Global Change Biology

Global Change Biology (2015) **21**, 4425–4435, doi: 10.1111/gcb.13031

Climate and landscape influence on indicators of lake carbon cycling through spatial patterns in dissolved organic carbon

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Abstract

Freshwater ecosystems are strongly influenced by both climate and the surrounding landscape, yet the specific pathways connecting climatic and landscape drivers to the functioning of lake ecosystems are poorly understood. Here, we hypothesize that the links that exist between spatial patterns in climate and landscape properties and the spatial variation in lake carbon (C) cycling at regional scales are at least partly mediated by the movement of terrestrial dissolved organic carbon (DOC) in the aquatic component of the landscape. We assembled a set of indicators of lake C cycling (bacterial respiration and production, chlorophyll a, production to respiration ratio, and partial pressure of CO_2), DOC concentration and composition, and landscape and climate characteristics for 239 temperate and boreal lakes spanning large environmental and geographic gradients across seven regions. There were various degrees of spatial structure in climate and landscape features that were coherent with the regionally structured patterns observed in lake DOC and indicators of C cycling. These different regions aligned well, albeit nonlinearly along a mean annual temperature gradient; whereas there was a considerable statistical effect of climate and landscape properties on lake C cycling, the direct effect was small and the overall effect was almost entirely overlapping with that of DOC concentration and composition. Our results suggest that key climatic and landscape signals are conveyed to lakes in part via the movement of terrestrial DOC to lakes and that DOC acts both as a driver of lake C cycling and as a proxy for other external signals.

Keywords: aquatic carbon cycling, boreal, climate, CO_2, dissolved organic carbon, fluorescence, lake, landscape, PARAFAC, Respiration, spatial structure

Received 4 May 2015 and accepted 14 June 2015

Introduction

Climate, precipitation, and atmospheric deposition patterns are changing in northern ecosystems, with major consequences for carbon (C) cycling in the aquatic component of the landscape. Such changes have led, for example, to increases in water temperature or loss of terrestrial organic carbon to aquatic ecosystems (Freeman *et al.*, 2001; Monteith *et al.*, 2007; Zhang *et al.*, 2010), resulting in enhanced carbon degradation in the sediments and surface waters of continental watersheds (Gudasz *et al.*, 2010; Lapierre *et al.*, 2013; Marotta *et al.*, 2014). The pathways underlying these patterns, however, remain unclear because patterns and processes are seldom studied simultaneously at large spatial scales across key environmental and climate gradients (Hanson *et al.*, 2014), and the responses of lake C cycling to individual drivers in experimental studies are hard to capture in survey studies across large spatial scales. Improved understanding of the factors that link environmental patterns to the aquatic C cycle in natural environments should enhance understanding and prediction of the direct and indirect impacts of changing climate on the aquatic ecosystem functioning.

The pathways linking landscape properties and lake functioning may in part be influenced by climate patterns, as suggested by the spatial coherence that has been observed in lake chemistry at different spatial scales. For example, temporal trends in climate have been shown to drive regional synchrony in lake properties such as primary productivity, nutrients, and dissolved organic carbon (DOC) concentrations (Baines *et al.*, 2000; Kling *et al.*, 2000; Pace & Cole, 2002). This coherence is stronger among than across lake chains (Sadro *et al.*, 2012), suggesting that connected lakes are

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more synchronous as they directly share common sources of various elements at the catchment level. Spatial coherence, however, has also been observed for unconnected lakes at large spatial scales (Weyhenmeyer, 2004; Blenckner *et al.*, 2007), and coherence appears to increase with lake order but decay as a function of latitudinal distance (Soranno *et al.*, 1999; Magnuson *et al.*, 2004). These examples show that beyond direct delivery of elements from one site to another, there is some form of spatial dependency in lake functioning, where nearby lakes are exposed to similar climate that presumably also translates into similar land cover and hydrology that dictates the flow of elements from land to water.

If the pathways underlying the temporal synchrony of nearby lakes respond to climate trends, spatially structured patterns in lake C cycling would be expected across climate gradients. There is little evidence of such patterns, however, because large-scale surveys and monitoring programs of lake water have traditionally been performed in response to acid rain or eutrophication issues, with measurements relevant to carbon cycling having a secondary role. As a consequence, the few large-scale studies on lake C cycling have typically focused on a single response variable, and individual evidence may appear contradictory. For example, concentrations and fluxes of CO_2 are reported to increase in large lakes at latitudes ranging from 0 to 60°N as mean water temperature decreased (Alin & Johnson, 2007), but concentrations of CO_2 in more than 7000 lakes around the world are independent of water temperature at the time of sampling (Sobek *et al.*, 2005). The latter were, however, weakly linked to mean annual temperature at the sampling site (Sobek *et al.*, 2005), and altitudinal and latitudinal gradients in climate and land cover have been related to dissolved organic carbon and nitrogen concentrations in lakes (Sobek *et al.*, 2007; Weyhenmeyer & Karlsson, 2009; Sadro *et al.*, 2012). Together, these findings are consistent with lake synchrony studies in that they suggest that large lakes (Alin & Johnson, 2007) respond more coherently to climate gradients that an heterogeneous ensemble of lakes (Sobek *et al.*, 2005) and that underlying climate settings and their potential effect on land cover may be more important than ambient temperature in explaining large-scale patterns in C cycling.

