

News date: 16th Nov., 2015

compiled by Dr. Alvarinho J. Luis

Northern lakes act as CO₂ chimneys in a warming world

Source: Uppsala Universitet

Summary: Many of the world's approximately 117 million lakes act as wet chimneys releasing large amounts of the greenhouse gas carbon dioxide into the atmosphere. The most recent estimates show that carbon dioxide emissions from the world's lakes, water courses and reservoirs are equivalent to almost a quarter of all the carbon dioxide produced by burning fossil fuels.

A new study published in *Nature Geoscience* shows that lakes in the northern hemisphere will probably release much more carbon dioxide due to global climate changes. The investigation, based on a large amount of data from over 5,000 Swedish lakes, has been led by Prof. Gesa Weyhenmeyer of Uppsala University, in collaboration with colleagues in Denmark, the Netherlands and Brazil.

There are two main CO_2 sources in water. It can either be produced by microorganisms, especially in lakes, or it can be washed into the water from the surrounding area. In order to make statements on future carbon dioxide emissions, it is very important to know where the carbon dioxide in the water comes from. Earlier test results from individual aquatic systems showed that CO_2 is mainly produced by micro-organisms in the lake itself. In the present study, researchers show for the first time that this applies to less than half of all Swedish lakes.

In most Swedish lakes, the CO_2 originates from the surrounding land from where it is washed into water courses and then into lakes which then release CO_2 into the atmosphere, says Weyhenmeyer.

The study also shows that the amount of CO_2 produced in the lakes and from the influx of CO_2 from the ground both increase in warmer and nutrient-rich climate zones. For this reason, changes in land use are also very significant. The study reports that a small lake in southern Sweden surrounded by farmland releases twice as much CO_2 as small lake in northern Sweden surrounded by forest.

There is a considerable risk that CO_2 emissions from lakes and water courses, especially those in northern Scandinavia, Canada and Russia, will increase as the climate warms up. And it is in these areas that most of the lakes are located,

says Weyhenmeyer.

Journal Reference: G. A. Weyhenmeyer, S. Kosten, M. B. Wallin, L. J. Tranvik, E. Jeppesen, F. Roland. Significant fraction of CO2 emissions from boreal lakes derived from hydrologic inorganic carbon inputs. *Nature Geoscience*, 2015; DOI: 10.1038/ngeo2582. pdf reprint follows.....

Significant fraction of CO₂ emissions from boreal lakes derived from hydrologic inorganic carbon inputs

Gesa A. Weyhenmeyer^{1*}, Sarian Kosten², Marcus B. Wallin^{1,3}, Lars J. Tranvik¹, Erik Jeppesen^{4,5} and Fabio Roland⁶

Annual CO₂ emissions from lakes and other inland waters into the atmosphere are estimated to almost entirely compensate the total annual carbon uptake by oceans¹⁻³. CO₂ supersaturation in lakes, which results in CO₂ emissions, is frequently attributed to CO₂ produced within the lake⁴⁻⁸. However, lateral inorganic carbon flux through watersheds can also be sizeable⁹⁻¹¹. Here we calculated lake surface water CO₂ concentrations and emissions using lake pH, alkalinity and temperature from a compilation of data from 5,118 boreal lakes¹². Autumn surface water CO₂ concentrations and CO₂ emissions from the 5,118 lakes co-varied with lake internal autumn CO₂ production. However, using a mass balance approach we found that CO₂ emission in the majority of lakes was sustained by inorganic carbon loading from the catchment rather than by internal CO₂ production. Small lakes with high dissolved organic carbon and phosphorus concentrations, shorter retention times and longer ice-free seasons had the highest CO₂ concentrations. CO₂ emissions from these small lakes was twice that of comparable lakes in colder regions, and similar to emissions from subtropical and tropical lakes. We conclude that changes in land use and climate that increase dissolved inorganic carbon may cause emission levels from boreal lakes to approach those of lakes in warmer regions.

Recent studies have shown that inland waters emit substantial amounts of carbon dioxide (CO_2) into the atmosphere^{1,2,13-15}. However, it is still uncertain to what extent the CO₂ is produced in terrestrial or in aquatic ecosystems. To answer this question, dissolved inorganic carbon (DIC) mass balances are needed. Mass balances have been applied to single lake ecosystems, showing that the dominant net CO₂ source in a lake can either be lake internal CO₂ production (for example, refs 16,17) or DIC inflow from lake external sources (for example, refs 10,18-21). Thus, dominance of both internal and lake external CO₂ production has been reported but it is still unknown how widespread a dominance of lake internal or lake external CO₂ production is. To fill this knowledge gap we applied DIC mass balances to 5,118 lakes in the boreal region using freely available freshwater data. The mass balances refer to autumn water chemical conditions before ice-on (see Methods), where DIC concentrations in dimictic lakes often peak for the second time of the year and CO₂ emissions from lakes are high²².

