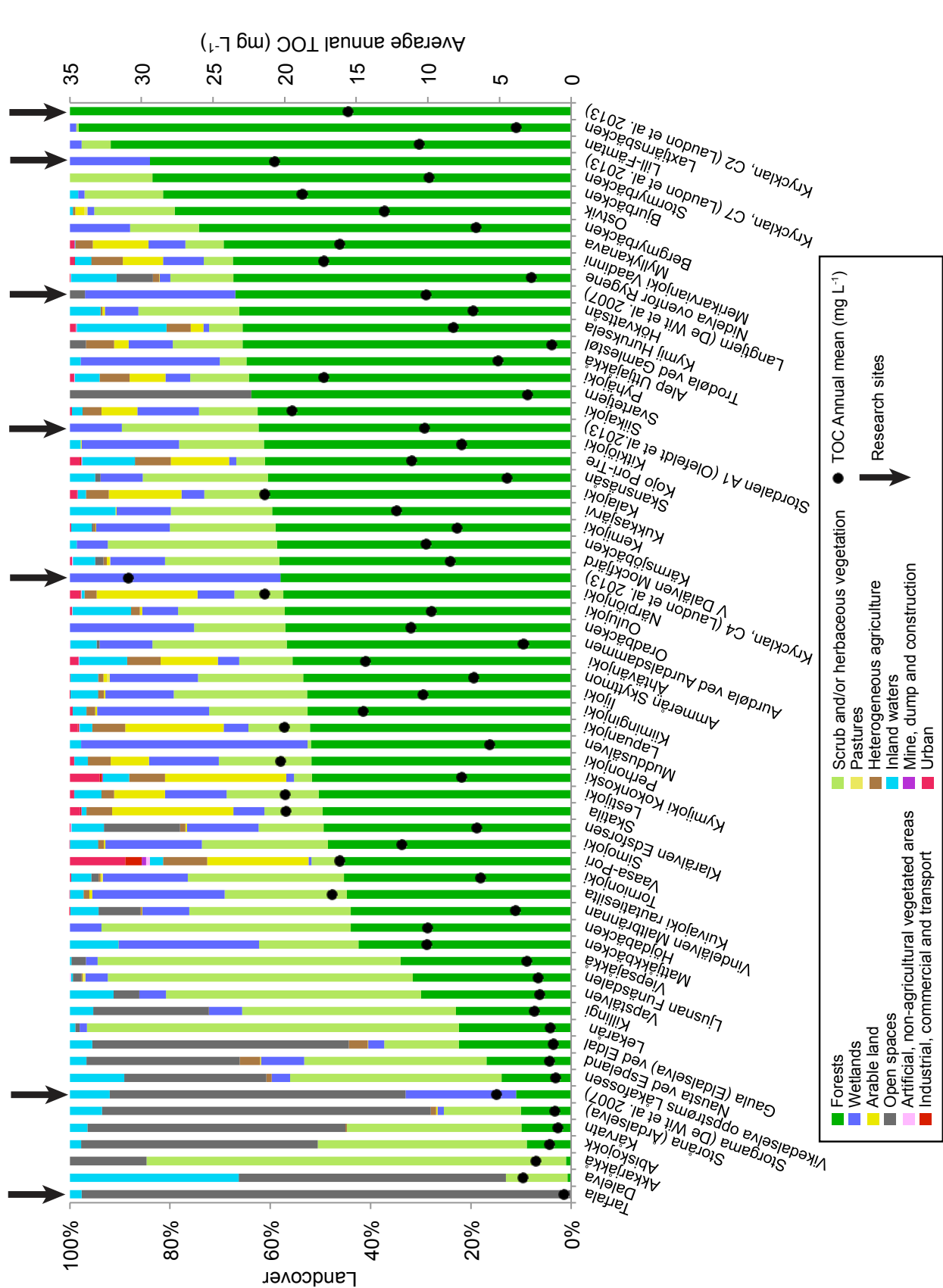
Figure 3: Land cover distributions and annual average TOC concentrations across 66 catchments in Norway, Sweden and Finland. Arrows along top indicate research catchment sites.

Table 3: Significant variables (with coefficients listed) for the multiple linear regression of monthly TOC concentrations (dependent variable) versus monthly climatic parameters, land cover and catchment area (independent variables). The color ranking 1-14 is a significant ranking where 1 (red) is the most significant predicting variable.