There is a temporal scale mismatch, however, when relating spatial patterns in static, multiyear climate averages and landscape properties to highly dynamic lake nutrient and gas concentrations as well as biological rates of production and degradation of organic carbon, which may vary at scales as short as hourly. As large-scale spatial surveys necessitate significant time investment on the field, integrating large data sets on lake processes typically involves interseasonal and interannual data with possible confounding temporal effects. There is thus always a fraction of this temporal signal that cannot be captured by static climate or landscape predictors when exploring large spatial scale patterns in lake processes. On the other hand, the presence of significant relationships between those static predictors and dynamic lake processes would imply that there is more variation among lakes than within lakes when large enough environmental gradients are considered, and thus that the spatial patterns in climate and landscape properties may align with, and perhaps explain the spatial patterns in lake carbon cycling. As climate and landscape properties (e.g., temperature, precipitation, topography) are highly structured in space over regional to global scales (IGBP-DIS, 1998; Hansen *et al.*, 2003; Hijmans *et al.*, 2005), a strong effect of these predictors should induce spatially structured patterns in lake C cycling at large spatial scales. Regional patterns have indeed been observed for nutrients and DOC concentrations in response to a combination of climate and landscape properties (Sobek *et al.*, 2007; Cheruvelil *et al.*, 2008; Seekell *et al.*, 2014). Moreover, there is evidence that regional patterns in DOC are reflected in lake C cycling due to its role as a substrate for degradation and as a proxy of the loadings of other terrestrial material, as observed in the surface water CO_2 concentrations (Lapierre & del Giorgio, 2012). Such regional structure suggests that static landscape and climate variables may be good predictors of dynamic lake processes at large spatial scales due to their role in constraining concentrations of carbon and nutrients within a relatively narrow range among lakes with comparable land cover. A major gap that remains unaddressed in the previous studies is how spatial patterns in DOC, both in terms of concentration and composition, may affect the large-scale spatial structuring of lake C cycling.

Here, we hypothesize that key lake properties and processes involved in C cycling in lake surface waters follow spatially structured patterns in response to large-scale gradients in climate and landscape properties and that DOC concentration and composition play a central role in conveying this response. To explore these hypotheses, we first determined the patterns in a set of indicators of lake C cycling and DOC across climate and landscape gradients. Spatial autocorrelation analyses were then performed on a large field data set composed of lake biogeochemical variables, combined with publicly available climatic and landscape properties to compare the spatial structure in lake processes to that of its potential drivers. Finally, we explored the individual and overlapping effect of climate, landscape, and DOC on this set of indicators of lake C cycling, using redundancy analyses and variance partitioning.

Materials and methods

Sampling

Over the course of four years, we surveyed lakes situated in seven distinct biogeographic regions of Québec, Eastern Canada (Fig. 1a), allowing for a multiscale comparison where the distance between lakes ranged from less than one kilometer to up to 1300 km. From 2009 to 2012, 239 individual lakes were sampled once or twice between May and August for a total of 280 observations; means have been used for multiple lake samples.

Water was sampled for chemical and gas analyses from the deepest measured point in the lake at 0.5 m from the surface. Water temperature was measured from a multiprobe (Yellow Springs Instruments, Yellow Springs, OH, USA). A portion of the water was filtered through 0.45 μm initial pore size cartridge filters (Sarstedt, OH, USA) and stored in 40-ml acid-washed glass vials for subsequent dissolved organic carbon (DOC) and optical measurements. Unfiltered water was added to 60-ml acid-washed glass vials for subsequent nutrient analyses. All samples were immediately stored in the cold and dark and kept under those conditions until laboratory analyses.

Biological and chemical analyses

DOC concentrations were measured on an OI 1010 TOC-TIC (Aurora, College Station, TX, USA) analyzer following sodium persulfate digestion. Total phosphorus (TP) was analyzed spectrophotometrically after persulfate digestion. Total nitrogen was analyzed as nitrates following alkaline persulfate digestion and measured on an Alpkem Flow Solution IV autoanalyzer.

Optical analyses

We used a combination of chemical (DOC concentration, see above) and optical analyses to explore the dynamics in the concentration and composition of DOC, respectively. A PARAFAC model (Stedmon *et al.*, 2003) was fit to identify and quantify fluorescence components, that is, groups of DOC that had similar optical properties and distribution across the lakes and that presumably shared common sources and sinks. Fluorescence intensity was measured on a Shimadzu RF5301 PC (Shimadzu, Kyoto, Japan), across excitation wavelengths of 250–450 nm (5 nm increments) and emission wavelengths of 280–600 nm (2 nm increments). Absorbance measurements were performed on a UV-visible Ultrospec 2100 spectrometer (Biochrom, Cambridge, UK) using a 2-cm quartz cuvette and corrected by subtracting nanopure water blanks. All measurements were performed within 2 weeks after collection. Fluorescence data were first corrected for inner-filter effect and standardized to Raman units using the FDOMCORR 1.6 toolbox (Murphy *et al.*, 2010) in MATLAB (MathWorks, Natick, MA, USA). The model was then fit on corrected data using the DOMFLUOR 1.7 toolbox (Stedmon & Bro, 2008). Composition of the DOC pool was assessed as the percent that each component contributed to the total fluorescence in a given sample. Additional details on the modeling procedure are provided in Lapierre & del Giorgio, (2014).

