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The role of terrestrial productivity and hydrology in regulating aquatic dissolved organic carbon concentrations in boreal catchments

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Abstract

The past decades have witnessed an increase in dissolved organic carbon (DOC) concentrations in the catchments of the Northern Hemisphere. Increasing terrestrial productivity and changing hydrology may be reasons for the increases in DOC concentration. The aim of this study is to investigate the impacts of increased terrestrial productivity and changed hydrology following climate change on DOC concentrations. We tested and quantified the effects of gross primary production (GPP), ecosystem respiration (RE) and discharge on DOC concentrations in boreal catchments over 3 years. As catchment characteristics can regulate the extent of rising DOC concentrations caused by the regional or global environmental changes, we selected four catchments with different sizes (small, medium and large) and landscapes (forest, mire and forest-mire mixed). We applied multiple models: Wavelet coherence analysis detected the delay-effects of terrestrial productivity and discharge on aquatic DOC variations of boreal catchments; thereafter, the distributed-lag linear models quantified the contributions of each factor on DOC variations. Our results showed that the combined impacts of terrestrial productivity and discharge explained 62% of aquatic DOC variations on average across all sites, whereas discharge, gross primary production (GPP) and RE accounted for 26%, 22% and 3%, respectively. The impact of GPP and discharge on DOC changes was directly related to catchment size: GPP dominated DOC fluctuations in small catchments (<1 km^2), whereas discharge controlled

DOC variations in big catchments (>1 km^2). The direction of the relation between GPP and discharge on DOC varied. Increasing RE always made a positive contribution to DOC concentration. This study reveals that climate change-induced terrestrial greening and shifting hydrology change the DOC export from terrestrial to aquatic ecosystems. The work improves our mechanistic understanding of surface water DOC regulation in boreal catchments and confirms the importance of DOC fluxes in regulating ecosystem C budgets.

KEYWORDS
boreal catchments, catchment size, discharge, DOC, GPP, landscape, RE, terrestrial productivity

1 | INTRODUCTION

Dissolved organic carbon (DOC) concentrations in freshwater ecosystems have increased in large areas of the northern hemisphere over the past few decades (Filella & Rodríguez-Murillo, 2014), which is called 'aquatic browning' (Roulet & Moore, 2006). From an ecological point of view, rising DOC has important implications on aquatic ecosystems. First, rising DOC may reduce aquatic primary production and affect predator–prey interactions (Kritzberg et al., 2020) due to the changes in the light climate. Second, it may also contribute to eutrophication in coastal ecosystems, further leading to hypoxia (Conley et al., 2011) and loss of biodiversity (Villnäs & Norkko, 2011). Additionally, societal impacts of rising DOC include increasing costs for purifying drinking water and reducing the aesthetic and recreational value of aquatic landscapes (Ekström, 2013; Freeman et al., 2004). Finally, DOC exports play an essential role in ecosystem carbon (C) budgets (Cole et al., 2007; Nilsson et al., 2008). Since much of terrestrial derived DOC that reaches surface waters will be converted to CO_2 by biotic and abiotic processes, increased DOC has the potential to mobilize large terrestrial C pools and affect C fluxes in both the atmosphere and the ocean (Mann et al., 2012; Öquist et al., 2014).

Proposed mechanisms behind rising DOC concentrations include decreasing acidification (Kang et al., 2018), land-use changes (Wilson & Xenopoulos, 2008) and climate change (Asmala et al., 2019; Ekström, 2013). Decreasing acidification has been identified as a significant driver behind the long-term DOC increases in catchments, either by itself (Ekström, 2013; Monteith et al., 2007; Vuorenmaa et al., 2006) or coupled with climate change (Burns et al., 2006; Evans et al., 2005). In addition, land management, such as the drainage of peatlands, may transform the hydrochemistry and hydrology (Holden et al., 2004), and DOC loads are typically high from the drained boreal peatlands (Asmala et al., 2019; Nieminen et al., 2021). However, the land-use changes or recovery from acid deposition do not always explain changing DOC concentrations. For example, increased DOC has also occurred in areas with low acid deposition and with increasing forest biomass, such as Krycklan in Sweden (Laudon & Sponseller, 2018).

Climate change impacts such as permafrost thaw (Tank et al., 2016), increased precipitation alone (Hongve et al., 2004) or in combination with higher temperature (Keller et al., 2008) have contributed to rising DOC concentrations. However, Freeman et al. (2004) stated that neither warming, increased discharge or the shifting trends in the proportion of annual rainfall arriving in summer can offer satisfactory explanations. However, the CO_2-mediated stimulation of primary productivity is responsible for increasing exported DOC from peatlands. Schlesinger and Andrews (2000) also confirmed that elevated CO_2 concentrations could potentially increase primary productivity leading to increased DOC inputs from terrestrial to aquatic systems. On the contrary, Ellis et al. (2009) found that elevated atmospheric CO_2 treatment decreased DOC concentrations, in environments where the concentrations of labile C limit decomposition. Ombrotrophic bogs were such environments. Finstad et al. (2016) and Pumpanen et al. (2014) brought forward the hypothesis that browning could be linked to changes in the productivity of forests. To a large extent, DOC in water bodies is derived from the microbial decomposition of organic matter (Schimel et al., 1994; Xu & Guo, 2018). Increases in the productivity of vegetation could then increase DOC via gradual increases in soil organic matter (SOM) or via a process called priming (He et al., 2020). Priming denotes changes in recalcitrant local soil C decomposition after new fresh C is available. Changes in decomposition due to priming can be positive or negative. Positive priming effects mean that increases in production lead to increasing decomposition of SOM and consequently increased DOC concentrations. Positive priming is attribute to increased microbial growth and activity (Kuzyakov, 2010; Linkosalmi et al., 2015). However, it is often associated to nutrient-poor conditions, under which microbes use the energy from rhizodeposition 'to mine nutrients in SOM', thereby releasing extra CO_2 (Sullivan & Hart, 2013). A negative priming effect means that increased production leads to a decrease in native SOM decomposition and subsequently DOC concentrations. Competition between the plant roots and rhizosphere organisms for mineral N and nutrient limitation of the rhizosphere have been proposed as reasons for negative priming (Dijkstra et al., 2013; Kuzyakov, 2002). Additionally, the so-called preferential substrate utilization may lead to a decrease in the decomposition of old C in the short term, since the microbes tend to utilize the fresh organic matter initially (Kuzyakov et al., 2000). Priming will lead to more rapid changes in DOC

since microbial biomass and functions change over days to weeks after a change in the supply of easily available carbon.

Studies by Pumpanen et al. (2014) and Finstad et al. (2016) examined the role of direct priming on DOC concentrations using proxy measurements. Finstad et al. (2016) showed that increasing Normalized Difference Vegetation Index (NDVI), a remote sensing–derived indicator of plant leaf area and productivity, was associated with DOC concentrations. Increases in NDVI led to increasing DOC concentrations. Pumpanen et al. (2014) showed that the average yearly DOC concentration depended on the forest GPP of the previous year. Both analyses suggest a role of priming, but the evidence presented is at a time scale that exceeds the response of microbial communities to changes in resources (years instead of days to weeks). Additionally, the effects of increasing soil organic carbon cannot be excluded in both studies.