For the mass balances we estimated lake internal CO₂ production, DIC inputs from lake external sources, DIC outflows and CO₂ emissions to the atmosphere. The internal CO₂ production (CO_{2 internal prod}) comprised three processes: CO₂ production at the sediment-water interface by microbial mineralization (CO_{2_sediment prod}; equation (3)), CO₂ production in the water column by microbial mineralization of dissolved organic carbon (CO_{2_water_prod}; equation (4)), and CO₂ production in the water column by photochemical mineralization (CO_{2 photo prod}; equation (5)). Among the three lake internal processes we found that CO_{2 water prod} was generally the largest contributor to lake internal CO₂ production in the 5,118 boreal lakes, which had a median dissolved organic carbon (DOC) concentration of 11 g m⁻³. $CO_{2_water_prod}$ in the lakes ranged between 1 and 772 mg C m⁻² d⁻¹ during autumn (median: $221 \text{ mg C m}^{-2} \text{ d}^{-1}$). This range is similar to the span of CO2_water_prod in another study of 15 highly varying boreal lakes $(0-700 \text{ mg C m}^{-2} \text{ d}^{-1}; \text{ ref. 23})$. In cold regions with low nutrient and DOC concentrations, CO2 water prod is usually lower. Jonsson et al.¹⁶ found, for example, that the net pelagic CO₂ production (that is, CO2_photo_prod and CO2_water_prod minus primary production) in 16 nutrient-poor subarctic lakes ranged between 2 and $34 \text{ mg C m}^{-2} \text{ d}^{-1}$. For our study lakes we estimated that CO_{2 water prod} contributed to the total CO₂ lake internal production during autumn by, on average, 74% (range: 3.4-91%). CO_{2 water prod} was related to the CO₂ surface water concentrations in the boreal lakes in autumn ($R^2 = 0.28$, p < 0.0001, n = 5, 118), which ranged from 0.3 to $5.7 \,\mathrm{g}\,\mathrm{C}\,\mathrm{m}^{-3}$ (Fig. 1). Performing a sensitivity analysis with a 20% DOC increase raised the median contribution of $CO_{2_water_prod}$ to $CO_{2_internal_prod}$ from 74 to 78%.

The second largest contributing process to internal CO₂ production was CO_{2_sediment_prod}, which during autumn ranged between 12 and 172 mg C m⁻² d⁻¹, with a median of 47 mg C m⁻² d⁻¹. The median CO_{2_sediment_prod} was comparable to the mean measured benthic respiration flux in boreal Canadian lakes (that is, 40.2 ± 8.4 mg C m⁻² d⁻¹; ref. 24). The contribution of CO_{2_sediment_prod} to total CO₂ lake internal production ranged between 4 and 60%, with a median of 17%. The contribution was rather insensitive to variations in total phosphorus (TP) concentrations and water temperatures. A 20% increase in TP concentrations, or a 1°C increase in water temperature, only slightly enhanced the median contribution of CO_{2_sediment_prod} to CO_{2_sediment_prod}

¹Department of Ecology and Genetics/Limnology, Uppsala University, Norbyvägen 18D, 752 36 Uppsala, Sweden. ²Department of Aquatic Ecology and Environmental Biology, Institute for Water and Wetland Research, Radboud University, Heyendaalseweg 135, 6525AF Nijmegen, The Netherlands. ³Department of Earth Sciences, Uppsala University, Villavägen 16, 752 36 Uppsala, Sweden. ⁴Department of Bioscience and Arctic Research Centre, Aarhus University, Vejlsøvej 25, 8600 Silkeborg, Denmark. ⁵Sino-Danish Centre for Education and Research (UCAS), Beijing 100049, China. ⁶Department of Biology, Juiz de Fora Federal University, 36.036-900 Juiz de Fora, Brazil. *e-mail: Gesa.Weyhenmeyer@ebc.uu.se

LETTERS



Figure 1 | CO₂ lake concentrations and DIC fluxes for 5,118 boreal lakes distributed across 16 temperature regions. a-e, Shown are normal quantile boxplots (see Methods) for each region for CO₂ concentrations (**a**), total phosphorus concentrations (**b**), total lake internal CO₂ production (**c**), CO₂ emission (**d**) and DIC input from lake external sources (**e**). For comparison, data from 75 shallow, mainly subtropical and tropical South American lakes are added (to the right of the dashed line; values from ref. 6). For abbreviations and flux estimates see the equations in Methods.

from 17 to 19%. The CO₂ concentrations in the surface waters of the lakes were not related to the water-temperature-dependent and TP-dependent CO_{2 sediment prod} ($R^2 = 0.00, p > 0.05, n = 5,118$).

Photochemical mineralization ($CO_{2_photo_prod}$) accounted for the third internal source for CO_2 in lake waters, estimated to range between 21 and 27 mg C m⁻² d⁻¹ in the 5,118 lakes. These values correspond to maximum DIC production rates in Swedish boreal lakes during the month of June according to recent studies²⁵. Thus, the $CO_{2_photo_prod}$ in this study is most probably overestimated. Despite this overestimation we found that the contribution of $CO_{2_photo_prod}$ to total CO_2 lake internal production was only of minor importance (median: 8.5%, range: 3.0–48%). A 20% increase in global radiation raised the median contribution of $CO_{2_photo_prod}$ from 8.5 to 10%.

