INTRODUCTION

Lakes and streams acting as recipients of carbon, nitrogen, and other nutrients transported from terrestrial ecosystems contribute to landscape greenhouse gas (GHG) balances emitting carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O). Freshwater N₂O has received minor interest compared to carbon gases, CO₂ and CH₄. Consequently, both the variability of N₂O concentrations and factors regulating N₂O fluxes from lakes at regional and global scales have remained poorly constrained and estimates of freshwater N₂O emissions are still uncertain due to sparse data (Deemer et al., 2016; DelSontro, Beaulieu, & Downing, 2018; Soued, del Giorgio, & Maranger, 2015). Majority of freshwater studies have focused on rivers and streams in N-rich agricultural environments (Beaulieu et al., 2011; Hu, Chen, & Dahlgren, 2016; Mulholland et al., 2008) excluding landscapes dominated by forests and peatlands, the most widely distributed ecosystems in the boreal zone.

The main processes involved in N₂O cycling are aerobic nitrification and anaerobic denitrification, which are regulated by several environmental factors like oxygen and organic matter content.
pH, temperature, and the availability of ammonium and nitrate (Butterbach-Bahl, Baggs, Dannenmann, Kiese, & Zechmeister-Boltenstern, 2013). In contrast to most previous studies, which have predominantly focused either on a few lakes and/or summer time measurements, we measured N₂O concentrations for the four seasons in the water columns of 112 lakes in Finland covering different lake types, locating between the latitudes 60°N and 67°N (Figure 1; Table 1). We examined how seasonal and spatial variation of N₂O concentrations in lakes was associated with the characteristics of lakes (area, maximum depth, water chemistry, temperature, oxygen content) and catchments (area, elevation, and land use cover) and compared spatiotemporal variation in N₂O concentration with that of CO₂ and CH₄ measured simultaneously with the N₂O (Juutinen et al., 2009; Kortelainen et al., 2006).

2 | MATERIALS AND METHODS

2.1 | Selection of the lakes and sampling

A subpopulation of 87 boreal lakes in Finland (Figure 1; Table 1) was randomly selected from the Nordic Lake Survey (NLS) database (Henriksen et al., 1998; Mannio, Räike, & Vuorenmaa, 2000). Besides N₂O lake chemistry and morphometry (lake area [LA], max depth), catchment characteristics (catchment area [CA], elevation), land use cover (agricultural land %, peat %, forest %, urban %), and climate-related variables (latitude, water temperature) were determined for each lake in order to identify key drivers contributing to seasonal variation of N₂O concentrations in boreal lakes. The

| TABLE 1 | Average catchment characteristics and lake water chemistry of autumn 1 m samples for N₂O lakes (randomly selected lakes sampled in 1998–1999) and for Nordic Lake Survey (NLS) lakes (sampled in 1995) |
| N₂O lakes, n = 87 | NLS, n = 87 |
| --- | --- |
| LA (km²) | Med 0.31 | Mean 3.3 | Min 0.04 | Max 44 | Med 0.22 | Mean 20 |
| CA/LA | 14 | 49 | 2.8 | 610 | NM | NM |
| Max depth (m) | 6.8 | 9.1 | 1 | 42 | NM | NM |
| Water (%) | 9.2 | 11 | 0.3 | 35 | 8.8 | 10 |
| Agric. land (%) | 4.2 | 7.6 | 0.03 | 39 | 2.3 | 5.6 |
| Peat (%) | 8.3 | 14 | 0 | 84 | 14 | 18 |
| Alkalinity (µmol/L) | 140 | 170 | −70 | 990 | 130 | 150 |
| pH | 6.5 | 6.6 | 4.2 | 7.7 | 6.7 | 6.6 |
| O₂ (%) | 82 | 79 | 47 | 95 | NM | NM |
| pCO₂ (µatm) | 1,100 | 1,500 | 330 | 4,500 | 860 | 990 |
| N₅tot (µg/L) | 500 | 560 | 170 | 2000 | 430 | 470 |
| Inorganic N (%) | 7.4 | 10 | 0.4 | 48 | 7.7 | 11 |
| P₅tot (µg/L) | 15 | 21 | 5.0 | 85 | 14 | 19 |
| Inorganic P % | 15 | 17 | 1.6 | 63 | 15 | 19 |
| TIC (mg/L) | 2.0 | 2.5 | 0.6 | 11 | 1.6 | 2 |
| TOC (mg/L) | 9.4 | 11 | 2.2 | 38 | 7.8 | 9 |
| Fe₅tot (µg/L) | 400 | 650 | 29 | 3,600 | 360 | 560 |

Abbreviations: CA, catchment area; LA, lake area; NM, not measured; TIC, total inorganic carbon; TOC, total organic carbon.
lakes were sampled once in winter, spring, summer, and autumn during 1998–1999 from four depths at the deepest point of the lake for \( \text{N}_2\text{O} \) concentrations and physical and chemical characteristics. For \( \text{CO}_2 \) and \( \text{CH}_4 \) analyses, a subpopulation of 177 lakes was randomly selected from the NLS data (Juutinen et al., 2009; Kortelainen et al., 2006). Gas analyses were carried out in the laboratories of University of Eastern Finland, due to limited capacity for \( \text{N}_2\text{O} \) analyses we had to exclude approximately one-third of the lakes, ending up with 87 randomly selected lakes for \( \text{N}_2\text{O} \) measurements. Majority of boreal lakes are located in forested catchments with a relatively minor human disturbance. Consequently, eutrophic lakes cannot be expected to be well represented in the randomly selected lake database. In order to include also some eutrophic lakes in the study, a subset of NLS lakes (\( n = 25 \)) with the highest total P concentrations was also sampled. Results from these lakes were included in the analyses to study the relationships between lake water chemistry and \( \text{N}_2\text{O} \) concentrations and flux estimates from different lake types but excluded from the population estimates.

Following similar approach as for \( \text{CH}_4 \) we used modified Finnish lake typology required for the ecological lake status classification by the EU Water Framework Directive to classify the lakes, for example, according to nutrient concentrations, color, size, and depth (Juutinen et al., 2009). In order to avoid very small subgroups, we divided the lakes into four subgroups: Nutrient-Rich and Calcareous (NRC, alkalinity >0.4 mmol/L, winter turbidity >5 FTU), Humic Small (HSm, color ≥30 Pt mg/L, area ≤5 km², or mean depth <3 m), Humic Large (HL, color ≤30 Pt mg/L, area ≥5 km²), and Clear Water Lakes (CL, color <30 Pt mg/L).