Here, we use the PARAFAC model described previously in the boreal biome (Lapierre & del Giorgio, 2014; Stubbins *et al.*, 2014). We summarize the main points emerging from those studies that are useful to interpret the results from this study. The model, based on 1349 samples from temperate and boreal aquatic environments, identified six different fluorescence components. Components 1–3 (C1–C3) were associated with biologically refractory and humic- and fulvic-like materials with high C : N ratios; component C3 was extremely sensitive

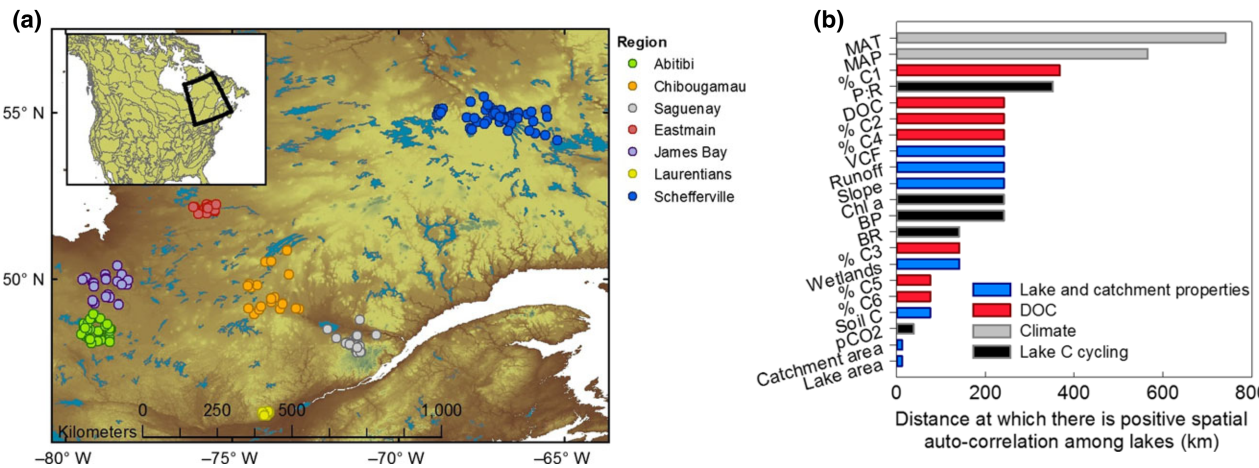

Fig. 1 The spatial structure in the measured variables across a large boreal landscape. (a) Distribution of the sampling sites across Québec biogeographic regions. (b) Spatial scales of variation of climate, landscape, and lake properties depicted as the distance at which spatial autocorrelation becomes nonsignificant. Climate and landscape variables are long-term mean annual averages. Percentage of components C1–C6 represent the percent contribution of a specific DOM group to total fluorescence in a sample; they are collectively an index of DOM composition. VCF stands for vegetation continuous field; it denotes vegetation density in the catchment.

to photochemical degradation. Components C4–C6 were associated with more biologically labile, freshly produced DOM with higher proportions of N (Stubbins *et al.*, 2014). Component C6 in particular had a fluorescence signature typical of protein-like material and was the strongest driver of DOC biological degradability across hundreds of boreal lakes, rivers, and wetlands (Lapierre & del Giorgio, 2014).

Indicators of lake C cycling

We explored 'lake C cycling' through key variables related to the production and degradation of organic carbon in lakes: bacterial respiration (BR), chlorophyll a (Chl a), partial pressure of CO_2 (pCO_2), bacterial production (BP), and production to respiration ratio (P : R); carbon storage would have been a desirable variable to include but was not available for the vast majority of the sampled lakes. Chl *a* concentrations were measured spectrophotometrically, in duplicates, following filtration on Whatman (GF/F) filters and hot ethanol (90%) extraction. Filters were sonicated prior to extraction. For all variables measured in duplicate, we used the mean value. Bacterial and total respiration (BR and TR, respectively) were determined from changes in oxygen concentrations during *in vitro* incubations in the dark and at site temperature. Oxygen was measured from filtered (2.7 μm) and unfiltered water (for BR and TR, respectively) in 500-ml sealed erlenmeyers using optical sensors (Fibox 3, PreSens, Regensburg, Germany) following Marchand *et al.* (2009) . The P : R ratio was measured as the ratio of Chl *a*-derived rates of PP to rates of TR: Estimates of pelagic PP rates were estimated from Chl *a* concentrations following Del Giorgio & Peters (1994). This transformation allowed us to convert algal biomass to rates of production in the same units as total respiration and thus to explore the patterns in production to respiration (P : R) ratios across lakes. Bacterial production (BP) was based on rates of radioactive leucine incorporation (Marchand *et al.*, 2009) in standardized conditions. Finally, surface water CO_2 concentration (expressed as the partial pressure of CO_2, $p$$CO_2$, in μatm) was measured *in situ* using an EGM-4 infrared gas analyzer (PP-systems, Boston, MA, USA), after equilibration by pumping water through a Liqui-Cel Mini Module (Charlotte, NC, USA) contactor membrane.

GIS analyses

The surface and catchment areas, as well as the elevation of the sampled lakes, were determined using ARCGIS v.10.1 (ESRI, Redlands, CA, USA) and the DEM derived from 1 : 50 000 maps. The sources of complementary geographic data are presented in Table 1. Here, we focus on long-term averages of climate and landscape properties, which we consider as temporally static for the time frame of this study, to emphasize the landscape-level response of the indicators of lake C cycling across large geographic gradients.