The connections between primary production and DOC concentrations in surface waters are not easy to detect. In large monitoring data sets, surface DOC concentrations may be caused by changes in vegetation and its management. For example, in the study of (Pumpanen et al., 2014), changes in forest age are easily confounded with changes in productivity. However, direct regressions of DOC concentrations and photosynthetic production are not reasonable since changes in productivity will affect changes in DOC concentrations at a delay of several days to several weeks. The delay is caused by the transport of fresh photosynthates to the roots, changes in decomposition and microbial biomass after a change in root exudation and hydraulic delays required to transport DOC from the soil to water bodies (Wen et al., 2020). This study tested the effects of variation in terrestrial productivity from eddy covariance (EC) data with high-frequency measurements of DOC concentrations in four Northern boreal watersheds. We used cross-spectral wavelet analyses as well as distributed-lag linear models (DLMs) to test and quantify the effects of GPP, ecosystem respiration (RE), net ecosystem production (NEP) and discharge on aquatic DOC concentrations.

2 | MATERIALS AND METHODS

2.1 | Study site

Four catchments with different sizes (small, medium and large) and landscapes (forest, mire and forest-mire mixed) were studied. The size and landscape of catchment were noted after site name.

Hereafter, the letters 'S', 'M' and 'L' signify the catchment sizes (small, medium and large), whereas the symbols 'forest', 'mire' and 'mix' reflect the land cover types (forest, mire and forest-mire mixed). Three sub-catchments locate in Krycklan, about 50 km northwest of the city of Umeå in northern Sweden (64°14′N, 19°46′E) (Figure S1). In Krycklan, C2[S-forest] is covered by forest with the size of 0.14 km^2; C4[S-mire] of 0.19 km^2 is covered by 40.4% of wetlands and the remainder is forest; C6[M-mix] of 1.3 km^2 is constituted by 72.8% of forest, 24.1% of wetland and 3.1% of lakes (Table 1). The climate is characterized as a cold temperate humid type with persistent snow cover during the winter season. The 30-year mean annual temperature (1981–2010) is 1.8°C, January −9.5°C, and July 14.7°C. The mean annual precipitation is 614 mm and mean annual runoff 311 mm, giving annual average evapotranspiration of 303 mm (Laudon et al., 2013). The 40-year average duration of winter snow cover is 167 days, but this has been decreasing over time (Laudon et al., 2021). Yli-Nuortti (NT[L-mix]) is a catchment nearby Nuorttiaapa measuring station and located in Värriö, Finland (67°44′N, 29°27′E) approximately 120 km north of the Arctic Circle close to the northern timberline (Figure S1). NT[L-mix] covers about 40 km^2 with 25% of peatlands, and 5% of the area is covered by alpine vegetation on the top of the fells while the rest of the catchment is dominated by pine forests on glacial tills (Table 1). There are no lakes above the measurement station. According to the statistics of the Finnish Meteorological Institute (1981–2012), the mean annual air temperature is −0.5°C. The mean temperature in January is −11.4°C and in July 13.1°C. The mean annual precipitation is 601 mm, mean annual runoff 212 mm and annual average evapotranspiration is 389 mm. The average number of days with snow cover is 205–225 days (Pohjonen et al., 2008).

2.2 | Sampling and laboratory dissolved organic carbon measurement

All water samples were sampled in the surface water (25 cm) by acid-washed LDPE bottles. In Finland (NT[L-mix]), we sampled monthly in winter and fall, fortnightly in spring and every week in summer (2018–2020). In Sweden (C2[S-forest], C4[S-mire] and C6[M-mix]), water samples were collected monthly during winter, every 2 weeks during summer and fall, and every third day during the spring flood (2016–2018). Water samples were filtered immediately after

TABLE 1 Site information

| Site ID. | Site name | Size (km^2) | Land cover % | | | | Land-use type | EC tower |
			Forest	Wetland	Lake	Alpine vegetation		
C2[S-forest]	Västrabäcken	0.14	100	0	0	0	Forest	Rosinedal
C4[S-mire]	Kallkälsmyren	0.19	59.6	40.4	0	0	Wetland	Degerö
C6[M-mix]	Stortjärnsbäcken	1.30	72.8	24.1	3.1	0	Mixed	Rosinedal
NT[L-mix]	Nuorttiaapa station	40.0	70.0	25.0	0	5.0	Mixed	SMEAR I

sampling by a filtration system made of glass using Whatman GF/F Glass Microfiber Filters (pore size 0.45 μm), which had been rinsed by the sample water before filtration. All samples were frozen until further DOC analysis.

In Finland, DOC concentrations were determined by thermal oxidation coupled with infrared detection (Multi N/C 2100, Analytik Jena, Germany) following acidification with phosphoric acid. In Sweden, after all samples were acidified with H_3PO_4, DOC concentrations were measured by catalytic oxidation combustion (Shimadzu TOC-5000, Kyoto, Japan) (Laudon et al., 2011).

2.3 | Prediction of dissolved organic carbon based on real-time spectral absorbance

To monitor the real-time spectral absorbance, in situ portable multi-parameter UV–Vis probes (spectro:lyser, S:CAN Messtechnik GmbH, Austria) were installed in Yli-Nuortti river on June 12, 2018, and in the Krycklan catchments on May 9, 2016. The spectro:lyser measures absorbance across the wavelengths from 220 to 732.5 nm at 2.5 nm intervals with a path length of 35 mm. The benefits of in-situ UV–Vis probe is to make high-frequency aquatic monitoring possible, especially during short-duration events or in remote areas (Avagyan et al., 2014; Rode et al., 2016; Zhu et al., 2020).

Principal component regression (PCR) was used to model the relationship between DOC concentration and absorbance. In the PCR model, absorbance values from 250 nm to 732.5 nm at 2.5 nm intervals (194 variables) were the independent variables. The dependent variables were the DOC concentrations measured in the lab from water samples collected in the respective days. The observations were split into a training and testing data set. The training set contained 75% of observations that were randomly selected from all samples (C2[S-forest], C4[S-mire], C6[M-mix] and NT[L-mix]), and the testing set contained the remaining 25% of observations. The PCR analyses were conducted with the 'pls' package (Mevik et al., 2019) in R (R Core Team, 2019). After the PCR model was built, hourly real-time spectral absorbances were used as input to predict hourly DOC concentrations. The hourly predicted DOC concentrations were aggregated into daily data for further analysis. The outlier values were automatically detected and corrected using the '*tsclean*' function of package *'forecast'* (Hyndman & Khandakar, 2008) in R (R Core Team, 2019).

2.4 | Water discharge

In Finland, water discharge was determined based on the continuous water depth measurements carried out by pressure sensors measuring the hydrostatic pressure (Levelogger, Solinst, Georgetown, Canada) in the bottom of the river, which was corrected by barometric pressure measurements (Barologger, Solinst, Georgetown, Canada). The water depth measurements were converted to flow rates using channel cross-section, water depth and manual flow rate measurements (Flow Tracker Handheld ADV, SonTek, CA, USA) carried out at sampling locations.

