

**Transport of  
dissolved carbon in a  
discontinuous  
permafrost region**

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# Subsurface release and transport of dissolved carbon in a discontinuous permafrost region

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## Abstract

Warming and thawing of permafrost and deepening of the active layer related to climate change have been reported in arctic and subarctic environments. These changes alter subsurface hydrological flow pathways and advection rates through the landscape, and thereby also affect the quantity and timing of hydrological transport of dissolved carbon. This study investigates hydrological carbon transport through the subsurface to streams and how it is affected by the distribution of subsurface hydrological pathways and travel times through the landscape. We develop a consistent mechanistic, pathway- and travel time-based modeling approach for release and transport of dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC). The model implications are tested against observations in the subarctic Abiskoajokken catchment (68°21' N, 18°49' E) as a field case example of a discontinuous permafrost region. The results show: (a) Essentially flow-independent concentration and load of the annually renewed and depleted DOC, due to the small characteristic respiration-dissolution time scale relative to the average travel time of water through the subsurface to the stream. (b) High flow-dependence of DIC load, due to its large characteristic weathering-dissolution time relative to the average subsurface water travel time to the stream, which also keeps the DIC concentration essentially flow-independent and less fluctuating in time than the DIC load.

## 1 Introduction

The hydrological transport of carbon has recently begun to be included in global carbon budgets (Cole et al., 2007) and ecosystem assessments acknowledge the role that the lateral movement of water through aquatic systems plays for dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) cycling (Tranvik et al., 2009). Subsurface transport of carbon is of particular importance in arctic and sub-arctic environments, as these systems undergo particularly large climate-driven changes, with potentially

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large positive climate feedbacks. Frozen taiga and tundra soils are believed to contain more than double the amount of carbon compared to the atmosphere (Tarnocai et al., 2009). These terrestrial high-latitude environments have traditionally been viewed as long-term sinks of carbon. Due to ongoing climatic warming, however, these systems can evolve such that they no longer serve as clear net carbon sinks and can potentially become large net carbon sources (Kling et al., 1991; ACIA, 2005; Fung et al., 2005; Schuur et al., 2008). In this important context, our knowledge is limited on how the source releases carbon in such high-latitude terrestrial systems and feeds DOC and DIC from the ground and into their streams and other aquatic ecosystems.

Arctic and sub-arctic landscapes present unique conditions for hydrological flow and solute transport. Subsurface flow pathways of water in many arctic environments, for example, are determined by permafrost characteristics and ground ice distributions in the landscape (Carey and Woo, 2001; Frampton et al., 2011; Sjöberg et al., 2012). As such, the terrestrial freshwater cycle in the Arctic is intimately coupled with the existence of permafrost (White et al., 2007; Woo et al., 2008), which largely determines the temporal dynamics and spatial pathways of water flow through the landscape (Kane et al., 1981; Bosson et al., 2012). The location and distribution of the flow pathways in turn influence the dissolved carbon release, transport and cycling through the landscape (MacLean et al., 1999; Petrone et al., 2006; McNamara et al., 2008). For example, frozen ground conditions in winter seasons may imply a deeper average distribution of flow pathways with a relative increase in mineral weathering (MacLean et al., 1999) and its byproducts such as DIC (Lyon et al., 2010). During the spring freshet and summer seasons, there may instead be a prevalence of near-surface flow through more organic rich soil horizons (Carey, 2003; Carey and Quinton, 2004) leading to increased concentrations of DOC in rivers and aquatic systems.

While these phenomena can be described in such a straightforward manner, 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 across arctic and sub-arctic landscapes. A Lagrangian

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approach based on the hydrological pathways and travel times of water and solute through the landscape and its underlying geological formations offers several advantages for such understanding and quantification. This approach has been applied in various settings and conditions; to transport in streams and drainage networks (Rinaldo and Marani, 1987; Rinaldo et al., 1991; Gupta and Cvetkovic, 2000, 2002), soils and aquifers (Bresler and Dagan, 1981; Shapiro and Cvetkovic, 1988; Rainwater et al., 1987; Destouni and Cvetkovic, 1991; Cvetkovic et al., 1998; Cvetkovic and Dagan, 1994; Ginn, 1999, 2001; Fiori et al., 2002), fractured rocks (Cvetkovic, 1991; Painter et al., 1998, 2002; Frampton and Cvetkovic, 2011), waste deposits of different types (Eriksson and Destouni, 1997; Eriksson et al., 1997; Rosqvist and Destouni, 2000), and several such subsystems linked in whole catchments (Destouni et al., 2010; Cvetkovic et al., 2012). Owing to its flexibility, such a Lagrangian framework can provide a hydrologically-consistent manner by which to explore the export of dissolved carbon from terrestrial arctic landscapes. By interpretation of measured DOC and DIC transport to the stream in the Swedish sub-arctic Abiskoajokken catchment (Lyon et al., 2010) used the same approach to quantify average rates ( $r$ , in terms of mass release per unit time and bulk soil volume [ $\text{M T}^{-1} \text{L}^{-3}$ ]) for organic and inorganic carbon dissolution into soil water.

The aim of the current study is to further resolve, distinguish and quantify the mechanisms and controls that determine the bulk average release rate  $r$  for DOC and DIC for current conditions. This study is setup as follows. First, we describe the Abiskoajokken catchment, which is here used as a specific permafrost region example, for which observed export patterns and hydrogeochemical signals of DOC and DIC are interpreted to infer their source inputs and transport through the subsurface hydrological pathways in this landscape with seasonal frost and permafrost. For this interpretation, we further link different previous modeling components to develop a pathway- and travel time-based modeling framework for diffuse source release and solute transport of relevance for dissolved carbon (Eriksson and Destouni, 1997; Lyon et al., 2010). The interpretation is based on comparison of the implications of the developed mechanistic modeling

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framework for DOC and DIC release and transport with available long-term hydrological observations and stream water chemistry data.