Statistical analyses

We scaled the variation in lake C cycling and its drivers using Moran's index (Moran's I) for spatial autocorrelation to quantify how similar lakes are to each other within a given distance interval in relation to any given variable (Fortin & Dale, 2005). Briefly, using the freeware application SAM 4.0 (Rangel *et al.*, 2010), we built a correlogram for each variable that represented average Moran's I for 20 groups (comprising between 2842 and 2844 pairs of lakes) of sites that were within a certain distance interval from one another. This number of groups, as well as the distance interval for each group, was determined by the software as a function of the number of sites and average distance between sites to optimize the distribution of inter-lake distance among groups. For each variable, we determined the maximum distance at which there was positive spatial autocorrelation between two lakes, that is, the average distance at which two lakes tend to become completely independent with respect to the variable in question. There were thus only 20 possible values for this distance. Considering the number of variables studied and that most variables were not spatially autocorrelated beyond a few hundred kilometers, Moran's I became nonsignificant at the same distance for several variables, resulting in the quantized distribution shown in Fig. 1b. Using different number of groups slightly changed the estimated distance at which spatial autocorrelation disappears, but did not change the overall patterns reported below.

Spatial patterns in different groups of variables were explored with principal component analyses, using JMP 10 software (SAS institute, Cary, NC, USA). The apparent regional patterns in the RDAs were validated using a linear discriminant analysis (LDA, performed in JMP 10) with predetermined regions (illustrated in Fig. 1a) as categories; the LDA results are derived from a Jackknifed classification matrix. Indicators of lake C cycling and DOC concentration and composition were then placed along climate gradients, as a function of mean annual air temperature (MAT) at the sampling sites. Because we did not anticipate any particular shape for these relationships, we ran a kernel smoother (Gaussian weighting function, alpha = 0.8, using JMP 10) on the normalized data to allow the fit to capture potential nonlinearity. A comparable approach has been used to detect nonlinear relationships between TP and Chl *a* across different regions in US lakes (Filstrup *et al.*, 2014).

Table 1 Source of the geographic data analyzed in this study

Variable	Source
Mean annual temperature, precipitation	WordClim, (Hijmans *et al.*, 2005)
Vegetation density in the catchment	Global forest monitoring program, (Hansen *et al.*, 2003)
Soil carbon content	The Atlas of the Biosphere, (IGBP-DIS, 1998)
% wetlands in the catchment	Geobase (Geobase, 2009)
Lake area	Geobase (Geobase, 2009)
Catchment area	Geobase (Geobase, 2009)
Catchment slope	Geobase (Geobase, 2009)
Mean annual runoff	UNH/GRDC, (Fekete *et al.*, 2000)

We explored how different categories of predictors explain the indicators of lake C cycling with redundancy analysis (RDA), using the VEGAN package (Oksanen *et al*., 2013) in R 3.0 (R Core Team, 2013). Variables were log-transformed to meet normality assumptions when needed. We performed forward selection until the model could not be improved to avoid overfitting and ensured that residuals of the selected models were normal. There were substantial amounts of co-linearity among the different predictors; thus, we performed variance partitioning to assess overlapping explanatory power by groups of predictors on predicted variables (Legendre & Legendre, 1998; Peres-Neto *et al*., 2006). Beyond representing a statistical issue, overlap in the variance explained can also be interpreted as different predictors sharing common underlying drivers in the landscape for survey studies. With prior knowledge of the theoretical links between variables, hypotheses can be tested. In this study, we explored how a proximal driver of indicators of lake C cycling (i.e., DOC concentration and composition) is likely integrating an indirect effect of climate and landscape properties around the studied lakes, as denoted statistically by the overlap in the variance explained by those three groups of variables on different processes.

Results

Spatial structure in climate, landscape, and lake properties

The variables studied here spanned a wide range in spatial structure. At the upper end of the spectrum, mean annual temperature (MAT) and precipitation (MAP) tended to vary at a very large scale with lakes spatially autocorrelated within a radius of 741 and 566 km of each other, respectively, indicating that MAT at any given site can be partly predicted from that observed at any other site situated within 741 km. At the other end of the spectrum were lake and catchment area, which varied at a local scale, that is, any pair of lakes situated more than 12 km apart tended to be completely independent in terms of those variables. Varying at an intermediate, regional scale were indicators of lake C cycling, for which lakes tended to be spatially autocorrelated at distances ranging between 38 and 351 km (Fig. 1b). Production to respiration (P : R) ratio and CO_2 concentration (expressed as pCO_2, see Materials and methods) occupy the upper and lower range, respectively, while bacterial respiration (BR), bacterial production (BP), and chlorophyll a (Chl a) were spatially autocorrelated for lakes situated within around 200 km from each other.

Concentration and composition of DOC were spatially structured at comparable regional scales. In particular, total DOC and humic- or fulvic-like components of DOC (C1–C4, see Material and Methods) tended to be spatially autocorrelated over larger scales than protein-like components, C5 and C6, which are typically considered labile and freshly produced. For those typically labile DOC groups, autocorrelation disappeared at a scale (75 km), comparable to the structure of soil organic carbon content in the top 50 cm (Fig. 1b). Landscape properties, such as vegetation density in the catchment (VCF, see Table 1), catchment slope (Slope), mean annual runoff (Runoff), and percent area covered by wetlands (% Wetlands), were located between those different DOM groups and tended to be autocorrelated at scales of 141–241 km (Fig. 1b).

Regional patterns in climate, landscape and, lake properties

The spatial autocorrelation in the measured variables resulted in strong regional patterns in terms of climate and landscape properties (Fig. 2a), DOC concentration and composition (Fig. 2b), and indicators of lake C cycling (Fig. 2c). Beyond visual examination, a linear discriminant analysis reveals that 93%, 87%, and 72% of the lakes were classified in the correct a priori determined sampling ‘region’ (see Fig. 1a) based on climate and landscape properties (Fig. 2a), DOC (Fig. 2b) and lake C cycling (Fig. 2c), respectively. Figure 3 further shows that the variability in DOC and indicators of lake C cycling is typically smaller within than among regions and that the different regions tend to align continuously, albeit nonlinearly along climate gradients; MAT systematically explained more than ambient water temperature. The shape of the relationships for the indicators of lake C cycling matched well with that of lake DOC, and all tended to shift around a few degrees from the freezing point. Both the concentration and the composition of the DOC changed along climate gradients as different fluorescence components varied in diverging directions (Fig. 3).

