



# Catchment-scale dissolved carbon concentrations and export estimates across six subarctic streams in northern Sweden

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**Abstract.** Climatic change is currently enhancing permafrost thawing and the flow of water through the landscape in subarctic and arctic catchments, with major consequences for the carbon export to aquatic ecosystems. We studied stream water carbon export in several tundra-dominated catchments in northern Sweden. There were clear seasonal differences in both dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) concentrations. The highest DOC concentrations occurred during the spring freshet while the highest DIC concentrations were always observed during winter baseflow conditions for the six catchments considered in this study. Long-term trends for the period 1982 to 2010 for one of the streams showed that DIC concentrations has increased by 9 % during the 28 yr of measurement while no clear trend was found for DOC. Similar increasing trends were also found for conductivity, Ca and Mg. When trends were discretized into individual months, we found a significant linear increase in DIC concentrations with time for September, November and December. In these subarctic catchments, the annual mass of C exported as DIC was in the same order of magnitude as DOC; the average proportion of DIC to the total dissolved C exported was 61 % for the six streams. Furthermore, there was a direct relationship between total runoff and annual dissolved carbon fluxes for these six catchments. These relationships were more prevalent for annual DIC exports than annual DOC exports in this region. Our results also

highlight that both DOC and DIC can be important in high-latitude ecosystems. This is particularly relevant in environments where thawing permafrost and changes to subsurface ice due to global warming can influence stream water fluxes of C. The large proportion of stream water DIC flux also has implications on regional C budgets and needs to be considered in order to understand climate-induced feedback mechanisms across the landscape.

## 1 Introduction

Tundra soils at northern latitudes contain 30–50 % of the global soil stocks of C (Gorham, 1991; Tarnocai et al., 2009), representing a pool at least twice as large as that of the atmosphere. This pool may potentially be released either as CO<sub>2</sub> or CH<sub>4</sub> (Dutta et al., 2006; Shaver et al., 2006; Lee et al., 2010) or by increased leaching losses of dissolved C (Frey and Smith 2005; Dutta et al., 2006; Frey and McClelland, 2009) due to the polar amplification of climate change and changes in precipitation patterns seen in the past decades. Changes have already manifested in northern latitude ecosystems in the form of thawing permafrost (Osterkamp 2007; Sjöberg et al., 2013), changes in hydrology (Peterson et al., 2002; Déry et al., 2005) and vegetation cover (Sturm et al., 2001), all potentially affecting C dynamics. At a landscape

level, leaching losses of soil C is an important component of the landscape C budget since it can contribute to a large part of the net C loss mainly attributed to lake respiration of terrestrial C (Cole et al., 2007; Karlsson et al., 2009; Lundin et al., 2013). Lake dissolved organic carbon (DOC) concentrations in high-latitude ecosystems have been shown to relate positively to terrestrial net primary production (NPP) (Jansson et al., 2008), and a warmer climate is likely to increase NPP (Kimball et al., 2007) and eventually enhance terrestrial DOC losses. Changes in temperature and hydrology could also liberate large amounts of previously inactive carbon, for instance due to permafrost thawing (Schuur et al., 2009; Dorrepaal et al., 2009; Klaminder et al., 2008) or priming effects related to vegetation changes (Fontaine et al., 2007).

Many tundra soils underlain by continuous or discontinuous permafrost are today experiencing an increase in the active layer thickness (Osterkamp, 2007) due to global warming. This may have profound effects not only on C losses but also on the forms of C lost (Vonk and Gustafsson, 2013). For instance, although there are indications of increased losses of DOC (Frey and Smith, 2005) from northern latitude ecosystems, changes in hydrological flow pathways may also alter the proportion between organic and dissolved inorganic carbon (DIC) export (Lyon et al., 2010a; Jantze et al., 2013). Loss of permafrost areas due to degradation (Zimov et al., 2006; Klaminder et al., 2008) or a deepening of the active layer may increase the importance of subsurface flow pathways (Striegl et al., 2005; Walvoord and Striegl, 2007; Lyon et al., 2009, 2010b). Striegl et al. (2005) found, for instance, that the summer DOC export decreased in the Yukon River when comparing 1978–1980 to 2001–2003. They attributed this decreased export to increased groundwater flow pathways, residence times and mineralization of DOC in the active layer. Walvoord and Striegl (2007) also found an upward trend in the groundwater contribution and thus DIC to stream flow in the Yukon River basin. They proposed that the increase in groundwater contributions were caused predominantly by climate warming and permafrost thawing (e.g., Lyon and Destouni, 2010) that enhances infiltration and supports deeper flow pathways.

These observations may have large consequences not only for landscape C budgets but also for the ecosystem functioning of the recipient aquatic ecosystems. Changes in the terrestrial DOC export to high-latitude lake ecosystems can alter light conditions within lakes and thus affect the relative contribution of the benthic and pelagic primary production as well as overall biomass production (Karlsson et al., 2009; Ask et al., 2009). Shifts in hydrologic flow pathways may also alter the proportion between DOC and DIC (Walvoord and Striegl, 2007). An increased export of DIC, mainly  $\text{HCO}_3^-$  and  $\text{CO}_2$  (g), may result in a negative feedback for atmospheric  $\text{CO}_2$  since  $\text{HCO}_3^-$  can be retained once it reaches the ocean (Bernier and Bernier, 1996). Since about two-thirds of the C found in  $\text{HCO}_3^-$  originates from respired soil  $\text{CO}_2$  globally (Bernier and Bernier, 1996), the retained

$\text{HCO}_3^-$  can be an important sink for terrestrial C and may counteract (to some extent) increased DOC leaching and respiration. An increased groundwater contribution may also play a significant part in  $\text{CO}_2$  losses due to degassing from aquatic ecosystems. Northern aquatic streams and lakes are generally supersaturated with  $\text{CO}_2$  (Kling et al., 1991; Jansson et al., 2003; Giesler et al., 2013) and a large part of this  $\text{CO}_2$  can be related to terrestrial soil respiration (Humborg et al., 2010). A degassing of this  $\text{CO}_2$  may thus contribute to a significant part of the landscape C budget (Lundin et al., 2013).