### 2.2 Calculation of gas fluxes

\( \text{N}_2\text{O} \) measurements were carried out by the headspace equilibration technique (McAuliffe, 1971). Ultra pure \( \text{N}_2 \) gas (30 ml) was added to 60 ml syringes containing 30 ml water, the syringes were then shaken vigorously for 3 min. The headspace gas concentration was quantified with a gas chromatograph (Hewlett Packard Series II and Shimadzu GC-14-A) equipped with an FI-detector.

Lake–atmosphere gas fluxes were estimated from the surface water gas concentrations according to the First Fick’s law of diffusion (Fick, 1855; Wanninkhof & Knox, 1996):

\[
F_{\text{gas}} = k_{\text{gas}} (C_{\text{gas}} - C_{\text{eq}}),
\]

where \( F_{\text{gas}} \) is the lake–atmosphere flux of \( \text{N}_2\text{O} \), \( k_{\text{gas}} \) is the gas transfer velocity (\( \text{m/day} \)), \( C_{\text{gas}} \) is the concentration of the gas in the surface water (\( \mu\text{mol/L} \)), and \( C_{\text{eq}} \) is the concentration of the gas (\( \mu\text{mol/L} \)) in equilibrium with the atmosphere.

The in situ equilibrium concentration of \( \text{N}_2\text{O} \) in the lake surface water was calculated according to Henry’s Law, with appropriate corrections for the in situ temperature, assuming atmospheric concentration of 324 ppb for \( \text{N}_2\text{O} \) (319 ppb in 2005 in Hyytiälä SMEAR station, Finland) and a 0.8 ppb increase per year (Ciais et al., 2013).

Since the data behind measured gas transfer coefficients (\( k \) values) are limited and the relationship between lake size and \( k \) values varies, we used three approaches to estimate lake–atmosphere gas exchange (Heiskanen et al., 2014; Holgerson, Farr, & Raymond, 2017; Vachon & Prairie, 2013). We calculated the gas fluxes for the 94 lakes that had \( \text{N}_2\text{O} \) concentrations measured in all four seasons. Some lakes are located in remote areas and were difficult to sample before and/or after ice melt.

First, following the approach by Heiskanen et al. (2014), \( k_{\text{N}_2\text{O}} \) value was calculated, using the average values of \( k\text{CO}_2 \) from a small Finnish lake during a 4 month period, which we transformed into the \( k_{\text{N}_2\text{O}} \):

\[
k_{\text{gas1}}/k_{\text{gas2}} = (S_{\text{gas1}}/S_{\text{gas2}})^{-n},
\]

where \( S_{\text{gas}} \) is the Schmidt number for a given gas (Jähne, Heinz, & Dietrich, 1987) and \( n \) is 1/2.

The wind speed was assumed to be 3 m/s, which is an average open water period wind speed at the height of 10 m for the inland measurement stations in Finland (Leinonen, 2000).

Secondly, we applied Holgerson et al. (2017) approach for each of our 94 lakes, that is, the size-class median estimates of \( k_{\text{600}} \) for 67 ponds and lakes across a size gradient measured with floating chambers or gas tracers.

Thirdly, we calculated \( k_{\text{600}} \) values using an empirical model based on wind speed (\( U_{10} \)) and LA in 21 Canadian water bodies (Vachon & Prairie, 2013):

\[
k_{\text{600}} = 2.51 + 1.48 U_{10} + 0.39 U_{10} \log_{10} LA.
\]

### 2.3 Estimation of annual fluxes and upscaling

First, following similar approach as for \( \text{CO}_2 \) (Kortelainen et al., 2006), we estimated the annual \( \text{N}_2\text{O} \) emission per surface area unit of each lake using the \( \text{N}_2\text{O} \) concentrations from the depth of 1 m during the four sampling occasions as follows: the winter 1 m concentrations were used for half a month period assuming that the \( \text{N}_2\text{O} \) accumulated under ice would be released to the atmosphere during the short spring circulation period. The spring \( \text{N}_2\text{O} \) concentrations at the depth of 1 m were used for a 1.5 month period and the summer 1 m concentrations for a 3 month period representing the summer stratification between May and August. The samples taken during the autumn circulation were used for the period of 2 months representing the time after breakup of the summer stratification and before the permanent ice cover (altogether 7 months of ice-free season). For comparison, we also estimated the evasion for the ice-free period by multiplying the median summer time \( \text{N}_2\text{O} \) flux with the duration of the ice-free period (7 months).

Secondly, we estimated the annual median \( \text{N}_2\text{O} \) evasion both for each lake size class separately (lake size-specific evasion) and for all size classes combined (the mean, median, and summer median of individual lakes) for Finnish and Boreal lakes. The randomly selected lakes, which could be sampled during all four seasons
(n = 71), were divided into the different size classes and the median annual evasion of each size class was multiplied with the area of the Finnish and boreal lakes belonging to the respective size class. Summing up the size class-specific estimates results in an estimate for Finland and boreal region, respectively. The rationale was that when upscaling is based on the evasion estimated specific to each of the four lake size classes, the impact of the numerous small lakes in our data on the regional estimate is not disproportionately large.

Furthermore, we estimated the contribution of N2O emission from lakes to that of forests both in Finland and in the boreal zone. For Finland, we used the lake surface area of 32,663 km² (Raatikainen & Kuusisto, 1990) and the forest area of 0.203 × 10⁶ km² (Vaahtera et al., 2018). For the boreal region, the lake surface area was estimated from MODIS data (1,422,448 km²), the forest area of 12.1 × 10⁶ km² was derived from Potter, Matson, Vitousek, and Davidson (1996).

2.4 Water chemistry

The lakes were sampled in each sampling occasion for dissolved oxygen, alkalinity, conductivity, pH, color, total nitrogen (Ntot), nitrate (+ nitrite) nitrogen defined as (NO3-N, nitrite had negligible contribution to the total amount of nitrate and nitrite), ammonium nitrogen (NH4-N), total phosphorus (Ptot), phosphate phosphorus (PO4-P), total organic carbon (TOC), and total iron (FeTOT). Water chemistry was analyzed from unfiltered samples in the accredited laboratories of the Regional Environment Centers. Ntot was determined by oxidation of the sample by combustion and measuring inorganic C by IR-spectrophotometry (National Board of Waters, 1981).