The effect of climate, landscape, and DOC on indicators of lake C cycling

Long-term averages in climate and landscape properties alone explained 32% of the variation in a multilinear combination of the five measured indicators of lake C cycling (Fig. 4a), based on an analysis of redundancy (RDA). Not all variables included in Fig. 1b were significant predictors, but long-term climate average (MAT and MAP) and several landscape properties (catchment slope, soil C, vegetation in the catchment, catchment area) had significant effects. There was very little variance explained by landscape properties that did not overlap with that explained by climate (Fig. 4a). Ambient water temperature at the moment of sampling was a weak but significant predictor of BP ($P = 0.002$), PP ($P < 0.001$), and P : R ($P < 0.001$), but did not

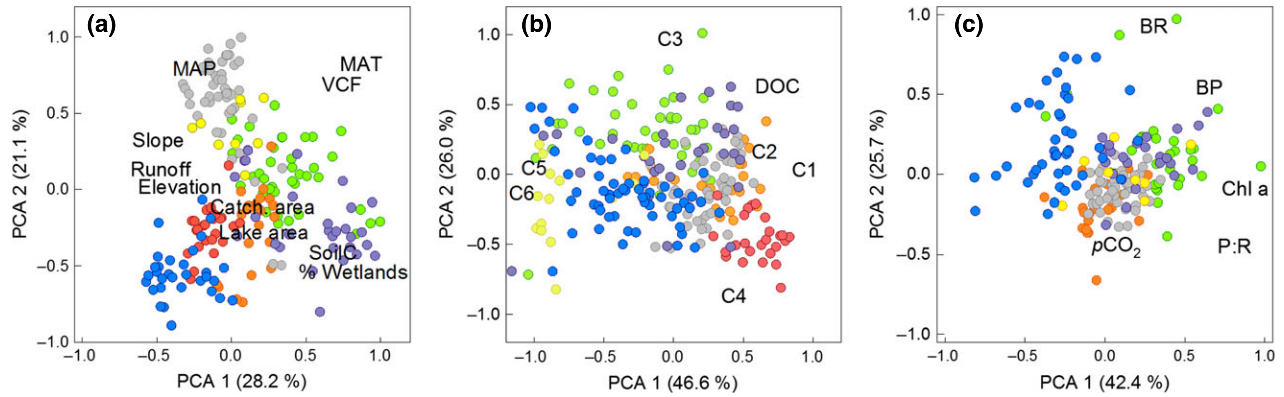

Fig. 2 Regional patterns in climate, landscape, DOC, and indicators of lake C cycling. Principal component analysis (PCA) of climate and landscape properties (a), DOC concentration and composition (b), and indicators of lake carbon cycling (c); color codes match map legend in Fig. 1 to illustrate the regional patterns. Components C1–C6 represent the percent contribution of a specific DOM group to total fluorescence in a sample; they are collectively an index of DOM composition.

significantly explain BR and pCO_2 ($P = 0.90$ and $P = 0.15$, respectively, see Fig. 3); water temperature was not a significant predictor in the RDA once the other predictors were included.

Including DOC concentrations and composition as predictors allowed to explain an additional 11% of the variance in the indicators of lake C cycling (Fig. 4b), with DOC concentration and C2, C5, and C6 fluorescence components being significant; DOC concentration and composition collectively explained 38% of the variance, which corresponds to the vast majority of the variance explained when all drivers are included (43%, Fig. 4b). This means that although climate and landscape properties explained, together, a significant amount of the total variance (Fig. 3a), there was almost no variance explained by climate and landscape properties that was not accounted for by DOC.

Discussion

This study provides empirical evidence of biogeochemical pathways linking large-scale environmental patterns to local aquatic carbon cycling through the delivery of organic carbon (C) from land to water, reflected here by the interplay between climate, landscape, and limnological variables with different spatial structures. We show that across an area of over 1 million km², some variables (mean annual temperature and precipitation; MAT and MAP, respectively) varied gradually along geographic gradients, while others, namely lake and catchment area, varied in a much less structured way and were nearly unpredictable over space. Between those extremes were a series of lake and landscape properties varying at intermediate regional scales, and the combined effect of those different drivers resulted in strong regional patterns in indicators of C cycling in lakes. Lake dissolved organic carbon (DOC) concentration and composition appear to represent proximal drivers that may link static climate and landscape properties to highly dynamic aquatic processes at regional scales.

Regional structure in indicators of lake C cycling and DOC

The major role of DOC on indicators of lake C cycling is not unexpected, as its impact on individual aspects of microbial processes is widely recognized. It is well established, for example, that the concentrations of DOC drive the rates of respiration, as well as primary and secondary production in lakes, or the ratio between both (Del Giorgio & Peters, 1994; Ask *et al.*, 2009; Thrane *et al.*, 2014; Seekell *et al.*, 2015). Likewise, it is known that DOC pools from contrasting sources and origins have different composition and degradability in aquatic environments (Wickland *et al.*, 2007; Fellman *et al.*, 2008; Lapierre & del Giorgio, 2014), and DOC composition has been proposed as a 'scalable sentinel response' to climate change (Williamson *et al.*, 2014), with effects on a large suite of aquatic processes (Solomon *et al.*, 2015). There were thus hints that several pathways would allow DOC to convey an effect of climate on aquatic processes, but little was known on how those different DOC pools respond to contrasting drivers in the landscape and how they interact in driving lake C cycling over large spatial and environmental gradients. The results presented here show that DOC allows to understand the spatial patterns in key processes involved in lake C cycling across a wide number and variety of natural aquatic environments situated