The color scale 1-14 is a significant ranking, where 1 is the most significant and predicting variable

Color legend: 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9-14

Group	Variable	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Annual av.
	p-value	1×10^{-18}	3×10^{-18}	3×10^{-17}	2×10^{-19}	2×10^{-18}	2×10^{-2}	1×10^{-17}	8×10^{-18}	4×10^{-1}	2×10^{-2}	2×10^{-18}	6×10^{-22}	6×10^{-1}
	intercept	-7	-14	-18	-24	7	-9	-19	14	-42	0.5	-3	2	-42
	Catchment area	-0.2	-0.2		-0.2	-0.1	-0.2	-0.2	-0.1	-0.2	-0.1		-0.1	-0.1
Land cover	Urban			-398							-62			
	Industrial, commercial and transport			961										
	Mine, dump and construction											2		
	Heterogeneous agriculture	-16				-18	-17	-20	27		-4		-23	
	Open spaces												-68	76
	Arable land			46										
	Forest													-9
PET	November		0.4											
	August												1.3	
	July	-0.3	-0.2			-0.4	-0.3						-0.6	0.3
	March												-0.7	
	February											0.4		
P	August	-1	-0.1	-0.8	-0.1		-0.8	-0.1	-0.2		-0.1	-0.1	-0.3	
	July						0.7						-0.8	
	April												-0.2	
	March												0.3	
Average min T	November	2		1	2		1	2						
	April				-2									
	January								-3					
Average max T	November												5	
	July	3	4	3	2		3	3		3	1		9	
	June													1
	March		-1			-2				-2			-2	
	January												-2	
Average T	November		1						4				5	
	August												-2	
	July					3						0.3		
	May			-2										
	April	-2						-2						

Figure 4: The range of average monthly TOC concentrations for the upper and lower quintile of forest coverage from the national monitoring sites in Norway, Sweden and Finland. Symbols are the monthly average values from the research catchments. Note that the TOC concentration for Stordalen A1 is the annual average concentration reported by Olefeldt et al. (2013).

Figure 5: Modeled DOC transport dynamics. The results show how changes in the release rate k (or characteristic release time 1/k) influences DOC concentration and mass flux from the soil–groundwater into the stream system.

Figure 6: Modeled DIC transport dynamics. Results are shown on log scale for single, variable, uniformly-distributed travel time both with an average value of 1 year but for different values of release rate k (or characteristic release time 1/k).

Paper II

In this paper we analyze how concentration and mass flux rate of DOC and DIC look under various relevant conditions for their release and transport from terrestrial to aquatic environments. To do this we use conceptual frameworks for dissolved C transport (see Box 1). For the case of DOC, the main conceptualization was that the average bulk concentration of organic C in the soil, unconsolidated sediments or bedrock originates from the bulk of organic matter in the organic horizon that can be annually renewed, dissolved and finally transported as DOC to the stream each year. For the case of DIC, we conceptualize that the average bulk concentration of DIC originates from weathering of the unconsolidated sediments or bedrock and that this material is not renewed. To check the validity of these conceptual modeling frameworks, we used a catchment-scale water travel time with an average of

1 year corresponding to the average travel time of water in Abiskojokka catchment (Lyon et al., 2010) as a base assumption. What this implies for the DOC export, which is driven by the short organic C cycle whereby vegetation grows, senesce and decomposes every year, is that that DOC release time should be on the order of 1 year or smaller. Basically, DOC release times should be smaller or equal to the average water travel time in the catchment (or 1 year in the case of Abiskojokka). Counter to the processes driving DOC export, weathering, which dominates DIC formation in the Abisko landscape, is a slow process relative to decomposition and release rates for organic C so that complete depletion is expected to take long times (i.e., on the order of geological time scales). Due to the slow nature of weathering processes, thus, it is anticipated that DIC has a dissolution time that should be greater than the average travel time in the catchment.