In Sweden, water discharge was computed hourly from water level measurements (using pressure transducers connected to Campbell Scientific dataloggers, USA, or duplicate WT-HR water height data loggers, Trutrack Inc., New Zealand). Rating curves were derived based on discharge measurements using salt dilution or time-volume methods (Laudon et al., 2011).

2.5 | Carbon fluxes

There are three measuring stations nearby our study sites where the C exchange between the terrestrial ecosystem and the atmosphere is continuously recorded by the EC technology (Medlyn et al., 2005). The EC data included GPP, RE and NEP. We assumed that NEP = −NEE (Black et al., 2007), and the value for RE and GPP was taken from day-time measurements (Aubinet et al., 2012).

In Finland, the Värriö measuring station SMEAR I (67°45′N, 29°36′E, 390 m asl) is close to NT[L-mix]. Most of the area is dominated by 60-year-old Scots pine (*Pinus sylvestris* L.) forests, in addition to which there are also large wetlands and deep gorges in the surroundings (Vehkamäki et al., 2004a, b). Flux data from SMEAR I was applied to NT[L-mix]. The flux data were collected from the Dynamic Ecological Information Management System (https://deims.org/b4713 11f-e819-4f6f-bbae-1ac86cd9777f). The processing pipeline differed from the two Swedish sites due to polar day (24 h sunlight) during the growing season. More details about the whole process for data quality control are presented in Kulmala et al. (2019).

In Sweden, the Rosinedalsheden station (64°10′N, 19°45′E, 145 m asl) is located in a forest stand that consists of naturally regenerated 80-year-old Scots pine (*P. sylvestris* L.), and the soil is a deep deposit of sand and fine sand. The ground vegetation is dominated by blueberries (*Vaccinium myrtillus* L.) and lingonberries (*Vaccinium vitis-idaea* L.). Degerö station (64°11′N, 19°33′E, 270 m asl) is situated on a highland between two major rivers, Umeälven and Vindelälven. The site is a nutrient-poor minerogenic mire dominated by flat mire lawn plant communities with bog mosses (*Sphagnum balticum*, *Sphagnum majus* and *Sphagnum lindbergii*) dominating the bottom layer. The field layer is dominated by the cottongrass (*Eriophorum vaginatum* L.) and cranberry (*Vaccinium oxycoccos* L.), bog-rosemary (*Andromeda polifolia* L.) and deergrass (*Trichophorum cespitosum* L.). Sedges (*Carex spp.*) occur more sparsely. C fluxes data from Rosinedalsheden were applied to C2[S-forest], and C6[M-mix] and C fluxes data from Degerö were used in C4[S-mire] (Table S1). C fluxes data from the two EC towers were obtained from the ICOS data portal (Drought 2018 Team & ICOS Ecosystem Thematic Centre, 2020). The data had been subjected to standardized quality control using the ONEFlux processing pipeline (https://github.com/icos-etc/ONEFlux), including spike detection, data flagging and friction velocity filtering (Papale et al., 2006). ONEFlux processing pipeline is described in more detail in Pastorello et al. (2020).

2.6 | Wavelet coherence analysis

To test the hypothesis that discharge, GPP, NEP and RE influence DOC concentration in catchments, we investigated the temporal correlations between discharge, GPP, NEP, RE and DOC concentration by wavelet coherence analysis during the growing season and whole experimental period. Wavelet analysis has been effectively applied in the geosciences and ecology, showing good localization properties in the time and frequency domain (Grinsted et al., 2004; Kumar & Foufoula-Georgiou, 1997; Vargas et al., 2011). Wavelet analysis aims to quantify the variance of a specific time series and correlations between different time series across multiple frequencies in time (Grinsted et al., 2004). We applied continuous wavelet transforms (CWTs) to show frequency-dependent behaviour for exploring the relationship between discharge, GPP, RE, NEP and aquatic DOC concentration. In the CWT, it is possible to detect if two time series tend to oscillate simultaneously, rising and falling together within a given time period (in phase, and therefore showing no time lags), or rise and fall out of phase within a given time period (therefore showing a time lag between them) (Vargas et al., 2011). A 95% confidence level for the CWT was done through Monte-Carlo simulation using 1000 times. In this study, wavelet analysis was done using 'WaveletComp' package (Schmidbauer & Roesch, 2018) in R (R Core Team, 2019).

2.7 | Distributed lag models

DLMs (Gasparrini, 2011) were applied to quantify the lag effects of discharge, GPP, NEP and RE on DOC in each site (C2[S-forest], C4[S-mire], C6[M-mix] and NT[L-mix]) separately. DLMs are linear regressions between weighted lagged values of independent variables and dependent variables. In our case, we assumed that lag times are long, and the values of the weights were specified using polynomial transformations of lags of the independent variables by building so-called cross-basis functions. In DLMs, fourth-degree polynomial cross-basis functions were built for each factor GPP, RE and NEP and second-degree for discharge. Then, DOC variations were predicted by linear combinations of the cross-basis of each factor. We used explorative analysis to determine the optimal length of the lags. Lag time for each variable was determined by the results of wavelet coherence analysis and adjustments of DLMs, 0–7 days lag time was chosen for discharge and 4–30 days for GPP and RE. Since our variables exhibit an annual cycle, we also added year as a factor variable and did not consider longer time lags. This part was done using the 'DLNM' package (Gasparrini, 2011) in R (R Core Team, 2019). The Akaike information criterion (AIC) and explanatory power (R^2) were used to select the best DLM model across all sites (Table 3). Finally, the distributed lag models (DLM 1–6) applied across all sites were defined as follows:

$$DOC = \beta_1 DIS_{lag} + \alpha Year \tag{1}$$

$$DOC = \beta_1 GPP_{lag} + \alpha Year \tag{2}$$

$$DOC = \beta_1 RE_{lag} + \alpha Year \tag{3}$$

$$DOC = \beta_1 NEP_{lag} + \alpha Year \tag{4}$$

$$DOC = \beta_1 DIS_{lag} + \beta_2 GPP_{lag} + \alpha Year \tag{5}$$

$$DOC = \beta_1 DIS_{lag} + \beta_2 GPP_{lag} + \beta_3 RE_{lag} + \alpha Year \tag{6}$$

where β is the lag effect of discharge (DIS), GPP, RE and NEP on DOC concentrations, DIS_{lag}, GPP_{lag}, RE_{lag} and NEP_{lag} are the mean cross-basis of discharge, GPP, RE and NEP during their lag times, respectively. We also tested the different effects caused by discharge, baseflow and direct runoff on DOC variations by DLM 1 across the sites, and the results showed that DOC is more related to discharge than baseflow or direct runoff (Table S2). Therefore, discharge was set as the effect of hydrology to DOC variations in DLM 5–6.