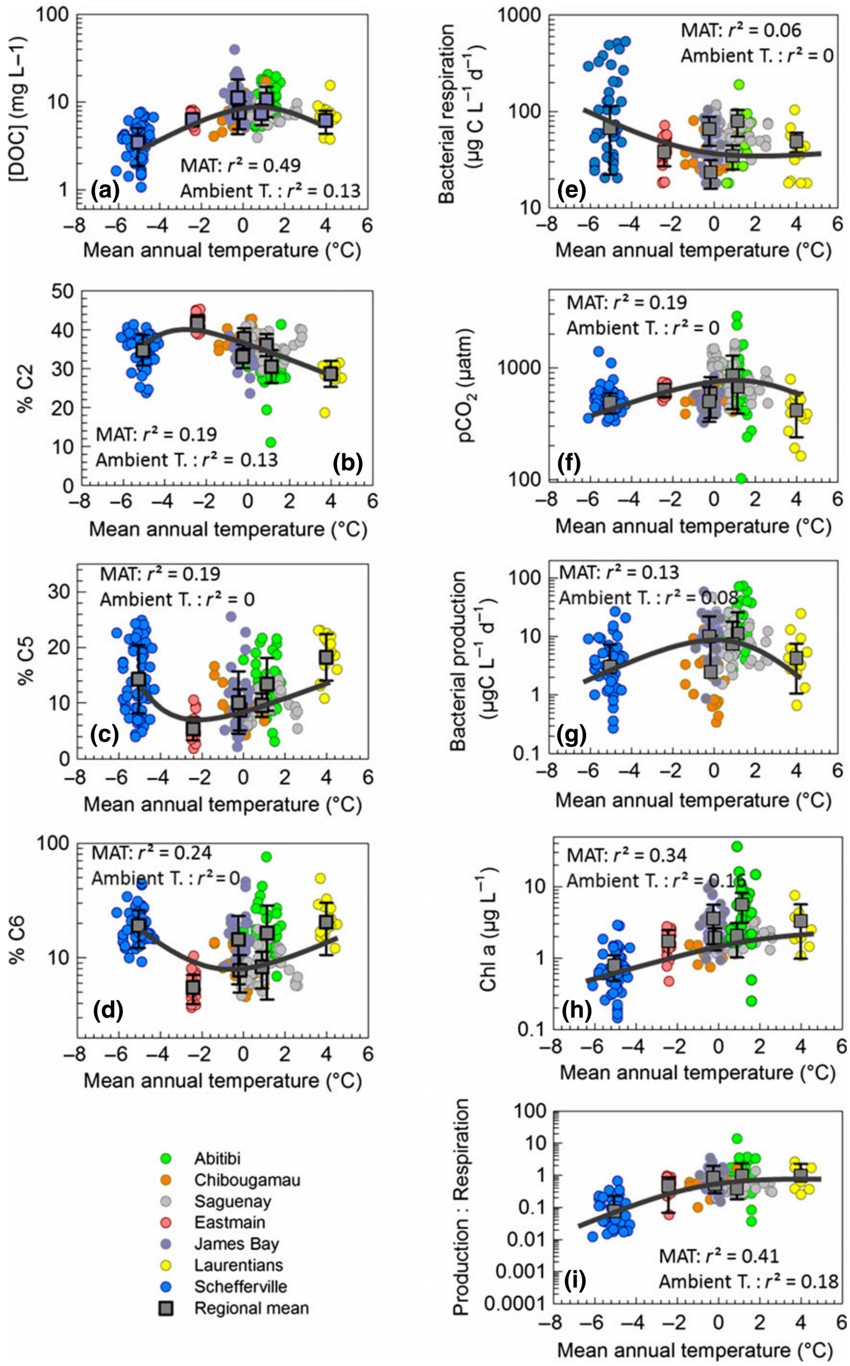

Fig. 3 DOC and indicators of lake C cycling patterns across climate gradients. Gray squares denote regional mean (± SD). Nonlinear trend fits the individual points, not the regional means. Coefficients of determination are reported for ambient water temperature (Ambient T.), but only the trend with mean annual temperature (MAT) is shown. Only the PARAFAC components that are significant predictors of the indicators of lake C cycling in the redundancy analysis are plotted for brevity.

across large latitudinal gradients, and perhaps less intuitively, that the spatial patterns in DOC are driving strong spatial structure in lake C cycling, resulting in strong regional patterns in lake functioning.

The presence of spatial autocorrelation in indicators of lake C cycling and DOC suggests that variables with strong spatial structure are driving those lake properties. In community ecology, spatial autocorrelation is often explained by connectivity and thus dispersal (Cottenie, 2005; Beisner *et al.*, 2006), such that 'space' in itself is a causal predictor of the spatial patterns. Similar patterns in lake biogeochemistry have been attributed