The modeling results for the DOC case showed that for a water travel time of 1 year, the lower average dissolution time ($1/k$) gives longer time to complete dissolution of the annually renewed C source of DOC. In addition, decreasing dissolution time leads to temporal fluctuation of DOC concentrations in the stream and longer time to reach stable average concentration (Figure 5). The modeling results for the DIC case showed that there was a much greater dissolution time for DIC compared to average water
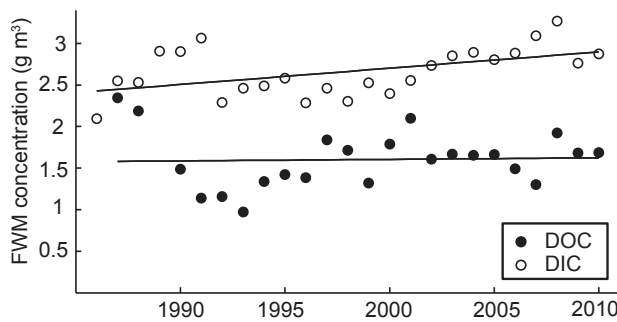
Figure 7: Observed dissolved carbon time series of annual flow-weighted mean concentration of DOC and DIC at the outlet of Abiskojokka catchment. The DIC concentration exhibits a significant positive trend.
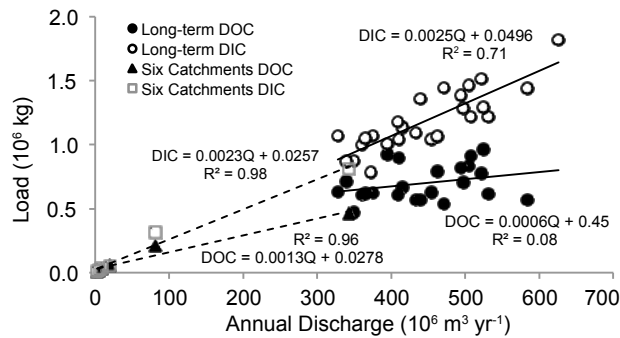
Figure 8: Annual DOC and DIC loads in relation to total annual stream flow for Abiskojokka based on available long-term data in comparison to the range of values observed for the six catchments considered in this study (here, filled triangles are DOC and open triangle are DIC). Solid trend lines are fit to the long-term data for Abiskojokka while dashed trend lines are fit to the annual data for the six catchments.
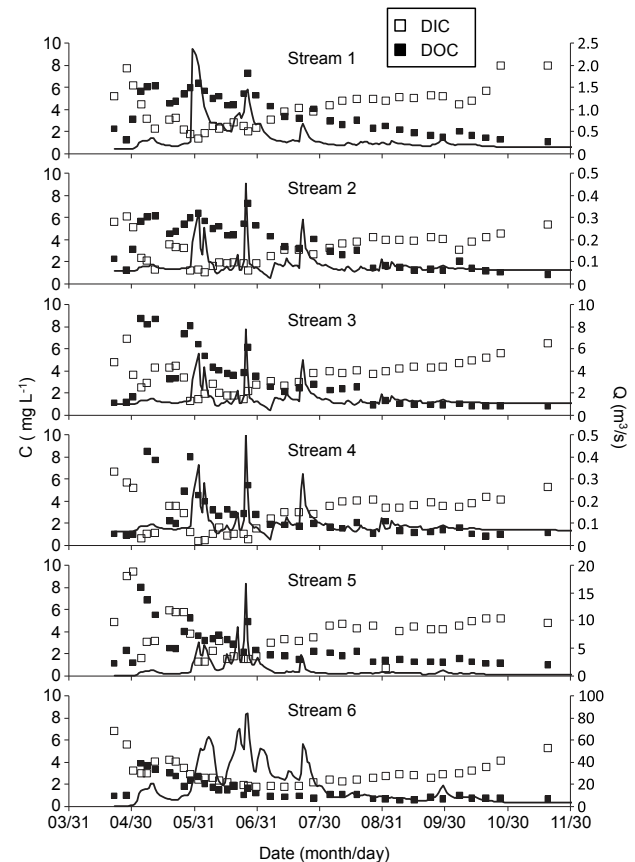

Figure 9: Observed annual variation for the year 2008 in stream water DOC and DIC concentrations in the six streams studied. The black line shows stream water flow (hydrograph) for each stream.

travel time. This implies that DIC concentration in stream water, under the conditions prevailing in Abiskojokka, is determined by the dissolution time rather than source mass fluctuations. Thus, the DIC should not exhibit temporal fluctuation effects, which is evident in the case for DOC concentration, consequently DIC mass flux should be dominated by specific discharge (Figure 6). Simply put, DIC mass flux should show high flow-dependence due to a relatively slow average DIC (weathering) release rate from its essentially constant geogenic source, which also keeps the DIC concentration essentially constant in time. Contrary, DOC should show flow-independence of both the concentration and the mass flux due to the annual renewal of organic matter (Figures 5 and 6).