In northern latitudes, such as the Swedish subarctic region, the distribution of hydrologic flow pathways can be seen as an important factor for catchment-scale C export. Lyon et al. (2010a) demonstrated this connection between flow pathway distribution and the travel time of water through a catchment and carbon export for the subarctic Abiskojoekken catchment in northern Sweden using a detailed distributed modeling approach. While such studies offer promise for estimation of future C loads through simulation, basic knowledge of how hydrologic responses will shift in the future due to climatic changes in arctic and subarctic areas is necessary. Such knowledge, however, is still lacking since research into the hydrologic processes in northern, cold regions is rather limited (Woo et al., 2008). Furthermore, knowledge of the coupled response of hydrology and C concentrations across scales and conditions is sparse for northern environments due to their inherent remoteness. Especially studies including both DOC and DIC are still scarce. To address these potential knowledge gaps, we investigated the annual catchment-scale C dissolved C concentration dynamics and associated implications for annual C loads from six subarctic catchments spanning across different landscape conditions in northern Sweden.

## 2 Methods

### 2.1 Study sites

We selected six streams across a subarctic landscape gradient in northern Sweden ( $68^{\circ}21'36''\text{N}$ ,  $18^{\circ}46'48''\text{E}$ ) located between the towns of Kiruna and Abisko (Fig. 1). The catchments and their streams are all north-facing and draining into the upper reaches of the Torne River system (Table 1). The long-term mean annual temperature in Abisko is about  $-1^{\circ}\text{C}$  (1961–1990; Åkerman and Johansson, 2008) but has been above  $0^{\circ}\text{C}$  in more recent decades (Callaghan et al., 2010). Precipitation in the region is around  $300\text{ mm yr}^{-1}$  in Abisko, increasing eastward to about  $424\text{ mm yr}^{-1}$  at Kiruna, which is located about 16 km northwest of the outlet of stream 1 considered in this study (1961–1990; Åkerman and Johansson, 2008). The vegetation in the region is dominated by deciduous forest at lower altitudes (*Betula pubescens Ehrh. ssp czerepanovii*) and dwarf shrub heath tundra at altitudes above

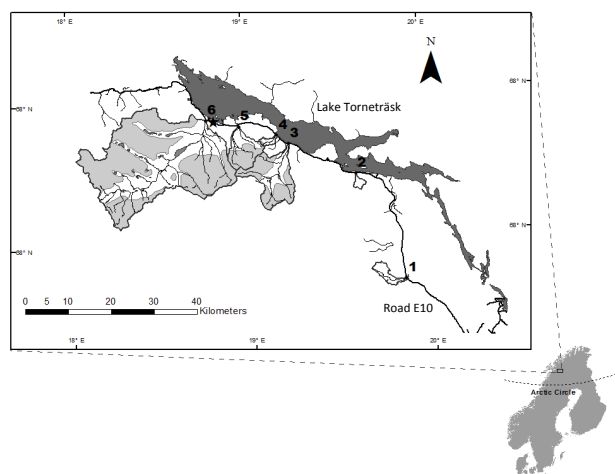
**Table 1.** Terrain characteristics for the six catchments considered in this study. The elevation, slope, and aspect are the mean for the whole catchment. Flow pathway length is defined as the distance from any given point in the catchment to the outlet of the catchment following the flow pathway defined by the steepest (topographic) gradient.

Stream	Common name	Area (km <sup>2</sup> )	Elevation	Elevation	Slope	Aspect	Flow pathway length (m)	Flow pathway length/ gradient (m/m)
			range (m)	(m)	(deg)	(% north)		
1	Homojokka	15.9	475–1013	575	5.6	11	762	16 567
2	–	5.2	362–811	651	9.2	36	999	7178
3	Pessijokka	99.5	360–1734	967	10	16	833	6319
4	–	5.8	366–928	756	9.5	33	754	4537
5	Miellajokka	51.5	384–1731	955	14.5	41	704	4688
6	Abiskoajokka	565.3	374–1793	956	13.1	22	816	4754

approximately 550 m. The soils are mainly upland soils and larger patches of peat soil are only found in the catchment of stream 1 where they account for 7 % of the catchment area. Humus layer thickness is generally less than 10 cm (Björk et al., 2007; Giesler et al., 2012). Podzols are commonly found at lower elevations (< 600 m a.s.l.) and cryoturbated soils at higher elevations (Becher et al., 2013). The permafrost in the Abisko region is considered discontinuous and has a non-random patchy distribution determined by site-specific factors that affect the microclimate (Johansson et al., 2006). Permafrost is found at lower altitudes on north-facing slopes and does exist as low as 350 m a.s.l. (Johansson et al., 2006; Åkerman and Johansson, 2008). Permafrost thickness increases with altitude from one or a few meters to many tens of meters and is common in the tundra zone (Johansson et al., 2006). The bedrock in the region is dominated by mica schist with segments of marble west and north of Abisko, whereas salic igneous rocks and quartzitic and phyllitic hard schists are predominant eastward. Only some westerly tributaries of stream 6 (Abiskoajokka) are potentially influenced by calcareous bedrocks but this is not expressed in stream Ca/Na ratios; the ratios being by far higher in more westerly situated streams which show both high Ca/Na ratios and high DIC concentrations as compared to the six streams in this study (Giesler et al., 2013).