2.5 Catchment characteristics

The CAs of the NLS lakes were determined from the topographic maps, and the catchment boundaries were digitalized and combined with land use data based on satellite images using the Arc View georeferencing software. Lake area, CA, catchment to LA ratio, latitude, and the proportion of peatland, forest on mineral soil, agricultural land, water (consisting of the upstream water bodies and the lake itself), and built-up area in the catchments were determined.

2.6 Statistical analysis

The relationships between the N2O concentrations and lake chemistry, morphometry, latitude, and catchment characteristics were examined using Pearson’s correlation coefficients using SAS 9.4 for Windows software. The variables were loge or square root transformed in order to normalize their distribution.

Stepwise multiple linear regression models predicting N2O concentrations were carried out using lake chemical, physical, and morphometric variables, climatic variables (temperature, latitude), and catchment properties as predictors. The cases with an absolute value of the studentized residual exceeding 3 were excluded and only the independent variables with p < .05 were included in the models.

Linear mixed models were used to take into account that each lake was sampled four times and from four depths (interdependence of within-lake sampling). We run a linear mixed model with NO3-N, depth, season as fixed factors, and lake as random factor. That is, the model analyzed the dependence of N2O concentration on NO3-N, and eliminated the effects of depth and season on the emerging N2O to NO3-N relationship.

To examine the relationship between mean NO3-N and mean N2O concentrations in different lake types across the four seasons, a nonlinear regression model was fitted (Equation 4, see Results). Seasonal mean values for each lake type (n = 16) were obtained by averaging all measured depths across respective lakes. To examine how representative the patterns and drivers of N2O dynamics found in our lakes were, we used Equation (4) to estimate N2O concentrations in the water samples from the independent nitrate-nitrite dataset of 874 randomly selected Finnish NLS lakes sampled during the autumn overturn in 1998–1999. The modeled results were compared to the measured N2O-N concentrations in our study lakes (n = 1,542 seasonal water samples from 112 lakes).

3 RESULTS

3.1 Seasonal and spatial variation in N2O

During the open water season, 71% of the surface water samples were saturated with respect to the atmospheric equilibrium value of N2O, that is, lakes were mostly sources of N2O. Nitrous oxide, similar to CO2, peaked in winter. In contrast, CH4 concentration was significantly higher in summer (Kruskal–Wallis 123.93, p < .001, Figure 2).

Seasonal variation of N2O concentrations followed the variation of nitrate—highest in winter and lowest in summer despite differences in concentration levels across lake type classification (Figure 3). Furthermore, nitrate was the best predictor for N2O concentrations in the entire data (Table 2; Figure 4) and across different seasons and depths despite the large variation in LA (from 0.04 to 44 km²), maximum depth (from 1 to 42 m), latitude (from 60°N to 67°N), and land use cover. Nitrate concentrations were highest in nutrient-rich, calcareous (NRC) lakes accompanied by highest N2O concentrations in winter and spring (Figure 3). In summer and autumn, N2O concentrations were more evenly distributed across the lake types. Depth profile distribution demonstrated similar N2O concentrations from surface to
bottom (Figure 5) in contrast to CO$_2$ and CH$_4$ which accumulated in bottom water (Juutinen et al., 2009; Kortelainen et al., 2006). There was no district correlation between N$_2$O and water pH. Both the highest and the lowest N$_2$O concentrations occurred around the median pH of 6.5 (Figure 6). Even though there was a weak correlation between oxygen and N$_2$O (Table 2), the variability in N$_2$O concentrations followed the variation of nitrate—highest in winter and lowest in summer despite differences in concentration levels across the lake type classification.

There was no district correlation between N$_2$O and water pH. Both the highest and the lowest N$_2$O concentrations occurred around the median pH of 6.5 (Figure 6). Even though there was a weak correlation between oxygen and N$_2$O (Table 2), the variability in N$_2$O concentrations across O$_2$ gradient was large especially before ice melt among all lake types (Figure 7). N$_2$O and nitrate concentrations increased with decrease in elevation (Figure 8a) accompanied by increasing coverage of agricultural land (Figure 8d), which resulted in higher nitrate and N$_2$O concentrations in large lakes (Figure 8b) often surrounded by more intensive human impact compared to small headwater lakes, which in the boreal zone are predominantly surrounded by forests and peatlands.
Lake chemistry predicted \( \text{N}_2\text{O} \) better than catchment land use cover. In linear multiple regression models, electron acceptors, nitrate and oxygen, and lake water temperature as independent variables predicted best \( (r^2 = .55, \text{stepwise procedure}) \) the \( \text{N}_2\text{O} \) in the entire data \( (n = 1,396, \text{all seasons and depths}) \). Nitrous oxide in bottom water \( (\text{all seasons}) \) was best predicted by nitrate and oxygen concentration \( (r^2 = .54) \). The surface water model \( (\text{all seasons}) \) had nitrate, temperature, and the percentage of agricultural land in the catchment as the independent variables explaining 58% of the variation in \( \text{N}_2\text{O} \). The best model for winter \( (\text{all depths}) \) explained 58% of the variability in \( \text{N}_2\text{O} \) selecting nitrate, latitude, and \( \text{pH} \) as the independent variables (Table 3). The linear mixed model results demonstrated that the significant relationship between nitrate and \( \text{N}_2\text{O} \) remained \( (p < .001) \) even after the influence of depth \( (p = .096) \) and season \( (p < .001) \) had been taken into account.

### 3.2 | \( \text{N}_2\text{O} \) evasion

We estimated the median \( \text{N}_2\text{O} \) evasion based on 71 lakes \( (<100 \text{ km}^2) \) as 0.009 g N m\(^{-2}\) LA year\(^{-1}\). The seasonal median fluxes were 0.002 g N/m\(^2\) LA at the thaw (during 0.5 months), 0.002 g N/m\(^2\) LA in spring (1.5 months), 0.001 g N/m\(^2\) LA in summer (3 months), and 0.003 g N/m\(^2\) LA in autumn (2 months). For the largest lakes \( (>100 \text{ km}^2) \), we used the evasion estimate from the 10–100 km\(^2\) lake size class.

