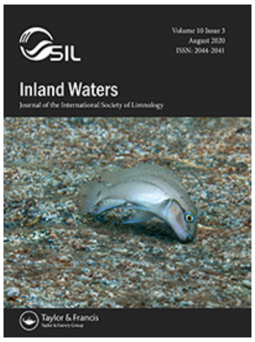

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Diverse drivers of long-term pCO_2 increases across thirteen boreal lakes and streams

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ABSTRACT

Understanding the mechanisms driving carbon dioxide (CO_2) concentrations in inland waters is important to foresee CO_2 responses to environmental change, yet knowledge gaps persist regarding which processes are the key drivers. Here we investigated possible drivers across 13 Swedish lakes and streams where the partial pressure of CO_2 (pCO_2) has increased over a 21-year period. Overall, we could not identify a single dominating mechanism responsible for the observed pCO_2 increase. In the 8 lakes, we found that pCO_2 increased, driven either by a possible dissolved organic carbon (DOC) stimulation of microbial mineralization or by water color primary production suppression. In streams, the dominating mechanism for a pCO_2 increase was either a change in the carbonate system distribution or a possible nutrient-driven decrease in primary production. This is the first study to demonstrate and explain consistent positive pCO_2 temporal trends in freshwater ecosystems, and our results should be taken into account when predicting future emission of CO_2 from inland waters.

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carbon dioxide; freshwater; inland water; mechanisms

Introduction

Inland waters are active components of the global carbon cycle by processing, transporting, and storing vast amounts of carbon (Cole et al. 1988, 2007, Lapierre and del Giorgio 2012). Because of the large amounts of carbon being processed in situ as well as in the surrounding catchment and imported as dissolved inorganic carbon (DIC) to inland waters, the majority of the world's inland waters are supersaturated with carbon dioxide (CO_2) and therefore net sources of CO_2 to the atmosphere (Cole et al. 1994, Raymond et al. 2013). The terrestrial input of carbon (C) to inland waters has been estimated at 1.9–5.1 Pg y^{-1} (Cole et al. 2007, Battin et al. 2009, Tranvik et al. 2009, Drake et al. 2018). Of this, 0.8–3.9 Pg of C is annually outgassed, mostly as CO_2 (Cole et al. 2007, Battin et al. 2009, Tranvik et al. 2009, Drake et al. 2018). The higher estimate indicates the significance of the outgassing flux from inland waters because it is even higher than the terrestrial C sink for anthropogenic emissions, which corresponds to ~2.8 Pg yr^{-1} (Canadell et al. 2007).

Although some of the mechanisms driving the partial pressure of CO_2 (pCO_2) in inland waters have been intensively studied, uncertainty remains regarding which processes are the key CO_2 drivers, in particular determining key drivers on a spatial versus temporal scale. Understanding the mechanisms driving inland water pCO_2 across both the spatial and the temporal scale is important to allow generalizations and predictions of inland water pCO_2 responses to environmental change (Seekell and Gudasz 2016, Nydahl et al. 2017). Overall, 4 key processes regulate pCO_2 in inland waters: (1) CO_2 production through microbial and photochemical mineralization of dissolved organic carbon (DOC; e.g., Tranvik 1992, Hope et al. 1996); (2) CO_2 consumption via primary production (e.g., Balmer and Downing 2011); (3) distribution changes within the carbonate system as a result of pH changes (e.g., Lazzarino et al. 2009); and (4) CO_2 input from the surrounding catchment driven by catchment hydrology (e.g., Jones and Mulholland 1998, Striegl and Michmerhuizen 1998, Palmer et al. 2001).

Among these processes, direct microbial mineralization of DOC has been suggested as a dominant driver of pCO_2 in lakes, in particular in boreal lakes (Hope et al. 1996, del Giorgio et al. 1997), with pCO_2 positively related to DOC (Sobek et al. 2003, Lapierre and del Giorgio 2012). DOC concentrations in surface waters have

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been increasing in the boreal region over the last decades (Monteith et al. 2007, Filella and Rodriguez-Murillo 2014). Several explanations for these increasing DOC trends have been proposed. Some argue that DOC trends are consistent with changes in temperature and rainfall (Worrall and Burt 2007, Eimers et al. 2008) while others propose that increasing DOC trends are the result of a reduction of anthropogenic sulfur in the atmosphere and a subsequent decrease in acid deposition (Evans et al. 2006, Vuorenmaa et al. 2006, Monteith et al. 2007). Increased surface water DOC concentrations could potentially lead to enhanced pCO_2 because of increased substrate availability for microbial mineralization. Microbial mineralization is also dependent on water temperature, resulting in increasing CO_2 production at higher temperatures (Gudasz et al. 2010).

Microbial mineralization is not only affected by DOC concentration and water temperature, the quality of the DOC can also be a key regulator and thereby influence CO_2 in the water column (Bodmer et al. 2016). The quality of DOC depends on its composition (Jaffe et al. 2008), which often is divided into humic-like or protein-like components (Kothawala et al. 2014). The composition of DOC is generally a function of its origin, with protein-like DOC generally derived from algae and in situ heterotrophic processes (i.e., autochthonous; McKnight et al. 2001), whereas humic-like DOC is predominantly derived from terrestrial vascular plants and soil organic matter (i.e., allochthonous; Miller and McKnight 2010). Autochthonous DOC generally has a higher nitrogen (N) content (atomic C:N 4–10) than does allochthonous DOC (C:N ≥ 20; Meyers and Ishiwatari 1993). Consequently, the C:N ratio could be used as a proxy indicator of DOC quality (Bernal et al. 2005). Likewise, because humic-like substances generally have more aromatic structures and are thus more colored than protein-like substances (Kothawala et al. 2014), absorbance at 420 nm (i.e., water color) divided by DOC concentration (water color/DOC) could also be used as a proxy indicator for DOC quality. Protein-like DOC is typically more degradable; however, even if the autochthonous DOC is rapidly degraded, the net result on pCO_2 would be negligible because the CO_2 recently fixed by primary producers would be returned to the water column with no additional CO_2 input. Conversely, an increased input of humic-like DOC (i.e., increased C:N ratios and/or water color/DOC) could lead to enhanced pCO_2 because of increased substrate for microbial mineralization.