3 | RESULTS

3.1 | Prediction of dissolved organic carbon by principal component regression model

We chose the first six components as the input variables into the PCR model. When applied to the training set, the DOC values from PCR calibration produced accurate estimates, as can be seen from the high explanatory power values of the model (R^2 = 0.93) and low root-mean-square deviation (RMSD = 3.38). PCR model showed even better performance when applied to the testing set, proved by the high explanatory power (R^2 = 0.95), low RMSD (RMSD = 2.95) and low mean bias error (0.12) (Table S3).

Daily DOC concentrations were predicted by PCR model based on the real-time spectral absorbances measurements in the field. Across all the sites, DOC concentrations were usually more stable and lower in the snow cover period compared with the growing season (Table S4). During the snow melt period, sudden increases are visible each year in C2[S-forest], C6[M-mix] and NT[L-mix], while there was a clear decrease in C4[S-mire] (Figure 1a). C4[S-mire] had the highest mean DOC concentrations (33.83 ± 7.95 mg l^{-1}) and the lowest coefficient of variation (CV = 23.49%) across 2016 to 2018. The lowest mean DOC values (4.51 ± 2.85 mg l^{-1}) and highest CV (63.12%) was found in NT[L-mix] from 2018 to 2020. The mean values of DOC were 18.95 ± 6.36 mg l^{-1} in C2[S-forest] (CV = 33.58%) and 17.80 ± 5.57 mg l^{-1} in C6[M-mix] (CV = 31.31%) across 2016 to 2018, respectively (Figure 1b).

3.2 | Wavelet coherence analysis between dissolved organic carbon and environmental factors

There was temporal synchrony between discharge (Figure 2a) and DOC during 1 to 30 days and 4 to 30 days between GPP (Figure 2b), NEP (Figure 2c), RE (Figure 2d) and DOC. However, the temporal synchrony was not continuous, and it was mainly restricted to the growing
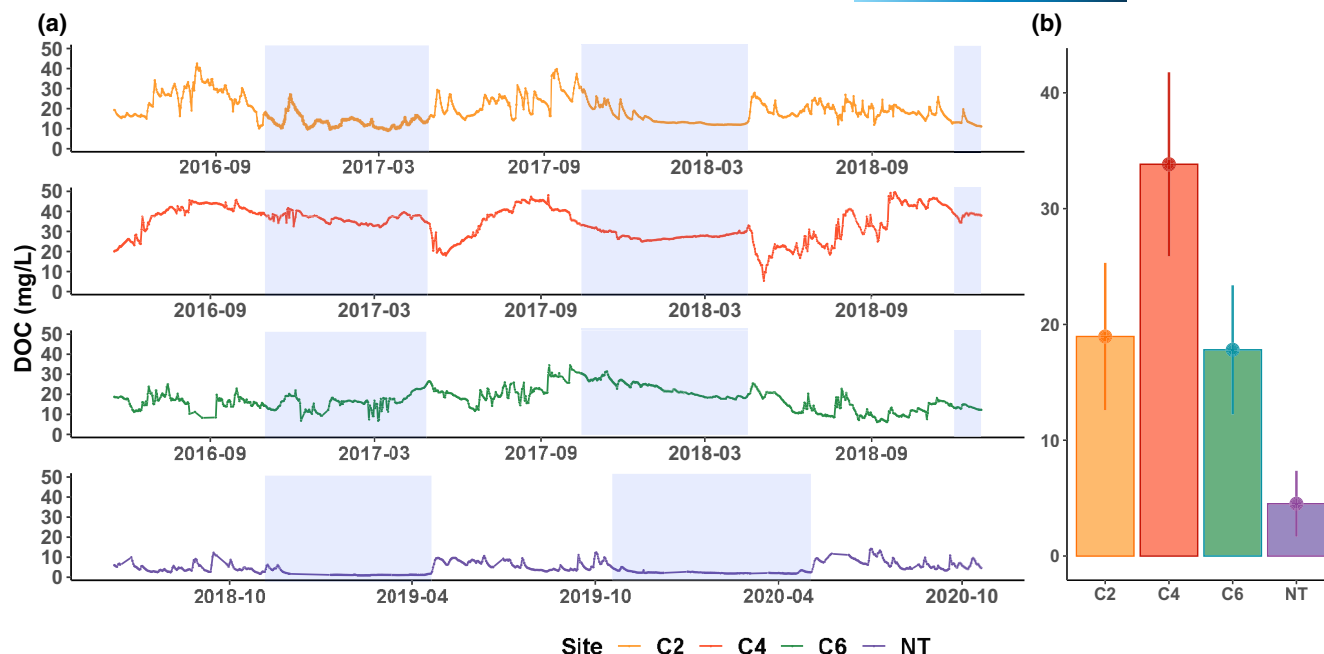
FIGURE 1 Daily dissolved organic carbon (DOC) variations (predicted by principal component regression model) across 2016 to 2019 in C2[S-forest], C4[mire], C6[mix], 2018 to 2020 in Nuorttiaapa Station (NT[mix]) (a) and the mean DOC concentration with standard deviation in each site during the whole experimental period (b). The different colours (orange, red, green and purple) represent different sites (C2[S-forest], C4[mire], C6[mix] and NT[mix]). The blue shades represent the snow cover periods

season. The temporal relations between discharge, GPP, NEP, RE and DOC concentration were unstable and varied between summers in different years and sites, and sometimes the temporal synchrony was not visible for all of them (Figure S2). The wavelet coherence analysis revealed that the environmental predictors affected DOC concentration and the effects always had a lag time (Figure 2).

3.3 | Relationships between dissolved organic carbon and environmental factors by distributed-lag linear models

The performances of DLMs improved with more time-lagged environmental factors, as shown by the increase of R^2 and decrease of AIC across all the sites. Among all the DLMs, DLM6 turned out to be the best one across all the sites, and it explained on average 62% of DOC variations. DLM6 in NT[L-mix] performed best with the highest R^2 (0.73), followed by C2[S-forest] ($R^2 = 0.69$) and C4[S-mire] ($R^2 = 0.65$), while C6[M-mix] had the lowest R^2 (0.42) (Table 2).

The cumulative responses of DOC concentration to a 10-unit increase of discharge were similar in C6[M-mix] (Figure 3c) and NT[L-mix] (Figure 3d). DOC concentration reached the first peak at 2 days lag, then decreased slightly and became the highest at 7 days lag (Figure 3c and d). In C2[S-forest], DOC concentration reached the peak at 3 days lag and then stayed relatively stable (Figure 3a). In C4[S-mire], DOC concentration responded negatively to the increase of discharge during the whole lag period (Figure 3b).

In C4[S-mire] (Figure 3f), C6[M-mix] (Figure 3g) and NT[L-mix] (Figure 3h), DOC concentration decreased immediately after a 10-unit increase of GPP and turned to revive slowly from 20, 25, 7 days lag, respectively. After a month lag, DOC concentration in NT[L-mix] (Figure 3h) returned to the original level but in C4[S-mire] (Figure 3f) and C6[M-mix] (Figure 3g) to lower than the initial level. Unlike the other three sites, DOC concentration in C2[S-forest] stayed relatively stable during the first half month, then started to rise and showed higher than the original level at a month lag (Figure 3e).