## 2 The Abiskoajokken catchment: site description and data

The Abiskoajokken catchment (68°21'36" N, 18°46'48" E) is situated, in the Abisko region, in a glacially eroded sub-arctic environment 200 km north of the Arctic Circle (Fig. 1). It has an area of 566 km<sup>2</sup> and the catchment ranges in elevation from 350 m to 1600 m a.s.l. (above sea level) with about 73 % of the catchment area situated above 800 m which is an elevation indicating probable presence of permafrost in the Abisko region according to Ridfelt et al. (2008). Abisko has a mean annual precipitation of 304 mm yr<sup>-1</sup> for 1961–1990 (Swedish Meteorological and Hydrological Institute – SMHI) although there is a strong gradient in precipitation with elevation in the catchment. Mean annual air temperature for the same time period was –0.8 °C (SMHI, station no. 18880, Abisko), and has since 2000 significantly exceeded 0 °C, which is a threshold for many ecological and cryospheric processes (Callaghan et al., 2010). Permafrost cover in the Abisko region is discontinuous (Johansson et al., 2006) and decreases in coverage along a gradient from west to east. The permafrost distribution is patchy and shows a strong relationship to elevation (Ridfelt et al., 2008). The presence and quality of permafrost varies on a small spatial scale under the influence of topography, snow cover amount and duration as well as local hydrological conditions (Kneisel, 2010).

The bedrock is dominated by mica schists and calcite marble (Kulling et al., 1960). While there is no detailed soil characterization available for the Abiskoajokken catchment, the median depth of unconsolidated sediments has been estimated to range between 5 m for deciduous forest to about 7 m for herbaceous vegetation in this region (Smedberg et al., 2009), and regional studies of nearby Kärkevagge show typically coarse-loamy soils with 5 % clay with slightly acidic soil pH (Darmody et al., 2000). The subalpine birch forest in the Abisko valley generally reaches 650 m a.s.l., and up to

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700 m a.s.l. on south-facing slopes. The ground is generally dry and the undergrowth is species poor. Above the birch forest the vegetation shifts to willow and dwarf birch dominated heaths. Above the heaths the vegetation transitions into dry shrubland and meadows with low herbs. Middle alpine grass heaths dominate at elevations higher than 1000 m a.s.l. (Rafstedt et al., 1985).

Publicly available long-term hydrological observations and stream water chemistry sampling make it possible to utilize the Abiskoajokken for this study. Monthly stream water chemical data including alkalinity ( $\text{mekv L}^{-1}$ ) and total organic carbon (TOC,  $\text{mg L}^{-1}$ ) are available for Abiskoajokken for the period 1982 to 2011 through a systematic monitoring program carried out by the Swedish University of Agricultural Sciences (SLU), Department of Environmental Assessment. We have then used the detailed stream water chemistry observations of alkalinity ( $\text{mmol L}^{-1}$ ), and DOC ( $\text{mg L}^{-1}$ ) and DIC ( $\text{mg L}^{-1}$ ) concentrations presented by Lyon et al. (2010) for the year 2008, in combination with the publicly available long-term monthly chemistry data, to develop long-term monthly concentrations of DOC and DIC. For Abiskoajokken there was a 1 : 1 relation between the long-term monitoring of monthly TOC and the detailed DOC observations of Lyon et al. (2010) ( $r^2 = 0.95$ ; see Fig. A1, Appendix A), such that DOC can be considered essentially equivalent to TOC for this system. For long-term DIC, there was further a strong linear relationship between the long-term monitoring observations of alkalinity and the detailed DIC observations of Lyon et al. (2010) ( $r^2 = 0.99$ ; see Fig. A2, Appendix A). This DIC relationship was used to translate the available long-term monthly alkalinity values into long-term monthly DIC concentrations.

Daily streamflow for Abiskoajokken is available through the Swedish Meteorological and Hydrological Institute (SMHI; Gage ID 957) from 1986 to 2010, monthly and annual total discharges ( $\text{m}^3 \text{yr}^{-1}$ ) were computed for this period. Annual mass flux of DOC and DIC were calculated from 1986 to 2010 as the annual sum of the monthly total mass flux (the product of concentration and monthly total water discharge). Annual flow-weighted mean concentrations ( $\text{kg m}^{-3}$ ) for DOC and DIC were calculated by dividing the respective annual mass flux by the total annual water discharge. The annual

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flow-weighted mean concentration for DOC from 1989 is an outlier and is therefore removed from the data set.