In relatively pristine boreal regions and forested landscapes, where nutrient concentrations remain low, DOC is a key driver of pCO_2 (Rantakari and Kortelainen 2008, Lapierre et al. 2013). However, the main drivers of CO_2 vary spatially, particularly in regions with contrasting levels of nutrients and alkalinity (Lapierre et al. 2017), and in more diverse landscape with larger coverage of peatlands or arable land the correlation between pCO_2 and DOC is weaker (Rantakari and Kortelainen 2008). Thus, on a spatial scale, the main driver for surface water pCO_2 can vary substantially (Lapierre et al. 2017), depending on the heterogeneity of the landscape.

Another important driver for pCO_2 in inland waters can be primary production, resulting in reduced CO_2 concentrations through photosynthesis. Primary production is commonly nutrient limited, with phosphorus (P) most frequently the limiting nutrient in inland waters (Schindler 1977, Elser et al. 1990). Consequently, P concentrations are commonly used as a proxy indicator for primary production in inland waters. Another factor regulating primary production, and hence pCO_2, in inland waters is water color (Jones 1992). Increased water color can constrain primary production as a large fraction of photosynthetically active radiation is absorbed by DOC and is thus not available for photosynthesis (Jones 1992). Increasing water color has been observed in surface waters in the boreal region, attributed to increasing DOC (Monteith et al. 2007, Haaland et al. 2010), which could lead to decreased CO_2 uptake through primary production, subsequently enhancing pCO_2.

Another important mechanism regulating pCO_2 in inland waters is the carbonate system (Stets et al. 2017), which is partially driven by changes in pH. Although many inland waters in the boreal region are recovering from acidification due to acid deposition reduction, pH has continued to decrease in some Swedish lakes and streams over the past 2 decades (Nydahl et al. 2017, Huser et al. 2018). This decrease in pH could be due to the increased DOC concentrations, with a subsequent increase in organic acidity, or to increased use of fertilizers in agriculture because inorganic nitrogenous fertilizers promote soil acidity (Collins and Jenkins 1996, Nohrstedt 2001, Lapierre et al. 2017). A decrease in pH would lead to increased pCO_2 as the distribution within the carbonate system shifts and the proportion between CO_2, bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) changes toward more CO_2. A recent study including >100 streams across the conterminous United States showed that carbonate buffering was the primary control on pCO_2 in surface waters (Stets et al. 2017). Likewise, pH was found to be the best predictor of lake surface water pCO_2 for >900 Florida lakes (Lazzarino et al. 2009).

Not only internal processes are important drivers for pCO_2 in inland waters; hydrological inputs (ground and surface water) that mobilize organic and inorganic C from the catchment soils are also important (Hotchkiss et al. 2015, Ledesma et al. 2015, Weyhenmeyer et al. 2015). These hydrological inputs are often driven by precipitation. Although precipitation has generally increased across Sweden over the past couple of decades, in some areas precipitation has decreased (Chen et al. 2015), possibly decreasing surface water discharge and increasing water retention time (WRT). The lake and stream internal CO_2 production is partly controlled by landscape WRT with more efficient production in waters with a long WRT (Hanson et al. 2011). Decreased precipitation would also lead to a higher relative contribution by groundwater to surface water with effects on the surface water chemistry (Carroll et al. 2018). Shallow groundwater is generally supersaturated with CO_2 and is a large contributor to surface water pCO_2, particularly in headwater streams (Jones and Mulholland 1998, Hotchkiss et al. 2015). In larger downstream rivers, both shallow and deep groundwater regulate water chemistry (Hagedorn et al. 2000) while in-stream metabolism concurrently increases in relative importance as a pCO_2 driver farther downstream (Hotchkiss et al. 2015). Despite being a minor volumetric source of water to lakes, groundwater is also a major driver of lake water chemistry because of the high concentration of dissolved species in groundwater (Schmidt et al. 2009, Shaw et al. 2013). Carbon input to inland waters from the surrounding terrestrial ecosystem can also be driven by the spatial variation in terrestrial net primary productivity (NPP; Maberly et al. 2013, Hastie et al. 2018). Previous studies have also found a relationship between surface water pCO_2 and land cover (Sobek et al. 2007, Hastie et al. 2018). Although the spatial variation in NPP and differences in land cover can be considered an underlying driver of pCO_2 in surface waters, the input of terrestrial C to inland waters is strongly regulated by hydrology.

A recent study of a dataset of 71 lakes and 30 streams found that 8 lakes and 5 streams demonstrated an increase in pCO_2 over the past 2 decades (Nydahl et al. 2017). Here we investigated the reasons behind the observed synchronous pCO_2 increase in these 13 boreal surface waters, focusing on 4 key driving processes: DOC mineralization, primary production, the carbonate system, and catchment hydrology. We tested the hypotheses that the increase in pCO_2 is due to (1) stimulation of microbial mineralization by increased DOC concentrations and/or increased temperature; (2) stimulation of microbial mineralization as a result of DOC quality changes toward more reactive DOC, subsequently enhancing C mineralization rates and CO_2 production; (3) decreased primary production with a consequent decrease in CO_2 bio-uptake; (4) a shift in the carbonate system toward a higher proportion of free CO_2 due to a pH decrease; and (5) a greater proportion of groundwater to surface water input and longer WRT due to decreased precipitation.

Methods

From a database of 101 Swedish lakes and streams with available time series since 1997, we selected the waters that previously had shown increasing surface water pCO_2 (Nydahl et al. 2017). The waters with increasing pCO_2 comprised 8 lakes and 5 streams and were distributed across the boreal and hemiboreal region of Sweden, with a dominance toward southern Sweden (Fig. 1). We focus here only on the systems that increased in pCO_2 to allow a more targeted analysis and a better understanding of the processes that could lead to higher CO_2 emissions from inland waters. The lakes were generally small with a median surface area of 0.37 km^2 and shallow with a median mean depth of 4.4 m. The catchment areas of the lakes were mainly forested (average 81.2%) with some agricultural land (average 5.2%). The catchment areas of the streams also had a large percentage of forest (average 54.5%); in the catchment of Vindbron, however, the greatest proportion of land use was semi-urban (41.1%), and in Akkarjåkka the largest land use type was tundra (54.5%). Many surface waters in Sweden have been limed since the late 1970s to counter surface water acidification caused by acid deposition (Henrikson et al. 1995); however, none of the waters included in this study have been limed.