After a 10-unit increase of RE, DOC concentrations in C2[S-forest] (Figure 3i), C6[M-mix] (Figure 3k) increased immediately. In comparison, DOC concentrations in C4[S-mire] decreased slightly at the initial stage and turned to increase at 10 days lag (Figure 3g). These three sites then showed much higher than the original level at a month lag (Figure 3g,i,k). DOC variations in NT[L-mix] were much smaller than in the other sites, and DOC concentration was slightly higher than the initial level after a month lag (Figure 3l).

3.4 | Contributions of environmental variables to dissolved organic carbon concentrations

Comparing all the sites, the independent contribution of discharge and GPP to DOC concentrations in DLM6 behaved differently as catchment size increased (discharge increased whereas GPP decreased; Table 3). The separated contributions of the environmental factors to the DOC variations (in DLM6) across the experimental period are visualized in Figure 4. Unlike the other
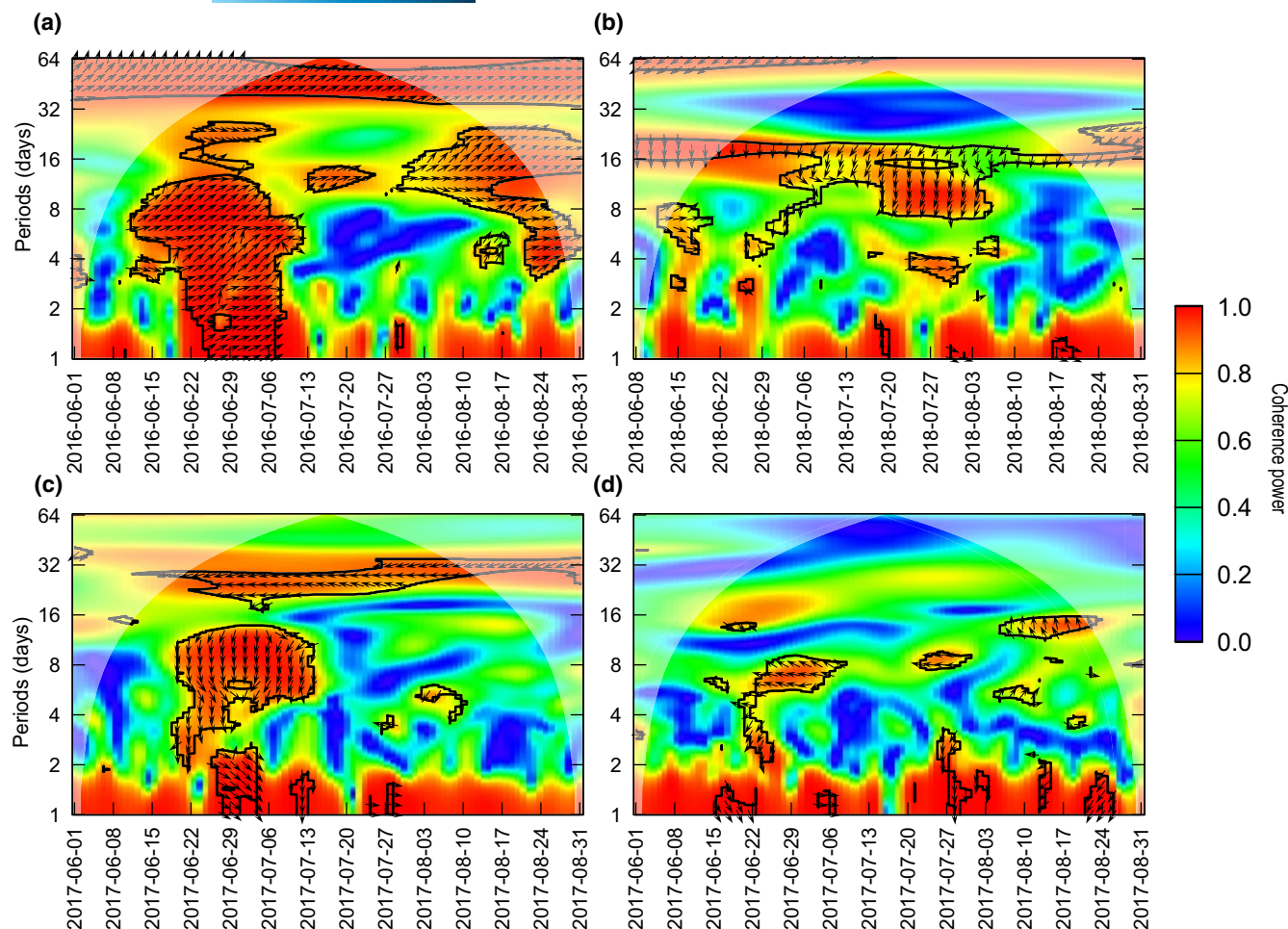
FIGURE 2 Wavelet coherence analysis between dissolved organic carbon concentrations and discharge (a), gross primary production (b), net ecosystem production (c), ecosystem respiration (d) in summer 2016, 2018, 2017 and 2017, respectively. The colours of power values range from blue (low values) to green (intermediate) to red (high values). The red parts inside the black border indicate significantly temporal coherence between the studied parameters ($p < .05$). The arrows show the leading and lagging relationship between the variables. The y-axis indicates the length of the time window in the wavelet coherence analysis (in days)

three sites, DOC variations in NT[L-mix] (Figure 4g) were controlled mainly by discharge leading to the negligible contributions of GPP and RE. In C2[S-forest] (Figure 4a), C4[S-mire] (Figure 4c) and C6[M-mix] (Figure 4e), RE contributed positively to DOC variations, but GPP and discharge acted differently among sites. GPP and discharge both contributed positively to DOC variations in C2[S-forest] (Figure 4a) but negatively in C4[S-mire] (Figure 4c), while in C6[M-mix] (Figure 4e), the former acted negatively and the latter positively. The dynamics of discharge, GPP and RE across years in each site are shown in Figure 4b,d,f,h. The mean values of discharge, GPP and RE during the experimental period were listed in Table 4.

4 | DISCUSSION

Our findings indicated that the contributions of GPP and discharge to aquatic DOC concentrations were closely related to catchment size. In small catchments (<1 km²), GPP dominated DOC variations,

whereas in bigger rivers (>1 km²), discharge is the most important driver of DOC concentrations. Additionally, GPP and discharge can have either positive or negative effects on DOC concentrations depending on the landcover type. RE was always positively related to DOC concentrations. The impacts of GPP, RE and discharge on DOC variations were always detected with delays from days to weeks.