The median evasion for the <0.1, 0.1–1, 1–10, 10–100 km\(^2\) lake size classes was estimated as 0.0047, 0.007, 0.018, and 0.02 g N/m\(^2\) LA, respectively (using \( k \) values from Holgerson et al., 2017). Nitrous oxide data were not available for lakes larger than 100 km\(^2\); for this lake size class, we used the median evasion estimate from the 10–100 km\(^2\) size class. \( \text{N}_2\text{O} \) evasion per surface area unit was highest in the largest lakes reflecting the distribution of nitrate concentrations. In contrast, \( \text{CO}_2 \) evasion estimates per surface area unit (Kortelainen et al., 2006) were largest in small lakes.

Total annual \( \text{N}_2\text{O} \) flux from Finnish lakes \( (\text{total LA } 32,663 \text{ km}^2) \) was estimated as 0.6–0.8 Gg N\(_2\text{O}-\text{N}/\text{year}, based on the areas of the lake size distribution by Raatikainen and Kuusisto (1990) and the evasion estimates for the different lake size classes (Figure 9). Both Holgerson et al. (2017) and Heiskanen et al. (2014) approaches resulted in an estimate of 0.6 Gg N\(_2\text{O}-\text{N}/\text{year} for Finnish lakes, when the median \( \text{N}_2\text{O} \) evasion estimates of different lake size classes were multiplied with the respective lake surface area distribution. The Vachon and Prairie (2013) approach resulted in a little bit larger estimate of 0.8 Gg N\(_2\text{O}-\text{N}/\text{year} (Table 4; Tables S1 and S2).

Freshwater N cycling integrates numerous simultaneous temperature-dependent microbiological processes. Also, our data demonstrated

| TABLE 2 | Correlation coefficient values between \( \text{N}_2\text{O} \) and lake (area) and catchment area, maximum depth, latitude, land use cover, elevation, water temperature, and water chemistry in all lakes (randomly selected and eutrophic lakes), in surface water (all seasons), in bottom (all seasons), and in winter (all depths) |
|---------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| ln LA | 0.183*** | NS | 0.248*** | 0.140* |
| ln CA | 0.294*** | 0.154* | 0.342*** | 0.267*** |
| ln Maximum depth | 0.076* | NS | NS | NS |
| Lake latitude | -0.136*** | -0.194*** | -0.139* | -0.228*** |
| Forest % | NS | 0.100* | NS | NS |
| √Peat % | NS | -0.123* | NS | -0.155** |
| √Field % | 0.211*** | 0.232*** | 0.190*** | 0.345*** |
| √Built-up % | NS | NS | NS | 0.168** |
| √Water % | -0.168*** | -0.209*** | -0.102* | -0.215*** |
| Lake elevation | -0.195*** | -0.158*** | -0.193*** | -0.334*** |
| Water temperature | -0.378*** | -0.558*** | -0.304*** | -0.239*** |
| ln N\(_{\text{tot}}\) | 0.0692* | 0.332*** | NS | NS |
| ln NH\(_4\) | NS | 0.377*** | NS | -0.178** |
| ln NO\(_3\) | 0.593*** | 0.582*** | 0.612*** | 0.635*** |
| ln P\(_{\text{tot}}\) | NS | 0.194*** | NS | 0.141* |
| ln PO\(_4\) | NS | 0.381*** | NS | 0.115* |
| ln TOC | 0.0737* | 0.110* | NS | NS |
| ln Water color | NS | 0.147* | NS | NS |
| ln O\(_2\) | 0.189*** | -0.105* | 0.206*** | 0.276*** |
| ln Fe\(_{\text{tot}}\) | NS | 0.192*** | NS | NS |
| ln Conductivity | 0.148*** | 0.260*** | NS | 0.210*** |
| ln Alkalinity | NS | 0.143* | NS | NS |
| pH | NS | -0.120* | NS | NS |

Abbreviations: CA, catchment area; LA, lake area; NS, not significant; TOC, total organic carbon. *\( p < .05; **p < .01; ***p < .001. \)
FIGURE 4  The relationship between NO$_3$-N and N$_2$O (a), and NO$_3$-N and N$_2$O/NO$_3$-N ratio (b). All lakes, seasons, and depths. Lake type identified, all data were log-transformed. Our data across all lake types and seasons showed strong positive correlation between nitrate and N$_2$O and strong negative correlation between nitrate and N$_2$O/NO$_3$-N ratio.

FIGURE 5  Distribution (median, first and third quartile) of N$_2$O across depth; randomly selected lakes, all seasons. Median N$_2$O was evenly distributed across depth.

FIGURE 6  N$_2$O distribution across pH, different lake types identified as in Figure 3; all lakes, depths, and seasons. Both the highest and the lowest N$_2$O concentrations occurred around the median pH of 6.5.

FIGURE 7  N$_2$O concentrations in different lake types before (a) and after ice melt (b) across oxygen saturation percentage; all lakes and depths. Oxygen was not a key driver for N$_2$O in Finnish boreal lakes.
**FIGURE 8** Distribution (median, first and third quartile) of \( \text{N}_2\text{O} \) across elevation (a) and the percentage of agricultural land in the catchment (d). Distribution (median, first and third quartile) of \( \text{NO}_3\text{-N} \) across lake area (LA) (b) and season (e). Distribution (median, first and third quartile) of \( \text{N}_2\text{O}/\text{NO}_3\text{-N} \) ratio across LA (c) and season (f), randomly selected lakes, all depths.

**TABLE 3** Selected linear stepwise multiple regression equations for \( \text{N}_2\text{O} \) (water chemistry, land use cover, climatic drivers, and catchment topography were used as predictors; randomly selected and eutrophic lakes)