### 3 A mechanistic framework for modeling of solute release and transport

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

$$c = r \frac{\tau}{\theta} \quad s = r \frac{q_s \tau}{\theta} \quad (1)$$

where  $r$  is the average release (dissolution) rate [ $\text{MT}^{-1}\text{L}^{-3}$ ] of DOC or DIC in the stream tube,  $\tau$  is the advective (water) travel time [ $\text{T}$ ] through the stream tube (essentially the time for 1 pore volume of water to flow through the stream tube),  $\theta$  is the average volumetric water content (-) [ $\text{L}^3$ -water  $\text{L}^{-3}$ -bulk] (equals the porosity under fully saturated, groundwater conditions), and  $q_s$  is the local specific groundwater discharge (Darcy flux) [ $\text{LT}^{-1}$ ] 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):

$$r = \frac{dc}{dt} = -kc_0^* \quad (2)$$

where  $t$  is running time [ $\text{T}$ ],  $c^*$  is the average bulk concentration [ $\text{ML}^{-3}$ -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 [ $\text{T}^{-1}$ ] (with  $1/k$  quantifying a characteristic time for the entire initial bulk concentration

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$c_0^*$  to dissolve). Inserting Eq. (2) into Eq. (1) (through  $r$ ) and solving for initial conditions  $c^*(0) = c_0^*$  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  $\tau \leq 1/k$  (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  $t \leq \tau$  (i.e., before 1 whole pore volume of water has flown through the stream tube)

$$\frac{\theta_c}{c_0^*} = kt; \quad \frac{\theta_s}{q_s c_0^*} = kt \quad (3a)$$

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

$$\frac{\theta_c}{c_0^*} = k\tau; \quad \frac{\theta_s}{q_s c_0^*} = k\tau \quad (3b)$$

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

$$\frac{\theta_c}{c_0^*} = k(\tau - 1) + 1; \quad \frac{\theta_s}{q_s c_0^*} = k(\tau - 1) + 1. \quad (3c)$$

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  $t \leq 1/k$  (i.e. before  $c_0^*$  has been entirely dissolved)

$$\frac{\theta_c}{c_0^*} = kt; \quad \frac{\theta_s}{q_s c_0^*} = kt \quad (4a)$$

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

$$\frac{\theta_c}{c_0^*} = 1; \quad \frac{\theta_s}{q_s c_0^*} = 1 \quad (4b)$$

- for  $\tau < t \leq \tau + 1/k$  (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)

$$\frac{\theta_c}{c_0^*} = k(\tau - 1) + 1; \quad \frac{\theta_s}{q_s c_0^*} = k(\tau - 1) + 1. \quad (4c)$$

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

$$\bar{c}_f = \frac{1}{\bar{q}_s} \int_0^{\infty} sg(\tau) d\tau; \quad \bar{s} = \bar{q}_s \bar{c}_f \quad (5)$$

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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  $\tau$  through the catchment to the stream.

Equations (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. Different types and forms of the pdf  $g(\tau)$  are then possible and relevant for different catchment conditions (Lindgren et al., 2004; Cvetkovic, 2011; Persson et al., 2011). In the following section, we exemplify and illustrate results from the catchment-average solution Eq. (5) for uniform  $g(\tau)$ , with  $0 \leq \tau \leq 2E[\tau]$  and  $E[\tau]$  being average  $\tau$ . This  $g(\tau)$  choice is made for simplicity, and use of other, more realistic  $g(\tau)$  functions, typically with greater  $\tau$  spreading over a catchment (Persson et al., 2011; Cvetkovic, 2011) may further enhance the main implications of spatial  $\tau$  variability that are shown and discussed in the following.

## 4 DOC and DIC release and transport

### 4.1 The DOC case

The aim of the calculation examples in this section is to demonstrate what the flow-independent dynamics of normalized (with  $q_s$  and/or  $\overline{q_s}$ ) concentration  $c$  and mass flux  $s$  from Eqs. (3)–(5) look like under various conditions of relevance for DOC release and transport. For DOC,  $c_0^*$  represents the bulk concentration of organic material in the organic horizon, which is annually renewed, dissolved and transported as DOC to streams each year. This annual periodicity implies that the characteristic DOC release time  $1/k$  should be on the order of 1 yr or smaller. The reported order of magnitude of mean advective travel time,  $\tau$ , through the soil-groundwater system of the Abiskojoekken catchment is also about 1 yr (Lyon et al., 2010), as has been reported for average  $\tau$  also in other regionally-relevant Swedish catchments (Darracq et al., 2010; Persson et al., 2011). Similar average  $\tau$  and  $1/k$  magnitudes (of around 1 yr) further imply that

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any and all of the different Eqs. (3)–(4) may apply to the DOC release and transport in the different individual stream tubes of a catchment that are included in the integration over the entire travel time pdf  $g(\tau)$  in Eq. (5).

The solutions Eqs. (3)–(4) show further that the (local and flux-averaged) concentration of DOC can be quite dynamic (see Fig. 2b), even for temporally constant  $\tau$ ,  $\theta$  and  $c_0^*$ . Specifically,  $c$  will change in time, by the moving through the different solution components of Eqs. (3)–(4) as  $t$  increases, even if  $\tau$ ,  $\theta$  and  $c_0^*$  remain the same, and regardless of how  $q_s$  changes over  $t$ . This temporal variability of  $c$  may in the DOC case also dominate the dynamics of local ( $s = c * q_s$ ) and catchment-average ( $\bar{s} = \bar{c}_f \bar{q}_s$ ) mass flux, so that it is relatively independent of the temporal variability of discharge into the stream (local  $q_s$  and catchment-average  $q_s$ ). We will return to investigate this hypothesis further using observation data from Abiskoajokken catchment, which are independent of the calculation results from Eqs. (3)–(5) exemplified and discussed in this section.

Figure 2 shows calculated dynamics of possible DOC transport, in terms of normalized concentration  $c\theta/c_0^*$  and mass flux  $s\theta/(q_s c_0^*)$  in a single stream tube (single  $\tau$ ; Fig. 2a), and  $\bar{c}_f \theta / c_0^*$  and  $\bar{s} \theta / \bar{q}_s c_0^*$  over a whole catchment (variable  $\tau$ ; Fig. 2b), for different (stream tube or catchment-average)  $\tau$  and release rate  $k$  (or characteristic release time scale  $1/k$ ). For any given travel time  $\tau$  (local or average; here 1 yr), lower average release rate  $k$  implies longer time  $1/k$  to complete dissolution of the annually renewed soil source of DOC ( $c_0^*$ ) as well as longer time to reaching a stable average  $c$  and  $s$  level (even though with possible fluctuations around these level for different  $\tau$  conditions; Fig. 2b).