The best distributed-lag linear model (DLM6) could explain on average 62% of aquatic DOC variations across the four catchments. In DLM6, discharge, GPP and RE accounted for 26%, 22% and 3% of DOC variations, respectively. The relationship between GPP and DOC concentration could be controlled by the priming mechanism (Guenet et al., 2010). While a meta-analysis showed only a small potential for a significant priming effect in aquatic ecosystems (Bengtsson et al., 2018; Catalán et al., 2015), here we mainly focused on the priming effect in the soil. As plants are usually nitrogen-limited in boreal ecosystems, they mostly rely on mycorrhiza for nutrient acquiring. Enhanced forest productivity results in more root exudates (more C) for mycorrhizal fungi to grow, increasing nutrients supply feeding back on trees (Heinonsalo et al., 2010; Pumpanen

TABLE 2 Performance of distributed-lag linear models (DLMs) showing the relationship between dissolved oraginc carbon (DOC) and environmental predictors across 2[S-forest], C4[mire], C6[mix] and NT[mix]

DLMs	DIS	GPP	RE	NEP	C2[S-forest] AIC	C2[S-forest] R²	C4[S-mire] AIC	C4[S-mire] R²	C6[M-mix] AIC	C6[M-mix] R²	NT[L-mix] AIC	NT[L-mix] R²
	Lag\day											
1. DOC = DIS_{lag} + year	0–7				6051.4	0.22	6374.8	0.25	5339.5	0.36	2693.6	0.72
2. DOC = GPP_{lag} + year		4–30			5464.7	0.52	5980.9	0.41	5337.2	0.28	3387.6	0.17
3. DOC = RE_{lag} + year			4–30		5415.3	0.55	5807.8	0.51	5352.8	0.26	3390.8	0.17
4. DOC = NEP_{lag} + year				4–30	5712.6	0.38	6187.3	0.27	5325.2	0.28	3405.0	0.15
5. DOC = DIS_{lag} + GPP_{lag} + year	0–7	4–30			5130.6	0.67	5778.1	0.53	5201.9	0.38	2621.4	0.72
6. DOC = DIS_{lag} + GPP_{lag} + RE_{lag} + year	0–7	4–30	4–30		5069.0	0.69	5531.9	0.65	5155.3	0.42	2608.6	0.73

Note: DOC concentrations were predicted by principal component regression (PCR) model; DIS means discharge.

et al., 2009). In most northern headwaters, the link between terrestrial and aquatic environments is dominated by riparian zone with high SOC and water content (Laudon & Sponseller, 2018; Lidman et al., 2017; McGlynn et al., 1999; Wen et al., 2020). Our results demonstrated that although the riparian organic C pool can continue to sustain DOC export at the present rate for several hundred years without supplement (Ledesma et al., 2015), new C input due to increased GPP does affect the export of DOC from riparian zone to aquatic systems.

In this study, we found that GPP dominated the DOC variations in small catchments (C2[S-forest] and C4[S-mire]) but not in the larger catchments. The importance of GPP was masked by discharge in medium- and large-scale catchments (C6[M-mix] and NT[L-mix]). The faded impact of GPP with increased catchment size may be due to several reasons. Firstly, as groundwater moves from uphill mineral soils through the riparian zone with large stores of SOM, major biogeochemical transformations occur within meters of the stream (Laudon & Sponseller, 2018). These transformations mean that the active area affecting the DOC export is limited and does not change as much with increased catchment size. Thus, with the same amount of DOC export, the dynamics of aquatic DOC concentrations must be more evident in small-scale catchments than in larger ones. Secondly, Tiwari et al. (2017) stated that biogeochemical transitions from small to mesoscale catchments are partially mediated by the increased relative contribution of deep groundwater inflow with increased drainage area. The principal hydrological pathway shifts from mainly surface flow paths in small streams to deeper organic-poor groundwater in larger-scale catchments. These changes in the flow path also explain why discharge masked the effect of GPP in C6[M-mix] and NT[L-mix]. Finally, during the spring flood which typically occurs in the months of May and June over 60% of the annual DOC flux may occur (Mann et al., 2012). Large-scale rivers have a larger contribution of their flow from groundwater and their DOC concentrations are buffered against changes of DOC inputs from soils (Shanley et al., 2002; Strohmenger et al., 2021). These differences further explain why the proportion of DOC driven by GPP decreases when catchment size increase.

The roles of GPP in controlling DOC variations among sites were also complicated. We would have expected a mostly positive contribution of GPP on DOC concentrations due to the priming effect. The idea is that increased GPP leads, with a lag, to higher microbial biomass and, with a longer lag, to an increase in DOC production. However, apart from C2[S-forest] catchment, our results showed GPP was negatively correlated with DOC contents in C4[S-mire], C6[M-mix] and NT[L-mix], which does not support our hypothesis that GPP has a positive priming effect on aquatic DOC concentrations. Especially in C-rich ecosystems, increases in labile C supply may lead to increases in microbial biomass that consumes most organic materials. In these ecosystems, DOC consumption by an increasing microbial biomass exceeds the production of DOC (Qiao et al., 2014). Ding et al. (2018) emphasized the role of the C/N ratio for priming and demonstrated that priming often requires sufficient N supply. When the two small-scale catchments (C2[S-forest] and
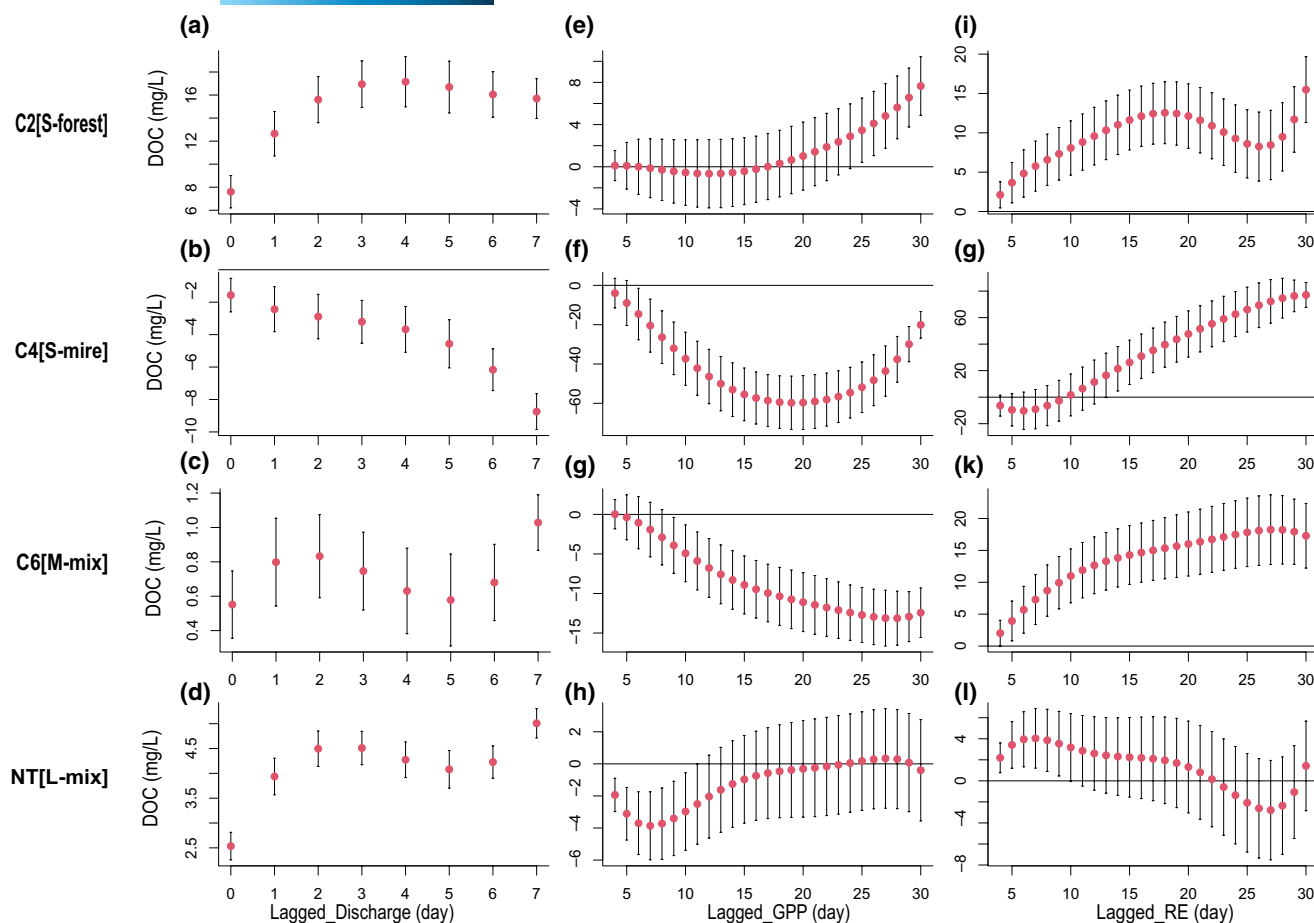
FIGURE 3 Cumulative lag-response of DOC concentrations to 10-unit increase in discharge (a–d), gross primary production (GPP) (e–h), ecosystem respiration (RE) (i–l) in C2[S-forest], C4[S-mire], C6[M-mix] and NT[L-mix] based on DLM6. The horizontal line at 0 represents the mean DOC concentrations

