



# Regional Variability and Drivers of Below Ice CO<sub>2</sub> in Boreal and Subarctic Lakes

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## ABSTRACT

Northern lakes are ice-covered for considerable portions of the year, where carbon dioxide (CO<sub>2</sub>) can accumulate below ice, subsequently leading to high CO<sub>2</sub> emissions at ice-melt. Current knowledge on the regional control and variability of below ice partial pressure of carbon dioxide (*p*CO<sub>2</sub>) is lacking, creating a gap in our understanding of how ice cover dynamics affect the CO<sub>2</sub> accumulation below ice and therefore CO<sub>2</sub> emissions from inland waters during the ice-melt period. To narrow this gap, we identified the drivers of below ice *p*CO<sub>2</sub> variation across 506 Swedish and Finnish lakes using water chemistry, lake morphometry, catchment characteristics, lake position, and climate variables. We found that lake depth and trophic status were the most important variables explaining variations in below ice *p*CO<sub>2</sub> across the 506 lakes. Together, lake morphometry and water chemistry explained 53%

of the site-to-site variation in below ice *p*CO<sub>2</sub>. Regional climate (including ice cover duration) and latitude only explained 7% of the variation in below ice *p*CO<sub>2</sub>. Thus, our results suggest that on a regional scale a shortening of the ice cover period on lakes may not directly affect the accumulation of CO<sub>2</sub> below ice but rather indirectly through increased mobility of nutrients and carbon loading to lakes. Thus, given that climate-induced changes are most evident in northern ecosystems, adequately predicting the consequences of a changing climate on future CO<sub>2</sub> emission estimates from northern lakes involves monitoring changes not only to ice cover but also to changes in the trophic status of lakes.

**Key words:** CO<sub>2</sub>; winter limnology; ice cover; carbon; nutrients; lake depth.

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**Author contributions** B.A.D. designed the study, analyzed the data, and wrote the manuscript; P.K. and M.R. provided Finnish data; S.S. provided Swedish data; and G.A.W. helped preparing Swedish data and designing the study. All co-authors substantially contributed to data evaluation and the writing of the paper.

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## INTRODUCTION

Substantial emissions of carbon dioxide (CO<sub>2</sub>) into the atmosphere make inland waters critical components of atmospheric CO<sub>2</sub> budgets (Cole and others 2007; Tranvik and others 2009; Raymond and others 2013). Northern latitude lakes, in the boreal and arctic region, play a particularly important role in atmospheric CO<sub>2</sub> budgets, as a recent estimate of CO<sub>2</sub> emissions from boreal and

arctic inland waters (between latitudes 50°–90°N) suggests that 0.15 Pg C y<sup>-1</sup> is evaded into the atmosphere, of which 0.11 Pg C y<sup>-1</sup> is from arctic and boreal lakes and reservoirs (Aufdenkampe and others 2011). CO<sub>2</sub> emissions into the atmosphere are strongly influenced by the partial pressure of carbon dioxide ( $p\text{CO}_2$ ) at the water–atmosphere interface. Many studies on  $p\text{CO}_2$  in arctic and boreal watersheds have been conducted on a catchment (Kling and others 1991; Kelly and others 2001; Laurion and others 2010) and regional scale (Humborg and others 2010; Weyhenmeyer and others 2012; Campeau and Del Giorgio 2014); however, most of these studies have a sampling bias towards the open water season. This sampling bias is particularly problematic in northern latitudes, as lakes that may be ice-covered for up to 7 months of the year (Prowse and others 2012) can accumulate a substantial amount of CO<sub>2</sub> below ice, subsequently leading to high CO<sub>2</sub> emissions into the atmosphere at ice-melt (Striegl and others 2001; Huotari and others 2009; Ducharme-Riel and others 2015). A recent study by Karlsson and others (2013) found that in twelve small lakes in subarctic Sweden the CO<sub>2</sub> emitted at ice-melt accounted for 12–56% of the annual CO<sub>2</sub> emitted from these lakes. However, on a regional scale, the contribution of CO<sub>2</sub> emitted at ice-melt in terms of annual CO<sub>2</sub> emissions has yet to be documented.

The growing interest in global CO<sub>2</sub> emission estimates from inland waters emphasizes our need to consider the dynamics of lakes in a landscape context. Across the boreal and arctic region, studies have shown latitudinal variations in lake water CO<sub>2</sub> during the open water season to be related to differences in catchment characteristics, lake morphometry, dissolved organic carbon (DOC), nutrient concentrations, and climate variables (Kelly and others 2001; Sobek and others 2003; Rantakari and Kortelainen 2005; Kortelainen and others 2006, 2013; Roehm and others 2009; Lapierre and del Giorgio 2012; Ducharme-Riel and others 2015; Finlay and others 2015). Differences in climate and catchment characteristics influence the loading of carbon and nutrients bound in organic matter (OM) to lakes, and in turn differences in lake size and shape affect stratification and oxygenation and therefore the utilization and transformation of OM, including microbial respiration of DOC into CO<sub>2</sub>. The interaction between DOC and nutrients in relation to DOC degradation and subsequent CO<sub>2</sub> is still unclear on a regional scale (Roehm and others 2009) as nutrients have been found to increase productivity, decreasing CO<sub>2</sub> via photosynthesis (for example, del Giorgio and others 1999; Hanson

and others 2003), but also to stimulate the degradation of DOC, increasing  $p\text{CO}_2$  via respiration (for example, Huttunen and others 2003; Smith and Prairie 2004; Ask and others 2012). Further, climate drivers related to winter conditions, for example, ice cover duration and snow cover, are commonly neglected in landscape-scale studies. Thus, in order to understand regional scale variability of below ice  $p\text{CO}_2$ , climate variables related to the winter period need to be included.

During the winter period, the physical structure of ice-covered lakes differs from the open water season as ice limits wind-induced lake mixing and gas exchange. In late winter, when ice has reached its maximum growth, mainly heat flux from sediments and penetration of solar radiation through the ice drives circulation and water column mixing (Kirillin and others 2012). Snow accumulation on ice-covered lakes further reduces light availability, minimizing water column mixing and primary production below ice (Belzile and others 2001). Over the ice cover period, minimized mixing can lead to stratification, that is, where surface and bottom waters become disconnected with warmer waters (4°C) found near the bottom of the lake. During winter, CO<sub>2</sub> has been found to build up in hypolimnetic bottom waters, indicating that sediment respiration is an important source of CO<sub>2</sub> to ice-covered lakes (Striegl and Michmerhuizen 1998; Kortelainen and others 2006). Across 15 temperate and boreal ice-covered lakes, Ducharme-Riel and others (2015) found that benthic-derived CO<sub>2</sub> had a relatively greater role in shallow lakes, likely due to the larger sediment surface area-to-water volume ratio and smaller distance between bottom sediments and surface waters in shallow lakes (for example, Kelly and others 2001). Thus, the role of benthic-derived CO<sub>2</sub> in below ice CO<sub>2</sub> accumulation should further be investigated on a regional scale across different lake morphometry types.

Because climate-induced changes and associated feedbacks are accelerated in northern environments, particularly during winter (Callaghan and others 2010), understanding the regional drivers of  $p\text{CO}_2$  across ice-covered lakes is not only important for understanding present-day CO<sub>2</sub> emissions from lakes but also for predicting the consequences of a changing climate and cryosphere to future CO<sub>2</sub> emissions. Thus, the main aim of this study was to identify the drivers of below ice  $p\text{CO}_2$  on a large regional scale across lakes, and within a lake between surface and bottom waters. We hypothesized that ice cover length is significantly related to variations of below ice  $p\text{CO}_2$  across lakes. We

further hypothesized that within a lake below ice  $p\text{CO}_2$  is significantly higher in bottom waters compared to surface waters because sediment respiration acts as an additional source of CO<sub>2</sub> to bottom waters. Additionally, we hypothesized that  $p\text{CO}_2$  below ice is significantly higher in small and shallow lakes compared to large and deep lakes due to differences in the dilution of CO<sub>2</sub> in the water column. To test our hypotheses, we compiled data on below ice water chemistry, lake morphometry, catchment characteristics, lake position (see methods), and climate for 506 ice-covered lakes across Sweden and Finland.