| Dataset      | n   | Dependent variable | Intercept | Parameter estimate | Parameter | Unit   | Model \( R^2 \) |
|--------------|-----|--------------------|-----------|--------------------|-----------|--------|-----------------|
| All data     | 1,396 | \( \ln \text{N}_2\text{O} \) = 2.31 ± 0.145 | \( \ln \text{NO}_3\text{-N} \) | µg/L | .41 |
|              |      |                    | 0.128     | \( \ln \text{O}_2 \) | %        | .51    |
|              |      |                    | -0.0303   | Temperature | °C      | .55    |
|              |      |                    |          | -0.496           | \( \sqrt{\text{Field \%}} \) | %      | .58    |
| Surface      | 397  | \( \ln \text{N}_2\text{O} \) = 2.74 ± 0.0952 | \( \ln \text{NO}_3\text{-N} \) | µg/L | .47 |
|              |      |                    | -0.0373   | Temperature | °C      | .56    |
|              |      |                    | 0.0377    |          | %        | .58    |
| Bottom       | 408  | \( \ln \text{N}_2\text{O} \) = 2.07 ± 0.258 | \( \ln \text{NO}_3\text{-N} \) | µg/L | .46 |
|              |      |                    | 0.496     | \( \ln \text{O}_2 \) | %        | .54    |
| Winter       | 379  | \( \ln \text{N}_2\text{O} \) = 123 ± 0.394 | \( \ln \text{NO}_3\text{-N} \) | µg/L | .52 |
|              |      |                    | -7.68     | \( \ln \text{Latitude} \) | °N      | .57    |
|              |      |                    | -0.191    | pH                 |         | .58    |
large differences in N$_2$O concentrations and estimated emissions among different lake types (Table 5; Table S2). Nevertheless, our data underline the key role of nitrate in regulating seasonal and spatial N$_2$O concentrations across boreal lakes. Nitrate explained as much as 78% of the variation in seasonal mean N$_2$O concentrations across all lakes and depths (Equation 4; Figure 10). While O$_2$ was the dominating...
Using Equation (4), the modeled \( N_2O \) concentrations for independent nitrate-nitrite dataset of 874 randomly selected Finnish NLS lakes reproduced similar median \( N_2O \) concentrations and an increasing trend by lake size further indicating that our lake dataset is representative to Finnish conditions and can be expected to represent lakes over larger boreal landscape (Figure 11).

Large lakes turned out to be disproportionately important \( N_2O \) sources among the lake population on the landscape level. Lakes larger than 10 km\(^2\) were estimated to contribute 77% of the total \( N_2O \) emission from Finnish lakes. In contrast, \( CO_2 \) evasion estimates demonstrated that lakes smaller than 10 km\(^2\) dominated landscape \( CO_2 \) evasion among the lake population while lakes larger than 10 km\(^2\) (representing 65% of the total LA distribution) represented only 45% of the estimated \( CO_2 \) evasion (Kortelainen et al., 2006). Our estimated \( N_2O-N \) emissions from lakes represent 17% of the \( N_2O-N \) emissions from boreal forests, the dominating ecosystem in Finland where lakes cover 10% of the total land area (Table 6).

### TABLE 6  Estimates of annual \( N_2O \) flux from Finnish and boreal lakes (calculated as median fluxes by lake size class, \( k \) values by Holgerson et al. (2017) compared to estimates of annual \( N_2O \) flux from boreal forest in Finland and in boreal zone (using the estimate of 17.6 mg \( N_2O-N \) m\(^{-2}\) year\(^{-1}\); Potter et al., 1996)

|                | Finland | Boreal region |
|----------------|---------|---------------|
| Lake \( N_2O-N \) flux (Gg/year) | 0.6     | 29            |
| Forest \( N_2O-N \) flux (Gg/year) | 4\(^a\) | 213\(^a\)     |
| LA (km\(^2\)) | 32,663\(^b\) | 1,422,448\(^d\) |
| Forest area (km\(^2\)) | 203,000\(^c\) | 21,200,000\(^d\) |
| Lake \( N_2O-N \) flux/Forest \( N_2O-N \) flux (%) | 17      | 14            |

Abbreviation: LA, lake area.
\(^a\)Potter et al. (1996).
\(^b\)Raatikainen and Kuusisto (1990).
\(^c\)Vahtera et al. (2018).
\(^d\)Lake area distribution estimated based on MODIS data, excluding lakes <0.1 km\(^2\).

\( N_2O - N \) (ng/L) = 374 e\(^{0.003 NO_3-N} \) (Equation 4)

\( N_2O \) concentrations were high in lakes located in low elevation catchments accompanied by large coverage of agricultural land (Figure 8a,d). Nutrient-rich, calcareous lakes had high \( N_2O \) concentrations, which were comparable with methane distribution...
The possible higher reduction of N_2O to N_2 as a result of denitrification, with high nitrate availability, favors N_2O accumulation in the dormant season. The variability in N_2O concentration was the largest around the median pH 6.5 (Figure 6), which may reflect optimal pH of the accumulation of N_2O, that is, net production of N_2O from nitrification and denitrification. Low pH inhibits the N_2O reductase which increases the N_2O to N_2 ratio in denitrification (Richardson, Felgate, Watmough, Thomson, & Baggs, 2009). Nitrification is further inhibited at high C:N ratios (Her & Huang, 1995), typical for boreal Finnish lakes and often accompanied by low pH (Kortelainen et al., 2013). Supportingly, Humic large lakes, including only seven lakes in our dataset, also had high N_2O concentrations (Figures 3 and 10).

Nitrous oxide peaked in winter similar to CO_2, while CH_4 concentration was significantly higher in summer (Figure 2). During the open water season, 29% of the surface water samples were under saturated with respect to the atmospheric equilibrium value of N_2O showing that lakes can also act as N_2O sinks, which supports results from Canadian freshwaters (Soued et al., 2015) and farm water bodies in the United States (Webb et al., 2019) underlining the uncertainty in the data presently used both in regional and global N_2O budgets. On average, N_2O concentrations were evenly distributed through the water column (Figure 5), which contrasts the vertical distribution patterns of CO_2 and CH_4 that accumulate above the sediment (Juutinen et al., 2009; Kortelainen et al., 2006). Comparable distribution of GHGs has been reported from 13 temperate lakes in Colorado Rocky Mountains (McCrackin & Elser, 2011).

Generally, N_2O production is limited by low N turnover and low N mineralization in the high latitude N limited ecosystems (Potter et al., 1996). In N limited boreal terrain, N turnover is rapid and internal N cycling across forested ecosystems dominates the spatial nitrate distribution, which often reflects more closely catchment land use cover and topography than N deposition—in spite of N deposition being the major N source (Palviainen, Lehtoranta, Ekholm, Ruoho-Airola, & Elser, 2011). Furthermore, estimated N_2O emissions per surface area unit were largest from large lakes (Figure 9) reflecting the distribution of nitrate concentrations. In contrast, concentrations and estimated emissions of CO_2 and CH_4 decrease with increase in LA (Denfeld, Kortelainen, Rantakari, Sobek, & Weyhenmeyer, 2016; Juutinen et al., 2009; Kortelainen et al., 2006).