From the Swedish national freshwater monitoring program, described by Fölster et al. (2014), we acquired water chemistry data from the lakes and streams from 1997 to 2017, freely available at http://www.slu.se/vatten-miljo. In this study we considered data on total organic carbon (TOC), pH, alkalinity, total phosphorous (TP), total nitrogen (TN), inorganic nitrogen (IN), absorbance at 420 nm measured in a 5 cm cuvette (water color), and water temperature. We used TOC concentrations as a proxy indicator for DOC concentrations because the particulate fraction of organic C in boreal and hemiboreal inland waters generally is <1% (Laudon et al. 2011). Additionally, TP was used as a proxy indicator for primary production (Wetzel 1992). Dissolved organic nitrogen (DON) was calculated by subtracting IN from TN. We used DOC:DON (C:N) ratios for DOC quality because these ratios are positively related to humic-like substances (Kothawala et al. 2014). We also used water color/DOC as a proxy indicator for DOC quality because a higher water color/DOC is equivalent to more color, thus indicating a higher proportion of humic-like substances. All lakes and streams included in this study were sampled at least 4 times per year during the study period, 1997 to 2017.

All samples were collected at a water depth of 0.5 m, except in more shallow streams where samples were taken closer to the surface. All samples were analyzed by the Swedish Board for Accreditation and Conformity

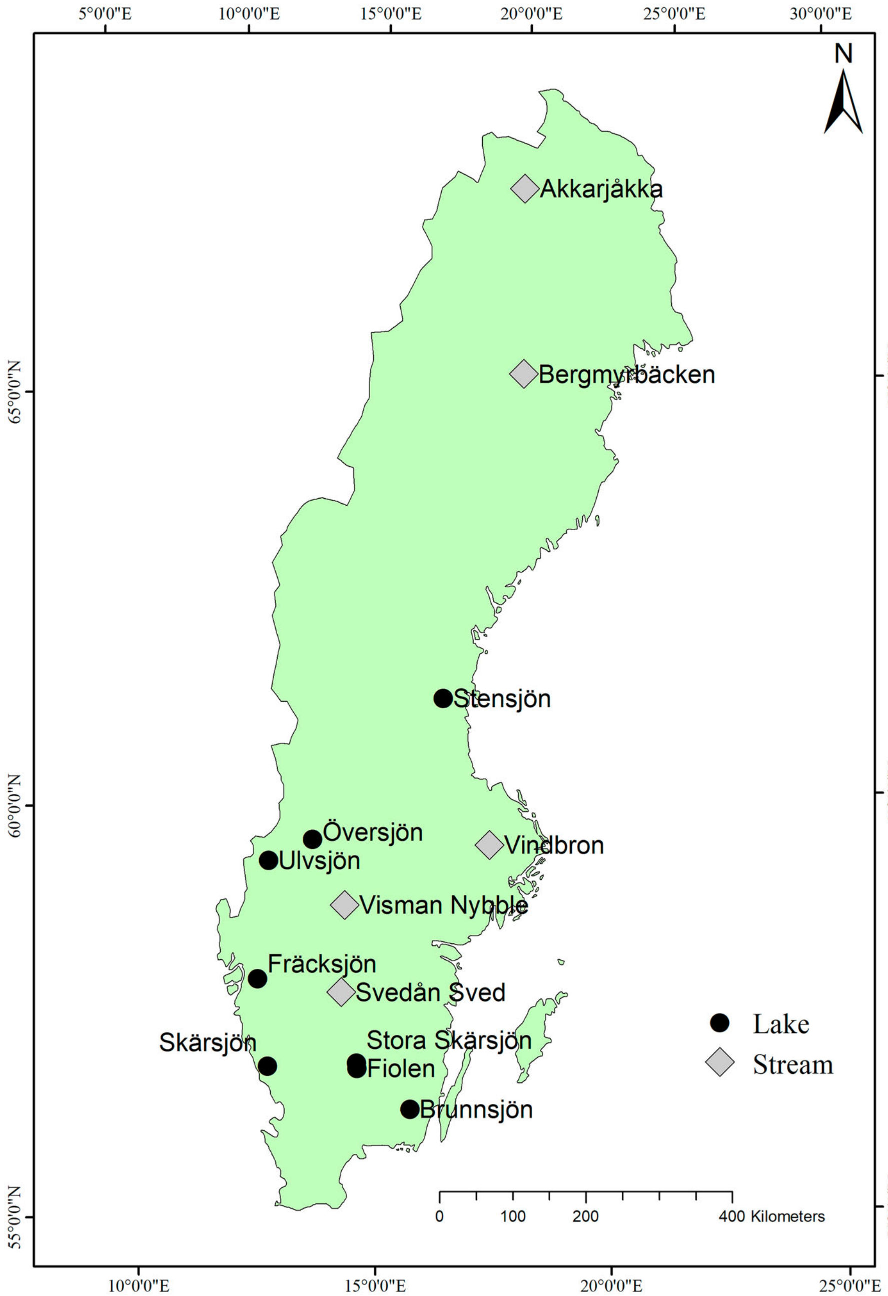

Figure 1. Location of the Swedish study lakes (circles) and streams (diamonds).

accredited laboratory at the Swedish University of Agricultural Sciences following standard limnological procedures. A detailed method description including analytical precision and range can be found at http://www.slu.se/en/departments/aquatic-sciencesassessment/laboratories/geochemical-laboratory/water-chemical-analyses/. All water chemistry analyses considered in this study were made based on unfiltered water, except absorbance which was based on filtered (0.45 µm) water.