## 2.2 Stream water sampling and hydrological measurements

For each of the six catchments, grab samples of water were taken from mid-April 2008 to the end of April 2009. Samples were taken more intensely during the spring freshet (2 to 3 to times per week) and weekly thereafter. From December to April only monthly samples were taken. These water samples were collected and stored for various future analyses. For stream 5 an autosampler (ISCO 6712, Teledyne ISCO, Lincoln, USA) was installed and daily samples were taken from 22 May to 10 October. Water samples from the ISCO



**Fig. 1.** The six catchments studied in the Abisko region. The grey areas are those above 880 m where mountain permafrost can occur more frequently.

autosampler were collected weekly and analyzed for alkalinity, pH and conductivity. The water samples collected for DOC analyses were filtered (0.45 µm Millex HA filter, Millipore) and thereafter acidified with hydrochloric acid. Water samples collected for silica analyses (only streams 1, 2, 4 and 5) were filtered through a 0.22 µm Whatman Nucleopore filter and acidified with nitric acid. These samples collected for DOC and silica analyses were stored in a cooler until further analyses. The water samples collected for alkalinity measurements were kept untreated in a cooler until analyses. Water samples collected for analysis of CO<sub>2</sub> concentration were collected in 60 mL plastic syringes. Three syringes with 30 mL of water and no air space were collected for each stream at each sampling occasion. The samples were analyzed within 4 h after sampling.

Stream flow hydrographs were developed for each of the six catchments considered in this study. For stream 6 (Abiskoajokka) considered in the study of Lyon et al. (2010a) and Jantze et al. (2013), daily stream flows for the entire hydrologic year 1 May 2008 through 30 April 2009 were measured by and available through the Swedish Meteorological and Hydrological Institute (SMHI; Gage ID 957). For the remaining sites, daily stream flow was observed from 1 May 2008 through 1 October 2008. Flow estimates were based on stage changes in the streams measured using Hobo water level loggers and empirical rating curves. All curves were assumed to be power laws (as is typical for natural streams) based on 4 to 6 observed flows per stream made over the course of the year. Fits for the power law rating curves to the flow observations had  $R^2$  values between 0.86 and 0.99. The daily stream flows for the remainder of the hydrologic year were approximated by scaling observed stream flows from stream 6 to each individual catchment using catchment area ratios. The influence of this approximation is assumed to be minimal since the 7 months where flows were approximated account for only about 10% of the annual flow (considering long-term observations at stream 6) and this low flow winter period is noticeably less dynamic compared to the spring freshet and summer high flows (Lyon et al., 2010a). To explore the potential impact of uncertainty in discharge measurements on subsequent annual load estimates we have assumed a  $\pm 15\%$  measurement error associated with the rating curves. While flow estimation errors can range from less than 5% to more than 40% (Guerrero et al., 2012), we consider this as a conservative error estimate associated with flow measurements given previous work (Lyon et al., 2012). In addition, the uncertainty from the chemical analyses ( $\pm 0.3 \text{ mg L}^{-1}$  for DOC and  $\pm 0.02 \text{ mg L}^{-1}$  for DIC) is added to the total error estimate of the loads.

### 2.3 Chemical analysis and load estimates

DOC was measured on a total organic carbon (TOC) analyzer (Shimadzu TOC-VcPH total organic carbon analyzer) coupled to a nitrogen module (total nitrogen analyzer) by the catalytic combustion technique. Silica was analyzed on an ICP-OES (inductively coupled plasma atomic emission spectrometer), Varian Vista Ax, providing an accuracy and precision better than 4% based on certified standard measurements. Alkalinity was measured using a Mettler Toledo automated titration system using a Metrohm Aquatrode Plus (6.0257/000) pH electrode (Metrohm AG, Switzerland). The samples were titrated to a pH 4.0 with 0.1 M HCl and then back-titrated to 5.6 using 0.1 M NaOH. Alkalinity was calculated from the difference in the amount of NaOH and HCl used and the sample volume. Analysis of  $\text{CO}_2$  concentration was done using a headspace equilibration technique (Cole et al., 1994). A 30 mL gas headspace was created, where after the syringes were shaken vigorously for 1 min they were left standing for 1 min for equilibration of the gas and water

phases. The concentrations of  $\text{CO}_2$  in the headspace were analyzed using an infrared gas analyzer (EGM-4; PP-Systems Inc.). The  $\text{CO}_2$  concentration in the water was calculated according to Åberg et al. (2007). DIC was calculated from alkalinity and  $p\text{CO}_2$  values using the simulations computer program PHREEQCI (Parkhurst and Appelo, 1999). DIC concentrations for the stream 5 ISCO samples were calculated by computing the relationship between alkalinity and DIC ( $\text{DIC (mg L}^{-1}) = 15.03 \times \text{alkalinity (mmol L}^{-1}) + 0.11$ ,  $r^2 = 0.99$ ,  $p < 0.001$ )

Annual DOC and DIC loads were estimated as the product of daily concentrations and stream flows. Since DOC and DIC concentrations were measured at nonuniform time intervals, linear interpolation was used to approximate daily concentrations from the observed concentrations. These daily concentrations were then multiplied by daily average flow amounts to estimate DOC and DIC mass flux coming from each of the catchments monitored in this study. These mass fluxes were summed to estimate annual load of DOC and DIC coming from each catchment. Additionally, the annual average flow weighted concentrations were determined to provide reference.

### 2.4 Hydrological characteristics in relation to DOC and DIC loads

As previous studies have explored the connection between hydrology and chemical fluxes in this region (i.e., Lyon et al., 2010a), we considered several simple hydrological and terrain analyses to capture the potential spatial variability of terrestrial hydrology and explore their relation to the annual DOC and DIC loads from these six catchments. The selection of characteristics considered was guided by previous work in the region (e.g., Lyon et al., 2010a; Karlsson, 2010) and other cold-region research. Stream discharge for each stream was analyzed to determine several basic statistics: the total annual discharge ( $\text{m}^3 \text{ yr}^{-1}$ ), annual runoff (specific discharge), average daily discharge ( $\text{m}^3 \text{ s}^{-1}$ ), and maximum and minimum daily discharge (Table 2). In addition, the ratio of maximum to minimum daily discharge was calculated for each catchment as this ratio has been seen to be a good proxy for the ratio between fast and slow flows within subarctic landscapes (i.e., Ye et al., 2009).