To put these calculated dynamics for DOC and DIC transport into context they were compared to a time series of annual flow-weighted mean DOC, DIC concentrations, mass flow rates as well as annual discharge from Abiskojokka for 1986-2010 (Figure 7). As expected from the mechanistic model we found that annual DIC mass flux showed high positive correlation with annual discharge and that DIC and DOC concentration as well as DOC mass flux rate essentially are flow-independent (Figure 8). For the time period 1986 to 2010 Abiskojokka exhibits a significant decreasing trend in total annual discharge and a significant positive trend in the annual flow-weighted average DIC concentration (Figure 7). The decreasing discharge trend is in line with the mechanistic model solution (Box 1) with increasing water travel times. Similar negative water discharge trends have previously been observed and modeled (Lyon et al., 2009; Dahlke et al., 2012: Sjöberg et al., 2013) and could primarily be attributed to thawing of permafrost (Dahlke et al., 2012). The DIC concentration increase is consistent with these increasing travel times and the mechanistic modeling framework.
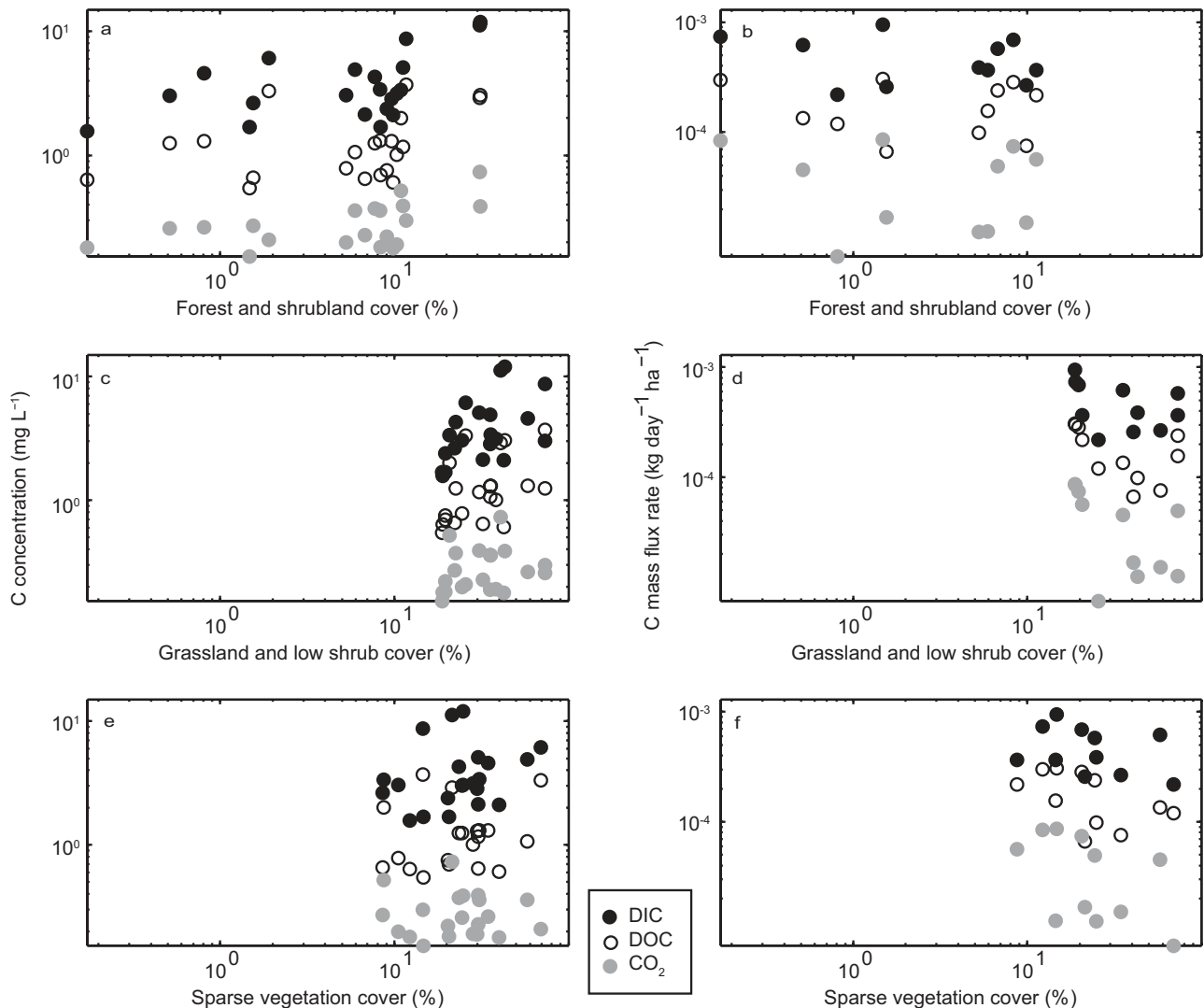
Figure 10: Dissolved C concentrations measured within sub-catchments of the Abiskojokka catchment. Concentration was measured at 32 sites; mass flux rate was estimated for 18 sites. a), c) and e) Dissolved C concentration vs. forest and shrubland cover, grassland and low shrub cover, and sparse vegetation cover, respectively. b), d) and f) Dissolved C mass flux rate vs. forest and shrubland cover, grassland and low shrub cover, and sparse vegetation cover, respectively.

Paper III

The aim of Paper III is to bridge potential knowledge gaps regarding the coupled response of hydrology and C concentrations across scales in sub-arctic catchments. This was addressed by investigating the mechanisms that control the dynamics of dissolved C in stream water using one year of water chemistry sampling in 6 sub-arctic streams together with long-term C export for 2 of those streams (Abiskojokka and Pessijokka).

Over one year of sampling, there were clear (and opposite) temporal patterns for DOC and DIC concentrations for all six streams studied in the Abisko region (Figure 9). While DOC concentration reached its peak during snowmelt with a 6- to 11-fold increase over baseflow values, DIC concentrations were at their lowest. The highest DIC concentrations, together with dissolved silica concentrations, EC and cation concentrations, were reached in late April (late spring) about 4 weeks before snowmelt. The temporal pattern of dissolved C concentration was surprisingly similar across these 6 catchments despite the large difference in catchment area (ranging between 5 and 566km^2) and annual discharge amounts.