From the available water chemistry data, we calculated concentrations of CO_2 using water temperature, alkalinity, and pH according to Weyhenmeyer et al. (2012). For the calculation, only positive alkalinity values and pH values >5.4 were used to avoid substantial CO_2 overestimation (see Nydahl et al. 2017). We also applied the tripotric model by Hruska et al. (2003) to estimate the dissociation of organic acid anions ($RDOO^-$) from measured pH and TOC, and thereby calculated a new value for alkalinity used to calculate CO_2 (Wallin et al. 2014, Nydahl et al. 2017). From the calculated CO_2 we determined $p CO_2$ (in µatm) using Henry's constant (as described by Weyhenmeyer et al. 2012). We used yearly median values rather than means to avoid the impact of outliers and minimize uncertainties in the $p CO_2$ calculations.

In addition to the water chemistry data, we downloaded hydrological data for precipitation, discharge, and groundwater storage for each subcatchment of the lakes and streams from the Swedish Meteorological and Hydrological Institute's Vattenweb (http://vattenweb.smhi.se/). For precipitation and discharge, average monthly values for all subcatchments were modeled using the hydrological model S-HYPE (Lindström et al. 2010). Groundwater in the moraine and glacifluvial sediments were also modeled using S-HYPE. The groundwater levels in the moraine and glacifluvial sediments were then averaged to provide one value for groundwater levels. Groundwater levels were modeled for every day of the year and reported as the percentage of the daily groundwater level during 1961–2017. A groundwater level of 0% represents the historically lowest groundwater level on the same day of the year, whereas a groundwater level of 100% is equivalent to the highest groundwater level on the same day of the year.

Statistical analyses

In this study we used 2 main statistical approaches, one to evaluate variables affecting surface water pCO_2 and one to evaluate changes over time. For the first approach we applied partial least squares (PLS) regressions on the whole dataset to quantify the effect of DOC, temperature, water color/DOC, C:N, pH, water color, TP, precipitation, discharge, and groundwater on surface water pCO_2. Data were normalized using log-transformation where necessary. Because we applied PLS on the whole dataset, we also included time as an x-variable. We performed 3 separate PLS analyses. First, we performed a PLS on all available lake and stream data combined, including water type (i.e., lake or stream) as an x-variable, totaling 11 x-variables. The other 2 PLS analyses were performed on the lake and stream data separately, totaling 10 x-variables each (x predictor variables listed in Table 1). PLS is a reliable technique when identifying relevant variables and their magnitude of influence, particularly if the sample size is small (Abdelhady et al. 2018). The variable importance for projections (VIPs), which are linear coefficient plots between the predictors and the response variable (i.e., pCO_2) across all model components, were used to identify the explanatory variables that contribute most to the models (i.e., the weight of each predictor). Here we considered VIP values >1.0 as significant explanatory variables (Eriksson et al. 1999). PLS analyses were performed using the software package SIMCA 15.0.2 (Umetrics AB, Umeå, Sweden). For the second approach we used the nonparametric Mann-Kendall trend tests to determine significant increases or decreases in DOC, temperature, water color/DOC, C:N, pH, water color, TP, precipitation, discharge, and groundwater during 1997–2017. Input data for the tests were yearly median values from 1997 to 2017. Significance was set at $p < 0.05$ for all Mann-Kendall trend tests.

Results

All 8 lakes and 5 streams showed a significant increase in surface water pCO_2 from 1997 to 2017 (Mann-Kendall trend test: $p < 0.05$). All waters were oversaturated with pCO_2 relative to the atmosphere; the long-term median ranged from 770 to 1236 μatm and 967 to 4124 μatm in the lakes and streams, respectively (Table 1). The long-term median DOC concentration ranged from 7.4 to 19.5 mg L^{-1} in the lakes and 1.9 to 17.4 mg L^{-1} in the streams (Table 1). The lakes were generally acidic, with a pH ranging from 5.7 to 6.9, while most streams had slightly higher pH ranging from 6.5 to 7.2 (Table 1). TP concentrations were generally low (<12.5 μg L^{-1}) in the waters, except for 2 streams where TP concentrations were 42 and 55 μg L^{-1} (Table 1).

Predictor variables for pCO_2 in lakes and streams

Using PLS to predict the synchronous increase in surface water pCO_2 in the studied lakes and streams, we found that TP, C:N, and DOC were important variables for explaining the variability in surface water pCO_2 when lakes and streams were combined in the same model. The first PLS component explained 19% of the total variance and showed that TP and DOC were positively correlated to pCO_2, whereas C:N was negatively correlated to pCO_2 (Fig. 2a). Overall, the PLS model for the lakes and streams extracted 3 significant components that collectively explained 59% of the variation in surface water pCO_2 (R^2Y) by using 50% of the variation in the environmental predictors (R^2X). The second and third PLS components explained 17% and 13% of the variance, respectively (Fig. 2a). Water type (i.e., lake and stream) was found on opposite sides in the PLS plot, although water type was not a significant explanatory variable for surface water pCO_2 (VIP < 1, Fig. 3a). The importance of TP, DOC, and C:N for explaining the variability in pCO_2 was further supported by the VIP scores, which for TP, DOC, and C:N were >1.0 (Fig. 3a). None of the 3 catchment hydrological variables—precipitation, discharge, or groundwater—had a significant impact on surface water pCO_2 in the model where lakes and streams were combined (Fig. 2a).

In our second PLS model we considered only those lakes that had a total of 10 x-variables (x predictor

Table 1. Median water chemical and catchment hydrological values for 8 Swedish lakes and 5 Swedish streams for the 21-year period from 1997 to 2017.