Adding up all three internal CO_2 production components we obtained an autumn $CO_{2_internal_prod}$ flux ranging between 46 and 880 mg C m⁻² d⁻¹ (median: 301 mg C m⁻² d⁻¹) in the 5,118 lakes. Our modelled $CO_{2_internal_prod}$ corresponded well to measured $CO_{2_internal_prod}$ in 16 lakes in Sweden (Supplementary Fig. 1). The $CO_{2_internal_prod}$ was positively related to CO_2 surface water concentrations in the lakes in autumn ($R^2 = 0.27$, p < 0.0001, n = 5,118), suggesting that $CO_{2_internal_prod}$ is an important source for CO_2 autumn concentrations in the lakes. However, the $CO_{2_internal_prod}$ was still too small to sustain our estimated $CO_{2_emission}$ during autumn (range of the $CO_{2_emission}$ with a



Figure 2 | Contribution of lake internal CO₂ production to CO₂ emission in 5,118 boreal lakes during autumn. Lake internal CO₂ production was modelled with equations (3)–(5) and CO₂ emission with equation (6) (Methods). The solid grey line shows the 1:1 relationship. Data points above the dashed grey line represent lakes in which the contribution of lake internal CO₂ production to CO₂ emission is less than 50%.

conservative gas transfer velocity: $125-1,905 \text{ mg C m}^{-2} \text{ d}^{-1}$, median: $448 \text{ mg C} \text{m}^{-2} \text{d}^{-1}$, and with a gas transfer velocity adjusted for lake surface area: $128-2,620 \text{ mg C m}^{-2} \text{ d}^{-1}$ with a median of $708 \text{ mg C m}^{-2} \text{ d}^{-1}$; equation (6) in Methods). Using the conservative CO_{2 emission} estimate, we found that, in autumn, CO_{2 emission} exceeded CO_{2 internal prod} in 83% of the lakes, and in 36% of the lakes $\mathrm{CO}_{2_internal_prod}$ contributed ${<}50\%$ to $\mathrm{CO}_{2_emission}.$ Using the CO_{2 emission} adjusted for lake surface area, we observed that the contribution of $CO_{2 \text{ internal prod}}$ to $CO_{2 \text{ emission}}$ was <50% in as many as 63% of the boreal lakes (Fig. 2). These results suggest that in the majority of boreal lakes a substantial additional external DIC input is needed to sustain the large autumn CO₂ emission, in particular because these estimates did not include any DIC losses by photosynthesis and outflows. Comparing our modelled CO_{2 internal prod} contribution against CO_{2 emission} we received a good agreement with measured values from 16 Swedish lakes, in which the contribution varied from 10 up to 150% (Supplementary Fig. 1). Values >100% reflect DIC losses by photosynthesis and outflows.

Estimating the external DIC input to lakes (DIC_{external}, equation (8) in Methods) we found a range between -92 and $6,057 \text{ mg C m}^{-2} \text{ d}^{-1}$ (median: $625 \text{ mg C m}^{-2} \text{ d}^{-1}$). Negative values (in 5% of the lakes) result most probably from an underestimation of DIC losses. DIC_{external} includes DIC from inflowing surface waters, DIC from groundwater inflows and DIC wet deposition onto the lake surface area. Neglecting DIC wet deposition and adding a 20% direct groundwater lake inflow to long-term mean surface water runoff we estimated (see Methods) that DIC concentrations in inflowing waters (surface and groundwater combined) would range between -8.4 and 79 g C m^{-3} (median: 6.3 g C m^{-3}). These concentrations are on average (median) five times higher than the calculated DIC concentrations in the lake surface water, with the highest deviations in the lakes with the longest lake water retention time (relationship between lake water retention time and the DIC_{external}/DIC_{lake_water} concentration ratio: $R^2 = 0.56 p < 0.0001$, n = 4,879; *n* applied to positive CO₂ inflow values only). The DIC concentrations in some of the lake inflows might seem high, most probably owing to an underestimation of water discharge, in particular the neglect of autumn peak and groundwater flows.

NATURE GEOSCIENCE DOI: 10.1038/NGE02582

LETTERS



Figure 3 | **DIC mass balances for four lake types. a**-d, Fluxes are shown for lakes with short (**a**,**c**) and long (**b**,**d**) water retention time (WRT) for lakes located in colder (ice-free season length \leq 180 days; **a**,**b**) and warmer (ice-free season length > 240 days; **c**,**d**) geographical regions (for flux estimations see equations in Methods). Given are median values for each lake type as well as 2.5 and 97.5 percentiles in parentheses (values in mg C m⁻² d⁻¹). The number of boreal lakes included are 1,436 in **a**, 105 in **b**, 1,008 in **c** and 311 in **d**. Also given are median values of each lake type for WRT, lake area and surface water concentrations of DIC, DOC and TP.

Lakes with a large influence of groundwater are consequently not adequately considered in our estimates. Because groundwater DIC concentrations in the boreal region commonly vary between 7 and 23 g C m⁻³ (ref. 26) we suggest that high $DIC_{external}$ estimates are a result of substantial DIC groundwater inputs.

We used $DIC_{external}$ to calculate the contribution of $CO_{2_internal_prod}$ to the total DIC input-that is, the sum of CO2_internal_prod and DIC_{external}. The CO_{2_internal_prod} to the total DIC input was highly variable, ranging from 3 to 144%, with a median of 43%. Only in 40% of the lakes was CO_{2 internal prod} the dominant source for the total DIC input which sustained the CO₂ emission. Our estimated contribution of CO2_internal_prod to the total DIC input depends on water discharge, DOC, TP, water temperature, global radiation, and the gas transfer velocity (equations (3)-(7)). The contribution was most sensitive to variations in the gas transfer velocity (Supplementary Fig. 2). However, although we considered a 50% uncertainty in the gas transfer velocity, the contribution of CO_{2_internal_prod} to the total DIC input remained less than 50% in the majority of lakes (Supplementary Fig. 2). Also decreasing lake water CO₂ concentrations by 50%, which can be caused by errors in CO₂ concentration calculation in lakes with a low alkalinity, did not change the main result of this study (Supplementary Fig. 2). As the main result of this study is rather insensitive to the variables that show a high seasonal variability we suggest that the main result of this study also holds for seasons other than the autumn season.