The flow-weighted DOC concentration for the six catchments ranged between 1.6 and 4.0 mg L^{-1} and the DIC concentration between 2.2 to 3.1 mg L^{-1}. By comparing the annual flow-weighted concentrations we can see that DOC fluctuated more than DIC, but that the opposite is true when comparing the annual DOC and DIC loads (Figure 8). Furthermore, the estimated total dissolved C (DOC + DIC) loads ranged from 2.1 to 5.1 g C m^{-2} yr^{-1} where DIC on average accounted for 56% of the total dissolved C load and DOC accounted for the remaining 44%. To put this one year of sampling into perspective, the long-term (1987-2008) average proportion of DIC to total dissolved C was 60% for Abiskojokka.

By estimating the seasonal as well as the annual trends of the long-term data for Abiskojokka and Pessijokka (28 years and 6years respectively) we find, for the annual data, an increasing linear trend in DIC concentration of about 9% for both streams. Similar patterns were shown for electrical conductivity (EC) and the weathering products Ca^{2+} and Mg^{2+}. This increasing trend in DIC (also found in Paper II) can be attributed to an increase in water travel times through the landscape, which is in line with the mechanistic modeling approach that was outlined in Lyon et al. (2010) and expanded in Paper II. The increasing DIC concentration was mainly found during the autumn to early winter months (i.e., September, November and December). No clear temporal trend was found in the annual DOC concentrations. Annual DOC and DIC export was tested for relationships with several terrain characteristics from the 6 catchments including elevation, slope, aspect as well as flow pathways and different stream flow characteristics among others the total annual discharge, daily average, daily maximum and daily minimum discharge. However, from the relationships explored between dissolved C and the hydrological measurements and catchment characteristics, the only significant linear relationship was found between DIC mass flux to the specific discharge across the six catchments.

Paper IV

In this paper we study the potential links between lithology, vegetation and hydrology to develop our understanding of C sources and controls of dissolved C along the aquatic conduit. To do this, we analyze DOC, DIC and CO2 concentrations and mass fluxes from a synoptic field campaign, during low summer flow period, in headwater systems and small catchments that drain into Abiskojokka.

The average DOC (1.2 mg L^{-1}) and DIC (3.4 mg L^{-1}) concentrations found across the headwater systems in the synoptic study confirmed that DIC is a major component of dissolved C in small arctic and sub-arctic rivers. In addition to the relative dominance of DIC in this landscape, these low DOC concentrations in relation to DIC are likely associated to the relatively low flow conditions during the sampling campaign. That is, during low flows only the deeper mineral soil loading zones are connected to the streams. This is consistent with the findings in Paper III and the theory put forward in Paper II.

Considering the spatial variability of C export, smaller catchments had relatively high variability in dissolved C concentrations compared to large catchments. This downstream reduction in variability with increasing catchment area was likely due to mixing of water sources. Thus, there was a wide range, from high and low DIC and DOC concentrations in small catchments with low specific discharge (*q*), whereas this range was reduced with increasing catchment area such that the DIC concentrations were relatively low in the larger catchments. The DOC and DIC spatial variability was also compared to catchment biogeophysical characteristics. Considering the role of vegetation (Figure 10), we found that catchments with high grassland and low shrub cover have significantly higher variability in DIC concentrations than catchments with low cover. Catchments with high acidic bedrock cover had significantly higher variability in DOC and DIC concentrations compared to catchments with low acidic bedrock cover. For the hydrological parameters considered, there was

significantly higher variability of all C concentrations for catchments with longer average water travel times compared to catchments with shorter travel time. This spatial variability of waterborne C under stable low flow conditions across the landscape reflects the complex connection between dissolved C and biogeophysical parameters. Thus, the spatial variability of the C export and the vegetation cover and lithology is linked to (1) C content in the unconsolidated sediments and bedrock, (2) carbon's ability to dissolve or become available for transport and (3) hydrological connectivity between the C source and the stream.

Relationships between the organic and inorganic dissolved C with the biogeophysical parameters were explored. These results show that overall, both organic and inorganic C concentrations increased with increasing vegetation cover. Of the studied vegetation classes in this paper, forest and shrubland cover had overall significant positive relations to C concentrations. Additionally, grassland and low shrub cover was significantly positively related to DOC and DIC concentrations. The parameters connected to weathering products of mineralogenic sediments and bedrock EC, alkalinity, silica and base cations (Ca^{2+}, K^+, Na^+ and Mg^{2+}) showed significant positive relationships to all C concentrations. Finally, all C concentrations showed positive relationship with travel times and these were significant for DOC and DIC concentrations. The multiple linear regression analysis indicated that the hydrological factors q and travel time were important for predicting C.