For release and transport through a stream tube with a single  $\tau$  value (Fig. 2a), the flow-normalized  $c$  and  $s$  do not fluctuate around their average levels. The corresponding catchment-average  $\bar{c}_f$  and  $\bar{s}$ , however, can clearly fluctuate in time even for temporally constant  $c_0^*$ ,  $\theta$  and  $q_s$ , which have here been used for normalization (Fig. 2b). The local (stream tube) and catchment-average results differ temporally due to the spatial  $\tau$  variability accounted for in the latter, which implies different evolution with increasing

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time  $t$  through the different solution components Eqs. (3)–(4) for different stream tubes in the catchment. The temporal fluctuations in the catchment-average results increase with decreasing average dissolution time below  $1/k < 1$  yr (see example  $1/k = 0.1$  yr compared with  $1/k = 1$  yr in Fig. 2b). For average dissolution time  $1/k \geq 1$  yr, the fluctuations are overall smaller than for  $1/k < 1$ , and disappear when  $1/k$  values reach integer multiples of the source renewal rate of 1 yr (see  $1/k = 1.9$  yr and  $1/k = 1$  yr in Fig. 2b).

## 4.2 The DIC case

For the case of DIC,  $c_0^*$  represents the average bulk mineral concentration in the whole soil-rock system under the surface of the catchment where weathering can occur and dissolve inorganic carbon. This is a relatively slow process (compared to respiration and dissolution rates for organic carbon), and complete depletion of  $c_0^*$  (which is not renewed in the DIC case) should be expected to take long time, which is quantified by a much greater  $1/k$  than the average  $\tau$  of 1 yr. Under such conditions, Eq. (3b) for  $\tau < t \leq 1/k$  is more or less generally applicable for most (if not all) prevailing subsurface flow and transport pathways (stream tubes) through a catchment. This means that the DIC concentration in the water flow into receiving streams is in this case predominantly determined by the average  $k\tau$  value (i.e. the relation  $\tau/(1/k)$  between average advective travel time and time to total dissolution). In other words, the flux-average concentration of DIC should not exhibit the added dynamic fluctuation effect evident for DOC (Fig. 2b) from the different solution components Eqs. (3)–(4), which apply to stream tubes with different  $\tau$  in the catchment-average solution Eq. (5). This expectation is confirmed by calculation results for large dissolution time scale  $1/k (\gg \text{average } \tau)$ , as should be relevant for DIC release and transport (Fig. 3).

As a consequence, the dynamics of catchment-average mass flux  $\bar{s} = \overline{c_f q_s}$  should for DIC be dominated by the temporal variability of local ( $q_s$ ) and catchment-average ( $\overline{q_s}$ ) discharge to the stream. Minor contributors to the behavior of  $\bar{s}$  are the hydrological parameters  $\theta$  and  $\tau$  and possibly a decrease in the bulk mineral source  $c_0^*$  as

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the weathering and dissolution progresses with time. The latter has for simplicity been neglected in the underlying dissolution Eq. (2) as this effect has been investigated and shown previously by Eriksson and Destouni (1997) and will not affect the main results and implications of the present study. The temporal variability effects of  $\theta$ ,  $\tau$  and  $c^*$  are expected to be much smaller than the effect of temporal variability in discharge ( $q_s$  and  $\overline{q_s}$ ). This implies that we expect the DIC mass flux measured in the field to be highly correlated with the measured stream discharge, while the DIC concentration should be much less variable in time and essentially uncorrelated with discharge. This DIC expectation will in the following section be tested against available data from Abiskoajokken catchment, along with the complementary hypothesis formulated above for DOC.

## 5 Interpreting observed data from the Abiskoajokken catchment

### 5.1 Time series analysis of observed data

Time series of annual flow-weighted DOC and DIC concentrations, annual DOC and DIC mass fluxes, and annual total discharge for the Abiskoajokken catchment were analyzed for trends (at a significance level of  $\alpha = 0.1$ ) using the generalized least squares approach (Box et al., 2008) after removal of autocorrelation with the Durbin Watson test, in agreement with the methodology put forward by Durbin and Watson (1950, 1951, 1971). In addition, the dependence of DOC and DIC export concentrations and mass fluxes on total annual flow discharge was assessed. The total annual water discharge exhibits a significant decreasing trend ( $p = 0.078$ ) over the period of observation in Abiskoajokken catchment (between 1986 and 2010; Fig. 4). For the same time period there is a significant positive trend ( $p = 0.060$ ) in the annual flow-weighted average DIC concentration (Fig. 5a), while the annual flow-weighted average DOC concentration is essentially constant. There are no significant trends in the total annual mass flux of either DIC or DOC over the period (Fig. 5b).

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## 5.2 Relating observed fluctuations to model implications

As expected from the mechanistic model implications, the fluctuations of annual flow-weighted average DIC concentration around its mean trend is essentially independent of the discharge fluctuations (Fig. 6a), while the fluctuations in annual DIC mass flux show high positive correlation with the fluctuations in annual discharge (Fig. 6b). While some small-scale spatial variability in weathering rates likely exists between higher altitudes and lower valley bottoms (e.g. Allen et al., 2001; Dixon et al., 2008), similar high mass flux correlation with discharge (i.e. Fig. 6b) has also been found in other studies for diffuse solute inputs from geogenic sources or subsurface sources that have been formed over time as legacies of earlier anthropogenic solute inputs at the land surface (Godsey et al., 2009; Basu et al., 2010). DIC loading from the sub-arctic landscape at the scale of the Abiskoajokken catchment, as such, appears to be associated with the diffuse and relatively slow DIC release from rather ubiquitous weathering across the entire catchment.