Generally, we observed that lakes located in the coldest, DOC- and TP-poor geographical regions and lakes with a long water retention time showed lowest $CO_{2_internal_prod}$ and $CO_{2_emission}$ during autumn. Moving towards warmer, DOC- and TP-richer geographical regions and moving from large to smaller lakes with a shorter water retention time, $\mathrm{CO}_{2_internal_prod}$ and $\mathrm{CO}_{2_emission}$ were substantially increased. Dividing lakes into four categories-namely, small, DOC- and TP-poor lakes with a short water retention time located in cold geographical regions; larger, DOC- and TPpoor lakes with a longer water retention time located in cold geographical regions; small, DOC- and TP-richer lakes with a short water retention time located in warmer geographical regions; and larger, DOC- and TP-richer lakes with a longer water retention time located in warmer geographical regions-we found clear differences in the DIC mass balance (Fig. 3). The highest CO_2 concentrations were observed in small, DOC-rich lakes with a short water retention time located in warmer geographical regions. The CO₂ emission from these lakes was more than twice as high as from similar lake types in cold geographical regions (Fig. 3) and comparable to CO₂ emissions from some subtropical/tropical lakes (Fig. 1).

LETTERS

We conclude that, across boreal lakes, maximum CO₂ concentrations and CO₂ emissions during autumn occur where the water retention time is short (<1 yr), TP concentrations are moderately high (>10 mg m⁻³ and <120 mg m⁻³), DOC concentrations are relatively high (>7 g m⁻³ and <36 g m⁻³) and the ice-free season length is >240 days (Fig. 3). In these lakes the lake internal CO₂ production is usually half the CO₂ emission (Fig. 3). Substantially lower lake internal CO₂ production and CO₂ emissions occur in DOC- and TP-poor lakes in geographical regions with an ice-free season length <180 days (Fig. 3). The total area of lakes in regions where these conditions are expected to dominate is about 5×10^5 km², according to most recent estimates²⁷ (that is, 10% of the total lake surface area on Earth). Assuming an average CO_2 emission of about $400 \text{ mg Cm}^{-2} \text{ d}^{-1}$ from these lakes (Fig. 3) gives a total emission of $73 \text{ Tg} \text{ C} \text{ yr}^{-1}$. If lake external and internal DIC fluxes increase in these lakes as a consequence of land use and climate change, CO₂ emissions from these lakes might become as large as those observed in lakes in warmer, DOCand nutrient-richer regions. Such possible future CO₂ emissions from lakes require close observations of CO₂ concentrations and their sources.

Methods

Methods and any associated references are available in the online version of the paper.

Received 21 April 2015; accepted 9 October 2015; published online 9 November 2015

References

- 1. Raymond, P. A. *et al.* Global carbon dioxide emissions from inland waters. *Nature* **503**, 355–359 (2013).
- Tranvik, L. J. et al. Lakes and reservoirs as regulators of carbon cycling and climate. *Limnol. Oceanogr.* 54, 2298–2314 (2009).
- Wanninkhof, R. et al. Global ocean carbon uptake: Magnitude, variability and trends. Biogeosciences 10, 1983–2000 (2013).
- Cole, J. J., Caraco, N. F., Kling, G. W. & Kratz, T. K. Carbon-dioxide supersaturation in the surface waters of lakes. *Science* 265, 1568–1570 (1994).
- Sobek, S., Tranvik, L. J. & Cole, J. J. Temperature independence of carbon dioxide supersaturation in global lakes. *Glob. Biogeochem. Cycles* 19, GB2003 (2005).
- Kosten, S. et al. Climate-dependent CO₂ emissions from lakes. Glob. Biogeochem. Cycles 24, GB2007 (2010).
- Duarte, C. M. *et al.* CO₂ emissions from saline lakes: A global estimate of a surprisingly large flux. *J. Geophys. Res.* 113, G04041 (2008).
- Duarte, C. M. & Prairie, Y. T. Prevalence of heterotrophy and atmospheric CO₂ emissions from aquatic ecosystems. *Ecosystems* 8, 862–870 (2005).
- Humborg, C. et al. CO₍₂₎ supersaturation along the aquatic conduit in Swedish watersheds as constrained by terrestrial respiration, aquatic respiration and weathering. *Glob. Change Biol.* 16, 1966–1978 (2010).
- Maberly, S. C., Barker, P. A., Stott, A. W. & DeVille, M. M. Catchment productivity controls CO₂ emissions from lakes. *Nature Clim. Change* 3, 391–394 (2013).
- McDonald, C. P., Stets, E. G., Striegl, R. G. & Butman, D. Inorganic carbon loading as a primary driver of dissolved carbon dioxide concentrations in the lakes and reservoirs of the contiguous United States. *Glob. Biogeochem. Cycles* 27, 285–295 (2013).
- 12. Swedish National Lake Inventory data downloaded from http://www.slu.se/vatten-miljo on July 2014.
- Cole, J. J. et al. Plumbing the global carbon cycle: Integrating inland waters into the terrestrial carbon budget. *Ecosystems* 10, 171–184 (2007).
- 14. Battin, T. J. et al. The boundless carbon cycle. Nature Geosci. 2, 598-600 (2009).