C4[S-mire]) were compared, the effect of forest GPP on catchment DOC was positive, while that of mire was negative. Laudon and Sponseller (2018) confirmed that the landscape always plays a vital role in downstream biogeochemical patterns. Liu et al. (2017) found a positive linear relationship between C input and priming, that priming increased from negative or no priming at low C input to strong positive priming at high C input. In our case, when comparing these two small catchments, the mean GPP values in C2[S-forest] (3.01 g Cm^{-2} d^{-1}) are much higher than in C4[S-mire] (0.78 g Cm^{-2} d^{-1}), which may also explain the positive contribution of GPP to DOC variations in C2[S-forest] and the negative contribution in C4[S-mire]. Bastida et al. (2019) suggested that priming effects tend to be negative in more mesic sites with higher SOC contents and positive in more arid locations with low SOC contents. Our results also support the association between SOC content and priming. Moreover, the large difference of aboveground plant biomass between C2[S-forest] and C4[S-mire] could be the third reason, as it has been noted that the level of rhizosphere priming effect is positively correlated with aboveground plant biomass (Huo et al., 2017). In our case, C2[S-forest] was totally dominated by forest while C4[S-mire] was covered mainly by wetland, and there is much more aboveground biomass in the former than the latter.

Unlike GPP, RE was consistently positively correlated with DOC exports. These correlations fit into the idea that below-ground microbial activity leads to decomposition of complex organic substances into monomers which form the bulk of the DOC (Schimel et al., 1994). The catchment essentially stores the produced DOC in soil water and on soil surfaces, leading to the continued accumulation of DOC until the next precipitation event occurs and flushes out the stored DOC. Hence, low hydrological connectivity implies a delay of DOC export such that the DOC we see today in the stream may often be the DOC produced a while ago (Wen et al., 2020).

The importance of discharge as a carrier of DOC from terrestrial ecosystems cannot be ignored. Approximately 80% of watersheds in the USA and France show significant relationships between the stream DOC concentration and discharge, either positive or negative. Whereas, the remaining watersheds show negligible concentration change with discharge (Wen et al., 2020). In our case, discharge alone could contribute from 9.9% to 68.7% of DOC variations across years in both positive and negative patterns. Dawson et al. (2002) found that the relationship between discharge and DOC can be strengthened if the data was split seasonally, and then discharge could predict from 58% to 81% of DOC values in different seasons. Clark et al. (2007) showed that discharge could explain 72% of DOC
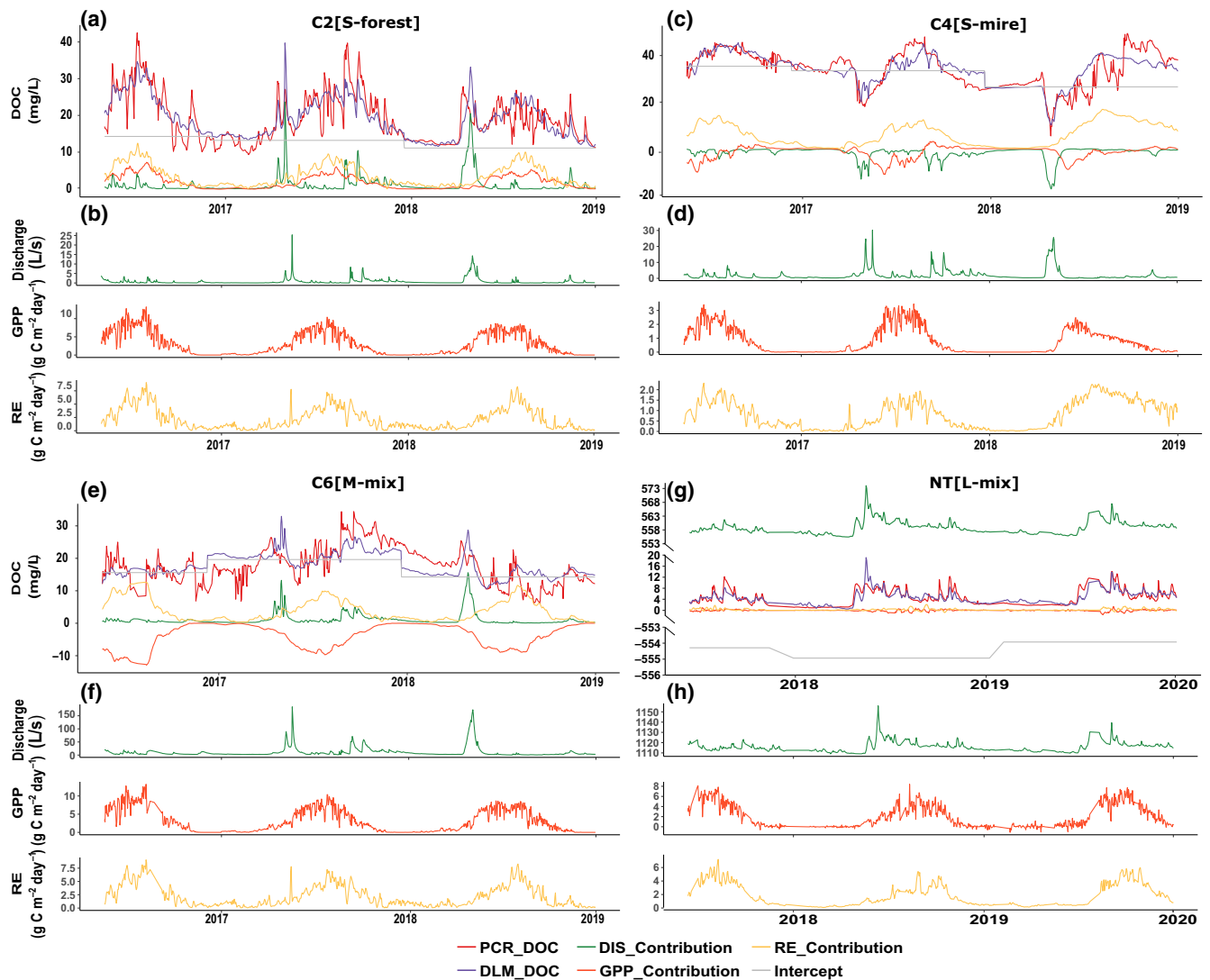
FIGURE 4 The independent contributions of discharge, gross primary production (GPP) and ecosystem respiration (RE) to dissolved organic carbon (DOC) based on DLM6 model in C2[S-forest] (a), C4[S-mire] (c), C6[M-mix] (e), NT[L-mix] (g). PCR_DOC was predicted by the PCR model as the response of DLM6, DLM6 model predicted DLM_DOC; DIS_Contribution was the independent contribution of discharge in DLM6 model; GPP_Contribution was the independent contribution of GPP in DLM 6 model; RE_Contribution was the independent contribution of RE in DLM6 model. Continuously measured environmental predictors (discharge, GPP and RE) across years in C2[S-forest] (b), C4[S-mire] (d), C6[M-mix] (f), NT[L-mix] (h)