## METHODS

### Study Region

Our study lakes were distributed along a north-south climate gradient between the latitudes 56°N and 69°N of boreal and subarctic/arctic region of Sweden and Finland, where permanent snow and ice cover duration ranges from 102 days in the south to 234 days in the north (Figure 1) and long-term average annual air temperatures range from +6.5 to −3.5°C. Lakes cover about 10% of the total area of Finland and 9% of the total area in Sweden (Raatikainen and Kuusisto 1990; Henriksen and others 1998). The topography of the study region is

relatively flat with higher elevations (up to 2100 m) in northwest of Sweden. The bedrock is predominantly Precambrian igneous and metamorphic rock. Land-cover patterns are similar in Sweden and Finland with highest agriculture area found in the south, extensive forest in the interior and tundra or open land in the north. In Finland, peatlands cover one-third of the land area, half of which have been ditched, mostly for forestry (Finnish Statistical Yearbook Forestry 1997).

### Database Description

The databases used in this study are available from the Swedish National Lake Inventory Programme (<http://www.slu.se/vatten-miljo>), and the published studies of Sobek and others (2003), Rantakari and Kortelainen (2005), and Kortelainen and others (2006), which together cover a broad geographical range spanning across Sweden and Finland and represent gradients in both trophic state and humic matter content: total phosphorus (TP) 11, 4–53 µg L<sup>−1</sup>; total nitrogen (TN) 460, 180–1400 µg L<sup>−1</sup>; and DOC 9, 3–21 mg L<sup>−1</sup> (all values are reported as median and 5 and 95 percentiles). The median lake area (LA) was 0.7 km<sup>2</sup> and more than 90% of the lakes were smaller than 100 km<sup>2</sup> (Table 1). Although most lakes were small, large lakes existed in the dataset (max LA of 1,538 km<sup>2</sup>), as Rantakari and Kortelainen (2005) included all lakes in Finland larger than 100 km<sup>2</sup>.

### $p\text{CO}_2$ Data

From the different databases, all lakes with below ice  $p\text{CO}_2$  measured during the ice cover period were included. The data were split into 5 groups, abbreviated Data<sub>Sweden</sub>, Data<sub>Swedendirect</sub>, Data<sub>Finland</sub>, Data<sub>FinlandTP</sub>, and Data<sub>Finlandlarge</sub> (Table 2), and described in the following.

Data<sub>Sweden</sub> ( $n = 224$ ) represents lakes from the Swedish National Lake Inventory Programme database from which  $p\text{CO}_2$  was calculated based on alkalinity (Alk), pH, water temperature ( $T_w$ ), and altitude (Alt) according to Weyhenmeyer and others (2012). To reduce the influence of acidification, recent liming, alkaline lakes, or algal bloom conditions which might bias  $p\text{CO}_2$  (for example, Humborg and others 2010), we excluded observations with  $\text{Alk} < 0$  or  $\geq 1$  mEq L<sup>−1</sup> or  $\text{pH} > 8$ . From this database, we selected below ice data by assuming that any sample collected between January and March with a surface water temperature  $\leq 4^\circ\text{C}$  was sampled below ice. If data for a particular lake were not available during January and March, we selected data from April only if the temperature

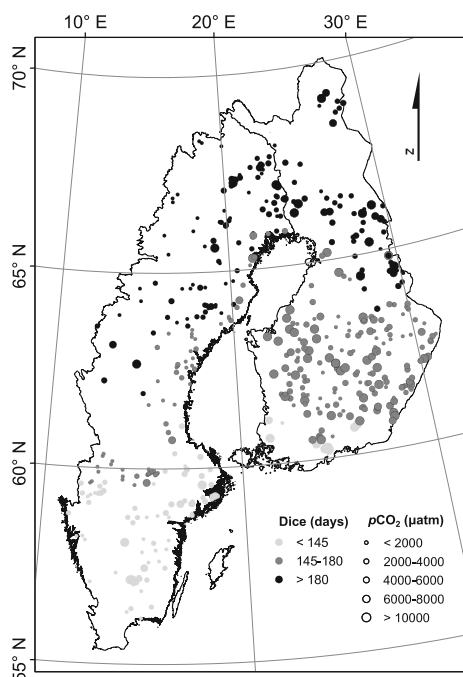


Figure 1. Map of Sweden and Finland depicting the locations of below ice surface water  $p\text{CO}_2$  ( $p\text{CO}_{2\text{surface}}$ ) and the corresponding ice cover duration ( $D_{\text{ice}}$ ).

**Table 1.** Distribution (Reported as Median, 5 and 95 Percentiles) of All Variables Used in the PLS<sub>all</sub> Analysis Including Below Ice Surface Water Chemistry, Lake Morphometry, Catchment Characteristics, Landscape Position, and Climate Parameters

Variables	<i>n</i>	Abbreviation	Median	5%	95%
Water chemistry					
Partial pressure of carbon dioxide (µatm) surface	506	$p\text{CO}_{2\text{surface}}^1$	2961	919	11162
Total phosphorus (µg L <sup>-1</sup> )	498	TP	11	4	53
Total nitrogen (µg L <sup>-1</sup> )	443	TN	460	180	1400
Dissolved organic carbon (mg L <sup>-1</sup> )	500	DOC	9	3	21
Conductivity (mS m <sup>-1</sup> )	498	Cond	5	2	11
Lake morphometry					
Lake area (km <sup>2</sup> )	506	LA	0.7	0.1	157
Volume (Mm <sup>3</sup> )	506	Vol	2.4	0.1	752
Average depth (m)	506	$Z_{\text{avg}}$	3.5	1.8	8.9
Shore line (km)	501	SL	5	1	437
Shoreline development	501	DL	2	1	10
Catchment characteristics					
Catchment area (km <sup>2</sup> )	464	CA	9	0.4	2170
Drainage ratio	464	DR	13	4	159
Agriculture (% of catchment)	450	Agr	0.6	0	19
Forest (% of catchment)	450	For	65	37	82
Wetland/peatland (% of catchment)	461	Peat	10	0	37
Urban (% of catchment)	450	Urb	0	0	2
Water (% of catchment)	450	Wat	11	2	27
Landscape Position					
Lake hydrology	380	LH	n/a	n/a	n/a
<i>X</i> -coordinate in WGS (°N)	506	<i>X</i> -coord	63	58	67
<i>Y</i> -coordinate in WGS (°E)	506	<i>Y</i> -coord	23	13	30
Altitude (m)	500	Alt	140	23	457
Climate variables					
Annual average temperature 1961–1990	506	$T_{\text{avg}}$	2.6	−1.4	5.5
Ice duration (days)	506	$D_{\text{ice}}$	166	121	207

<sup>1</sup> *Y*-variable used in PLS<sub>all</sub>.

**Table 2.** Description of the Five Data Groups Used in This Study

	Data <sub>Sweden</sub>	Data <sub>Swedendirect</sub>	Data <sub>Finland</sub>	Data <sub>FinlandTP</sub>	Data <sub>Finlandlarge</sub>
Country	Sweden	Sweden	Finland	Finland	Finland
$p\text{CO}_2$ method	Alk/pH	Direct	TIC/pH	TIC/pH	TIC/pH
Sample frequency	Multiple	One	One	One	Multiple
Sample month	Jan–Apr	Feb–Apr	Mar–Apr	Mar–Apr	Mar–Apr
Surface lakes (n)	224	42	175	28	37
Surface depth (m)	0.5–2	1	1	1	1
Bottom lakes (n)	55	32	165	22	37
Bottom depth (m)	1	1–2	0.2	0.2	1

Bottom depth is reported as the sampling depth above the sediment.

requirement of  $\leq 4^\circ\text{C}$  was met. We used surface water samples (in most cases at 0.5 m and in all cases  $< 2$  m) and bottom water samples (1 m above the deepest point of the lake). Whenever a lake was sampled more than once during the ice cover per-

iod (that is, within an ice cover period or across years), we calculated the maximum and median value for the ice cover period.

Data<sub>Swedendirect</sub> ( $n = 42$ ) is an additional set of Swedish lakes from Sobek and others (2003) from

which  $p\text{CO}_2$  was directly measured on an infrared gas analyzer. During the ice cover period, these lakes were sampled once between February and April 2001, surface water at 1 m below the ice and bottom water at 1–2 m above the sediment.