- 15. Aufdenkampe, A. K. *et al.* Riverine coupling of biogeochemical cycles between land, oceans, and atmosphere. *Front. Ecol. Environ.* **9**, 53–60 (2011).
- Jonsson, A., Karlsson, J. & Jansson, M. Sources of carbon dioxide supersaturation in clearwater and humic lakes in northern Sweden. *Ecosystems* 6, 224–235 (2003).
- Åberg, J., Jansson, M. & Jonsson, A. in International Association of Theoretical and Applied Limnology, Vol 29, Pt 2, Proceedings International Association of Theoretical and Applied Limnology—Proceedings Vol. 29 (ed. Jones, J.) 577–579 (2005).
- Whitfield, C. J., Seabert, T. A., Aherne, J. & Watmough, S. A. Carbon dioxide supersaturation in peatland waters and its contribution to atmospheric efflux from downstream boreal lakes. *J. Geophys. Res.* 115, G04040 (2010).
- Stets, E. G., Striegl, R. G., Aiken, G. R., Rosenberry, D. O. & Winter, T. C. Hydrologic support of carbon dioxide flux revealed by whole-lake carbon budgets. *J. Geophys. Res.* 114, G01008 (2009).
- Jones, R. I., Grey, J., Quarmby, C. & Sleep, D. Sources and fluxes of inorganic carbon in a deep, oligotrophic lake (Loch Ness, Scotland). *Glob. Biogeochem. Cycles* 15, 863–870 (2001).
- Striegl, R. G. & Michmerhuizen, C. M. Hydrologic influence on methane and carbon dioxide dynamics at two north-central Minnesota lakes. *Limnol. Oceanogr.* 43, 1519–1529 (1998).
- 22. Dillon, P. J. & Molot, L. A. Dissolved organic and inorganic carbon mass balances in central Ontario lakes. *Biogeochemistry* **36**, 29–42 (1997).
- Algesten, G. *et al.* Contribution of sediment respiration to summer CO₂ emission from low productive boreal and subarctic lakes. *Microb. Ecol.* 50, 529–535 (2005).
- Brothers, S. M., Prairie, Y. T. & del Giorgio, P. A. Benthic and pelagic sources of carbon dioxide in boreal lakes and a young reservoir (Eastmain-1) in eastern Canada. *Glob. Biogeochem. Cycles* 26, GB1002 (2012).
- Koehler, B., Landelius, T., Weyhenmeyer, G. A., Machida, N. & Tranvik, L. J. Sunlight-induced carbon dioxide emissions from inland waters. *Glob. Biogeochem. Cycles* 28, 696–711 (2014).
- Öquist, M. G., Wallin, M., Seibert, J., Bishop, K. & Laudon, H. Dissolved inorganic carbon export across the soil/stream interface and its fate in a boreal headwater stream. *Environ. Sci. Technol.* 43, 7364–7369 (2009).
- Verpoorter, C., Kutser, T., Seekell, D. A. & Tranvik, L. J. A global inventory of lakes based on high-resolution satellite imagery. *Geophy. Res. Lett.* 41, 6396–6402 (2014).

Acknowledgements

Financial support was received from the Swedish Research Council (VR), the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (FORMAS), the Knut and Alice Wallenberg Foundation and the European Union (C-CASCADES project). This work is part of and profited from the networks financed by Nordforsk (CRAICC and DOMQUA), National Science Foundation (GLEON) and the Norwegian Research Council (Norklima ECCO). S.K. was supported by NWO-VENI grant 86312012. E.J. was supported by the MARS project (Managing Aquatic ecosystems and water Resources under multiple Stress) funded under the 7th EU Framework Programme, Theme 6 (Environment including Climate Change), Contract No.: 603378 (http://www.mars-project.eu). Many thanks go to the Swedish Environmental Protection Agency and the staff of the laboratory of the Department of Aquatic Sciences and Assessment for financing, sampling and analysing thousands of water, and to the Swedish Meteorological and Hydrological Institute for making meteorological data freely available.

Author contributions

G.A.W. designed the study. All authors contributed with data and/or equations, and all authors made substantial contributions to the methods, the results, the discussion and the conclusions.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to G.A.W.

Competing financial interests

The authors declare no competing financial interests.