Basic terrain analysis was also considered as a proxy of terrestrial hydrology (Table 2). A raster digital elevation model (DEM) with a pixel resolution of 50 m was used for analysis of topographic characteristics. Flow direction was calculated using a D8 routing algorithm (O'Callaghan and Mark, 1984). Basic topographic features, including catchment area, average elevation, average slope, average aspect and flow pathway lengths, were calculated for each catchment within the System for Automated Geoscientific Analyses (SAGA) geographical information system (GIS). Flow pathway lengths here are the average length of all the estimated hydrologic flow pathways delineated from the DEM

**Table 2.** Stream flow characteristics for the six catchments considered in this study.

Stream	Total annual discharge	Total runoff	Average daily discharge	Max daily discharge	Min daily discharge	Max/Min discharge
	( $10^6 \text{ m}^3 \text{ yr}^{-1}$ )	(mm)	( $\text{m}^3 \text{ s}^{-1}$ )	( $\text{m}^3 \text{ s}^{-1}$ )	( $\text{m}^3 \text{ s}^{-1}$ )	(–)
1	8.3	519	0.3	2.37	0.19	12.43
2	5.0	962	0.2	0.46	0.12	3.69
3	81.2	816	2.5	17.97	2.07	8.69
4	2.8	480	0.5	0.51	0.03	18.63
5	20.3	394	0.6	16.76	0.41	40.39
6	342.6	606	10.6	84.63	5.27	16.07

over an entire catchment. In addition, we considered the ratio of flow pathway lengths to gradients (i.e., land surface slopes) as this potentially serves as a good proxy for hydrologic flux through the landscape and has been seen to be a dominant control of the residence time of water within catchments (i.e., McGuire et al., 2005; Lyon et al., 2010b).

## 2.5 Long-term chemistry in relation to DOC and DIC loads

To put the catchment sampling and observed DOC and DIC loads in perspective, we compare the annual average values measured in this current study with existing long-term sampling. Publicly available long-term monthly stream water chemical data (including alkalinity, cations/anions, and total organic carbon (TOC) are available for stream 6 for the period 1982 to 2011 and for stream 3 for the period 2000–2006 through a systematic monitoring program carried out by the Swedish University of Agricultural Sciences (SLU), Department of Environmental Assessment.

We have used the DOC and DIC concentrations collected in this current study in combination with the publicly available long-term monthly chemistry data to develop long-term monthly concentrations of DOC and DIC. For stream 6, there was a strong 1:1 relation between the long-term monitoring of monthly TOC and the detailed DOC observations ( $r^2 = 0.950$ ), such that DOC can be considered essentially equivalent to TOC for this system. For long-term DIC, there was also a strong linear relationship between the long-term monitoring observations of alkalinity and the detailed DIC observations ( $r^2 = 0.996$ ). Similar relationships were established for stream 3. The DIC relationships were used to translate the available long-term monthly alkalinity values into long-term monthly DIC concentrations while the DOC relationships were used to translate long-term monthly TOC into long-term monthly DOC concentrations. The software MINITAB (version 16, Minitab Inc.) was used to decompose time series data. The trend was found after subtracting the seasonal component from observations (here assumed to be additive and monthly, i.e., there is a subtraction or addition

for each month) and making a regression with the observation date.

## 3 Results

### 3.1 Observed variations in stream water DIC and DOC

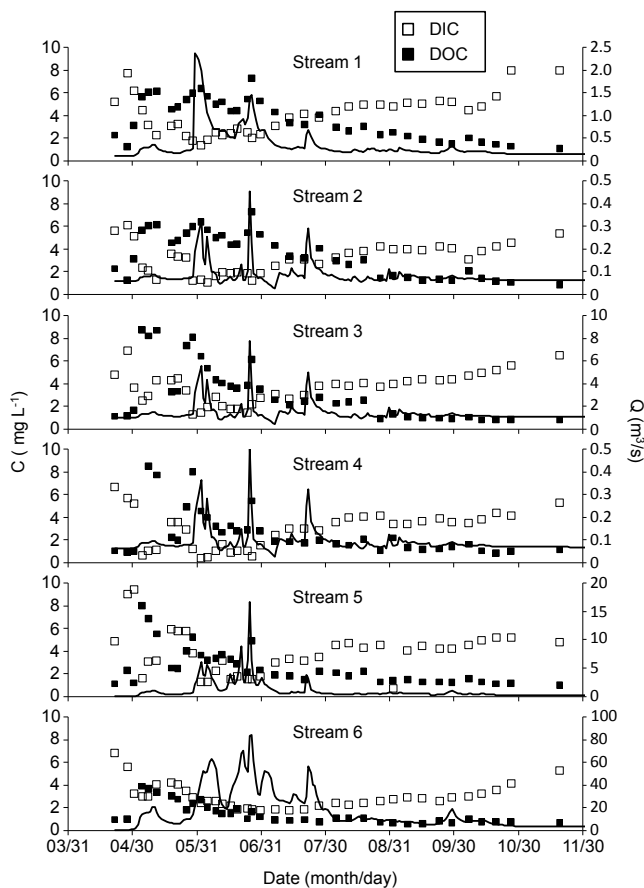
Stream water concentrations of DOC and DIC showed opposite temporal patterns across all streams considered in this study (Fig. 2). The variations amongst the six streams were remarkably similar although the discharge (Table 2) and the catchment sizes were widely dissimilar (ranging from 5.2 km<sup>2</sup> to 565.3 km<sup>2</sup>). DOC concentrations were generally highest at the first snowmelt peak of the spring freshet although peaks in DOC concentration also were noted during later high flow events. The increase in DOC concentration from baseflow to the first snowmelt peak was between 6- to 11-fold with the highest increase occurring in streams 3 and 4. DIC concentrations, on the other hand, were highest during the winter with the highest concentrations observed in late spring (Fig. 2). During the spring, freshet concentrations decreased and the lowest concentrations were generally observed during peak flow conditions. DIC concentrations were strongly positively related to Si (dissolved silica) concentrations (Fig. 3).