Data<sub>Finland</sub> ( $n = 175$ ) represents below ice  $p\text{CO}_2$  data from Kortelainen and others (2006) which were collected once during the winters of 1998–1999, with lakes sampled at the end of the winter stratification (approximately between the end of March and April).  $p\text{CO}_2$  was calculated from total inorganic carbon (TIC), pH, and  $T_w$ , using Henry's law constants corrected for temperature and atmospheric pressure (Plummer and Busenberg 1982). Headspace TIC was measured with gas chromatography, where water samples were acidified to convert all TIC to CO<sub>2</sub>. Water samples were collected at the deepest point of the lake, surface waters were sampled at a depth of 1 m and bottom waters were sampled at 20 cm from sediment surface. Because many of the Finnish lakes smaller than 100 km<sup>2</sup> are shallow, bottom water measurements made 20 cm from sediment surface were used to ensure that the difference between surface and bottom waters was represented.

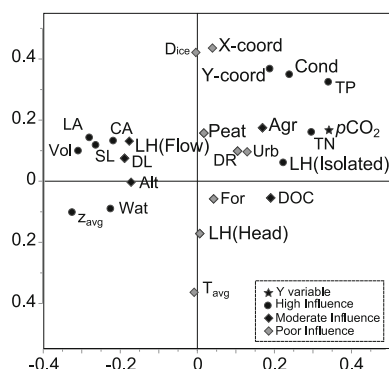
Data<sub>FinlandTP</sub> ( $n = 28$ ) is a subset of lakes sampled by Kortelainen and others (2006), which represent eutrophic lakes in the Nordic lake survey with the highest TP. Because a majority of Finnish lakes, as well as boreal lakes, are located in forested catchments with relatively minor human disturbance, eutrophic lakes are rather rare. Therefore, data from these lakes were included in analyzing the relationship between below ice  $p\text{CO}_2$  and other variables (Table 1; Figures 1, 2, 3, 4) but were

excluded from comparisons between Sweden and Finland (Table 3) and from the estimation of CO<sub>2</sub> emission because keeping them in the analysis would exert too strong an influence by quite a small population of lakes. The sampling techniques and  $p\text{CO}_2$  calculations follow Data<sub>Finland</sub>.

Data<sub>Finlandlarge</sub> ( $n = 37$ ) represent the largest lakes in Finland and are from Rantakari and Kortelainen (2005). The  $p\text{CO}_2$  was calculated from TIC, pH, and  $T_w$ , using Henry's law constants corrected for temperature and atmospheric pressure (Plummer and Busenberg 1982). TIC in the water was measured with a carbon analyzer. Below ice data were collected during the winters of 1998–1999, with lakes sampled at the end of the winter stratification (approximately between the end of March and April). Samples were collected at the deepest point of the lake, surface waters were sampled at a depth of 1 m, and bottom waters were sampled 1 m above the sediment. Lakes were sampled at least twice during the ice cover period, and thus a maximum and median value was used for these lakes. Data from these lakes were excluded from comparisons between Sweden and Finland because Data<sub>Sweden</sub> only included two lakes with an LA greater than 100 km<sup>2</sup>.

We acknowledge that the methodological differences between the five groups of data (Table 2) could cause deviation in  $p\text{CO}_2$  between groups, probably mainly between calculated versus measured  $p\text{CO}_2$  in acidic and organic-rich lakes (Abril and others 2014; Wallin and others 2014). We therefore examined a possible methodological bias by comparing  $p\text{CO}_2$  between Data<sub>Sweden</sub> and Data<sub>Sweden direct</sub>. We also compared Data<sub>Sweden</sub> and Data<sub>Finland</sub> to evaluate possible methodological issues between the Swedish and Finnish datasets (see “Results” section).

Altogether, surface water data from 506 ice-covered lakes were available for our analyses. Of the 506 lakes, some lakes did not have measurements from bottom waters and therefore a sub-database, of lakes with both surface and bottom water measurements, was created ( $n = 311$  lakes). To differentiate between below ice samples collected from surface waters and bottom waters, we used abbreviations  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$ , respectively.

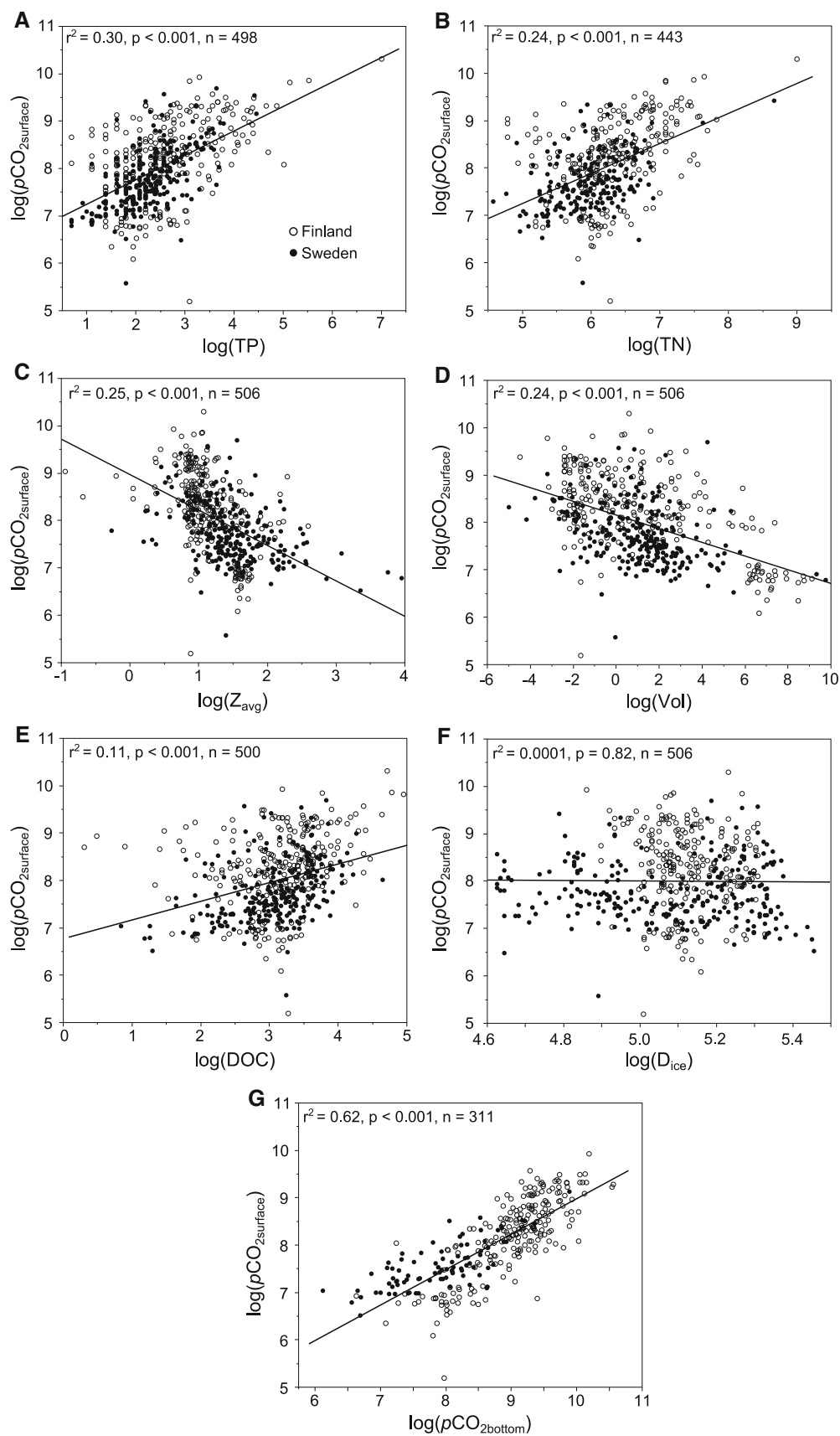


**Figure 2.** Partial least squares loading plot of below ice  $p\text{CO}_{2\text{surface}}$  observations for Sweden and Finland (PLS<sub>all</sub>;  $n = 506$ ). The loading plot depicts the correlation structure between  $p\text{CO}_{2\text{surface}}$  (Y-variable) and X-variables (for explanation of variable abbreviations, see Table 1). The greater the distance a variable is from the origin, the greater its overall influence (see Table 4 for VIP scores).