Our results underscore simultaneous (Miettinen et al., 2015) long-term CO_2, CH_4, and N_2O measurements from freshwaters in order to better understand major controls of landscape GHG evasion. Freshwater GHG flux measurements have predominantly been carried out in summer, while our study underscores the importance of dormant season N_2O measurements. The lower the temperature the higher N_2O concentrations, which in our data underlie the link between seasonal variability of nitrate and N_2O concentrations, that is, lower concentrations in warm growing season compared to cold dormant period. Elevated N_2O emissions during winter snow cover period have been measured also, for example, in cropland (Groppman et al., 2001). Higher N_2O emission was shown to coincide with a greater number of freeze-thaw cycles that broke up soil macro aggregates and increased soil inorganic N pool (Ruan & Robertson, 2017). In a northern hardwood forest reduced N uptake by fine roots due to soil freezing was concluded to be the primary regulator of increasing nitrate season did not prevent N_2O accumulation (Figure 7). In summer, low nitrate content and high N_2O/NO_3-N ratio resulted from nitrate being consumed in primary production, denitrification, and other microbial processes. In freshwaters, 0%-4% of N is generally released as N_2O in denitrification (Mulholland et al., 2008; Seitzinger, 1988; Silvennoinen, Liikanen, Torssonen, Stange, & Martikainen, 2008).

The explanation power of our statistical models for N_2O concentrations in lakes (Table 3) is comparable to the power of the models developed for terrestrial N_2O emissions (Leppelt et al., 2014; Pärn et al., 2018). In the lake dataset from boreal southern Norway and Sweden, N_2O concentrations correlated positively with nitrate in summer (Yang et al., 2015). In contrast, association between nitrate and N_2O has been weak in temperate lakes. No significant correlation between nitrate and N_2O could be found across temperate headwater Irish lakes in summer (Whitfield, Aherne, & Baulch, 2011). Furthermore, N_2O fluxes could not be predicted by any measured environmental variables in aquatic network in temperate Quebec, Canada, where 40% of the relatively pristine inland waters were undersaturated in N_2O in summer (Soued et al., 2015).

### 4.2 Landscape scale patterns

Key processes and feedbacks of landscape scale GHG fluxes have remained poorly quantified. Dynamics of N_2O in our lakes at landscape scale did not follow those of CO_2 (Kortelainen et al., 2006) and CH_4 (Juutinen et al., 2009; Figure 1). Large lakes dominate the lake surface area distribution in Finland (Raatikainen & Kuusisto, 1990). Furthermore, estimated N_2O emissions per surface area unit were largest from large lakes (Figure 9) reflecting the distribution of nitrate concentrations. In contrast, concentrations and estimated emissions of CO_2 and CH_4 decrease with decrease in LA (Denfeld, Kortelainen, Rantakari, Sobek, & Weyhenmeyer, 2016; Juutinen et al., 2009; Kortelainen et al., 2006).

The N_2O/NO_3-N ratio correlated negatively with nitrate (Figure 4b) being lowest in large lakes (Figure 8c) which had higher nitrate content than the small lakes. Increase in nitrate availability favors denitrification rate in lakes, but the efficiency of denitrification declines with increasing N inputs (Mulholland et al., 2008; Rissanen, Tiirola, Hietanen, & Ojala, 2013). Seasonal variation in N_2O/NO_3-N ratio was also strong—highest in summer and lowest in winter (Figure 8f). Low temperature decreases the activity of N_2O reductase more than the activity of other reductases in denitrification (Veraart, de Klein, & Scheffer, 2011) which together with high nitrate availability favors N_2O accumulation in the dormant season. The possible higher reduction of N_2O to N_2 as a result of low oxygen content (Richardson et al., 2009) in dormant
export. Increasing nitrate and N$_2$O concentrations during dormant period might thus ultimately reflect declining N uptake in terrestrial ecosystems resulting in increasing nitrate concentrations in downstream freshwaters (Campbell, Socci, & Templer, 2014). Freeze-thaw-related N$_2$O fluxes were shown to be a major component of annual N$_2$O emissions also in boreal peatlands of Northeast China (Cui et al., 2016).

Freshwater ecosystems have often been ignored in consideration of the landscape GHG fluxes. We used global warming potential (GWP; GWP$_{100}$ = 265; Ciais et al., 2013) to estimate the effect of N$_2$O evasion from Finnish lakes on the climate as CO$_2$ equivalents. Our estimate (0.6 Gg N$_2$O-N/year; Table 4) represents 35% (the 25%-75% quartiles being 29%-41%) of the GWP of Finnish lake diffusive CH$_4$ emissions (Juutinen et al., 2009). It is estimated that ebullition may even double the methane emissions (Bastviken, Cole, Pace, & Tranvik, 2004; Juutinen et al., 2009; Wik et al., 2014). When the uncertainty related to methane ebullition is taken into account (i.e., scenarios with and without ebullition included), our N$_2$O estimate represents 15%-41% of the GWP of Finnish lake CH$_4$ emissions. Assuming similar atmospheric impact for boreal lakes in general gives an emission estimate of 29 Gg N$_2$O-N/year.

Annual mean and median N$_2$O flux estimates from our lakes including seasonal data were surprisingly close to each other (Table 5) while estimates based only on summer measurements underestimated annual N$_2$O emissions (Table 4). Furthermore, the different annual N$_2$O flux estimates resulting from the three approaches for gas transfer coefficients ($k$ values) and their dependence on lake size (Heiskanen et al., 2014; Holgerson et al., 2017; Vachon & Prairie, 2013) demonstrate that there is uncertainty due to limited measured $k$ values, especially for large lakes. Recently, Webb et al. (2019) showed widespread unexpected undersaturation (67%) of N$_2$O in eutrophic farm water bodies in the United States. In our data, 29% of the samples were undersaturated, majority of these were sampled during summer.

Our data demonstrated large seasonal variation of nitrate and N$_2$O in boreal lakes and the important role of winter in annual emission estimates, the neglect of which results in underestimation of annual N$_2$O flux estimates. During recent mild winters, the ice cover period has been shorter and warm autumns have delayed the freezing day. Assuming 1 month shorter ice cover period (we extended autumn to 3 months, since especially autumn temperatures have been rising during recent years) resulted in 15% larger evasion estimates. Climate change scenarios predict increasing temperature and precipitation for northern Europe (Ciais et al., 2013) accompanied by increasing frequency of freeze-thaw events which have been shown to result in enhanced nitrate and N$_2$O fluxes (Cui et al., 2016). Rising temperature has further been shown to result in earlier spring snow melt floods throughout northeastern Europe (Blöschl et al., 2017) which contributes to seasonal distribution of nitrate transport from land to lakes and further to the overall role of lakes as N$_2$O sources in the boreal landscape.