TABLE 3 Contributions of time-lagged environmental variables (discharge, [GPP] and ecosystem respiration [RE]) to dissolved organic carbon (DOC) concentrations across four catchments based on DLM6

Site	Size (km²)	Discharge		GPP		RE		Year	R^2
C2[S-forest]	0.14	9.9%	+	53.2%	+	2.5%	+	3.6%	69.3%
C4[S-mire]	0.19	14.3%	−	30.8%	−	3.9%	+	15.6%	64.5%
C6[M-mix]	1.3	15.0%	+	2.2%	−	6.1%	+	18.6%	41.9%
NT[L-mix]	40	68.7%	+	1.0%	ns	1.1%	ns	2.3%	73.1%

Note: '+'\'−' indicate positive or negative relations with DOC concentrations, respectively.

concentrations during autumn storm events, but they also showed a poor relationship during other seasons. Instead of grouping data by season to remove this component of the annual variation (Clark et al., 2007; Dawson et al., 2008), in our DLM models, we included 'year'

as a factor variable without splitting data to keep the continuity and concerned the delay effect of discharge on DOC concentration.

It has been well documented that the hydrological connectivity to the stream versus the distribution of SOC ultimately dictates the

TABLE 4 Mean values and ranges of discharge, gross primary production (GPP) and ecosystem respiration (RE) during the experimental period across four sites

Site ID.	Size (km²)	Discharge (l s⁻¹)		GPP (gC m⁻² d⁻¹)		RE (gC m⁻² d⁻¹)	
		Mean	Range	Mean	Range	Mean	Range
C2[S-forest]	0.14	0.73 ± 1.67	0–25.36	3.01 ± 3.11	0–13.22	2.19 ± 2.00	0–9.05
C4[S-mire]	0.19	1.87 ± 3.24	0.06–30.23	0.78 ± 0.91	0–3.47	0.79 ± 0.66	0–2.32
C6[M-mix]	1.3	10.88 ± 19.95	0.41–184.09	2.94 ± 3.10	0–12.59	2.13 ± 1.97	0–8.95
NT[L-mix]	40	1115.71 ± 4.99	1109–1156	1.94 ± 2.25	0–8.40	1.83 ± 1.65	0.08–7.21

dynamic of DOC concentrations in soil and stream water, leading to different discharge-DOC relationships (Covino, 2017; Wen et al., 2020). In our study, the discharge–DOC relationship shifted from dilution pattern in wetland dominated catchment (C4[S-mire]) to flushing pattern in forest and mixed catchment (C2[S-forest], C6[M-mix] and NT[L-mix]), which is in line with Laudon et al. (2011). The transport of DOC losses from the bulk soil has been linked to hydrological processes in response to precipitation events and changing flow paths through different soil horizons containing contrasting amounts of organic matter (Dawson et al., 2008; Hinton et al., 1998). The contrasts in hydrological functioning of these sites (C4[S-mire] vs. C2[S-forest], C6[M-mix] and NT[L-mix]) is demonstrated by different flow paths. In wetland-dominated streams (C4[S-mire]), event water at the rising stage of hydrograph runoff as overland flow due to frozen wetland surface or saturation, leading to a dilution in DOC concentrations (Laudon et al., 2004). In the other three sites (C2[S-forest], C6[M-mix] and NT[L-mix]), SOC is enriched in uplands, and most of the runoff from forested areas reaches the stream via subsurface flow paths carrying new activated SOC (Bishop et al., 2004). Therefore, DOC concentrations were high at high flow.

Seasonal variations of DOC concentrations were also observed across the sites in our study. DOC concentrations were usually more stable and lower during the snow cover period compared with the growing season. We assumed that there are several mechanisms driving the low wintertime DOC concentrations. Firstly, DOC sources (litterfall) are relatively limited during winter which partially caused the low wintertime DOC (Jutras et al., 2011; Liu et al., 2014). Worrall et al. (2004) found a pulse of DOC from senescing vegetation at the end of the growing season, which further supports this idea. Secondly, the lower temperatures lead to decreased biological activity, lower decomposition of available organic matter and lower solubility of DOC in wintertime (Dawson et al., 2008). Finally, frozen soil during the wintertime may have reduced the movement of terrestrial organic matter from upland zones and riparian areas to streams, consequently resulting in large fluctuations of DOC concentrations during the snow melt season. (Pacific et al., 2010).

This study demonstrated the importance of terrestrial productivity interacting with discharge in controlling the variations of DOC concentrations in boreal catchments. Forest productivity dramatically promotes DOC contents in small-sized catchments (<1 km²), while the importance of GPP is masked by discharge with increased catchment size. Moreover, our statistical analysis indicated that

priming and the land cover (proportion of forest) should be included in process-based DOC models. Overall, our investigation revealed that terrestrial greening and altering hydrology following climate change may have a major impact on DOC concentration in aquatic ecosystems, as well as reaffirming the importance of DOC fluxes in controlling ecosystem C budgets, which is generally disregarded.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study will be openly available in DRYAD repository at https://doi.org/10.5061/dryad.wpzgmsbp9.

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