## Additional Variables

In addition to below ice  $p\text{CO}_2$ , we used data on pH,  $T_w$ , Alk, conductivity (Cond), TN, TP, and total organic carbon (TOC). TOC in boreal lakes usually contains  $97 \pm 5\%$  DOC (von Wachenfeldt and



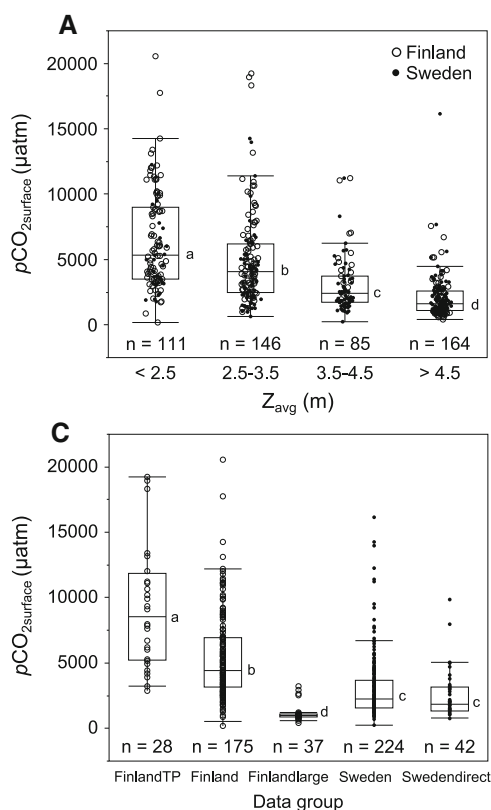


◀ **Figure 3.** Relationship between Swedish (*black circles*) and Finnish (*open circles*) below ice  $p\text{CO}_{2\text{surface}}$  and **A** TP ( $\log y = 6.7 + 0.5 \log x$ ), **B** TN ( $\log y = 4.1 + 0.6 \log x$ ), **C**  $Z_{\text{avg}}$  ( $\log y = 9.0 + 0.75 \log x$ ), **D** Vol ( $\log y = 8.2 + 0.14 \log x$ ), **E** DOC ( $\log y = 7.2 + 0.4 \log x$ ), **F**  $D_{\text{ice}}$  ( $\log y = 8.2 + 0.05 \log x$ ), and **G**  $p\text{CO}_{2\text{bottom}}$  ( $\log y = 1.5 + 0.75 \log x$ ). All data were log-transformed.

Tranvik 2008), thus TOC in this study can be seen as the equivalent to DOC. Water sampling and analyses were performed by the accredited water analysis laboratory at the Swedish University of Agricultural Sciences and by the accredited labo-

ratories of the Finnish Regional Environment Centers, according to standard limnological methods. Sobek and others (2003) carried out manual water sampling and analysis according to certified methods.

Further, GIS-derived data on lake morphometry, catchment characteristics, landscape position, and climate variables were included. Lake morphometry and catchment characteristics were acquired from topographic maps combined with land-use data on satellite images using the Arc View georeferencing software (for example, Kortelainen and others (2006)), for Finnish lakes, and from the



**Figure 4.** Distribution (median, 1st and 3rd quartile) of Swedish (*black circles*) and Finnish (*open circles*) below ice  $p\text{CO}_{2\text{surface}}$  between **A** average depth classes, **B** lake area classes, and **C** data groups. One outlier ( $> 20,000 \mu\text{atm}$ ) was removed for clarity. Wilcoxon each pair test results are letter-coded, where groups not sharing a letter are significantly different.

**Table 3.** Below Ice Surface Water Chemistry and Lake Morphometry Across Lakes in Sweden (Data<sub>Sweden</sub> and Data<sub>Swedendirect</sub>) and Finland (Data<sub>Finland</sub>) Reported as Median, 5 and 95 Percentiles

	Sweden <i>n</i> = 266	Finland <i>n</i> = 175
$p\text{CO}_{2\text{surface}}$ ( $\mu\text{atm}$ )	2168 (1060–7605)	4397 (1780–11441)
TP ( $\mu\text{g L}^{-1}$ )	9 (3–31)	12 (4–53)
TN ( $\mu\text{g L}^{-1}$ )	404 (177–820)	510 (180–1500)
DOC ( $\text{mg L}^{-1}$ )	9 (3–18)	9 (2–24)
Cond ( $\text{mS m}^{-1}$ )	4 (2–10)	4 (2–13)
LA ( $\text{km}^2$ )	0.7 (0.04–10)	0.2 (0.04–17)
Vol ( $\text{Mm}^3$ )	3 (0.1–83)	1 (0.1–66)
$Z_{\text{avg}}$ (m)	4 (2–11)	3 (2–7)
Alt (m)	203 (20–541)	116 (47–240)

Swedish National Lake Inventory Programme, as regularly released by the Swedish Meteorological and Hydrological Institute (SMHI, <http://www.smhi.se>), for Swedish lakes). Lake morphometry comprised data on lake surface area (LA; km<sup>2</sup>), volume (Vol; Mm<sup>3</sup>), average depth ( $Z_{avg}$ ; m) (calculated as Vol/LA), lake shoreline length (SL; km), and shoreline development length (DL) (calculated as the SL/ $\sqrt{2\pi LA}$ , Wetzel 2001). For lakes whose Vol was not in the registries, the Vol was determined using a calibrated lake volume estimate for Swedish lakes according to Sobek and others (2011) ( $\ln Vol = 1.39 + 1.12 * \ln LA$ ). For Finnish lakes, we recalibrated the Swedish estimate using data from 58 Finnish lakes ( $\ln Vol = 1.11 + 1.09 * \ln LA$ ;  $R^2 = 0.91$ ;  $n = 58$ ;  $p < 0.0001$ ).

Catchment characteristics included catchment area (CA; km<sup>2</sup>), drainage ratio (DR), % wetland/peatland in catchment (Peat), % agriculture in catchment (Agr), % urban in the catchment (Urb), % forest in catchment (For), and % water in the catchment including the lake itself and upstream water bodies (Wat). The drainage ratio (DR) was determined by dividing catchment area by lake area.

As an indicator of landscape position, in addition to altitude (Alt; m),  $X$ -coordinate ( $X$ -coord; °N), and  $Y$ -coordinate ( $Y$ -coord; °E), we defined lake hydrology (LH), following the protocol described in Martin and Soranno (2006), by assigning each lake to one of three categories; isolated, that is, have no connecting stream or lake (LH (Isolated)), headwater (LH(Head)), or flow through (LH(Flow)). Using ArcGIS (Version 10.1), each lake was assigned a category for the landscape position metric using the Swedish (VIVAN 2007, 298,215 lakes and 933,675 streams) and Finnish (53,511 lakes and 40,051 streams) network of rivers and lakes for flow-based modeling database. LH measures the overall surface hydrological position of a lake by incorporating connection both to lakes and streams. Altitude was calculated from a raster-based digital terrain model (DTM).