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**CONFLICT OF INTERESTS**

The authors declare no competing financial interests.

**DATA AVAILABILITY STATEMENT**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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**REFERENCES**

Bastviken, D., Cole, J., Pace, M., & Tranvik, L. (2004). Methane emissions from lakes: Dependence of lake characteristics, two regional assessments, and a global estimate. *Global Biogeochemical Cycles, 18*, GB4009. https://doi.org/10.1029/2004GB002238

Beaulieu, J. J., Tank, J. L., Hamilton, S. K., Wollheim, W. M., Hall, R. O. Jr., Mulholland, P. J., ... Thomas, S. M. (2011). Nitrous oxide emission from denitrification in stream and river networks. *Proceedings of the National Academy of Sciences of the United States of America, 108*, 214–219. https://doi.org/10.1073/pnas.111446108

Blöschl, G., Hall, J., Parajka, J., Perdigão, R. A. P., Merz, B., Almeida, A., ... Živković, N. (2017). Changing climate shifts timing of European floods. *Science, 357*, 588–590. https://doi.org/10.1126/science.aan2506

Butterbach-Bahl, K., Baggs, E. M., Dannenmann, M., Kiese, R., & Zechmeister-Boltenstern, S. (2013). Nitrous oxide emissions from soils: How well do we understand the processes and their controls? *Philosophical Transactions of the Royal Society B: Biological Sciences, 368*, 20130122. https://doi.org/10.1098/rstb.2013.0122

Campbell, J. L., Socci, A. M., & Templer, P. H. (2014). Increased nitrogen leaching following soil freezing is due to decreased root uptake in a northern hardwood forest. *Global Change Biology, 20*, 2663–2673. https://doi.org/10.1111/gcb.12532

Ciais, P. C., Bala, S. G., Bopp, L., Brovkin, J., Canadell, J., Ciais, P., ... Thornton, P. (2013). Carbon and other biogeochemical cycles. In T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, & J. Stott (Eds.), *Climate change 2013: The physical science basis*. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. (pp. 465–570). Cambridge, UK and New York, NY: Cambridge University Press.

Cui, Q., Song, C., Wang, X., Shi, F., Wang, L., & Guo, Y. (2016). Rapid N$_2$O fluxes at high level of nitrate nitrogen addition during freeze-thaw events in boreal peatlands of Northeast China. *Atmospheric Environment, 135*, 1–8. https://doi.org/10.1016/j.atmosenv.2016.03.053
Deemer, B. R., Harrison, J. A., Li, S., Beaulieu, J. J., DelSontro, T., Barros, N., ... Vonk, J. A. (2016). Greenhouse gas emissions from reservoir water surfaces: A new global synthesis. BioScience, 66, 949–964. https://doi.org/10.1093/biosci/biw117

DelSontro, T., Beaulieu, J. J., & Downing, J. A. (2018). Greenhouse gas emissions from lakes and impoundments: Upscaling in the face of global change. Limnology and Oceanography Letters, 3, 64–75. https://doi.org/10.1002/lol2.10073

Denfeld, B., Kortelainen, P., Rantakari, M., Sobek, S., & Weyhenmeyer, G. A. (2016). Regional variability and drivers of below ice CO2 in boreal and subarctic lakes. Ecosystems, 19, 461–476. https://doi.org/10.1007/s10021-015-9944-z

Fick, A. (1855). V. On liquid diffusion. The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science Series 4, 10(63), 30–39. https://doi.org/10.1080/14786445508419262

Groppman, P. M., Driscoll, C. T., Fahey, T. J., Hardy, J. P., Fitzhugh, R. D., & Tierney, G. L. (2001). Colder soils in a warmer world: A snow manipulation study in a northern hardwood forest ecosystem. Biogeochemistry, 56, 135–150. https://doi.org/10.1023/A:1013039830232

Heiskanen, J. J., Mammarella, I., Haapanala, S., Pumpanen, J., Vesala, T., Juutinen, S. A., … Martikainen, P. J. (2006). Sediment respiration and environmental controls of denitrification across different geographical scales. Aquatic Microbial Ecology, 25, 1–16. https://doi.org/10.3354/ame01619

Hu, M., Chen, D., & Dahlgren, R. A. (2016). Modeling nitrous oxide emissions from rice waters: A global assessment. Global Change Biology, 22, 3566–3582. https://doi.org/10.1111/gcb.13351

Jäähne, B., Heinz, G., & Dietrich, W. (1987). Measurements of the diffusion coefficients of sparingly soluble gases in water. Tellus B: Chemical and Physical Meteorology, 39, 80–91.

Juutinen, S., Rantakari, M., Kortelainen, P., Huttenen, J. T., Larmola, T., Alm, J., … Martikainen, P. J. (2009). Methane dynamics in different boreal lake types. Biogeoosciences, 6, 209–223. https://doi.org/10.5194/bg-6-209-2009

Juutinen, S. A., Rantakari, M., Kortelainen, P., Huttenen, J. T., Mattsson, T., Alm, J., … Martikainen, P. J. (2006). Sediment respiration and lake trophic state are important predictors of large CO2 evasion from small boreal lakes. Global Change Biology, 12, 1554–1567. https://doi.org/10.1111/j.1356-2846.2006.00116.x

Kortelainen, P., Rantakari, M., Pajunen, H., Huttenen, J. T., Mattsson, T., Juutinen, S., … Martikainen, P. J. (2013). Carbon evasion/accumulation ratio in boreal lakes is linked to nitrogen. Global Biogeochemical Cycles, 27, 363–374. https://doi.org/10.1002/gbc.20036

Leinonen, L. (Ed.). (2000). Air quality measurements. SVT Official Statistics of Finland. Helsinki: Finnish Meteorological Institute.

Leppelt, T., Dechow, R., Gebbert, S., Freibauer, A., Lohla, A., Augustin, J., … Strömgren, M. (2014). Nitrous oxide emission budgets and land-use driven hotspots for organic soils in Europe. Biogeochemistry, 11, 6595–6612. https://doi.org/10.5194/bg-11-6595-2014

Mannio, J., Räike, A., & Vuorenmaa, J. (2000). Finnish lake survey 1995: Regional characteristics of lake chemistry. Internationale Vereinigung Für Theoretische Und Angewandte Limnologie: Verhandlungen, 27, 362–367. https://doi.org/10.1080/03660770.1998.11901254

Mcauliffe, C. C. (1971). Gas chromatographic determination of solutes by multiple phase equilibration. Chemical Technology, 1, 46–51.