Methods

Data and variables. From large databases comprising thousands of boreal and hemiboreal lakes (downloaded from http://www.slu.se/vatten-miljo), we selected 5,118 lakes for which data were available on lake water temperature (T_w) , alkalinity (Alk; only positive values were chosen), pH (only values > 5.4 were used to avoid incorrect estimates of CO2 concentrations), dissolved organic carbon (DOC), total phosphorus concentrations (TP), altitude of the lake site, lake area (LA), lake volume (V), and size of catchment area excluding the area of the study lake (CA). From the data material we calculated concentrations of dissolved inorganic carbon (DIC) and CO2 (in µM) based on alkalinity, pH and water temperature (for a detailed description of the calculation process see Weyhenmeyer et al.²⁸). We multiplied CO₂ in μ M by a factor of 12.01 to obtain CO₂ in mg C m⁻³. According to Abril et al.²⁴ the CO2 calculation process might result in overestimated CO2 concentrations in acidic, organic-rich lakes. Many of the lakes in the boreal region belong to this category. Thus, our observed patterns could potentially be biased by an influence of organic acids on the alkalinity determination. However, when we analysed CO2 concentrations and mass balances for different lake types we did not find any significant differences in pH between the lake types (non-parametric Wilcoxon test: p > 0.05). As our main results are based on median values we minimize the uncertainties in CO2 concentration values, and possible overestimations are counter-acted by choosing a conservative CO₂ emission (see below). In addition, we perform a sensitivity analyses on the influence of CO_2 concentration errors on the main message of the study (Supplementary Information).

The 5,118 lakes, classified as boreal lakes, were distributed across the entire boreal and hemiboreal region of Sweden. The lakes were generally small (median lake area: 0.2 km2), shallow (median mean lake depth: 3.2 m), nutrient-poor (median TP concentrations: 10 mg m⁻³), and humic (median DOC: 10.5 g m⁻³; median pH: 6.6). The water samples were taken at a water depth of 0.5 m and analysed by the Certified Water Analyses Laboratory at the Swedish University of Agricultural Sciences according to standard limnological methods during the past 40 years. The samples were from the early autumn period when the water column in lakes is mixed and water temperatures are around 4 °C. The early autumn period is the time when the national freshwater inventory takes place. From 1990 to 2005 the national freshwater inventory was carried out during the autumn period in up to 5,000 waters every fifth year. Since 2007 the inventory takes place on an annual basis, but in much fewer waters. At least every seventh year sites are re-sampled. For most inland waters only one autumn sample per year was available. In the few cases where several autumn values during a year were available, we chose the autumn value where water temperatures were closest to 4 °C. Because our sites were re-sampled a few times since 1990, we used the median of available autumn values. The number of available autumn values for each site varied, but because year-to-year variation in autumn water chemistry at around 4 °C water temperature usually remains small compared to the spatial variation²⁸, we consider the median to be suitable for the modelling of spatial variation over a large scale.

Using GIS we overlapped the lake database with the database on meteorological variables from the Swedish Meteorological and Hydrological Institute at http://www.smhi.se and downloaded site-specific (that is, at the sampling point) long-term means (1961–1990) of annual precipitation ($P_{1961-1990}$), annual surface water runoff ($R_{1961-1990}$), annual mean air temperature (MAT₁₉₆₁₋₁₉₉₀; adjusted for altitude by -0.6 °C per 100 m according to Livingstone *et al.*³⁰), and annual global radiation (RAD₁₉₆₁₋₁₉₉₀). Using MAT₁₉₆₁₋₁₉₉₀ we calculated the duration of ice-free days at the lake sites as:

$$D_{T>0} = 365.25 - \frac{365.25}{\pi} \arccos\left(\frac{\text{MAT}_{1961-1990}}{-a \cdot \text{MAT}_{1961-1990} + b}\right)$$
(1)

where $D_{T>0}$ is the long-term mean of the duration of ice-free days at the lake site (in days), 365.25 is the length of a year (in days), MAT₁₉₆₁₋₁₉₉₀ is the altitude-adjusted long-term mean of site-specific annual mean air temperature (in °C), and *a* and *b* are constants with the values 0.66 and 14.32, respectively. Equation (1) has been calibrated and validated for Swedish conditions³¹. MAT₁₉₆₁₋₁₉₉₀ was also used as a measure of the temperature region to which a lake belonged.

We also calculated the lake water retention time for each lake:

$$WRT_{1961-1990} = \frac{V}{R_{1961-1990} \cdot CA}$$
(2)

where WRT₁₉₆₁₋₁₉₉₀ is the long-term mean of the water retention in a lake (in days), V is the lake volume (in m³), R₁₉₆₁₋₁₉₉₀ is the long-term mean of the surface water runoff at the outflow of the lake (in m d⁻¹; the runoff was originally in m yr⁻¹ which we transferred to m d⁻¹ by dividing R₁₉₆₁₋₁₉₉₀ by $D_{T>0}$), and CA is the size of the catchment area excluding the area of the study lake (in m²).

DIC input and output fluxes. For each lake we applied a DIC mass balance for autumn conditions with estimates on DIC input and DIC output. We estimated three types of lake internal CO₂ production (CO_{2_internal_prod}): CO₂ production at the sediment–water interface by microbial mineralization (CO_{2_sediment_prod}); CO₂

production in the water column by microbial mineralization of dissolved organic carbon ($CO_{2,water_prod}$); and CO_2 production in the water column by photochemical mineralization ($CO_{2,sediment_prod}$). To model $CO_{2,sediment_prod}$ we applied a simple predictive model using 219 measurements from eight Swedish boreal lakes, published in Gudasz *et al.*³²:

$$CO_{2 \text{ sediment prod}} = e^{(a \cdot \ln TP + b \cdot \ln T_w + c)}$$
(3)

where $\text{CO}_{2,\text{seliment,prod}}$ is the sediment mineralization (in mg C m⁻² d⁻¹), TP is the *in situ* total phosphorus concentration in the lake surface water (in µg l⁻¹), T_w is the *in situ* water temperature above the sediments (in °C), and *a*, *b* and *c* are constants with the values 0.47, 0.78 and 1.5, respectively. The model performance for the 219 measurements was good ($R^2 = 0.62$, p < 0.0001, n = 219 with an insignificant intercept and a regression slope of 1.0). The range of TP concentrations used for the model development corresponded to 11.4–46.7 µg l⁻¹. These concentrations cover the range of TP concentrations in the 5,118 boreal lakes well.