This interpretation is consistent with the recent work relating the easily-weatherable carbonaceous bedrock (specifically Ca/Na ratio) with DIC in streams across spatial scales for this region (Giesler et al., 2012). As such, the large average dissolution time scale ( $1/k \gg$  average  $\tau$ ) relevant for weathering and its byproducts (i.e. DIC) makes the mechanistic solution component Eq. (3b) predominant. The DIC concentration is controlled at a similar stable level, as determined by average  $k\tau$ , throughout most or all the flow and transport pathways (stream tubes) through the catchment. Not only  $k$  but also average  $\tau$  is then to a large degree independent of the temporal fluctuations of discharge into the stream, because the whole flow and transport process through the soil-groundwater system to the stream, which determines average  $\tau$ , is not much affected by (or correlated with) the most downstream discharge ( $\bar{q}_s$ ) and its temporal fluctuations precisely at the stream interface.

As also expected from the mechanistic model implications, and counter to the DIC behavior, the fluctuations of both the annual flow-weighted DOC concentration (Fig. 6a)

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and the annual DOC mass flux (Fig. 6b) are essentially uncorrelated with the discharge fluctuations over the same period. We recall here that the basis for expecting this discharge independence is the above-discussed added dynamic effect of spatially variable  $\tau$  in the DOC case (which is not the case for DIC, for which  $1/k \gg$  average  $\tau$ ). This additional dynamic effect is partly attributed to the variable nature of decomposition rates for organic material and the subsequent release rate for DOC, which are coupled to numerous factors such as temperature, vegetation type and soil moisture distribution (Clark et al., 2005; Hobbie et al., 2000; Lyon et al., 2011; Seibert et al., 2009; Sjögersten and Wookey, 2004; Winterdahl et al., 2011). This interpretation is consistent with several studies highlighting explicitly the connection between sub-annual variations in soil water DOC (akin to the DOC release time scale) and stream concentrations (e.g. Seibert et al., 2009; Clark et al., 2005).

Further, by quantifying the DOC concentration fluctuation around its temporal mean or median value, and comparing it to the fluctuation implied by the mechanistic model solution for different  $1/k$  values, we can get a rough estimate of what the prevailing average DOC release time scale  $1/k$  may actually be in Abiskoajokken catchment. Characteristic fluctuations around the mean are calculated here as the range between maximum and minimum value in relation to the mean, whereas fluctuations around the median are calculated as the difference between the 90 and 10 percentile in relation to the median. For the mechanistic model, an  $1/k$  value of about 0.1 yr then gives relative fluctuations of normalized flux-average DOC concentration (and normalized mass flux) of about 40% around the mean, and 22% around the median value (Fig. 2b). This is consistent with the relative fluctuations of annual flow-weighted average DOC concentration in the observed time series data, which are 43 and 27% around the mean and median value, respectively (Fig. 5a). Furthermore, a DOC release time scale of 0.1 yr matches the time period of the spring freshet while a release time scale of up to around 0.4 yr corresponds to the period of the wet summer season with shallow flow pathways for Abiskoajokken (Lyon et al., 2010). This is in line with previous conceptual understanding of DOC export from these landscapes (Smedberg et al., 2006).

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Considering again the observed annual flow-weighted average DIC concentration, it exhibits lower fluctuations around both its mean (22 %) and median (14 %) concentration value than (the corresponding 43 and 27 % of) the DOC concentration (all calculated from the time series in Fig. 5a). While the observed DOC mass flux fluctuations (from the time series in Fig. 5b) are then similar (36 and 27 % around the mean and the median mass flux value, respectively) to those of the DOC concentration (of 43 and 27 %, respectively), the corresponding DIC mass flux fluctuations of 44 and 23 % are higher than the DIC concentration fluctuations (of 22 and 14 %). The temporal variability in DIC mass flux can largely be explained by the temporal discharge variability (Fig. 6b) in combination with the higher DIC mass flux fluctuations relative to the DIC concentration fluctuations. These findings, thus, support the modeling expectation of the discharge ( $q_s$ ) temporal variability being greater and more important for the DIC mass flux than the temporal variability of the other involved hydrological parameters ( $\theta$ ,  $\tau$ ). The hydrological parameters ( $\theta$ ,  $\tau$ ) should similarly affect both the mass flux and the concentration according to the DIC-relevant mechanistic model Eq. (3b).

### 5.3 Observed data trends in relation to model framework

Taken together, the relationship between observed mass flux and concentration fluctuations and the outlined mechanistic model framework provides a lens through which to directly interpret the aforementioned longer-time trends (or lack thereof) in DOC/DIC concentrations and loads (Figs. 4 and 5). Starting with the significant longer-term trend that the discharge exhibits over the observation period in Abiskoajokken, this reflects a temporally sustained flow decrease over the whole catchment and, most importantly, over sufficiently long times (instead of being just short-term fluctuations) to also affect the average  $\tau$  through the catchment. The decreasing discharge trend is consistent with previously reported shifts in both observed (Lyon et al., 2009; Dahlke et al., 2012) and modeled (Sjöberg et al., 2012) hydrological flows, primarily due to thawing of permafrost and secondarily due to shifts in the hydroclimatic forcing (mainly temperature increases) (Dahlke et al., 2012).