Discussion

Objective A: Potential links between water-borne C and landscape characteristics

Land cover and biophysical characteristics play a major role for the movement of C from the terrestrial to aquatic systems. In these boreal and alpine regions of northern Sweden, the combination of complex vegetation patterns and the topography has great influence on water chemistry. With Objective A we attempt to distinguish the roles that vegetation, geology and hydrology play for the waterborne carbon in the landscape. This landscape driver approach has been used across different scales in the boreal and tundra environments in northern latitudes (Laudon et al., 2011 and references therein). Starting off with the coupling between inorganic carbon and mineralogenic sediments/lithology, Tank et al. (2012a) investigated the relationship between DIC flux and landscape variables. They found that the major control of DIC flux in the six largest Arctic basins is the lithology together with runoff, permafrost extent and glacial coverage (the latter factors are known to be changing with climate). The permafrost coverage, especially continuous permafrost, has been shown to decrease the fluxes of weathering products since permafrost

acts an impermeable layer that limits the interaction between the bedrock and mineralogenic soils with surface waters (e.g. Frey et al., 2007). Therefore, DIC fluxes together with other weathering products, are expected to increase with increasing runoff and decreasing permafrost extent (Walvoord and Striegl, 2007; Frey et al., 2007; Striegl et al., 2007), which is in line with the increasing DIC flux in Abiskojokka over the last 28 years (Paper III). Previous studies have also shown a strong connection between carbonate-rich bedrock to DIC in stream water (Blum et al., 1998; Tank et al 2012b) however we did not find such a clear relationship in Abiskojokka (Paper III-IV). Nonetheless, we find a clear strong relationship between DIC concentration and weathering products such as dissolved silica and base cations Ca^{2+}, K^+, Mg^{2+} and Na^+, which can be said to represent lithology. Considering that DIC load is positively related glacial coverage (Tank et al., 2012a; Striegl et al., 2007), due to enhanced erosion, we will possibly find high-alpine research sites such as the glaciated Tarfala catchment increasingly important during the coming century because of predicted glacial retreat (IPCC, 2014; Radić et al., 2014).

Moving on to vegetation, where the boreal environments consisting of forest, interspersed by shrubs and peatland, is of interest due to the connection of such land covers with C cycling. In addition to being an integral role of the C budget (Cole et al., 2007), dissolved C also plays an important role in the biogeochemistry and aquatic ecosystems in freshwater. Across this thesis, we find a significant positive relationship between forest, shrubs and grasslands with TOC, DOC and DIC (Figure 4 and 10). The relationship of forest and dissolved C is consistent with previous studies in forested catchments (e.g. Laudon et al., 2011) and is expected. What is interesting is that the sparse vegetation cover in catchments at higher altitude, with relatively small amounts of living biomass show similar positive relationship to DOC and DIC concentration as low-lying forested and shrub covered catchments, where biomass is abundant. This result is in line with other local Abisko findings that soils at higher altitudes have greater C storage than at lower altitudes due to the slower decomposition rates (Sjögersten and Wookey, 2009) and that organic material has been buried through cryoturbation (Becher et al., 2013).

Our results from the detailed synoptic investigation of dissolved C during late summer further show that DOC and DIC both can be positively related to several biophysical variables. Although we did find significant positive relationships between vegetation and C export (Figure 4) we had initially expected stronger trends than those observed due to the connection between vegetation and the organic matter in the soil (Hongve, 1999; Sjögersten and Wookey, 2009). We also found that high forest and/or peatland cover could not explain TOC export (Paper I). Altogether this demonstrates that neither vegetation nor geology alone can explain the dissolved organic and inorganic C in stream water in this northern landscape. In order

to understand the mechanisms and dynamics of waterborne C it is thus important to understand both surface and sub-surface hydrological processes, such as flow pathway distributions and hydrological conductivity, in combination with C release rates and loading zone distributions (sources) where mixing and C transport occurs.

Objective B: Spatial variability of dissolved C across Scandinavia

With Objective B, we aim to characterize the spatial variability of dissolved C in sub-Arctic streams. At the small scale, we found high variability of both organic and inorganic C concentrations within the sub-catchments to Abiskojokka. That variability tends to decrease with increasing catchment area, forest cover and in general the complexity in the landscape mosaic (Paper I and Paper IV). This downstream reduction is mainly due to mixing of water sources with the increasing landscape heterogeneity. Previous studies have found large variability of q and DOC concentration in headwater streams across the landscape (Lyon et al., 2012; Temnerud and Bishop, 2005 and references therein). The high variability may partly be explained by the travel time of water as we found that travel time is a significant predictor of DOC concentration during the synoptic campaign. Our findings demonstrate a significant spatial variation in C concentration across Abiskojokka catchment that appears connected to the local geology and travel time of water, indicating that the stream biogeochemistry is dominated by subsurface characteristics under low flow conditions. In addition to the landscape characteristics of land cover and lithology that were in focus of Paper I and Paper IV, terrain and stream flow characteristics were investigated in Paper III. The range of DOC and DIC mass fluxes was surprisingly small despite differences in terrain characteristics, such as catchment area, elevation range and slope, and hydrological characteristics that were investigated across these 6 catchments. To estimate accurate transport of DOC, DIC and CO_2 we therefore stress the need to work across scales and landscape types to better inform future modeling efforts and help complement existing datasets. Further, combining the studies of C sources with the subsurface hydrological processes is essential for understanding waterborne C export in permafrost regions.