### 3.2 Comparing to long-term trends in DOC and DIC

For the available long-term data, we estimated trends with linear regression and estimated a seasonal component by assuming an additive value for each month (i.e. 12 values per year, to catch the seasonality of the concentrations). This resulted in models where fits were compared to observed data. The quality of these fits is reported as MAPE (mean absolute percentage error), MAD (mean absolute deviation) and MSD (mean square deviation). We found an increasing linear trend for DIC concentration in stream 6 (Fig. 4, Table 3). The increase corresponded to about 9 % for the 28 yr of measurements. The trend was mainly related to an increase during the autumn/early winter months. When looking at individual

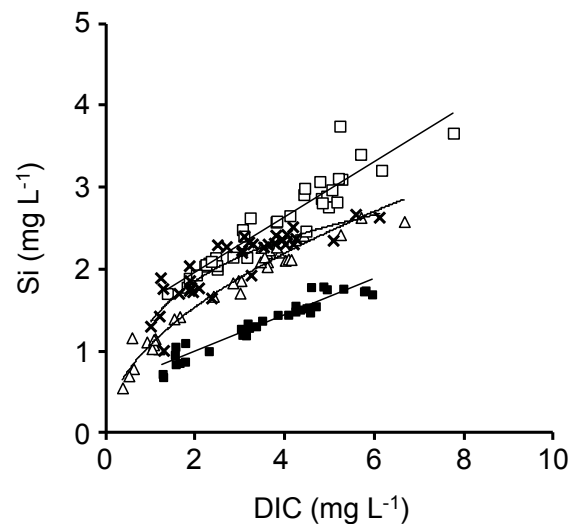
**Table 3.** Time series analyses (fitted trend and seasonal component) for stream 3 (2000–2006) and 6 (1982–2010). Here, MAPE is mean absolute percentage error, MAD is mean absolute deviation, MSD is mean square deviation, and Cond is conductivity. Yearly trends are given as  $\text{mg CL}^{-1}$  (DOC and DIC) or  $\text{mS m}^{-2}$  (conductivity).

	Stream 6			Stream 3		
	DOC	DIC	Cond	DOC	DIC	Cond
Yearly trend	-0.005	0.012	0.039	0.059	0.070	0.105
MAPE (%)	41.9	11.2	9.0	25.9	9.3	8.1
MAD	0.53	0.03	0.42	0.39	0.03	0.40
MSD	0.617	0.001	0.341	0.316	0.002	0.370



**Fig. 2.** Annual variation (2008) in stream water DOC and DIC concentrations in the six streams studied. The black line shows stream water flow.

months we found significant linear increases with time in DIC for September ( $r^2 = 0.26$ ,  $p = 0.006$ ), November ( $r^2 = 0.14$ ,  $p = 0.048$ ) and December ( $r^2 = 0.23$ ,  $p = 0.01$ ). Also, stream 3 showed an increase in DIC concentration of around 9% for the six years of measurement (Fig. 4). Conductivity showed the same patterns as DIC (Fig. 4) and weathering products such as Ca and Mg were always strongly related to DIC and showed similar increasing trends for both streams (data not shown). No clear trends in DOC concentration were

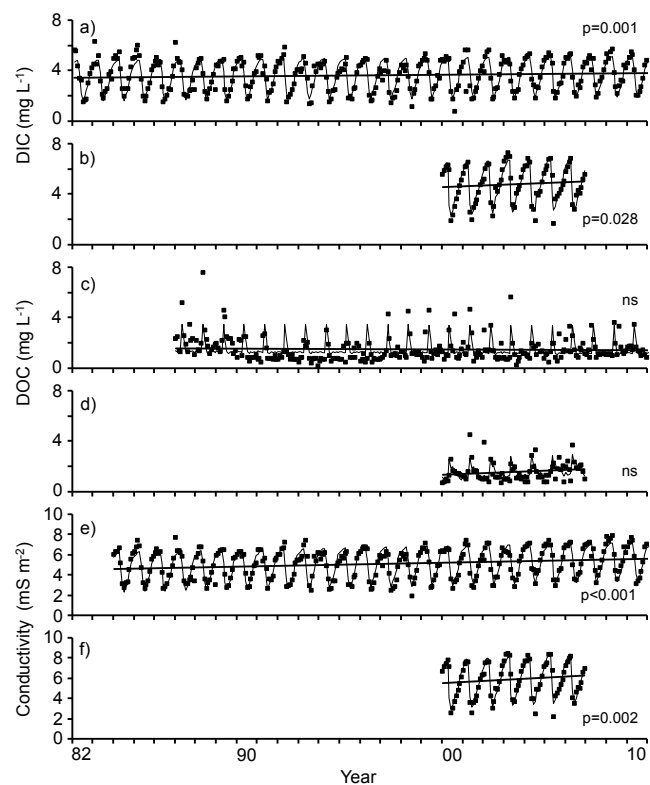


**Fig. 3.** The relationship between stream water concentrations of dissolved silica (Si) and DIC in stream 1 (open squares;  $r^2 = 0.90$ ,  $p < 0.001$ , linear), stream 2 (crosses;  $r^2 = 0.77$ ,  $p < 0.001$ , logarithmical), stream 4 (open triangles;  $r^2 = 0.95$ ,  $p < 0.001$ , power), and stream 5 (filled squares;  $r^2 = 0.89$ ,  $p > 0.001$ , linear). The relationships were best explained by either linear or curve-linear functions.

found for the streams and the fitted values in the time series analyses were poorly explained (Table 3).

### 3.3 C fluxes across the six streams

Estimates of C (DIC+DOC) loads for the six catchments ranged from 2.1 to 5.1  $\text{g C m}^{-2} \text{yr}^{-1}$  and DIC accounted for an average of 56% of the estimated C load. Flow-weighted annual DOC concentrations showed more variability across the six catchments than the flow-weighted annual DIC concentrations (Table 4). The opposite is true, however, when considering the annual DOC loads compared to the annual DIC loads. DIC loads ranged from 1.02 to 3.26  $\text{g C m}^{-2} \text{yr}^{-1}$  while DOC loads ranged from 0.82 to 2.29  $\text{g C m}^{-2} \text{yr}^{-1}$  across the six catchments (Table 3). Concentrations from daily measurements of DIC for stream 5 were strongly linearly correlated to DIC values from the



**Fig. 4.** Long-term trends for stream 6 (Abiskojojokka) and stream 3 (Pessijokka) in DIC (a) and (b), DOC (c) and (d), and conductivity (e) and (f). The abbreviation ns denotes not significant trend.