Furthermore, the DIC concentration trend is consistent with increasing  $\tau$  in the most relevant mechanistic model solution Eq. (3b) for DIC, which thus explains the increasing DIC concentration as a significant discharge and associated  $\tau$  trend effect. The DIC-relevant Eq. (3b) also shows that the related discharge decrease and  $\tau$  increase trends counteract each other in the mass flux expression, and should leave the DIC mass flux essentially unchanged, as is also independently observed in the field (Fig. 5b). This is the reason why the annual flow-weighted average DIC concentration is affected by this trend in annual discharge. For the DOC concentration, however, the dynamic effect induced by the spatial  $\tau$  variability is sufficiently dominant to override and mask this trend of decreasing annual discharge and related increasing average  $\tau$ , so that the annual flow-weighted average DOC concentration does not exhibit any significant trend over the observation period.

## 6 Conclusions

The results of this study show that the release of organic and inorganic carbon and export dynamics of DOC and DIC function on widely different timescales in the investigated field case of Abiskoajokken catchment. Furthermore, they depend differently on the subsurface conditions and spatiotemporal variability of different hydrological-hydrogeological (HH) and solute-specific (SS) factors in the catchment.

The overall export concentration and mass flux levels of both DOC and DIC depend on: (I) The respective (SS) bulk concentration ( $c^*$ ) in the subsurface source zones of DOC and DIC. (II) The relation between the (HH) average advective solute travel time ( $\tau$ ) and (SS) characteristic release time ( $1/k$ ) from and through the source zones of DOC and DIC release along the subsurface transport pathways to the receiving stream. Furthermore, for both the concentration and the mass flux of DOC (III) the spatial (HH) variability of travel times ( $\tau$ ) among the different subsurface flow and transport pathways to the stream controls the fluctuations around the overall DOC levels. In contrast, the fluctuations in DIC mass flux (IV) depend to large degree on the temporal (HH)

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variability of discharge into the stream ( $q_s$ ), while the smaller DIC concentration fluctuations ( $V$ ) depend on and reflect the effects of temporal (HH) variability in other flow and transport parameters ( $\theta, \tau$ ).

By distinguishing in this way the role of different solute-specific and hydrological-hydrogeological factors, with the latter being similar for different solutes in the same catchment, these results can facilitate further and improved quantification and projection of how climate-driven and hydrologically mediated and propagated changes in catchments of permafrost regions can affect the magnitude and dynamics of DOC and DIC export to streams. The present study has combined relatively long data time series with mechanistic model conceptualization and hypothesis formulation to identify and quantify the dominant solute release and transport processes for the main dynamic features in the time series. This approach can more generally help in clarifying the connections between stream chemistry dynamics of DOC, DIC and other types of solute, and the hydroclimatic controls and conditions along the diffuse subsurface sources and pathways that feed into the stream networks of regional catchments in different parts of the world.

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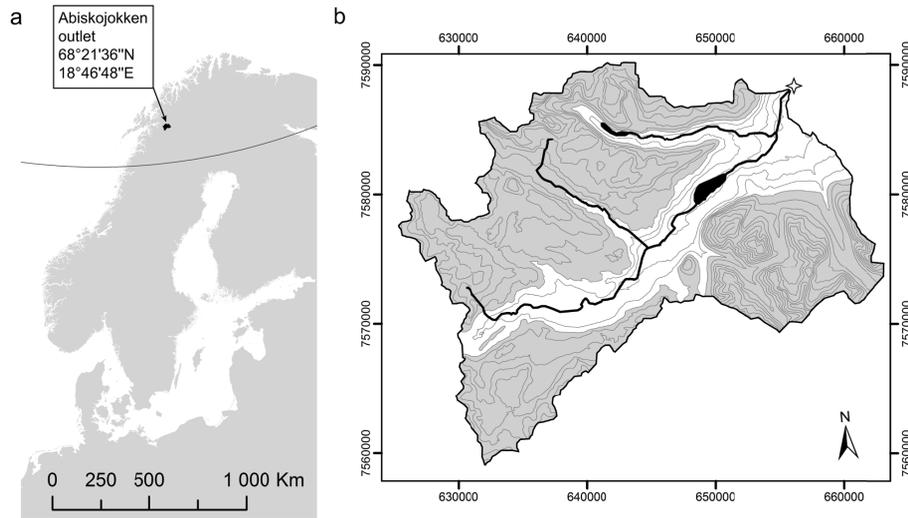
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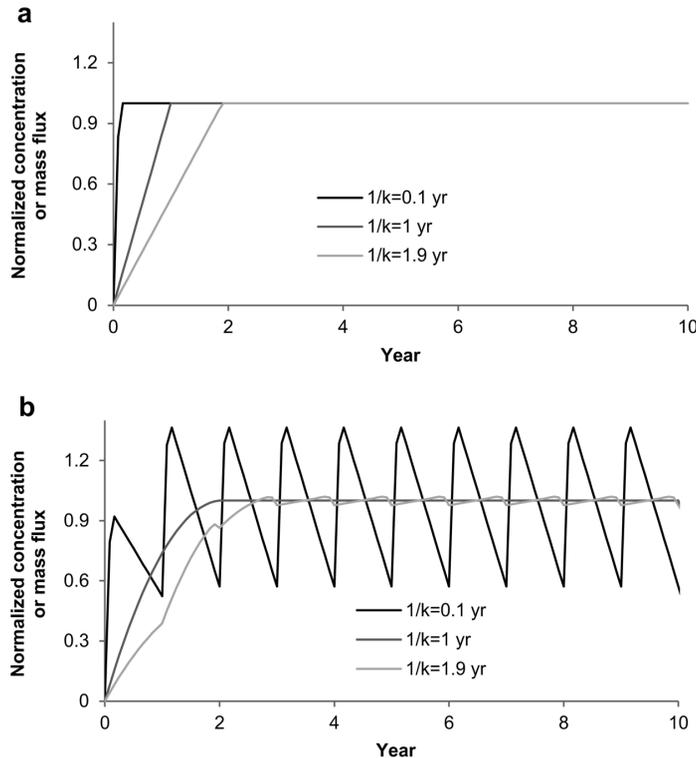
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**Fig. 1.** (a) Map of Scandinavia with Abiskojokken catchment in northern Sweden (marked in black) and the Polar circle (marked by line). (b) Site map of Abiskojokken catchment with 73 % of the catchment situated above 800 m a.s.l. (shaded grey), the outlet is marked by a star (68°21' 36" N, 18°46' 48" E). The coordinate system is given in SWEREF 99.