Results from both the synoptic campaign in headwaters and streams in Paper IV as well as the one year sampling in the 6 catchments and the long term monitoring in Paper III confirm that DIC is a major component of the dissolved C in sub-Arctic rivers across Sweden (Lyon et al., 2010) and in the pan-arctic drainage basin (Gordeev et al., 1996; Striegl et al., 2007; Tank et al., 2012b). The partitioning between DOC and DIC are thus different in boreal and tundra environments, since DOC here makes up the largest part of dissolved C in boreal systems (Wallin et al., 2010). The high DIC concentrations compared to DOC concentrations in Paper IV are likely related to the relatively low flow conditions during the sampling campaign in August. This time of the year, in late summer, is thus the time of the year when the active layer is likely at its deepest (Åkerman and Johansson, 2008; Sjöberg et al., 2013) and mineral-rich groundwater from the relatively deep flow pathways maintain the streamflow. It is clear that in landscape C budgets the net ecosystem exchange of both organic and inorganic C are in the same range. Therefore it is important, especially in these tundra environments, to include both DOC and DIC in carbon cycling estimates as they have interlinked release and transport processes. The terrestrial landscape is the major DOC source, through decomposition of organic matter, microbial residue or root respiration (Giesler et al., 2006). Degradation and root respiration together contribute to the formation of carbonic acid, which in turn promotes weathering, and thus the release of DIC (Berner and Berner, 1996).

Objective C: Catchment scale mechanisms controlling the dynamics of waterborne C

In objective C we resolve the waterborne C dynamics, mechanisms and their controls on C concentration and loads on catchment-scale. Looking at the temporal scale in Paper I, it is notable that the annual controls on C fluxes differ from the seasonal ones. Winterdahl et al. (2014) found that the intra-annual variability of DOC was best predicted by discharge, temperature and month of the year. In the regional-scale overview of TOC the boreal and tundra environments (Paper I) we find similar results, that climate on the monthly scale, July average maximum temperature and August precipitation, are the most significant in predicting monthly TOC concentration. On the annual scale, forest and open space cover are important, significant predictors of the annual average TOC (Table 3). On the monthly scale we further find that DIC concentration starts increasing during autumn to reach the maximum in early spring, while DOC concentration reaches its maximum during snowmelt season when shallow flow pathways prevail. These dynamics are likely due to the seasonally frozen ground in the winter, promoting deep flow pathways, in combination with the domination of snow as precipitation that leads up to high DOC release during the spring freshet. Thus the dynamics of dissolved C are controlled by the seasonality of these high-latitude climates and this consistency is in line with previous carbon transport work in the region (e.g., Lyon et al, 2010), whereas the relatively static land cover functions as the C source.

As expected from the mechanistic model output in Paper II we find that the temporal variability is smaller for flow-weighted average DIC concentration around its mean compared to DOC concentration and independent of the discharge fluctuations (Papers II-III). For the six studied catchments in the Abisko region we find that the annual DIC mass flux shows high positive correlation to annual discharge (Figure 8), this trend was found regardless of the spatial

variability in weathering rates across the catchment, e.g. between higher altitudes and lower valley bottoms (Allen et al., 2001; Dixon et al., 2008). The high correlation between solute mass flux and discharge has also been found in other studies with this type of diffuse solute input from geogenic sources as well as from legacies from anthropogenic fertilization at the land surface (Godsey et al., 2009; Basu et al., 2010). As indicated in the discussion of objective B DOC concentration and mass flux did not show any connection to estimated hydrological characteristics. Previous studies in permafrost influenced catchments have found decreases in DOC concentration over time (Striegl et al., 2005; Walwoord and Striegl, 2007; McClelland et al., 2007). Striegl et al. (2005) attribute such a decrease in DOC concentration to increases in flow pathway length, water residence time and microbial mineralization of DOC in the soil active layer and groundwater. The microbial mineralization and sorption of DOC in mineral soils plays an important role for understanding the dynamics of dissolved C in stream water. However, whether there is a need for other hydrological measurements or proxies to capture the DOC trends or that DOC concentration is less sensitive to shifts in water flow pathways compared to DIC concentration in the Abisko region remains unclear.

Conclusion

In general, this thesis has identified influences from the sub-arctic landscape on the waterborne C. These controls and mechanisms were found in the landscape and the hydrological characteristics across time and space.