McCrackin, M. L., & Elser, J. J. (2011). Greenhouse gas dynamics in lakes receiving atmospheric nitrogen deposition. Global Biogeochemical Cycles, 25, GB4005. https://doi.org/10.1029/2010GB003897

Miettinen, H., Pumpen, J., Heiskanen, J., Aaltoinen, H., Mammarella, I., Ojala, A., … Rantakari, M. (2015). Towards a more comprehensive understanding of lacustrine greenhouse gas dynamics – Two-year measurements of concentrations and fluxes of CO2, CH4 and N2O in a typical boreal lake surrounded by managed forests. Boreal Environment Research, 20, 75–89.

Mulholland, P. J., Helton, A. M., Poole, G. C., Hall, R. O., Hamilton, S. K., Peterson, B. J., … Thomas, S. M. (2008). Stream denitrification across biomes and its response to anthropogenic nitrate loading. Nature, 452, 202–205. https://doi.org/10.1038/nature06686

National Board of Waters. (1981). Methods of water analysis employed by the Water Administration (in Finnish with English summary). Helsinki: National Board of Waters, 136 pp.

Palviainen, M., Lehtoranta, J., Ekholm, P., Ruoho-Airola, T., & Kortelainen, P. (2015). Land cover controls the export of terminal electron acceptors from boreal catchments. Ecosystems, 18, 343–358. https://doi.org/10.1007/s10021-014-9832-y

Pärn, J., Verhoeven, T. A., Butterbach-Bahl, K., Dix, N. B., Ullah, S., Aasa, A., … Mander, Ü. (2018). Nitrogen-rich organic soils under warm well-drained conditions are global nitrous oxide emission hotspots. Nature Communications, 9,1135. https://doi.org/10.1038/s41467-018-04197-6

Potter, C. S., Matson, P. A., Vitousek, P. M., & Davidson, E. A. (1996). Process modeling of controls on nitrogen trace gas emissions from soils worldwide. Journal of Geophysical Research, 101, 1361–1377. https://doi.org/10.1029/95JD02028

Raatikainen, M., & Kuusisto, E. (1990). The number and surface area of the lakes in Finland. Terra, 102, 97–110.

Rekolainen, S., Mitikka, S., Vuorenmmaa, J., & Johansson, M. (2005). Rapid decline of dissolved nitrogen in Finnish lakes. Journal of Hydrology, 304, 94–102. https://doi.org/10.1016/j.jhydrol.2004.07.024

Richardson, D., Felgate, H., Watmough, N., Thomson, A., & Baggs, E. (2009). Mitigating release of the potent greenhouse gas N2O from the nitrogen cycle – Could enzymic regulation hold the key? Trends in Biotechnology, 27, 388–397. https://doi.org/10.1016/j.tibtech.2009.03.009

Rissanan, A. J., Tiirola, M., Hietanen, S., & Ojala, A. (2013). Interlake variation and environmental controls of denitrification across different geographical scales. Aquatic Microbial Ecology, 69, 1–16. https://doi.org/10.3354/ame2016

Ruan, L., & Robertson, G. P. (2017). Reduced snow cover increases wintertime nitrous oxide (N2O) emissions from an agricultural soil in the Upper US Midwest. Ecosystems, 20, 917–927. https://doi.org/10.1007/s10021-016-0077-9

Saitzinger, S. P. (1998). Denitrification in freshwater and coastal marine ecosystems: Ecological and geochemical significance. Limnology and Oceanography, 33, 702–724.

Silvennoinen, H., Liikanen, A., Torssonen, J., Stange, F., & Martikainen, P. J. (2008). Denitrification and N2O effluxes in the Bothnian Bay (Northern Baltic Sea) river sediments as affected by temperature under different oxygen conditions. Biogeochemistry, 88, 63–72. https://doi.org/10.1007/s10533-008-9194-7

Soued, C., del Giorgio, P. A., & Maranger, R. (2015). Variability in N2O fluxes across boreal aquatic networks. Nature Geoscience, 9, 116–120.

Vaaherä, E., Aarne, M., Ihalainen, A., Mäki-Simola, E., Peltola, A., Torvelainen, J., … Ylitalo, E. (2018). Finnish forest statistics. Helsinki, Finland: Natural Resources Institute Finland (Luke), 189 pp.

Vachon, D., & Prairie, Y. T. (2013). The ecosystem size and shape dependence of gas transfer velocity versus wind speed relationships in

https://doi.org/10.1007/978-3-540-81387-9
Veraart, A. J., de Klein, J. J. M., & Scheffer, M. (2011). Warming can boost denitrification disproportionately due to altered oxygen dynamics. *PLoS ONE, 6*, 1–6. https://doi.org/10.1371/journal.pone.0018508

Wanninkhof, R., & Knox, M. (1996). Chemical enhancement of CO₂ in natural waters. *Limnology and Oceanography, 41*, 689–697. https://doi.org/10.4319/lo.1996.41.4.0689

Webb, J. R., Hayes, N. M., Simpson, G. L., Leavitt, P. R., Baulch, H. M., & Finlay, K. (2019). Widespread nitrous oxide undersaturation in farm waterbodies creates an unexpected greenhouse gas sink. *Proceedings of the National Academy of Sciences of the United States of America, 116*(20), 9814–9819. https://doi.org/10.1073/pnas.1820389116

Whitfield, C. J., Aherne, J., & Baulch, H. M. (2011). Controls on greenhouse gas concentrations in polymeric headwater lakes in Ireland. *Science of the Total Environment, 410*, 217–225. https://doi.org/10.1016/j.scitotenv.2011.09.045

Wik, M., Thornton, B. F., Bastviken, D., MacIntyre, S., Varner, R. K., & Crill, P. M. (2014). Energy input is primary controller of methane bubbling in subarctic lakes. *Geophysical Research Letters, 41*, 555–560. https://doi.org/10.1002/2013GL058510

Yang, H., Andersen, T., Dörsch, P., Tominaga, K., Thrane, J. E., & Hessen, D. O. (2015). Greenhouse gas metabolism in Nordic boreal lakes. *Biogeochemistry, 126*, 211–225. https://doi.org/10.1007/s10533-015-0154-8

**SUPPORTING INFORMATION**

Additional supporting information may be found online in the Supporting Information section.

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