For the estimation of the $\rm CO_{2,water_prod}$ we used a simple regression equation developed for the $\rm CO_{2,water_prod}$ ($\rm CO_{2,intenal_prod}$) in Swedish boreal lakes³³:

$$CO_{2_water_prod} = a \cdot DOC - b$$
 (4)

where $CO_{2_water_prod}$ is the mineralization in the water column (in mg C m⁻² d⁻¹), DOC is the *in situ* dissolved organic carbon concentration in the lake water (in mg l⁻¹), and *a* and *b* are constants with the values 28.10 and 73.92, respectively. The equation produces negative values for DOC concentrations <2.7 mg l⁻¹. We had only very few lakes with DOC concentrations <2.7 mg l⁻¹. For these few lakes we set DOC to 2.7 mg l⁻¹.

As a third lake internal source for CO_2 we estimated the photochemical mineralization as a direct response to solar radiation exposure according to Vähätalo *et al.*³⁴:

$$CO_{2_photo_prod} = a \cdot RAD_{1961-1990} - b$$
(5)

where $CO_{2_photo_prod}$ is the photochemical mineralization in the lake water column (in mg C m⁻² d⁻¹), RAD₁₉₆₁₋₁₉₉₀ is the site-specific long-term mean of annual global radiation (in MJ m⁻² d⁻¹); the global radiation was originally in kWh m⁻² yr⁻¹, which we transferred to MJ m⁻² d⁻¹ by multiplying RAD₁₉₆₁₋₁₉₉₀ by a factor of 3.6 and dividing it by 365 days), and *a* and *b* are constants with the values 0.067 and 0.037, respectively.

As DIC output from a lake we considered the CO₂ emission (CO_{2_emission}) and the DIC outflow (DIC_{outflow}). DIC can also be lost from the lake water column by photosynthesis and calcium carbonate precipitation. DIC losses during autumn by these processes are most likely very small in the study lakes as both the alkalinity and total phosphorus concentrations had generally low values during autumn (median alkalinity: $0.1\,\text{mEq}\,l^{-1}$; range: $0.01-2.1\,\text{mEq}\,l^{-1}$; median TP: $10\,\mu\text{g}\,l^{-1}$; range: $2-60\,\mu\text{g}\,l^{-1}$). We therefore set DIC losses by photosynthesis and calcium carbonate precipitation to zero. This assumption is further supported by measurements on lake primary production in Swedish boreal lakes that corresponded on average to $5.9\,\text{mg}\,\text{C}\,\text{m}^{-2}\,d^{-1}$ reported by ref. 35 and to $6.7\,\text{mg}\,\text{C}\,\text{m}^{-2}\,d^{-1}$ reported by ref. 36 up to a maximum of 201 mg C m^ $^{-2}\,d^{-1}$ measured in one single boreal lakes³⁶. The average primary production flux is very low compared to all other DIC fluxes in Swedish boreal lakes³⁵. For other lakes, however, DIC losses by photosynthesis can be substantial, which would raise the total DIC loss in the mass balance.

For the determination of the $\text{CO}_{2-\text{emission}}$ we used a conservative number for the gas transfer velocity k_{600} . According to Crusius and Wanninkhof³⁷ k_{600} corresponds to 0.61 m d⁻¹ in small lakes, assuming a wind speed of 3.5 m s⁻¹. We further adjusted k_{600} by a water temperature dependent diffusion coefficient following Jähne and colleagues³⁸. The adjustment of k_{600} resulted in k values <0.61 m d⁻¹, with a median for all lakes of 0.35 m d⁻¹. The k values can be considered very conservative. Recent upscaling approaches have, for example, used k values between 0.54 and 1.90 m d⁻¹ (ref. 1). Because k varies with lake area, we also used k values adjusted for lake surface area following Read and colleagues³⁹. These values were then further adjusted for temperature. The k values adjusted for lake area and temperature (k_{adj}) were much larger than 0.35 m d⁻¹, ranging between 0.28 and 1.15 m d⁻¹, with a median of 0.63 m d⁻¹. We estimated the CO₂ emission with both k and k_{adi} according to:

$$CO_{2_{emission}} = (CO_{2_{water}} - CO_{2_{equilibrium_air}}) \cdot k$$
(6)

where $CO_{2-\text{emission}}$ is the CO_2 emission from a lake (in mg C m⁻² d⁻¹), $CO_{2-\text{water}}$ is the CO_2 concentration in the lake surface water (in mg C m⁻³), $CO_{2-\text{equilibrium},air}$ is the CO_2 in equilibrium with the air (in mg C m⁻³; to determine $CO_{2-\text{equilibrium},air}$ we used the water-temperature-adjusted Henry constant and 1,013 bar, adjusted for altitude for each lake²⁸), and *k* is the gas transfer velocity (in m d⁻¹). When we show CO_2 emissions in figures we always used k_{adj} .