Objective A: Potential links between waterborne C and landscape characteristics

This thesis has shown that TOC, DOC and DIC are significantly and positively related to many of the studied biophysical variables including forest, shrubs, grasslands, peatland, lithology, catchment area and discharge. These results demonstrate that neither land cover nor geology alone can explain the waterborne C in stream water in these boreal and tundra environments. Additionally, on the annual scale land cover may serve as a proxy for organic C in the landscape. Regarding the hydrology, we further showed that it is important to understand both surface and sub-surface hydrological processes such as flow pathway distributions in combination with distribution of C sources and its release rates in order to understand the mechanisms and dynamics of waterborne. Altogether, these results highlight the complexity of waterborne C processes.

Objective B: Spatial variability of dissolved C across Scandinavia

Overall, DIC is an important component of the C flux in these sub-arctic catchments that makes up the major part of dissolved C (DOC + DIC). There is a great variance of aquatic C export across space with respect to different landscape units and characteristics in the sub-arctic alpine catchments. This thesis demonstrates significant spatial variation in C concentration across Abiskojokka catchment. We further found high variance across different catchments implying connection with the local geology and the travel time of water; thus, stream biogeochemistry is dominated by subsurface characteristics during low, stable flow conditions. In all, to estimate accurate export of dissolved C we emphasize the need to work across scales and landscape types to create a baseline of knowledge for future modeling efforts and help complement existing datasets. Ultimately, for permafrost environments it is especially important to combine studies of C sources with the subsurface hydrological processes for understanding the waterborne C export.

Objective C: Catchment scale mechanisms controlling the dynamics of waterborne C

This thesis shows that stream water concentrations of DOC and DIC show opposite temporal patterns. The highest DIC concentrations occur during the late winter when deep water flow pathways prevail and DOC concentration peaks during spring flood. There is further consistency between climate variables (summer temperatures and precipitation) and TOC concentration on the monthly scale. For C transport mechanisms, the high flow-dependence of DIC load is due to a relatively slow average DIC (weathering) release rate from its essentially constant geogenic source, which keeps the DIC concentration essentially constant in time. The relative flow-independence of both the concentration and the load DOC is due to the annual renewal (respiration) of the soil source of DOC.

Final remark: A narrative on future perspectives

In an exercise at a PhD course that I attended we were asked to come up with future research ideas. We were encouraged to think outside the box and there were no limitations of costs and the most amazing ideas and machines were invented. Using the same approach in this part of the thesis I have come to think of one of the central issues when working within research of biogeochemistry and hydrology. Specifically, this is the large effort of field work and taking samples across large areas in short time spans to capture similar conditions (fundamentally

the method employed in Paper IV). Further, I started to think about the complicated procedure to analyze all these (sensitive) water samples for different geochemistry. The current status of biogeochemistry and hydrology would greatly improve if the already existing multi-parameter instruments and sampling machines were developed and combined. As a result the new instrument would not only measure physical parameters and automatically sample both stream and groundwater, but also instantly analyze different types of samples. We would be able to put the instrument in remote areas that are time consuming and difficult to access. The aim to understand the dynamics, controls and mechanisms of dissolved C is pushing the forefront (and pushing the boundaries of current data) in hydrology and biogeochemistry today. To enable higher spatial and temporal resolution monitoring would greatly advance the current state of this research field. To exemplify, these new instruments could be placed along streams in small catchments with fairly homogeneous land cover, especially in transition zones of e.g. vegetation (tundra to forest or forest to peatland) and terrain transition (e.g. steep to flat) to capture different C sources as well as flow pathways.

A next step in this research field is to improve the holistic picture and distinguish the interconnection between DIC and DOC. Here CO_2 plays an important role. The transformation from both DIC and DOC into CO_2 through the carbonate balance and decomposition of organic matter is thus the common denominator for both inorganic and organic C. To capture this holistic picture, my dream research scenario would be to sample both groundwater and stream water along several stream transects together with measurements of gaseous C emissions from the open water surface. The goal would be to capture and distinguish the release of DOC from soils, possible mineralization and degradation of DOC together from the groundwater influx of DIC and CO_2. As stream water chemistry can be viewed as a mixture of upstream C sources this setup could allow us to disentangle the integration of the upstream processes. The high-alpine Tarfala catchment could here serve as a field experiment site for the tundra environments that could enable an end-member focus on DIC and CO_2 to distinguish the effect that the subsurface water movements and water conductivity have on C export. Finally, we would be able to capture the relative importance of biotic and abiotic controls and how these may shift with natural and anthropogenic changes.

Another issue relevant for future research perspectives is the lack of maps covering subsurface characteristics. Imagine the huge step that the hydrology research community working in permafrost environments would take if we could map the flow pathways and the hydraulic conductivity. These subsurface characteristics (as demonstrated throughout this thesis) directly influence solute transport. Thus, to further improve upon exisitng techniques such as electromagnetic induction (EMI)

or accuracy of GRACE satellite data seems like a viable pathway forward to enhance our current knowledge on the spatial variability of subsurface characteristics in the Arctic landscape. Detailed information on hydraulic properties of permafrost and active layer in specific and subsurface properties in general would be a great step forward for hydrology and biogeochemistry science in Arctic and sub-Arctic environments.

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