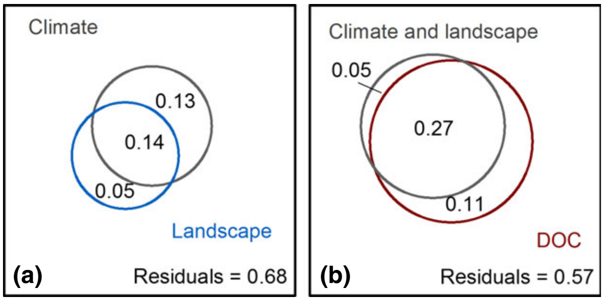

Fig. 4 Partitioning the variance explained in indicators of lake C cycling by climate, landscape, and DOC in redundancy analyses. For both analyses, predicted variables are BR, BP, P : R, Chl a, and pCO_2. Only the significant predictors from a forward selection redundancy analyses have been included. (a) 'Climate' predictors include mean annual temperature and precipitation, and 'Landscape' predictors include catchment slope, soil C, vegetation in the catchment and catchment area. The same predictors are included in 'Climate and landscape' in (b), where 'DOC' includes DOC concentration and %C2, %C5, and %C6. All fractions illustrated are significant.

to hydrologic connectivity among relatively close groups of lakes (Soranno *et al.*, 1999; Webster *et al.*, 2000; Sadro *et al.*, 2012), but regional patterns in DOC concentrations are independent of hydrologic connectivity over larger areas (Seekell *et al.*, 2014). Here, different components of the DOC pool are indeed moving through the aquatic networks in the landscape, but over a very large area and in a nonconservative manner; that is, they are gradually removed from the environment and replaced by local inputs (Mattsson *et al.*, 2005; Weyhenmeyer *et al.*, 2012; Lottig *et al.*, 2013). Moreover, the sampled lakes are almost never hydrologically connected across the study areas. Thus, hydrologic connectivity is not expected to play a large role in explaining the similarity of nearby lakes at cross-regional scales reported here, and our results suggest that nearby lakes have comparable DOC and C cycling because they lie in landscapes with comparable climate and landscape properties.

It is interesting to note that total DOC and the humic-like DOC components C1–C4 tended to be spatially structured at larger scales (Fig. 1b), coherent with their presumed (mostly) terrestrial origin in the sampled lakes (Wilkinson *et al.*, 2013; Lapierre & del Giorgio, 2014) and the spatial structure measured in most landscape properties (Fig. 1b). This terrestrial DOC has been shown to control large-scale patterns in lake primary production in northern lakes (Seekell *et al.*, 2015) and here chlorophyll a concentrations and production: Respiration ratios had the strongest spatial structure among the processes involved in lake C cycling (Fig. 1b); they are also the variables that most strongly followed climate gradients, along with DOC concentration (Fig. 3).

Not all the DOC pools covaried in the same direction across the study area, however, as components C5 and C6 actually followed the opposite pattern compared to C2 and total DOC (Fig. 3). These DOC pools also had the weakest spatial structure, close to that of variables such as pCO_2 and bacterial respiration (Fig. 1b). Components C5 and C6 have been associated with locally produced material (Stedmon & Markager, 2005; Lapierre & Frenette, 2009) in systems with long residence time such as most of the lakes studied here, which suggests that these components have a more local behavior and respond less strongly to spatial signals in the landscape. It is also possible that having weaker spatial structure, these DOC pools (as well as bacterial respiration and pCO_2) have a relatively stronger temporal signal that is not adequately captured by static (at least in the way they have been measured for the purpose of this study) climate and landscape predictors, which could be more strongly driven by short-term variation in weather ((Hudson *et al.*, 2003; Evans *et al.*, 2005). It is thus not surprising that the overall explained variation in lake C cycling (Fig. 4) may arguably seem low compared to levels explained in studies conducted in more homogenous settings, which may be explained in part because some predicted variables have confounding temporal effects not taken into account here and in part because the redundancy analysis (RDA, Fig. 4) cannot capture the nonlinear responses of lake C cycling that has been observed over climate gradients (Fig. 3). Nonetheless, the presence of spatially structured patterns in highly dynamic lakes properties and processes suggests that lake C cycling responds strongly to large-scale drivers such as climate, although the response of different processes is not homogeneous and is further mediated by local and temporally variable aspects of lake functioning. We further argue that DOC (concentration and composition) may capture a portion of temporally variable weather and hydrologic signals on indicators of lake C cycling, which cannot be explained by long-term climate averages.

The indirect effect of climate

Based on experimental evidence, spatial gradients in climate could have been expected to have a strong influence on the measured indicators of lake C cycling, considering the direct effect of temperature on various metabolic pathways (Apple *et al.*, 2006; Gudasz *et al.*, 2010; Yvon-Durocher *et al.*, 2012). Water temperature, however, was weakly or not related to the different lake processes, and it was not a significant predictor in the

RDA performed on the indicators of lake C cycling. This lack of direct effect of temperature can partly be explained by the mismatch in terms of environmental gradients covered. While variables such as DOC concentration and composition and several landscape properties varied by roughly 1 to 1.5 orders of magnitude across the study area, comparable to the range of variability observed in the measured indicators of lake C cycling, water temperature at the moment of sampling (late May to August) barely varied twofold (12.7–23.1 °C, 2.5th and 97.5th percentiles, respectively, Fig. 5), corresponding to a difference in temperature of roughly 10 °C. Even considering a metabolic Q10 ranging from 1 to 4 (Apple *et al*., 2006) for the biological processes measured here could not account for the ranges observed across the sampled lakes, and again, rates of C production and degradation were not linearly increasing with MAT and were weakly or not related to ambient water temperature. It would be thus unlikely that the relatively narrow ranges (compared to other predictors) in water temperature across the study area can explain the large ranges in lake processes involved in C cycling.