Climate variables, ice duration ( $D_{ice}$ ), and average annual air temperature ( $T_{avg}$ ) were assigned for each lake. Average annual air temperature for each lake was based on an averaged 1961–1990 temperature value (from SMHI for Sweden and Finnish meteorological institute (FMI, <http://www.ilmatiiteenlaitos.fi>) for Finland). Although regional ice cover data are available for Sweden and Finland, the scale of the data is coarse and therefore we used a more robust measure of ice cover duration for each individual lake. The number of days a lake is covered by ice ( $D_{ice}$ ) was calculated using an

air temperature function, which was calibrated and validated for Swedish lakes (Weyhenmeyer and others 2013):

$$D_{ice} = \frac{365.25d}{\pi} \times \arccos\left(\frac{T_m}{T_a}\right), \quad (1)$$

where  $d$  is days,  $T_m$  is the altitude-adjusted average air temperature, and  $T_a$  is the altitude-adjusted average air temperature amplitude.  $T_m$  and  $T_a$  were estimated (Weyhenmeyer and others 2013; for abbreviations see above) as

$$T_m = T_{avg} - \frac{Alt \times 0.6}{100} \quad (2)$$

$$T_a = -0.66 \times T_m + 14.32. \quad (3)$$

Although the primary aim of this study was to investigate regional scale patterns, we also addressed the temporal dimension by estimating the number of days a lake was ice-covered prior to sampling ( $S_{ice}$ ). This was done by subtracting the predicted ice-on date from the sampling date. Ice-on data for Swedish lakes were obtained from SMHI and for Finnish lakes from Finland's Environmental Administration (<http://www.ymparisto.fi/>), both providing regional ice-on dates for small and medium/large lakes. For lakes that were sampled more than once during the ice cover period ( $Data_{Sweden}$  and  $Data_{Finlandlarge}$ ),  $S_{ice}$  was calculated for the maximum  $pCO_2$ .

## Statistical Evaluation

In order to identify the drivers of below ice  $pCO_{2surface}$  and below ice  $pCO_{2bottom}$ , partial least square regression (PLS) was used. PLS, a method for relating how  $X$  correlates to  $Y$  by a linear multivariate model, offers a more robust technique compared to other multiple linear regression analyses as data can have missing values, they can co-correlate, and they do not need to be normally distributed (Eriksson and others 2006). In PLS,  $X$ -variables are classified according to their relevance in explaining  $Y$ , abbreviated as VIP values (Wold and others 1993). We considered VIP scores  $\geq 1.0$  as highly influential, between 0.8 and 1.0 moderately influential, and  $< 0.8$  less influential. The performance of the PLS model was expressed as  $R^2Y$ , representing how much of the variance in  $Y$  is explained by  $X$ , and  $Q^2Y$ , which is a measure of the predictive power of the PLS model. In the PLS models, data were log(10)-transformed if they were highly skewed (skewness  $> 2.0$  and min/max  $< 0.1$ ). An observation was excluded from the model if it fell outside the 99% confidence region



of the model (that is, hotelling  $T^2$ ) (Eriksson and others 2006). PLS modeling was carried out in the SIMCA-P 13.0 software (Umetrics AB, Umeå, Sweden).

We ran an initial PLS using data from all lakes with below ice  $p\text{CO}_{2\text{surface}}$  observations (out of 506 lakes, 9 observations fell outside the 99% confidence range of the model and therefore were removed), termed PLS<sub>all</sub>. A total of 22  $X$ -variables were included in the PLS<sub>all</sub> model with  $p\text{CO}_{2\text{surface}}$  set as the  $Y$ -variable (Table 1); because Alk, pH,  $T_w$ , and TIC were used to calculate  $p\text{CO}_2$ , they were removed from the PLS analyses to avoid autocorrelation. We ran an additional PLS model, using the maximum  $p\text{CO}_{2\text{surface}}$  for all lakes as the  $Y$ -variable and including an additional  $X$ -variable,  $S_{\text{ice}}$ , accounting for the days a lake was ice-covered prior to sampling.

A subset of 311 lakes, having both  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  data, was used to investigate if drivers of below ice  $p\text{CO}_{2\text{surface}}$  were different from the drivers of  $p\text{CO}_{2\text{bottom}}$ . Two separate PLS models were run for surface waters (PLS<sub>surface</sub>; out of 311 lakes 5 observations fell outside the 99% confidence range and were removed from the model) and bottom waters (PLS<sub>bottom</sub>; out of 311 lakes 6 observations fell outside the 99% confidence range and were removed from the model) with  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  set as the  $Y$ -variable, respectively.

Further statistical calculations were carried out in JMP, version 11.0.0. For determining the relationship between below ice  $p\text{CO}_{2\text{surface}}$  and below ice lake chemistry, lake morphometry, and ice cover variables, Pearson's correlation coefficients were used, where all the input data were log-transformed due to non-normal distribution of the data (Shapiro–Wilk test:  $p < 0.05$  indicating data are non-normally distributed). To test if below ice  $p\text{CO}_{2\text{bottom}}$  was significantly higher than  $p\text{CO}_{2\text{surface}}$ , we applied a matched-pair  $t$  test with log-transformed data where below ice  $p\text{CO}_{2\text{bottom}}$  and  $p\text{CO}_{2\text{surface}}$  were paired for each lake ( $n = 311$  lakes). To determine whether below ice  $p\text{CO}_{2\text{surface}}$  differed between data groups (Data<sub>Sweden</sub>, Data<sub>Swedendirect</sub>, Data<sub>Finland</sub>, Data<sub>FinlandTP</sub>, and Data<sub>Finlandlarge</sub>), mean lake depth (<2.5, 2.5–3.5, 3.5–4.5, >4.5 m), and lake area classes (<0.1, 0.1–1, 1–10, >10 km<sup>2</sup>), we applied non-parametric Wilcoxon tests and Wilcoxon each pair test where a significant difference between a class is reached when  $p < 0.05$ .

## RESULTS

### Below Ice $p\text{CO}_2$ in Surface Waters

Of the below ice  $p\text{CO}_{2\text{surface}}$  reported for the 506 lakes sampled, 504 were supersaturated in CO<sub>2</sub>. Highest below ice  $p\text{CO}_{2\text{surface}}$  was found in small eutrophic Finnish lakes. In Finland (that is, Data<sub>Finland</sub>), below ice  $p\text{CO}_{2\text{surface}}$  was on average about twice as high as in Sweden (Table 3). Also below ice nutrients in surface waters (median TP of 9 and 12 µg L<sup>-1</sup> and TN of 404 and 510 µg L<sup>-1</sup>, for Sweden and Finland, respectively) were higher in Finland than in Sweden, while DOC was similar (median DOC of 9 mg L<sup>-1</sup> for both countries). Further, Finnish lakes were generally smaller and shallower, while the Swedish lakes covered a larger altitude range (Table 3).

When we modeled variations in below ice  $p\text{CO}_{2\text{surface}}$  across all 506 Finnish and Swedish lakes (PLS<sub>all</sub>), we received a good model predictability ( $Q^2 = 0.58$ ) with two components able to explain 60% of the variation in  $p\text{CO}_{2\text{surface}}$  ( $R^2Y = 0.60$ ). In the PLS<sub>all</sub> model, the first component (that is, horizontal axis) explained 53% of the variation in  $p\text{CO}_{2\text{surface}}$ , representing lake morphometry and water chemistry variables (Figure 2). Lake morphometry ( $Z_{\text{avg}}$ , Vol, LA, SL, DL) was negatively related to  $p\text{CO}_{2\text{surface}}$ , whereas water chemical variables (TP, TN, Cond, DOC) were positively related to  $p\text{CO}_{2\text{surface}}$ . The second component (that is, vertical axis) represented regional climate (i.e.,  $D_{\text{ice}}$  and  $T_{\text{avg}}$ ) and latitude and only explained 7% of the variation in below ice  $p\text{CO}_{2\text{surface}}$ . When ice cover duration prior to sampling ( $S_{\text{ice}}$ ) was included as an additional  $X$ -variable in the PLS for the prediction of maximum  $p\text{CO}_{2\text{surface}}$ , we found that the model remained similar, without an influence of  $S_{\text{ice}}$  on the model ( $Q^2 = 0.59$ ,  $R^2Y = 0.61$ ).

