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Catchment properties as predictors of greenhouse gas concentrations across a gradient of boreal lakes

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Boreal lakes are the most abundant lakes on Earth. Changes in acid rain deposition, climate, and catchment land use have increased lateral fluxes of terrestrial dissolved organic matter (DOM), resulting in a widespread browning of boreal freshwaters. This browning affects the aqueous communities and ecosystem processes, and boost emissions of the greenhouse gases (GHG) CH₄, CO₂, and N₂O. In this study, we predicted biotic saturation of GHGs in boreal lakes by using a set of chemical, hydrological, climate, and land use parameters. For this purpose, concentrations of GHGs and nutrients (organic C, -P, and -N) were determined in surface water samples from 73 lakes in south-eastern Norway covering wide ranges in DOM and nutrient concentrations, as well as catchment properties and land use. The spatial variation in saturation of each GHG is related to explanatory variables. Catchment characteristics (hydrological and climate parameters) such as lake size and summer precipitation, as well as NDVI, were key determinants when fitting GAM models for CH₄ and CO₂ saturation (explaining 71 and 54%, respectively), while summer precipitation and land use data were the best predictors for the N₂O saturation, explaining almost 50% of deviance. Our results suggest that lake size, precipitation, and terrestrial primary production in the watershed control the saturation of GHG in boreal lakes. These predictions based on the 73-lake dataset was validated against an independent dataset from 46 lakes in the same region. Together, this provides an improved understanding of drivers and spatial variation in GHG saturation in boreal lakes across wide gradients of lake and catchment properties. The assessment highlights the need to incorporate multiple explanatory parameters in prediction models of GHGs for extrapolation across the boreal biome.

KEYWORDS

catchment, lake interactions, boreal lake, methane biogeochemistry, land use, land cover change, brownification

1 Introduction

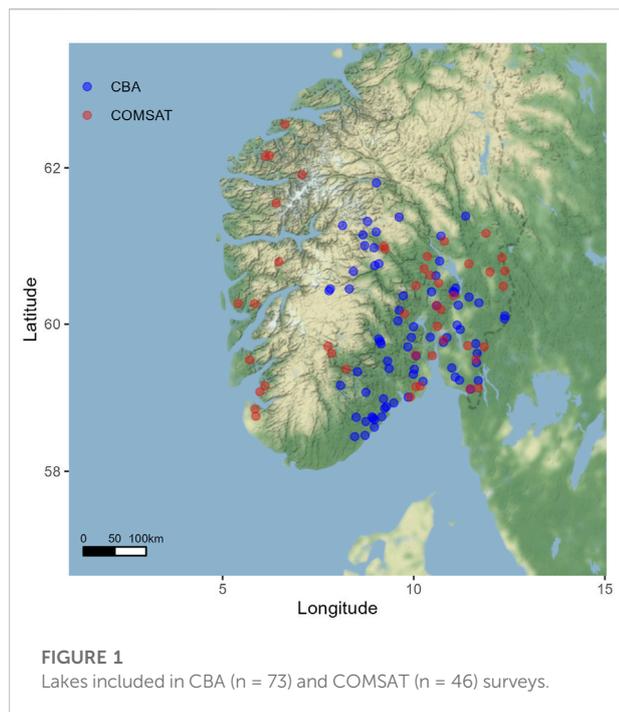
Boreal ecosystems are expected to be among the most affected biomes by climate change (Ruckstuhl et al., 2008). Regarding inland waters, boreal lakes are the most abundant lakes on Earth (Schindler, 1998). These lakes receive high loading of allochthonous dissolved organic matter (DOM) from the catchment (Dillon and Molot, 1997; Tranvik et al., 2009). As such, these lakes are crucial biogeochemical reactors: on the one hand, they sequester DOM by burial into lake sediments, while on the other hand, they are generally net heterotrophic and act as major conduits for greenhouse gas (GHGs) emissions (Hessen et al., 1990, 2017; Bastviken et al., 2004; Tranvik et al., 2009; Yang et al., 2015; de Wit et al., 2018).

In boreal watercourses the lateral fluxes of colored DOM have increased during the past 2–3 decades, mainly due to reduced deposition of acid rain (de Wit et al., 2007; Monteith et al., 2007), climate change (Mattsson et al., 2005), afforestation and land use changes (Palviainen et al., 2016; Škerlep et al., 2020). The increasing DOM is causing a widespread “browning” of these freshwater ecosystems (Finstad et al., 2016). In the boreal biome, changing climate will result in increased precipitation and runoff (Tranvik et al., 2009). In some regions of the northern hemisphere, like Fennoscandia (the common term for Finland, Norway and Sweden) in general, the proportion of forest cover has increased, leading to larger biomass pools (Fang et al., 2014). This increase in terrestrial biomass (“greening”) has boosted the amount of allochthonous DOM that may be entering into surface waters (Larsen et al., 2011b), with increased rainfall and runoff further enhancing DOM catchment export (Tranvik et al., 2009; de Wit et al., 2016). Browning strongly affects light attenuation (Karlsson et al., 2009; Thrane et al., 2014; Alleesson et al., 2021), nutrient dynamics (Dillon and Molot, 2005; Creed et al., 2018) and thus also primary productivity (Solomon et al., 2015; Hessen et al., 2017; Lau et al., 2021). The combined effect of increased DOM and increased light attenuation implies a decrease in photosynthesis, thus promoting the heterotrophy of these systems and the net emissions of GHGs derived from microbial mineralization of DOM (Yang et al., 2015).