System site	pCO_2 (µatm)	DOC (mg L^{-1})	Temperature (°C)	Water color/ DOC	C:N	pH	Water color	TP (µg L^{-1})	Precipitation (mm $month^{-1}$)	Discharge (m^3 s^{-1})	Ground-water (%)
Lake											
Brunnsjön	1421 (+)	19.5 (nc)	18.1 (nc)	0.021 (nc)	27.8 (nc)	5.71 (nc)	0.402 (nc)	12 (nc)	62.9 (nc)	0.138 (+)	33.2 (nc)
Fiolen	770 (+)	7.6 (+)	14.2 (nc)	0.009 (nc)	15.4 (+)	6.68 (+)	0.064 (nc)	11 (nc)	71.8 (nc)	0.061 (nc)	50.5 (nc)
Fräcksjön	1213 (+)	10.0 (+)	12.8 (–)	0.014 (nc)	23.1 (nc)	6.43 (nc)	0.135 (+)	9 (nc)	76.9 (–)	271 (nc)	49.7 (nc)
Skärsjön	718 (+)	4.5 (nc)	11.6 (nc)	0.006 (nc)	11.5 (+)	6.86 (nc)	0.026 (nc)	9 (nc)	104.5 (nc)	0.252 (nc)	60.8 (nc)
Stensjön	912 (+)	7.4 (+)	11.8 (+)	0.016 (nc)	29.5 (+)	6.39 (nc)	0.116 (nc)	6 (nc)	59.8 (nc)	0.030 (nc)	46.0 (nc)
Stora Skärsjön	1036 (+)	5.0 (+)	15.9 (nc)	0.014 (+)	17.3 (+)	6.77 (nc)	0.064 (+)	9 (+)	74.9 (nc)	0.176 (nc)	50.1 (nc)
Ulvsjön	1236 (+)	8.4 (nc)	7.2 (nc)	0.014 (nc)	22.9 (+)	6.05 (nc)	0.114 (+)	7 (nc)	68.5 (nc)	0.390 (nc)	51.9 (nc)
Översjön	1100 (+)	6.8 (nc)	7.1 (nc)	0.011 (nc)	24.8 (nc)	5.82 (+)	0.079 (+)	6 (nc)	63.3 (nc)	0.195 (nc)	53.0 (nc)
Stream											
Akkarjåkka	866 (+)	1.9 (–)	3.5 (nc)	0.014 (nc)	11.3 (+)	7.12 (nc)	0.027 (nc)	7 (–)	63.3 (nc)	12.5 (nc)	56.5 (nc)
Bergmyrbäcken	1040 (+)	6.0 (–)	4.1 (–)	0.021 (nc)	26.7 (+)	6.79 (+)	0.132 (–)	5 (–)	61.4 (nc)	0.171 (–)	56.5 (–)
Svedån Sved	967 (+)	7.6 (nc)	6.3 (nc)	0.021 (–)	23.5 (nc)	6.93 (nc)	0.162 (nc)	9 (–)	57.6 (nc)	0.499 (nc)	51.8 (nc)
Vindbron	4124 (+)	15.0 (nc)	7.3 (nc)	0.011 (nc)	6.1 (nc)	7.58 (–)	0.170 (nc)	55 (nc)	47.9 (nc)	5.350 (nc)	43.0 (nc)
Visman Nybble	2713 (+)	17.4 (+)	11.1 (nc)	0.016 (nc)	10.9 (nc)	6.49 (–)	0.267 (nc)	42 (nc)	57.2 (nc)	1.305 (nc)	44.7 (nc)

The sign within parentheses refers to results from Mann-Kendall trend tests where + is a significant increase, – is a significant decrease, and nc refers to no change: $p < 0.05$.

variables listed in Table 1). The first PLS component explained 31% of the variation and showed that pH and temperature were negatively related to pCO_2, whereas water color and water color/DOC were positively related to lake pCO_2 (Fig. 2b). The VIP scores further confirmed the importance of the variables pH, temperature, water color, and water color/DOC for explaining the variability in lake pCO_2, which all had VIP scores >1.0 (Fig. 3b). Overall, the PLS model explained 51% of the variation in pCO_2 (R^2Y) by using 61% of the variation in the environmental predictors (R^2X), divided over 4 significant components. The second PLS component explained 16% of the variation, whereas the third and fourth components only explained 8% and 7%, respectively.

In our third PLS model we considered only stream data, resulting in a model with 10 x-variables (x predictor variables listed in Table 1). The first PLS component explained 24% of the total variance and showed that TP and DOC were positively related to stream surface water pCO_2, whereas C:N was negatively related to pCO_2 (Fig. 2c). Overall, the model resulted in 3 significant components explaining 74% of the variation in stream pCO_2 (R^2Y), using 43% of the variation in the explanatory variables (R^2X). The second PLS component explained 12% of the variance, whereas the third PLS component explained 7% of the total variance (Fig. 2c). The dependency of stream pCO_2 on TP, DOC, and C:N was further confirmed by the VIP scores, which all were >1.0 (Fig. 3c).

Changes in lake water chemical variables since the late 1990s

During the 21-year study period, 1997–2017, pCO_2 increased significantly in the study lakes, but the interannual variability in pCO_2 was markedly high in all 8 lakes (Fig. 4). We found that 6 of the 8 lakes had significant trends in at least one of the variables we used as proxy indicators for DOC mineralization (i.e., DOC, temperature, water color/DOC, and C:N). Half of the 8 lakes increased in DOC (Fig. 4, Table 1), one of which also increased in temperature, and one showed a reduction in temperature (Table 1). Regarding our measures for DOC quality, both water color/DOC and C:N increased in Stora Skärsjön. We also observed enhanced C:N in 4 other lakes, but water color/DOC remained constant in the remaining 7 lakes.

We observed significant trends for at least 1 of the 2 primary production proxy indicators (TP and water color) in 4 of the 8 lakes (Fig. 3, Table 1). Water color increased in Fräcksjön, Stora Skärsjön, Ulvsjön, and Översjön (Table 1). TP increased only in one lake, Stora Skärsjön (Fig. 5, Table 1).

We found that the distribution within the carbonate system changed in 2 of the 8 lakes, as demonstrated by increased pH in these systems (Fig. 4, Table 1).

We observed only a few significant changes with regard to catchment hydrological drivers of surface water pCO_2 (precipitation, discharge, and groundwater) in the catchments of our study lakes. Precipitation decreased in the catchment of Fräcksjön while discharge increased in the catchment of Brunnsjön. Groundwater levels did not change in any of the lake catchments (Fig. 5, Table 1).