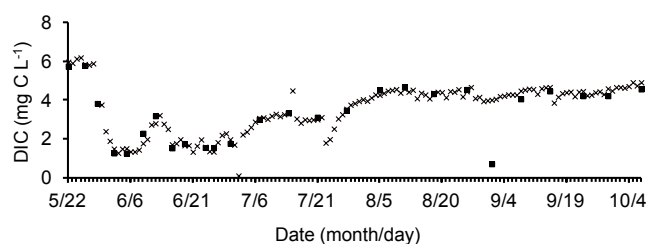
grab sampling ( $r = 0.89$ ,  $p < 0.001$ ;  $N = 26$ ). Flux estimates from the about three months of daily sampling compared well with the estimates based on the grab sampling (Fig. 5); 0.77 and 0.78 ( $\text{g C m}^{-2}$ ) respectively. The deviation in the flux estimate between the two sampling regimes was thus less than 2 %.

We also explored linear relationships between several predicting variables (landscape characteristics in Table 1 and stream discharge characteristics in Table 2) and the annual DOC and DIC exports for the six monitored catchments in this study. Of the predicting variables considered, only DIC showed a linear relationship with the total runoff (specific discharge) across these catchments ( $r^2 = 0.85$ ,  $p = 0.008$ ). There was also a tendency for a linear relationship between DIC and the flow path length and max/min discharge ( $r^2 = 0.64$ ,  $p = 0.06$  for both), although not significant, and for DOC and total runoff ( $r^2 = 0.62$ ,  $p = 0.06$ ).

## 4 Discussion and concluding remarks

### 4.1 Seasonal patterns in stream water DOC and DIC

The observed seasonal variation in stream water DIC and DOC concentrations coincides with a source shift from a sub-



**Fig. 5.** Comparison of daily DIC concentrations in stream 5 (crosses) and grab samples (filled squares) from the same stream.

**Table 4.** Flow-weighted annual DOC and DIC concentrations and mass fluxes for the six catchments considered in this study. Uncertainties are based on an assumed  $\pm 15\%$  measurement error associated with the rating curve and the uncertainty from the chemical analyses ( $\pm 0.3 \text{ mg L}^{-1}$  for DOC and  $\pm 0.02 \text{ mg L}^{-1}$  for DIC).

Stream	Flow-weighted concentrations		Mass fluxes	
	DOC	DIC	DOC	DIC
	(mg L <sup>-1</sup> )		(g C m <sup>-2</sup> yr <sup>-1</sup> )	
1	4.7 ± 0.9	3.1 ± 0.7	1.8 ± 0.4	2.1 ± 0.3
2	3.4 ± 0.7	2.9 ± 0.6	2.3 ± 0.7	3.3 ± 0.5
3	4.0 ± 0.8	3.0 ± 0.7	2.1 ± 0.6	3.2 ± 0.5
4	3.2 ± 0.6	2.4 ± 0.5	1.0 ± 0.3	1.2 ± 0.2
5	3.2 ± 0.8	2.2 ± 0.5	1.1 ± 0.3	1.0 ± 0.2
6	1.6 ± 0.5	2.4 ± 0.4	0.8 ± 0.3	1.4 ± 0.2

surface dominated flow during baseflow conditions, i.e., autumn/winter, to surface dominated flow pathways during the spring freshet. This is in accordance with hydrograph separations between shallow and deep groundwater from stream 6 (Lyon et al., 2010a) and with the observations of increased silica concentrations during baseflow conditions (Fig. 3). Silica is likely to reflect weathering inputs into the streams and should increase with a more groundwater dominated flow, as has been observed in many arctic streams and rivers (Frey et al., 2007 and references therein). The reverse is true for stream water DOC concentration, which increases during the snowmelt when shallower groundwater flow pathways dominate (Lyon et al., 2010a). Such shifts are commonly observed in watersheds with seasonal snowpacks in arctic (Carey 2003), alpine (Hood et al., 2005) and boreal streams (Laudon et al., 2004) and our data are in line with these results.

The extent and distribution of permafrost is typically seen to influence stream water DOC fluxes. Carey (2003) found that the spring freshet contributes to more than 50 % of the DOC export as well as to the spring snowmelt contribution of DOC (69 %) in the high-permafrost area of the Yukon Territory, Canada. Also, in Alaskan permafrost areas, the spring freshet was found to account for 51 % of the annual

DOC export in a high-permafrost watershed, while it was otherwise less than 20% of the annual DOC flux in low-permafrost watersheds (Petrone et al., 2006; MacLean et al., 1999). The increase in stream water DOC concentrations from baseflow to the snowmelt peak further differed between high-permafrost and low-permafrost catchments, with the increase being 12-fold in the former compared to 6-fold in the latter (Petrone et al., 2006). Carey (2003) suggested that permafrost dominated hillslopes potentially have a larger DOC reservoir and that permafrost dominated hillslopes are more effective at delivering DOC to the stream due to increased lateral flow. We found similar differences in the DOC increase going from baseflow conditions to the spring snowmelt as Petrone et al. (2006) and our data resemble mostly the high-permafrost area behavior found in these previous Canadian and Alaskan studies.

There is, however, currently no detailed information available on the areal extent of the permafrost in our studied catchments. Mountain permafrost determined mainly by air temperatures is found approximately above 880 m a.s.l. (Jeckel 1988; Johansson et al., 2006), but can probably occur at lower elevations on north-facing slopes that are less exposed to solar radiation (Johansson et al., 2006). The extent of the permafrost is also dependent on the snow depth, which is a critical factor for permafrost formation (King, 1986; Sepälä, 1986). This may be important in areas with less winter precipitation such as the Miellajokka (stream 5) catchment that receives less snow than areas more westward or eastward (Klaminder et al., 2008; Åkerman and Johansson, 2008). Clearly, there is need for detailed subsurface and geophysical investigations to better control detailed estimates of permafrost distributions.