Overall, TP was the most influential variable, followed by lake morphometry ( $Z_{\text{avg}}$ , Vol, LA, SL), TN, Cond, CA, LH(isolated), and Y-coord (Table 4). TP alone was able to explain 30% of the variation in  $p\text{CO}_{2\text{surface}}$  (Figure 3A). Also TN (Figure 3B),  $Z_{\text{avg}}$  (Figure 3C), and Vol (Figure 3D) had a high explanatory power. DOC only had a moderate influence on the PLS<sub>all</sub> model (Table 4), and by directly relating DOC to  $p\text{CO}_{2\text{surface}}$ , we found a weak positive relationship (Figure 3E). Average depth was negatively related to TN ( $r^2 = 0.09$ ,  $p < 0.001$   $n = 500$ ), TP ( $r^2 = 0.08$ ,  $p < 0.001$   $n = 500$ ), and DOC ( $r^2 = 0.07$ ,  $p < 0.001$   $n = 500$ ).  $D_{\text{ice}}$  was not an influential variable for the model

**Table 4.** Variable Importance in the Projection (VIP) Scores for Partial Least Squares (PLS) Models

	Variables	PLS <sub>all</sub>	PLS <sub>surface</sub>	PLS <sub>bottom</sub> <sup>1</sup>
Water chemistry	TP	<b>+1.7</b>	<b>+1.3</b>	<b>+1.5</b>
	TN	<b>+1.4</b>	<b>+1.3</b>	<b>+1.9</b>
	DOC	+0.9	+0.7	+0.9
	Cond	<b>+1.2</b>	<b>+1.1</b>	<b>+1.3</b>
Lake morphometry	LA	<b>-1.3</b>	<b>-1.4</b>	<b>-1.1</b>
	Vol	<b>-1.5</b>	<b>-1.5</b>	<b>-1.2</b>
	Z <sub>avg</sub>	<b>-1.5</b>	<b>-1.5</b>	<b>-1.6</b>
	SL	<b>-1.3</b>	<b>-1.4</b>	<b>-1.0</b>
	DL	-0.9	<b>-1.1</b>	+0.7
Catchment characteristics	CA	<b>-1.0</b>	<b>-1.2</b>	<b>-1.0</b>
	DR	+0.5	+0.5	+0.5
	Agr	+0.8	+0.7	+0.8
	For	+0.3	+0.3	+0.7
	Peat	+0.3	-0.2	+0.2
	Urb	+0.6	+0.7	+0.7
	Wat	<b>-1.0</b>	<b>-1.1</b>	<b>-1.0</b>
	LH (isolated)	<b>+1.1</b>	<b>+1.3</b>	<b>+1.2</b>
Landscape position	LH (head)	-0.3	-0.3	+0.1
	LH (flow)	-0.8	<b>-1.2</b>	<b>-1.0</b>
	X-coord	+0.7	+0.6	+0.7
	Y-coord	<b>+1.0</b>	<b>+1.0</b>	<b>+1.5</b>
	Alt	-0.8	-0.7	-0.7
Climate	T <sub>avg</sub>	-0.6	+0.3	-0.3
	D <sub>ice</sub>	+0.7	+0.3	+0.3

VIP scores are classified according to the relevance of X-variable in explaining Y; VIP scores  $\geq 1.0$  were considered highly influential (bolded), between 0.8 and 1.0 moderately influential, and  $\leq 0.8$  less influential. Signs, - and +, indicate negative and positive relationships to  $p\text{CO}_2$ , respectively. The highest VIP score for each model is bold italic.

<sup>1</sup> Y-variable  $p\text{CO}_{2\text{bottom}}$

performance (Table 4), and when relating  $D_{\text{ice}}$  to  $p\text{CO}_{2\text{surface}}$  we found no relationship (Figure 3F). However,  $D_{\text{ice}}$  was significantly positively related to Cond ( $r^2 = 0.30$ ,  $p < 0.001$ ,  $n = 498$ ), DOC ( $r^2 = 0.14$ ,  $p < 0.001$ ,  $n = 500$ ), TN ( $r^2 = 0.13$ ,  $p < 0.001$ ,  $n = 443$ ), and TP ( $r^2 = 0.01$ ,  $p < 0.01$ ,  $n = 498$ ).

Median below ice  $p\text{CO}_{2\text{surface}}$  was significantly different across varying lake areas (Wilcoxon test:  $\chi^2 = 85$ ,  $p < 0.0001$ ,  $n = 506$ ) and average depths (Wilcoxon test:  $\chi^2 = 180$ ,  $p < 0.0001$ ,  $n = 506$ ). According to the Wilcoxon each pair test, median below ice  $p\text{CO}_{2\text{surface}}$  was significantly different between each lake area and average depth class, that is,  $p\text{CO}_{2\text{surface}}$  was higher in shallow lakes ( $Z_{\text{avg}} < 2.5$  m) compared to deep lakes ( $Z_{\text{avg}} > 4.5$  m) (Figure 4A) and in small lakes ( $\text{LA} < 0.1 \text{ km}^2$ ) compared to large lakes ( $\text{LA} > 10 \text{ km}^2$ ) (Figure 4B).

### Below Ice $p\text{CO}_2$ in Bottom Waters and Its Relation to $p\text{CO}_2$ in Surface Waters

From a subset of 311 lakes having  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  measurements, two separate PLS mod-

els were run for surface waters (PLS<sub>surface</sub>) and bottom waters (PLS<sub>bottom</sub>) with  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  set as the Y-variable, respectively. The PLS<sub>surface</sub> model explained 64% ( $R^2Y = 0.64$ ) of the variation in  $p\text{CO}_{2\text{surface}}$  and model predictability was good ( $Q^2 = 0.61$ ). The model predictability of PLS<sub>bottom</sub> was similar ( $Q^2 = 0.64$ ) and explained 67% ( $R^2Y = 0.67$ ) of the variation in  $p\text{CO}_{2\text{bottom}}$ . The major difference between PLS<sub>bottom</sub> and PLS<sub>surface</sub> was that  $p\text{CO}_{2\text{bottom}}$  was slightly more influenced by water chemical variables, in particular TN, and  $p\text{CO}_{2\text{surface}}$  was slightly more influenced by lake morphometric variables, in particular  $Z_{\text{avg}}$  (Table 4).

When relating  $p\text{CO}_{2\text{bottom}}$  to  $p\text{CO}_{2\text{surface}}$ , we found that 62% of the variation in  $p\text{CO}_{2\text{surface}}$  could be explained by  $p\text{CO}_{2\text{bottom}}$  (Figure 3G). We found that the residuals of the regression (residuals  $\log(p\text{CO}_{2\text{surface}})$ ) were related to lake morphometry (Vol:  $r^2 = 0.11$ ,  $p < 0.001$ ,  $n = 311$ ; LA:  $r^2 = 0.10$ ,  $p < 0.001$ ,  $n = 311$ ) supporting the concept that  $p\text{CO}_{2\text{surface}}$  is a function of  $p\text{CO}_{2\text{bottom}}$  and the recipient volume of water.  $p\text{CO}_{2\text{bottom}}$  differed significantly from  $p\text{CO}_{2\text{surface}}$  (matched-pair  $t$  test

result:  $t = 22.7$ ,  $p < 0.05$ , number of pairs = 311) with median  $p\text{CO}_{2\text{bottom}}$  more than twice as high as the median  $p\text{CO}_{2\text{surface}}$  (7187 and 3206  $\mu\text{atm}$ , respectively). Further, when relating maximum  $p\text{CO}_2$  to  $S_{\text{ice}}$ , we found a weak positive relationship for  $p\text{CO}_{2\text{surface}}$  ( $r^2 = 0.01$ ,  $p = 0.037$ ,  $n = 506$ ) and a stronger positive relationship for  $p\text{CO}_{2\text{bottom}}$  ( $r^2 = 0.13$ ,  $p < 0.01$ ,  $n = 311$ ).

## Comparison Between Data Groups

Comparing all five data groups, we found that  $p\text{CO}_{2\text{surface}}$  was significantly different between groups (Wilcoxon test:  $\chi^2 = 199$ ,  $p < 0.0001$ ,  $n = 506$ ). When comparing  $\text{Data}_{\text{Swedendirect}}$ , i.e., directly measured  $p\text{CO}_2$ , with  $\text{Data}_{\text{Sweden}}$ , that is, calculated  $p\text{CO}_2$ , we did not find a statistically significant difference (Wilcoxon each pair test:  $p > 0.05$ ). In contrast, we found a significant difference between the Swedish dataset,  $\text{Data}_{\text{Sweden}}$ , and the Finnish dataset,  $\text{Data}_{\text{Finland}}$ , (Wilcoxon each pair test:  $p < 0.0001$ ) with higher below ice  $p\text{CO}_{2\text{surface}}$  found for  $\text{Data}_{\text{Finland}}$  (Figure 4C).