Changes in stream water chemical variables since the late 1990s

During the 21-year study period, the interannual variability in pCO_2 was markedly high in 2 streams,

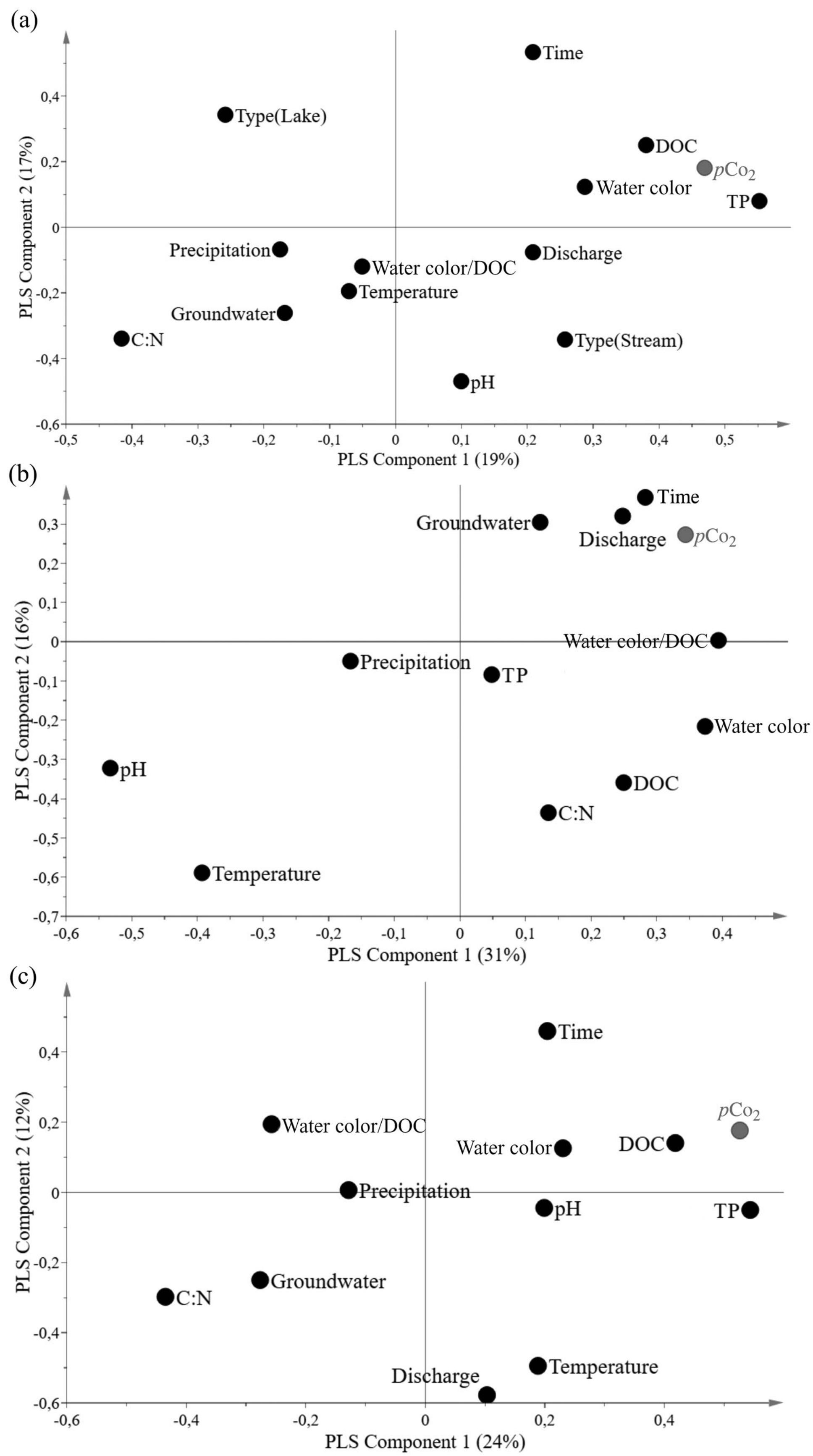

Figure 2. Loading plots of the partial least squares regression (PLS) analyses for (a) all surface waters, (b) only lakes, and (c) only streams as given by the PLS weights. The graphs represent the correlation structures between the x-variables (11 x-variables for all surface waters, 10 x-variables for only lakes, and 10 x-variables for only streams) and pCO_2. The y-variable (pCO_2) is shown in gray; the explanatory variables are shown in black. Only the first and second components are shown in the graphs for clarity.