#### 4.2 The role of shifting flow pathways for DIC and DOC concentrations

The observed temperature increase in the Abisko region during the last decades (e.g., Callaghan et al., 2010) could affect stream water C concentrations similar to previous observations from the Yukon River (Walvoord and Striegl, 2007) and other arctic streams (Frey and McClelland, 2009). Our long-term data does indeed suggest that there has been an increase in stream water DIC concentrations and that these changes are mainly related to the autumn period. There are a number of arguments favoring that the observed changes are related to changes in water flow pathways such as those seen in time from the long-term monitoring. For instance, recession flow analysis based on long-term flow records from stream 6 suggests that there has been an increase in the effective aquifer depth in the catchment that could be related to permafrost thaw (Lyon et al., 2009), while analyses of the annual discharges matching the period considered here show decreases in annual total discharge (Jantze et al., 2013). Furthermore, when considering the long-term annual DOC and DIC loads, Jantze et al. (2013) reported no significant trends

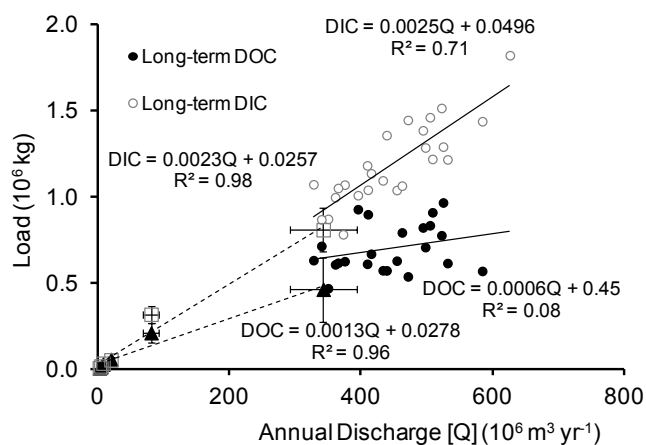
in the total annual mass flux of either DIC or DOC over the periods considered but a significant decreasing trend in total annual discharge for stream 6. Together, this decreasing trend in discharge and increasing trend in DIC concentration can be shown to be consistent with increasing water travel times through the landscape using a mechanistic modeling approach like that outlined in Lyon et al. (2010a) and Jantze et al. (2013). There are no direct observations of active layer changes from upland soils in the area but it seems likely that these also are affected similar to the observations from the permafrost mires (e.g., Callaghan et al., 2010). The increase in DIC especially during the autumn months is in line with this assumption since the active layer is deepest during this time period (Åkerman and Johansson, 2008). We interpret this change in DIC to be a result of an increased contribution of deeper groundwater, like those indicated by the detailed generic simulations of Frampton et al. (2011) of permafrost thawing effects on flow and flow pathways under long-term climate change, rather than to changes in external inputs (e.g., Sjöberg et al., 2013).

There is not a concomitant decrease in DOC as has been observed from several studies from other permafrost influenced watersheds (Striegl et al. 2005; Walvoord and Striegl, 2007; McClelland et al., 2007). Such a decrease has been attributed to increase in hydrological residence time and microbial breakdown of DOC that would otherwise be released to streams (Striegl et al., 2005). This current study found a lack of connection between traditional residence time proxies (like flow pathway length to gradient ratio) and DOC loads across sites, but this is likely attributed to the lack of subsurface information (hydrological conductivity) in these proxies. Therefore, this proxy fails to capture the speed at which water effectively moves through the terrestrial system. The lack of clear connection between DOC and discharge (i.e., Fig. 6, considering stream 6's long-term annual data relative to short-term data across all sites) could also be attributed to increased interactions between DOC and mineral surfaces due to sorption. The latter process is probably important and contributes to the build-up of mineral soil C with higher precipitation, i.e., increased soil infiltration in tundra soils (Klaminder et al., 2008). This suggests that DOC concentrations may be less sensitive to shifts in flow pathways as opposed to other factors. A possible explanation for this insensitivity might be that the relative difference in DOC release is minor in the mineral soil in contrast to the surface soils that contribute to the DOC release during the spring freshet.

#### 4.3 The relationship between DOC and DIC concentrations and fluxes

Overall, annual measurements for the six streams suggest that DIC is an important component of the C flux in these subarctic catchments. The DIC accounted on average for 56% of the stream water C export and the proportion





**Fig. 6.** Annual DOC and DIC loads in relation to total annual stream flow for stream 6 (Abiskojojokka) based on available long-term data (here, filled circles are DOC loads and open circles are DIC loads) 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 stream 6 while dashed trend lines are fit to the annual data for the six catchments.

between DIC and DOC agrees well with long-term average for Abiskojojokka (1987–2008), which is 60 %. This separates these subarctic catchment–stream systems from Scandinavian boreal forest streams, for which previous reports have shown that DIC accounts for only about 19 % of the total C export (Wallin et al., 2010). A similar proportion of DIC to the total C flux is also found in the large Siberian Rivers, especially in the east (data in Raymond et al., 2007 and Tank et al. 2012) and Alaskan rivers (Striegl et al., 2007). A comparison of literature values from permafrost-affected catchments in high-latitude areas (Table 5) also suggest that the concentration values we report for DOC are roughly within the same range as other permafrost-affected streams/rivers in high-latitude areas whereas our DIC concentrations seem to be in the lower range of the reported values (Table 5). It should be stressed that the catchments we studied are dominated by upland soils and that only stream 1 contains areas with wetland soils, which will affect stream DOC concentrations (Frey and Smith, 2005). A regional survey undertaken in mid-September 2008 of streams in the Abisko region (Giesler et al., 2013) also shows that the DIC concentrations we find in our six streams for the same time period are about 7 times lower than the highest value reported in the regional survey (average  $4 \text{ mg L}^{-1}$  versus  $32 \text{ mg L}^{-1}$ ). This is also in accordance with the geology of the stream catchments where the influence of calcareous bedrocks is probably low. Our values are also comparable to the average stream value ( $6 \text{ mg C as HCO}_3^-$ ) from permafrost-affected areas in west Siberia (Frey et al., 2007).