## DISCUSSION

### Drivers of Below Ice $p\text{CO}_2$ on a Spatial Scale

Using a multivariate approach comparing 506 ice-covered lakes across Sweden and Finland, we were able to identify key variables influencing below ice  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$ . Lake morphometry ( $Z_{\text{avg}}$ , Vol, LA, SL) and lake water chemistry (TP, TN, Cond) were most important in explaining variations of below ice  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  across lakes. Lake morphometric variables were situated opposite to water chemical variables along the first component axis in the PLS loading plot, suggesting that these variables are tightly negatively associated with each other and have more influence on  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  than regional scale climate variables such as ice cover length and air temperature (that is, variables that lie along the secondary principal component axis; Figure 2). Negative relationships between water chemistry and  $Z_{\text{avg}}$  reflect that small shallow lakes have proportionally higher chemical concentrations during winter compared to large deep lakes.

The identified drivers of below ice  $p\text{CO}_2$  are well known drivers for water chemical concentrations and for  $p\text{CO}_2$  during the open water season (for example, Kelly and others 2001; Sobek and others 2003). Lake morphometric variables reflect the degree of dilution, water mixing, catchment, and

lake internal loading. Although these processes are minimized during the ice cover period, they are very important in determining initial  $p\text{CO}_2$  in the water column before ice cover disconnects the catchment and the atmosphere from the lake. Many processes determine initial  $p\text{CO}_2$  in the water before the ice cover period begins, one being the intensity and length of autumn water turnover. For example, a complete autumn turnover prior to ice-on vents CO<sub>2</sub> from the lake, resulting in similar  $p\text{CO}_2$  throughout the water column (López Bellido and others 2009). If ice-on comes early, an incomplete autumn turnover will result in elevated  $p\text{CO}_2$  during winter (particularly in bottom waters). In addition, high precipitation prior to ice-on transports OM from the catchment to the lake, enhancing DOC and nutrient availability in the lake and hence conditions for degradation during ice cover (for example, Rantakari and Kortelainen 2005). Huotari and others (2009) support these ideas, as they found that longer autumn turnover resulted in lower CO<sub>2</sub> below ice, and a wet autumn resulted in elevated below ice CO<sub>2</sub> compared to a dry autumn.

Therefore, potentially our below ice  $p\text{CO}_2$  variations could simply reflect spatial  $p\text{CO}_2$  variations that already occur during the open water season. However, below ice  $p\text{CO}_{2\text{surface}}$  with a median of 2168  $\mu\text{atm}$  in Swedish lakes and a below ice  $p\text{CO}_{2\text{bottom}}$  with a median of 2853  $\mu\text{atm}$  in Swedish bottom waters (in Finland 4397 and 9943  $\mu\text{atm}$ , respectively, Table 3) were substantially higher than the median  $p\text{CO}_2$  of less than 1500  $\mu\text{atm}$  that was observed in the same regional area during the open water season (Weyhenmeyer and others 2012). Consequently, CO<sub>2</sub> is most probably further produced within the lake during the ice cover period.

CO<sub>2</sub> can be produced below ice cover by microbial mineralization of OM, mainly in the sediment where the availability of OM and nutrients regulates the microbial CO<sub>2</sub> production (del Giorgio and others 1999; Kortelainen and others 2006). Because we observed a positive correlation between  $p\text{CO}_{2\text{bottom}}$  and TP, TN, and DOC, we suggest that microbial mineralization in the water column and sediments frequently occurs below ice cover in our study lakes. Because additional nutrients and DOC can be released from the sediments into bottom waters under changed bottom water redox conditions (Mortimer 1941; Gonsior and others 2013; Yang and others 2014), microbial respiration in bottom waters might even be enhanced (Peter and others, submitted). Our study suggests that

nutrients have a positive effect on  $p\text{CO}_{2\text{bottom}}$  in ice-covered lakes; however, if this relationship is causal (enhanced DOC degradation) or correlative (sediment release of both nutrients and  $\text{CO}_2$ ), it needs to be investigated further.

Additional  $\text{CO}_2$  below ice compared to the open water season can also result from groundwater/weathering inputs (Striegl and Michmerhuizen 1998; Humborg and others 2010), here indicated by a positive correlation between  $p\text{CO}_{2\text{bottom}}$  and Cond. Although groundwater can be an important source of  $\text{CO}_2$ , particularly during early and late ice cover, it is likely that most of the groundwater entering the lake is from shallow soil water flow, opposed to deep groundwater flow. As this shallow soil water is generally below  $4^\circ\text{C}$ , it entrains the surficial layer of the lake and usually does not mix with bottom waters (Kirillin and Terzhevik 2011). Further, deep groundwater flow through sediments into lake bottom waters is less probable, confirmed by Pulkkanen and Salonen (2013) who found that the contribution of groundwater to deep lake waters during winter was minimal in five ice-covered Finnish lakes.

Our results did not support the hypothesis that below ice  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  is significantly related to variations in ice cover duration across lakes. In general, regional scale climate (that is,  $T_{\text{avg}}$ ) effects on below ice  $p\text{CO}_{2\text{surface}}$  or  $p\text{CO}_{2\text{bottom}}$  were lacking. These results were unexpected because ice cover length and temperature are known to regulate landscape-scale processes (that is, growing season length). A similar case of a lacking direct temperature effect along a large spatial gradient on lake  $p\text{CO}_2$  was found during the open water season for a global lake database (Sobek and Tranvik 2005), and it was reasoned that the lack of relationship was due to the fact that the temperature sensitivity of a process depends on the substrate supply (Pomeroy and Wiebe 2001). On a regional scale, substrate supply can be quite variable, demonstrated by the wide range of TP, TN, and DOC concentrations found in this study (Table 1), whereas on a local or individual lake scale the variation in substrate supply is smaller. Therefore, on a local and also on a temporal scale we might expect ice cover length and climate to play a much more important role in influencing below ice  $p\text{CO}_2$ . In this study, we may have missed some localized climate variability as snow cover and ice thickness data were not available for most of the lakes. Nonetheless, in a study of below ice  $p\text{CO}_2$  by Sobek and others (2003), localized climate variables (air temperature, precipitation, ice thickness, and snow cover) were less influential on be-

low ice  $p\text{CO}_2$  compared to physiochemical and morphometric variables.

Our analyses had one temporal component included and this was the duration of ice cover prior to sampling ( $S_{\text{ice}}$ ). We found that  $S_{\text{ice}}$  had a stronger relationship with  $p\text{CO}_{2\text{bottom}}$  compared to  $p\text{CO}_{2\text{surface}}$ . In a recent study, Denfeld and others (2015) found that in a small boreal lake below ice  $\text{CO}_2$  did not constantly increase throughout the winter period, but remained constant during the late winter period. This suggests that in some lakes there might only be a small change in surface water  $\text{CO}_2$  in late winter below ice, and therefore might explain why we did not find a strong relationship between  $S_{\text{ice}}$  and  $p\text{CO}_{2\text{surface}}$  in this study. However, this may not be true for all lakes (for example, Huotari and others 2009), particularly in bottom waters, as sediment respiration is likely to continue to contribute to  $\text{CO}_2$  accumulation throughout the whole winter (for example, Ducharme-Riel and others 2015). This suggests that on a temporal scale, ice cover days prior to sampling may be more important to  $p\text{CO}_{2\text{bottom}}$ , whereas other factors, such as substrate supply and light below ice, may be more important in determining  $p\text{CO}_{2\text{surface}}$  in late winter.

### Geographical Differences Including Hot Spots of Below Ice $p\text{CO}_2$

In agreement with our hypothesis, below ice  $p\text{CO}_2$  was generally highest in small shallow lakes and in bottom waters of lakes, suggesting that these waters are hot spots of gas accumulation during the ice cover season. Small shallow lakes and bottom waters have earlier been identified as biogeochemical hotspots during the open water season (Hanson and others 2007; Humborg and others 2010; Weyhenmeyer and others 2012), whereas this study further stresses their importance in carbon cycling during the ice cover period. Small shallow lakes dominate the Swedish (Sobek and others 2011) and Finnish landscape (Kuusisto and Hakala 2007) and are the most abundant lake type on earth (Downing and others 2006; Verpoorter and others 2014). Further, small shallow lakes are important systems to monitor in a changing cryosphere as they are particularly sensitive to changes in temperature and precipitation (Rautio and others 2011).