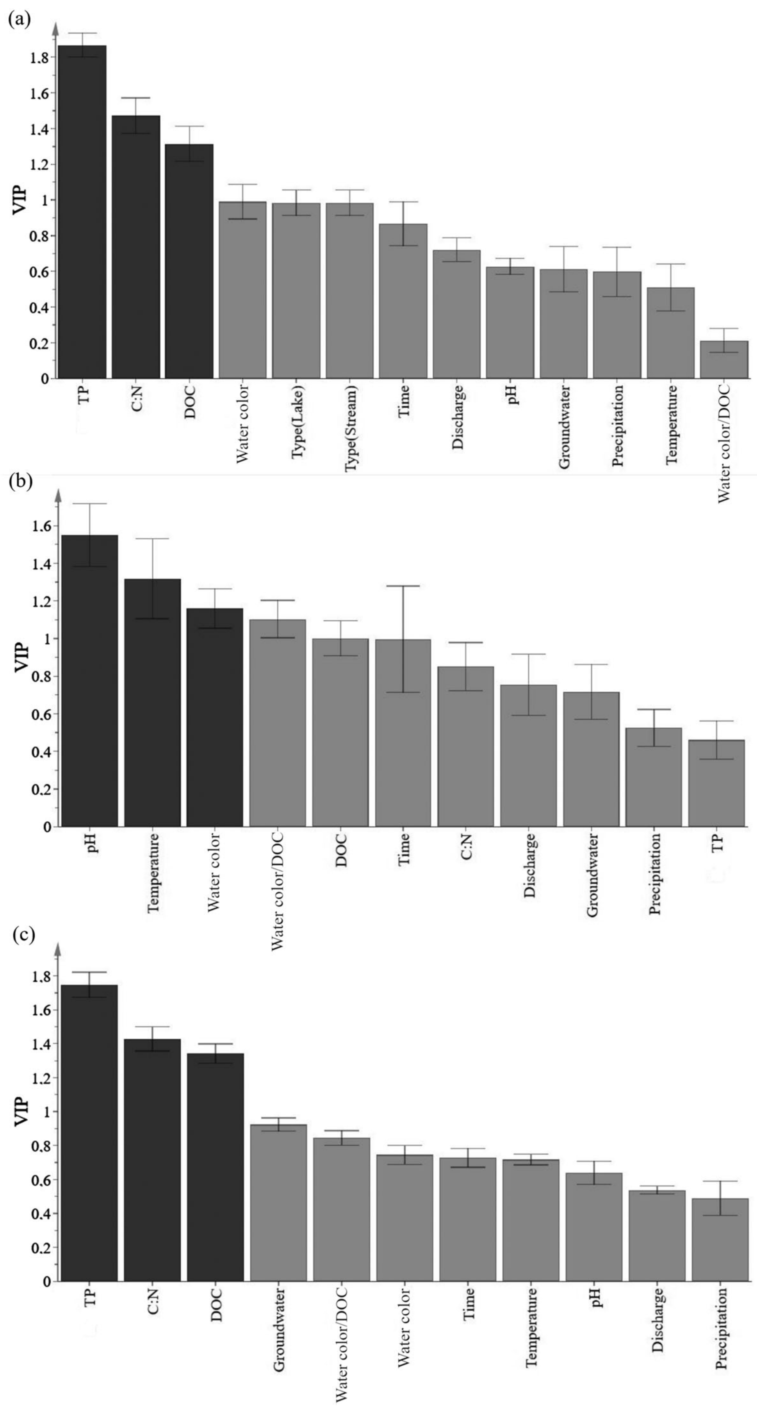

Figure 3. Importance of the 10 environmental input variables for the prediction of pCO_2 in (a) all surface waters ($n = 2439$), (b) only lakes ($n = 1000$), and (c) only streams ($n = 1439$) using PLS regression. The higher the VIP values the more important is the input variable for the model performance. VIP values >1 are considered important for the model performance. Dark gray bars represent variables with VIP >1; light gray bars represent variables with VIP <1.

Vindbron and Visman Nybble, but was lower in the other 3 study streams (Fig. 4). Four of the 5 study streams showed a significant change in at least one of the variables influencing DOC mineralization (DOC, temperature, water color/DOC, C:N); however, no obvious trend allowed us to elucidate if one of the DOC mineralization driving variables was a particularly strong driver of pCO_2 in the streams (Fig. 5, Table 1).

The proxy indicators for primary production, TP and water color, changed significantly in 3 of the 5 streams (Table 1). Both TP and water color decreased in Bergmyrbäcken, and TP decreased in Akkarjåkka and Svedån

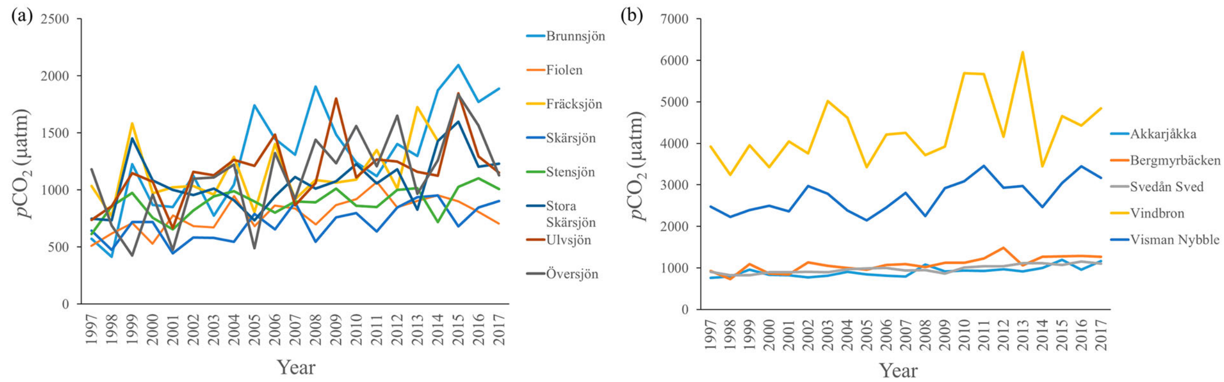

Figure 4. Time series of annual median partial pressure of carbon dioxide (pCO_2) in Swedish (a) lakes and (b) streams from 1997 to 2017 that increased significantly in pCO_2 during this period.

Sved (Table 1). No other changes in our primary production proxy indicators were observed in the streams.

We found that in 3 of the 5 streams the distribution within the carbonate system changed, as demonstrated by changes in pH (Fig. 4). One of the streams increased in pH, whereas in 2 of the streams pH decreased (Table 1).

As for the lakes, few changes were observed for the hydrological variables in the streams. Only in the catchment of Bergmyrbäcken were significant changes observed with decreased discharge and groundwater level (Table 1, Fig. 5).

Discussion

The dominating mechanism driving the observed synchronous pCO_2 increase across a variety of Swedish lakes and streams was highly variable and clearly differed between lakes and streams (Fig. 1a). Considering the temporal scale, which was the main focus of this study, the difference between the important drivers in lakes and streams became even clearer.

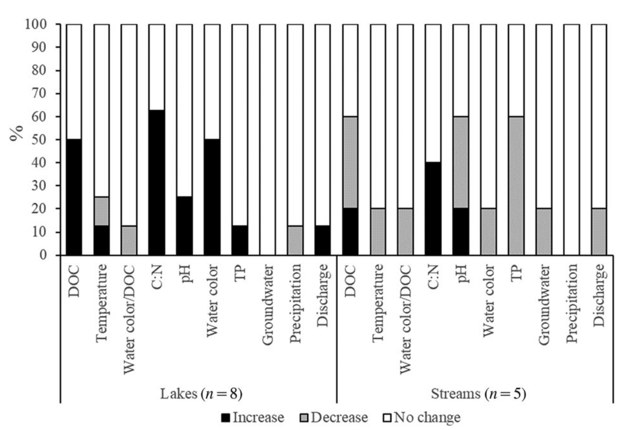

Figure 5. Percentage of significant increase, decrease, or no change in dissolved organic carbon (DOC), temperature, water color/DOC, C:N, pH, water color, total phosphorus (TP) and catchment area groundwater, precipitation and discharge in surface waters of Swedish lakes ($n = 8$) and streams ($n = 5$) during 1997 to 2017.