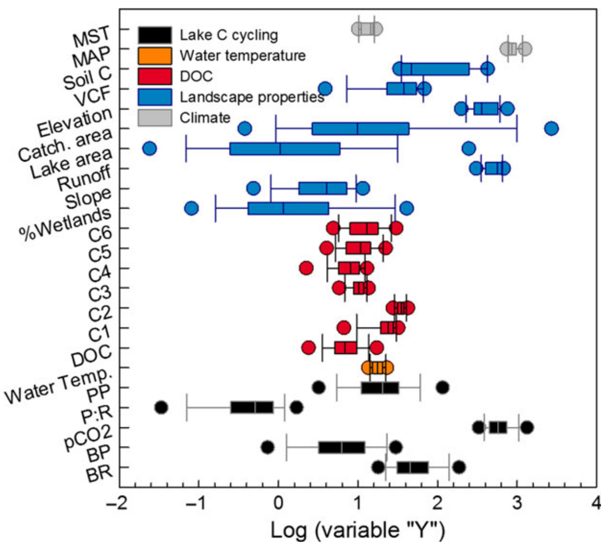

Fig. 5 Range of variation of the reported variables across the sampled lakes. All variables are presented on a log-scale to emphasize the orders of magnitude of variability; one unit on the 'X' scale represents one order of magnitude variation. MAP denotes mean annual precipitation, and MST denotes mean summer temperature. We used MST rather than mean annual temperature (MAT) for this analysis to avoid negative values that could not be logged and to have a range of temperature that is comparable to the water temperature reported; using one or another would not meaningfully alter the range in terms of order of magnitude (logMST range: 0.97–1.25; logMAT (+1) range: 0.59–1.16). Points represent the 2.5th and 97.5th percentiles, and vertical lines represent the 10th, 25th, 50th, 75th and 90th percentiles.

It is interesting to note that the mean annual temperature at the sampled sites was almost symmetrically distributed around the freezing point, ranging from −5.7 to 3.9 °C (2.5th and 97.5th percentiles, respectively). In this regard, the number of days when temperatures exceeded 0 °C was the strongest predictor of DOC concentrations in a large-scale survey of boreal lakes (Weyhenmeyer & Karlsson, 2009), presumably because it integrates air temperature, growing season length as well as ice on and off, and snowmelt. Furthermore, Laudon *et al.* (2012) have shown that the balance between production, decomposition, and transport of terrestrial DOC to aquatic environments tends to shift around mean annual temperatures of 0 °C, due to abrupt changes in the landscape that result in a combination of changing hydrology, primary productivity, and vegetation type in the catchment. Mean annual temperature thus appears to set regional baselines in lake DOC, where within-region variance was lower than among regions. Such pattern has been observed for DOC concentrations in Sweden across highland and lowland regions (Seekell *et al*., 2014), and shifts in DOC optical properties and lake biogeochemistry have been observed as landscape shifted from subalpine to alpine in high altitude lakes (Sadro *et al*., 2012). Here, we capture a combination of the latter patterns across large latitudinal gradients for hundreds of lakes distributed across an area covering more than 1 million km^2.

The latitudinal gradients in climate across our study area were indeed reflected in proxies of terrestrial primary productivity (i.e., vegetation density in the landscape, Fig. 2a), consistent with a general decrease in soil carbon content along a south–north gradient (IGBP-DIS, 1998), and dominant vegetation in the catchment varied from deciduous and mixed forest in the southernmost Laurentians region to scarce spruce–moss forest in the northernmost Schefferville region (Geobase, 2009). Moreover, an inflexion in the nonlinear trends of DOC and indicators of lake C cycling against MAT almost systematically occurred within a couple of degrees from the freezing point across the different regions (Fig. 3). Thus, the absence of a direct effect of midsummer water temperature on indicators of lake C cycling across the study area is not incompatible with the fairly strong effect of 'climate' on the same variables, which would rather be indirect and conveyed by drivers other than temperature. In this regard, DOC has a more obvious causal effect than climate (considering the absence of a temperature effect) or landscape properties on lake processes, and there was a significant overlap in the effect of those different drivers on indicators of lake C cycling (Fig. 4b). Although the large number of predictors included in our analyses and the high overlap in their effect on the response variables does not allow a robust interpretation of the individual effect of any single predictor, the overall pattern suggests that it is mainly DOC that is driving lake C cycling at that scale and that the effect of climate and landscape properties is expressed through both quantitative and qualitative aspects of DOC.

Our results suggest that key climatic and landscape signals are conveyed to lakes in part via the movement of terrestrial DOC to lakes and that DOC acts both as a driver of lake C cycling and as a proxy for other external signals. Changes in temperature and precipitation in boreal landscapes have led to increases in the production of organic carbon in terrestrial ecosystems and in the transfer of soil carbon to aquatic ecosystems (Freeman *et al.*, 2001; Erlandsson *et al.*, 2008), and this 'browning' trend (Roulet & Moore, 2006) may intensify in the coming decades as warming proceeds (Larsen *et al.*, 2011). The indirect effect of climate combined with the strong regional structure in aquatic processes suggests that gradual and diffuse regional changes in boreal landscapes could have a stronger effect on lake C cycling than more localized changes in land use. We further speculate that the in-lake responses may be the strongest in landscapes where mean annual temperature is within a few degrees of the freezing point, where the warming rates are among the fastest around the globe.

Acknowledgements

This work is part of the Carbon Biogeochemistry in Boreal Aquatic Systems (CarBBAS) research chair, co-funded by the Natural Science and Engineering Research Council of Canada (NSERC) and Hydro-Québec. We thank A. St-Pierre, A. Parkes, and the CarBBAS team for the contribution to the field and laboratory components of this study, and we thank S. Cardoso, T. DelSontro, and A. Heathcote for constructive discussions and comments on early version of this manuscript.

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