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**Fig. 2.** DOC transport dynamics. **(a)** Results for a single stream tube with a single travel time,  $\tau = 1$  yr. **(b)** Results for an ensemble of stream tubes with variable (uniformly distributed)  $\tau$ , and average  $\tau = 1$  yr. The results show how the release rate  $k$  (or characteristic release time  $1/k$ ) affects DOC concentration and mass flux from the soil-groundwater into the stream system.

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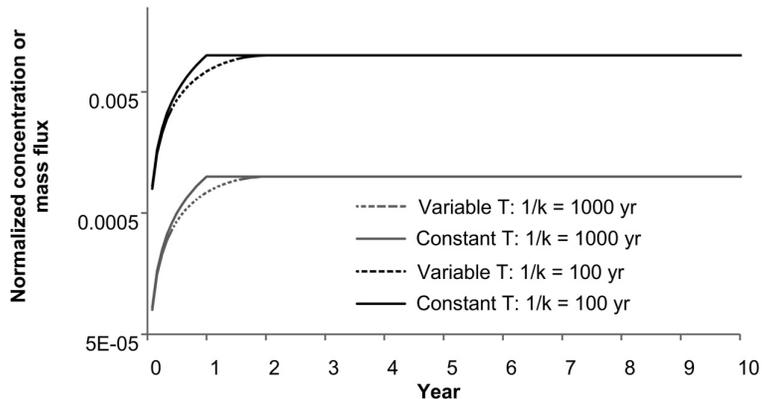
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**Fig. 3.** DIC transport dynamics. Results are shown on log-scale for single travel time ( $\tau = 1$  yr; solid curves) and variable, uniformly distributed  $\tau$  (with average value of 1 yr; dashed curves), for different release rates  $k$  (or characteristic release times  $1/k$ ).

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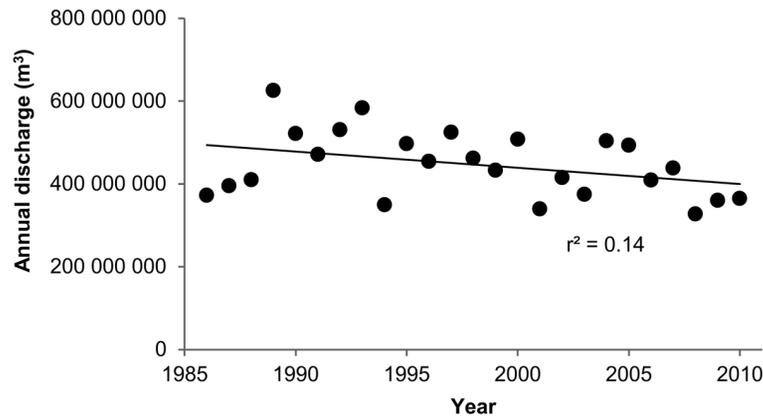
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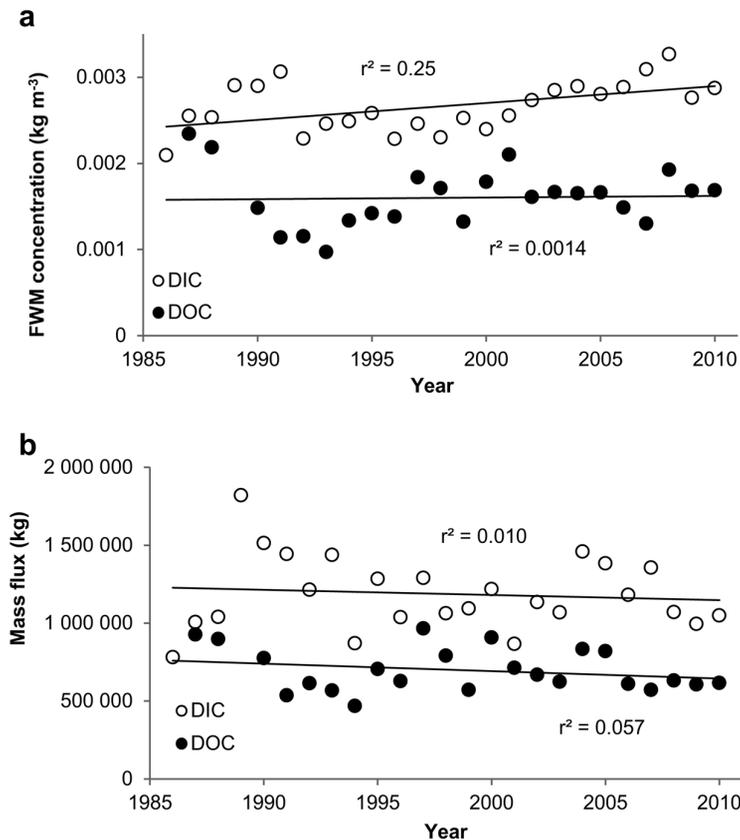


**Fig. 4.** Annual discharge between 1986 and 2010 at the outlet of Abiskoajokken catchment. The discharge exhibits a significant ( $p = 0.078$ ) negative trend.