Although the uncertainty in the absolute flux estimates is large, mainly due to uncertainties in the discharge esti-

mates, the relative proportion between DOC and DIC is most likely rather robust since it is less dependent on changes in the absolute discharge. There are few flux estimates available on DIC from permafrost-affected areas and are mainly for larger rivers. Striegl et al., 2007 report values ranging from  $2.5$  to  $8.7 \text{ g C m}^{-2} \text{ yr}^{-1}$  from the Yukon, Tanana and Porcupine rivers in Alaska. Flux estimates of DIC from the Scandinavian boreal forest about  $700 \text{ km}$  south of Abisko in the Krycklan catchment report loads ranging from  $-0.7$  to  $1.4 \text{ g C m}^{-2} \text{ yr}^{-1}$ ; the negative value indicating a net loss between upstream and downstream measured sites. The DIC export in several sub-catchments of a wetland-influenced catchment in the Abisko area, the Stordalen mire complex, ranged from  $0.6$  to  $3.0 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Olefelt et al., 2013) and the range in yearly DIC export from the Abiskojojokken stream (Fig. 6) ranged between  $1.5$  and  $3.2 \text{ g C m}^{-2} \text{ yr}^{-1}$ . The flux estimates we report for DIC are thus reasonably well in agreement with both long-term variations and wetland-influenced catchments in our study area but are less than loads reported from larger rivers in Alaska and are higher than loads reported for the Scandinavian boreal forest. Our DOC fluxes are roughly in the same range as those reported from streams in permafrost-affected areas; Carey (2003) reported a flux of  $1.6 \text{ g C m}^{-2} \text{ yr}^{-1}$  and Petrone et al. (2006) reported values between  $1.1$  and  $2.3 \text{ g C m}^{-2} \text{ yr}^{-1}$  for three catchments. Our export values are, however, considerably lower than many boreal forest streams (see compilation of DOC export data in Olefeldt et al., 2013).

Annual DIC loads for the six catchments considered in this study provide a continuation of the linear relationship between DIC and discharge (Fig. 6) such that they are more-or-less consistent with the mass fluxes estimated from the long-term data at stream 6. Counter to this, while there is some consistency, it is not possible to relate the DOC export from the six catchments to the long-term DOC relationship between annual load and annual stream discharge. There is a clear potential for nonlinear behavior and considerable scatter that does not allow for directly relating annual discharge to DOC export based on the data considered here. The relationships in Fig. 6 are consistent with the view of inorganic carbon loadings being derived across the entire catchment (Basu et al., 2010; Jantze et al., 2013) while additional factors (e.g., biological) and flow pathway variability through the catchment potentially influence the annual export of DOC.

#### 4.4 Implications for the regional C export

It is clear that DIC export is at least in the same order as DOC in the stream water C export in the studied streams. Notably, our values are in the lower range as compared to many streams and rivers in comparable permafrost-affected landscapes (Table 5). In landscape C budgets, both organic and inorganic C stream fluxes should be considered since both their origin and their effect on the net ecosystem exchange of C is largely the same. With the assumption that

**Table 5.** Literature data on concentration ranges in permafrost-affected northern streams. The lower DOC value and the higher DIC value reflect base flow conditions whereas the other values are for spring peak flow conditions.

Source	Location	Extent of permafrost	Range	
			DOC (mg L <sup>-1</sup> )	DIC
Striegl et al. (2007)	Porcupine River, Alaska	discontinuous	2–12	13–51
	Tanana River, Alaska	discontinuous	1–5	21–41
	Yukon River (Eagle), Alaska	discontinuous	2–8	18–31
	Yukon River (Stevens Villaga), Alaska	discontinuous	2–11	18–35
	Yukon River (Pilot Station), Alaska	discontinuous	3–11	18–49
Cai et al. (2007)	Chena River, Alaska	discontinuous	3–14	11–27
Carey (2003)	Granger Basin, Yukon territory	discontinuous	2–4	
Tye and Heaton (2007)	Stream 1, Ny-Ålesund, Svalbard	continuous	3–5	6–38
	Stream 2, Ny-Ålesund, Svalbard	continuous		12–29
	Stream 3, Ny-Ålesund, Svalbard	continuous	2–3	14–25
Petronne et al. (2006)	High, Caribou–Poker Creeks Watershed, Alaska	discontinuous	3–21	
	Medium, Caribou–Poker Creeks Watershed, Alaska	discontinuous	3–11	
	Low, Caribou–Poker Creeks Watershed, Alaska	discontinuous	3–8	
This study	Stream 3, Abisko	discontinuous	1–8	1–7
	Stream 4, Abisko	discontinuous	1–11	1–6
	Stream 5, Abisko	discontinuous	1–8	2–10
	Stream 6, Abisko	discontinuous	1–4	2–7

most of the DOC we see in streams is of terrestrial origin, the DOC is likely a result of degradation of plant or microbial residues or direct inputs via root exudation (Giesler et al., 2006). Both degradation of soil organic matter and root respiration (Berner and Berner, 1996) will contribute to the formation of carbonic acid (H<sub>2</sub>CO<sub>3</sub>), and promote weathering and thus formation of the DIC (Berner and Berner, 1996; Humborg et al., 2010). Hence, at least part of the DIC can probably be attributed to terrestrial C, which within rather recent times has been fixed via plant photosynthesis, with the remaining part of DIC originating from carbonate weathering. The overall effect of DIC on landscape C budgets is, however, still unclear and further studies are needed to elucidate its role for CO<sub>2</sub> emissions from the aquatic ecosystems as well as the partitioning between the contribution from carbonate versus silicate weathering, the latter contributing to DIC formed from respiratory CO<sub>2</sub> compared to the former where carbonate also contributes to the DIC formation.

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