Across Sweden and Finland, we found that below ice  $p\text{CO}_2$  variation was more pronounced longitudinally (east–west) than latitudinally (north–south). Longitudinally, comparing below ice water chemistry and lake morphometry between Sweden and



Finland (Table 3), we found that  $p\text{CO}_2$ , TP, and TN were higher in Finland. Finnish lakes had twice as high median  $p\text{CO}_{2\text{surface}}$  and three times as high median  $p\text{CO}_{2\text{bottom}}$  compared to Swedish lakes. Part of this variation may be due to differences in lake selection, sampling approaches, and methodology (Table 2). For example, Finnish lakes could have higher  $p\text{CO}_2$  because they were on average sampled later into the ice cover period, and for bottom waters some Finnish lakes ( $\text{Data}_{\text{Finland}}$  and  $\text{Data}_{\text{FinlandTP}}$ ) were sampled closer to the sediment (0.2 m above) than Swedish lakes (1–2 m above) (Table 2). If the selection of lakes, sampling approaches, and methods between the two countries were exactly the same, the observed differences in below ice water chemistry between Sweden and Finland might have been smaller, yet it is likely that Finnish lakes would still have higher below ice  $p\text{CO}_2$  because of different lake characteristics. Previous comparisons between the two countries during the open water period have found that Swedish lakes have higher  $p\text{CO}_2$  than Finnish lakes (Weyhenmeyer and others 2012). However, for Finnish lakes the average winter CO<sub>2</sub> has been reported to be five times that of the average summer CO<sub>2</sub> (Kortelainen and others 2006), while the difference between winter and summer CO<sub>2</sub> may rather be around a factor of 2 for Swedish lakes (Sobek and others 2003; median below ice  $p\text{CO}_2$  of 2244  $\mu\text{atm}$  in this study and median open water  $p\text{CO}_2$  of 1048  $\mu\text{atm}$  reported in Weyhenmeyer and others (2012)). The fact that Finnish lakes were on average smaller and shallower than Swedish lakes (Table 3; Figure 4A) likely resulted in the observed higher below ice  $p\text{CO}_2$  and nutrients found in Finland. Concerning the differences in methods for determination of  $p\text{CO}_2$ , we did not find a significant difference between directly measured and calculated  $p\text{CO}_2$  for Swedish lakes, indicating that any potential methodological bias to the data could not be detected statistically.

Latitudinally, across Sweden and Finland, the inputs of DOC, TP, and TN from the catchment are smaller in the north compared to the south (for example, Kortelainen and others 1997) and are related to ice cover length. In northern Scandinavia, low temperatures and a shorter growing season limit litter production in the catchment and seasonally frozen soils limit OM loading to adjacent surface waters (Laudon and others 2012). Because lake water chemistry was related to  $p\text{CO}_{2\text{surface}}$  and  $p\text{CO}_{2\text{bottom}}$  in our PLS models, a latitude influence on below ice  $p\text{CO}_2$  was expected. Nevertheless, in our PLS models, latitude ( $X$ -coord) did not have a large influence on below ice  $p\text{CO}_2$ . A possible explanation is that clear water lakes (which can be

found in northern Sweden) may have relatively higher  $p\text{CO}_2$  during the winter, as benthic algae produced during the open water season can act as an additional source to degradation (Karlsson and others 2008). Further, although land-cover types generally differ with latitude across Sweden and Finland (for example, highest agriculture area found in the south), the land-cover type in the catchment (Agr, For, Peat, and Urb) was not an important variable in explaining below ice  $p\text{CO}_2$ . The OM input to lakes is likely a mixture of different sources based on the proportion and connectivity of the different land-cover types in the catchment, and therefore a single important land-cover type may not be individually important on below ice  $p\text{CO}_2$ .

### Implications of Below Ice $p\text{CO}_2$ on CO<sub>2</sub> Emissions at Ice-Melt

To provide a broad estimate of the importance of the ice-melt period across Swedish and Finnish lakes, we estimated the CO<sub>2</sub> emission during the ice-melt period. Based on the relationship between below ice  $p\text{CO}_{2\text{surface}}$  and lake area (Figure 4B), we estimate that the CO<sub>2</sub> emission across Swedish and Finnish lakes during the ice-melt period corresponds to 1.2–1.5 Tg C  $\text{y}^{-1}$  (based on lake area size classes <0.1, 0.1–1, 1–10, >10 km<sup>2</sup>, lake area adjusted  $k_{600}$  from Raymond and others (2013), lake area-specific below ice  $p\text{CO}_{2\text{surface}}$  of this study (median  $p\text{CO}_{2\text{surface}}$  and maximum  $p\text{CO}_{2\text{surface}}$  for lower and upper range, respectively), and an ice-melt period lasting for 19 days according to Denfeld and others (2015) for a small boreal lake in Sweden). At ice-melt, CO<sub>2</sub> accumulated in bottom waters can be emitted to the atmosphere by bottom water convective turnover. If we assume that CO<sub>2</sub> accumulated in the bottom waters during ice cover is emitted at ice-melt, the CO<sub>2</sub> emission during the ice-melt period would double to 2.5–2.9 Tg C  $\text{y}^{-1}$ . It is likely that these estimates are conservative as  $p\text{CO}_2$  measurements were made in late winter prior to ice-melt and do not account for the dynamic ice-melt period when CO<sub>2</sub> from the catchment can be sourced to the lake (Miettinen and others 2014; Denfeld and others 2015). Nevertheless, the amount of 1.2–2.9 Tg C  $\text{y}^{-1}$  emitted from Swedish and Finnish lakes at ice-melt is comparable to estimates of average Holocene C accumulation in boreal lake sediments, 2–3 Tg C  $\text{y}^{-1}$  (Kortelainen and others 2004), that is, annually, as much C was estimated to be emitted from Swedish and Finnish lakes at ice-melt as was estimated to annually accumulate across all boreal lakes during the Ho-



locene. Increased human activity during recent decades has, however, resulted in increasing accumulation rates (Anderson and others 2013).

The CO<sub>2</sub> emitted from Swedish and Finnish lakes at ice-melt contributed to 18–30% of the annual CO<sub>2</sub> emission, of which the open water CO<sub>2</sub> emission corresponded to 6.63 Tg C y<sup>-1</sup> (based on lake area size classes <0.1, 0.1–1, 1–10, >10 km<sup>2</sup>, lake area adjusted  $k_{600}$  from Raymond and others (2013), open water lake area  $p\text{CO}_{2\text{surface}}$  from Humborg and others (2010) for Swedish lakes and from Kortelainen and others (2006) and Rantakari and Kortelainen (2005) for Finnish lakes, and an open water period lasting for 199 days, that is, the median open water period in this study). A contribution of 18–30% annual CO<sub>2</sub> flux from Swedish and Finnish lakes at ice-melt is similar to previous estimates (22%) for Finnish lakes, based on different assumptions (Kortelainen and others 2006), and is a bit less than previously found for individual lakes (ranging from 3 to 80% in 15 temperate and boreal lakes and from 12 to 56% in 12 subarctic Swedish lakes, Ducharme-Riel and others (2015) and Karlsson and others (2013), respectively), indicating that CO<sub>2</sub> emission during ice-melt in individual lakes may have an even stronger impact on annual CO<sub>2</sub> emission from lakes than estimated in this study.

## CONCLUSION

Our study demonstrates that lake water chemistry and lake morphometry were more important factors in determining below ice  $p\text{CO}_2$  on a regional scale than climate variables (air temperature and ice cover duration). CO<sub>2</sub> accumulates in ice-covered lakes, most probably due to nutrient- and OM-driven microbial mineralization in the sediments, and CO<sub>2</sub> inputs from the catchment prior to ice-melt. We conclude that on a regional scale, carbon cycling in ice-covered lakes and subsequent CO<sub>2</sub> emission at ice-melt are important components of annual CO<sub>2</sub> emission estimates. Thus, given the potential for significant ecosystem changes to ice-covered lakes, adequately integrating the ice cover period in global CO<sub>2</sub> emission estimates involves monitoring changes not only to ice cover but also to changes in the trophic status of lakes.

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