Effects of DOC mineralization on pCO_2

Half of our 8 study lakes increased in DOC simultaneously with pCO_2 during the 21-year study period, suggesting that microbial mineralization has been an important driver for pCO_2 in these lakes (Tranvik 1992, Hope et al. 1996). By contrast, the remaining 4 lakes were in agreement with a recent study showing that DOC concentrations and pCO_2 were uncoupled over time (Nydahl et al. 2017). We also found highly varying trends in the streams, where DOC concentrations increased, remained unchanged, or even decreased. Hence, we cannot fully confirm or disprove our hypothesis that pCO_2 increased due to stimulation of microbial mineralization by increased DOC concentrations. However, DOC concentrations seemed to be more important for CO_2 production in lakes than in streams, likely because of the longer WRT in lakes, allowing more time for C transformation processes to occur.

Our finding that changes in DOC could be more important in lakes than in streams is further supported by our finding that changes in the origin of the DOC were more pronounced in our study lakes. In 5 of the lakes, C:N increased, suggesting that the proportion of allochthonous DOC increased. The C:N ratio also increased in 2 of the 5 study streams. This increase in allochthonous DOC may have resulted in enhanced pCO_2 in these waters (D'Amario and Xenopoulos 2015, Bodmer et al. 2016).

Effects of primary production on pCO_2

None of the 13 waters included in our study had a concurrent decrease in TP and increase in water color. One of the lakes increased in TP while remaining constant in the other 7 lakes. If primary production was the key mechanism explaining the observed pCO_2 increase in our study waters, we would expect to see a decrease in TP because P concentrations could be a limiting factor for primary production (Schindler 1977, Elser et al. 1990). Consequently, this increase or lack of change in TP in the lakes suggests that the observed pCO_2 increase in our study lakes was not due to a nutrient-controlled decrease in primary production. In eutrophic lakes, primary production could exert strong controls on pCO_2, whereas in oligotrophic lakes, microbial mineralization likely has a stronger control on pCO_2 (Cole and Caraco 2001, Pacheco et al. 2014). All of our study lakes were oligotrophic, further supporting our finding that the observed pCO_2 increases were not due to decreased primary production resulting from nutrient limitation.

Stream size could play an important role in determining the importance of primary production controls on pCO_2 in streams. Much of the CO_2 in headwater streams is originally produced in the surrounding catchment and transported via groundwater to the streams (Wallin et al. 2013) while in-stream metabolism becomes more important farther downstream (Hotchkiss et al. 2015). All our study streams are higher-order streams (catchment areas between 21 and 1254 km^2), and 3 of the streams showed a reduction in TP. Consequently, a decreased primary production possibly resulted in the observed pCO_2 increase in these streams.

In 4 of the lakes, water color increased; hence, despite no change in TP, the darkening of the water in these 4 lakes could have reduced the primary production. As water color increases, less light is available for primary production, and thus CO_2 bio-uptake would decrease and pCO_2 subsequently increase (Jones 1992, Thrane et al. 2014, Nydahl et al. 2019). However, teasing apart the effect that increased DOC may have on primary production from the effect that increased DOC can have on bacterial mineralization is difficult. In addition, increased terrestrial DOC input could also lead to enhanced loading of terrestrial DIC with a subsequent increase in pCO_2.

Effects of changes in the carbonate system on pCO₂

An increase in pH would theoretically lead to a decrease in pCO_2; however, pH increased in 2 of the 8 study lakes, most probably a result of the recovery from acidification that many Swedish inland waters have experienced due to reduced atmospheric sulfur deposition (Fölster and Wilander 2002, Vuorenmaa et al. 2006). Despite the increase in pCO_2, pH did not change in the other 6 lakes. Consequently, other processes causing the pCO_2 to increase were generally more important than distribution changes within the carbonate system for the long-term pCO_2 trends in our study lakes. We also found an increase in pH in one stream. Conversely, pH decreased in 2 of the streams, and hence distribution changes within the carbonate system could potentially explain the observed pCO_2 increase in these streams.

Effects of changes in hydrology on pCO₂

Our hypothesis that pCO_2 has increased as a result of an increased groundwater input could not be confirmed in this study because groundwater levels remained constant during the 21-year study period in all study catchments, except Bergmyrbäcken where groundwater levels decreased. Thus, groundwater level changes are probably not the main driving force for any observed increase in pCO_2 in our study waters. Likewise, changed precipitation is probably not a key driver of the observed pCO_2 increase. Discharge increased in the catchment of Brunnsjön; hence, WRT would have decreased and thus could not explain the observed pCO_2 increase. However, no other key pCO_2 driving mechanism—DOC mineralization, primary production, or distribution changes within the carbonate system—changed in Brunnsjön during the 21-year study period; therefore, we cannot establish the key driver behind the observed pCO_2 increase in Brunnsjön. Discharge decreased in the stream Bergmyrbäcken, and although in situ C transformation processes are less important for CO_2 production in streams than in lakes (Jonsson et al. 2003, Hotchkiss et al. 2015), the decreased discharge may still have led to the observed pCO_2 increase.

Conclusions

Our detailed analyses of pCO_2 variation in 8 lakes and 5 streams in boreal Sweden during a 21-year period clearly demonstrates that no single main driver is behind the observed synchronous long-term pCO_2 increase in these inland waters. In lakes, stimulation of microbial mineralization by increased DOC and/or a change in the origin of the DOC toward more allochthonous DOC, which led to increased water color and subsequently to suppressed primary production as a response of light limitation, were found to be the more common drivers. In streams, the more dominating mechanisms were either carbonate system distribution changes due to decreased pH or a possible decreased primary production due to nutrient limitation. Increased in situ processing was more strongly pronounced in systems with high residence times (i.e., lakes), whereas streams are more variable and less consistent over time because of shorter water residence times. Thus, we conclude that the changes in long-term pCO_2 in lakes were largely DOC-driven, whereas changes in streams, although less pronounced, were more related to geochemical and hydrological aspects. To the best of our knowledge, this is the first study to show and explain consistent pCO_2 temporal trends in freshwater ecosystems, and our results should be taken into consideration when predicting future large scale CO_2 emissions from inland waters.

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