LETTERS

Finally, we determined the DIC_{outflow} according to:

$$DIC_{outflow} = R_{1961-1990} \cdot (CA + LA) \cdot DIC_{water}$$
(7)

where DIC_{outflow} is the DIC outflowing from a lake (in mg C m⁻³ d⁻¹), R₁₉₆₁₋₁₉₉₀ is the long-term mean of the surface water runoff at the outflow of the lake (in m d⁻¹; the runoff was originally in m yr⁻¹, which we transferred to m d⁻¹ by dividing RAD₁₉₆₁₋₁₉₉₀ by $D_{T>0}$), CA is the size of the catchment area excluding the area of the study lake (in m²), LA is the area of the lake (in m²), and DIC_{water} is the DIC concentration in the lake surface water (in mg C m⁻³). To make the DIC_{outflow} comparable to the other fluxes in mg C m⁻² d⁻¹, we multiplied mg C m⁻³ d⁻¹ by the lake volume and divided by the lake area.

With the estimated DIC input and output we finally determined the last unknown DIC flux, which is the DIC flux into a lake from lake external sources:

$$DIC_{external} = CO_{2_emission} + DIC_{outflow} + DIC_{internal_loss} - CO_{2_internal_prod}$$
(8)

where DIC_{external} is the DIC input into a lake from lake external sources (that is, DIC from inflowing surface waters, DIC from inflowing groundwater and DIC from wet deposition onto the lake surface area (in mg C m⁻² d⁻¹)), CO_{2_emission} is the CO₂ emission from a lake (equation (6) with gas transfer velocity adjusted for lake surface area), DIC_{outflow} is the DIC outflow from a lake (equation (7)), DIC_<u>internal_loss</u> is the lake internal DIC loss by photosynthesis and calcium carbonate precipitation, which we set to zero in the nutrient-poor, low-alkaline boreal lakes, and CO_{2_internal_prod} is the total lake internal CO₂ production (that is, the sum of CO_{2_isdiment_prod} (equation (3)), CO_{2_water_prod} (equation (4)) and CO_{2_photo_prod} (equation (5))). To receive DIC concentrations in inflowing surface waters (in mg C m⁻³) we multiplied the DIC_{external} by the lake area (in m²) and divided it by the product: (R₁₉₆₁₋₁₉₉₀ + 0.2 · R₁₉₆₁₋₁₉₉₀) · CA (in m³ d⁻¹), where the term 0.2 · R₁₉₆₁₋₁₉₉₀ is a proxy for groundwater inflows.

Statistics. Owing to our data material having non-normal distributions we used statistical methods that are insensitive to non-normal distributions, and we always used ln-transformations. Whenever averages are given, median values were calculated. Ranges always refer to 2.5–97.5 percentiles. Boxplots are normal quantile boxplots with 25th and 75th percentiles, and an upper quartile

 $+1.5\times$ (interquartile range) and a lower quartile $-1.5\times$ (interquartile range). All statistical tests were carried out in JMP, version 11.0.

References

- Weyhenmeyer, G. A., Kortelainen, P., Sobek, S., Müller, R. & Rantakari, M. Carbon dioxide in boreal surface waters: A comparison of lakes and streams. *Ecosystems* 15, 1295–1307 (2012).
- Abril, G. *et al.* Technical Note: Large overestimation of p_{CO(2)} calculated from pH and alkalinity in acidic, organic-rich freshwaters. *Biogeoscience* 12, 67–78 (2015).
- Livingstone, D. M., Lotter, A. F. & Walker, I. R. The decrease in summer surface water temperature with altitude in Swiss Alpine lakes: A comparison with air temperature lapse rates. *Arct. Antarct. Alp. Res.* 31, 341–352 (1999).
- Weyhenmeyer, G. A., Peter, H. & Willen, E. Shifts in phytoplankton species richness and biomass along a latitudinal gradient-consequences for relationships between biodiversity and ecosystem functioning. *Freshwat. Biol.* 58, 612–623 (2013).
- Gudasz, C. et al. Temperature-controlled organic carbon mineralization in lake sediments. Nature 466, 478-481 (2010).
- Algesten, G. *et al.* Contribution of sediment respiration to summer CO₂ emission from low productive boreal and subarctic lakes. *Microb. Ecol.* 50, 529–535 (2005).
- Vähätalo, A. V., Salkinoja-Salonen, M., Taalas, P. & Salonen, K. Spectrum of the quantum yield for photochemical mineralization of dissolved organic carbon in a humic lake. *Limnol. Oceanogr.* 45, 664–676 (2000).
- Algesten, G. *et al.* Role of lakes for organic carbon cycling in the boreal zone. *Glob. Change Biol.* **10**, 141–147 (2004).
- Seekell, D. A. *et al.* The influence of dissolved organic carbon on primary production in northern lakes. *Limnol. Oceanogr.* 60, 1276–1285 (2015).
- Crusius, J. & Wanninkhof, R. Gas transfer velocities measured at low wind speed over a lake. *Limnol. Oceanogr.* 48, 1010–1017 (2003).
- Jähne, B., Heinz, G. & Dietrich, W. Measurement of the diffusion coefficients of sparingly soluble gases in water. J. Geophys. Res. 92, 10767–10776 (1987).
- Read, J. S. et al. Lake-size dependency of wind shear and convection as controls on gas exchange. *Geophys. Res. Lett.* 39, L09405 (2012).