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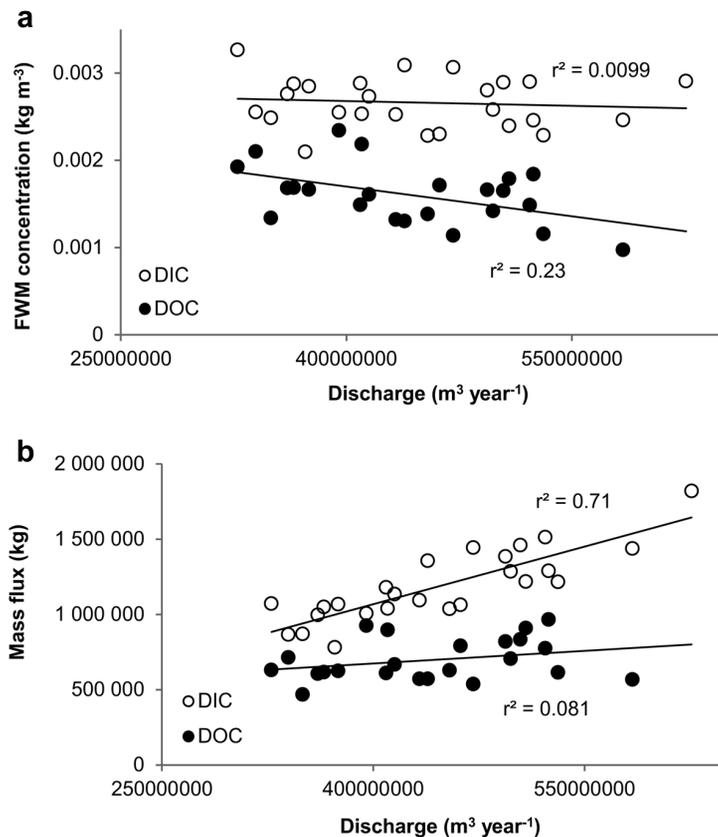
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**Fig. 5.** Dissolved carbon time series. **(a)** Annual flow-weighted mean (FWM) concentration of DOC and DIC at the outlet of Abiskoajokken catchment. The DIC concentration exhibits a significant ( $p = 0.060$ ) positive trend. **(b)** Annual flow-weighted (FW) mass flux of DOC and DIC at the outlet of Abiskoajokken catchment.

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**Fig. 6.** Dissolved carbon discharge relationships. **(a)** Annual flow-weighted mean (FWM) of DOC and DIC versus annual discharge at the outlet of Abiskoajokken catchment. **(b)** Annual flow-weighted (FW) mass flux of DOC and DIC versus annual discharge at the outlet of Abiskoajokken catchment.

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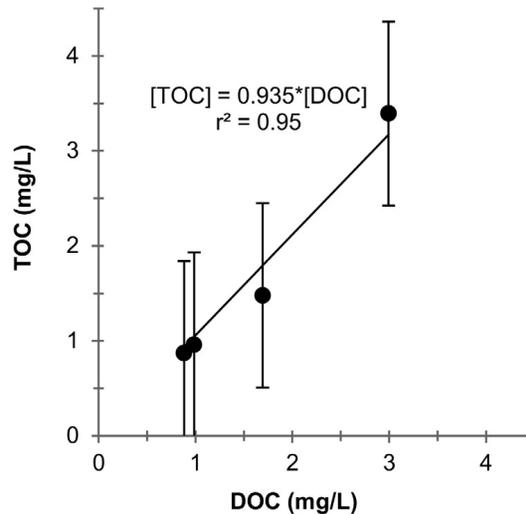
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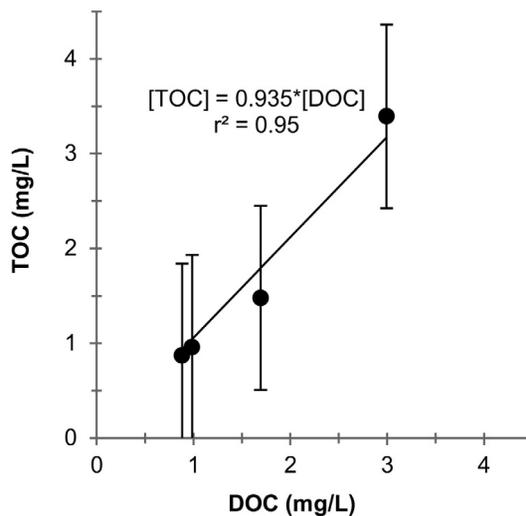


**Fig. A1.** Concentrations of TOC and DOC for Abiskoajokken outlet, measured within a range of 3–5 days. TOC concentration was measured on a monthly basis whereas DOC was measured between 2 and 9 times per month. The measurements were conducted between April to July 2008 and the bars show minimum and maximum TOC values measured for the same month.

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**Fig. A2.** Daily concentrations of alkalinity and DIC for Abiskoajokken outlet for entire April 2008.

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