A number of studies have addressed the main drivers of carbon dioxide (CO₂) and methane (CH₄) concentrations in boreal lakes, and how they are linked to DOM and lake productivity (Huttunen et al., 2003b; Bastviken et al., 2004; Juutinen et al., 2009; de Wit et al., 2018; Jahr, 2021) as well as catchment properties (Huttunen et al., 2003b; Li et al., 2020). Fewer studies have addressed nitrous oxide (N₂O) in boreal lakes. N₂O is an intermediate in the nitrification and denitrification (Trogler, 1999; Butterbach-Bahl et al., 2013) whose production depends on the availability of organic matter (OM), oxygen

availability, and reactive nitrogen concentration (Yang et al., 2015; Kortelainen et al., 2020; Clayer et al., 2021). Hydrological parameters, such as lake size or runoff, have been recognized as key underlying factors determining which are the main biogeochemical C processes that are governing the production of GHGs (Jones et al., 2018). In the boreal zone, small lakes have been shown to have relatively higher sedimentation and GHG emission rates than larger lakes (Juutinen et al., 2009; Kankaala et al., 2013). Typically, small lakes are also characterized by higher concentrations of allochthonous OM (Xenopoulos et al., 2003; Einola et al., 2011) and shorter water residence times (Vachon et al., 2017). The residence time has also a strong influence on the quality of DOM. The longer the residence time, the more refractory the DOM becomes, and thus the proportion of C sequestered in the sediments increases relative to what is released as CO₂ or CH₄ (den Heyer and Kalf, 1998). Most of this production is microbial, yet for all gases the concentrations and emissions are also affected by uptake and conversions by methanogens, denitrifiers, and autotrophs, but also by abiotic factors affecting microbial activity (i.e., temperature, redox conditions). These biotic processes are labelled biogenic and, therefore, traceable (Jones and Grey, 2011). For CO₂, the concentrations will also be affected by other processes as photooxidation and in cases also inputs from tributaries and groundwater, yet integrated over the water column of boreal lakes, the biogenic activities generally are by far the most important (Hessen et al., 1990; Larsen et al., 2011a; Alleesson et al., 2021).

The goal of this study was to model biotic saturation of GHGs in boreal lakes by using a set of chemical, hydrological, climate, and land use parameters. To date, only a limited number of studies have explored the influence of such factors in GHG saturations from boreal lakes. For this reason, we constructed predictive models for GHG saturation from this survey performed in 2019 (73 lakes) and tested them against an independent dataset of 46 lakes sampled from the epilimnion of lakes in mid-summer 2011 by somewhat different protocols but covering a similar area of southern Norway (Yang et al., 2015). This validation by an independent dataset is a novel take, and both these datasets differ from previous studies on basically lowland sites by encompassing an unusual range in DOM, from very clear, ultraoligotrophic alpine lakes to really brown lowland sites, surrounded by bogs and coniferous forests. The wide span in catchment properties is also reflected in the strong gradient in forest cover, precipitation and temperatures, helping to sort out the relative contribution from these potential drivers on the various GHGs. We tested the hypothesis that differences in GHG saturation can be predicted by water chemistry, catchment properties (hydrological and climate parameters),



and land use variables. This approach aids to better predict the effect of on GHG emissions from pristine boreal lakes in climate change scenarios by combining well standardized watershed variables.

2 Material and methods

2.1 Field survey

Surface water samples were collected during autumn 2019 from 73 lakes in south-eastern Norway. This lake survey was conducted by the Centre for Biogeochemistry in the Anthropocene (CBA) at University of Oslo, hence these lakes are labelled “CBA dataset” hereafter. The lakes were selected as a subset of lakes monitored in a concomitant 1000-lake survey of the whole of Norway, conducted by the Norwegian Institute of Water research (NIVA). This synoptic survey repeated previous campaigns conducted in 1986 and 1995 (Henriksen et al., 1998). The CBA dataset span a wide range of water quality properties (e.g., DOM, nutrients) (Crapart et al., 2021), catchment properties, and land use (Lie, 2021). Sampling was performed in late fall, during or after lake overturn. Samples were collected approximately 4 m from the shore by means of a sampling rod with a sampling beaker. Composite water (≈ 2 L) was collected in a sampling bucket, with as minimal physical disturbance. Where feasible, the outlet of the lake was used as a sampling point. Due to logistical reasons, this was the only feasible way to sample all the lakes of this study. The lake edge may be more responsive to

external inputs (i.e., precipitation, sunlight), likely acting more as a hotspot for GHG turnover compared to deeper sections of the lake. However, we believe that such bias is compensated in our models by including watershed hydrological characteristics (Section 2.4). In general, we assume that in these quite wind-exposed lakes, the gases are equally distributed in the epilimnion, but in sites with ebullition from the deepest part, we cannot exclude a non-homogenous distribution. In order to validate the models calibrated using the CBA dataset, a second dataset was used. This dataset, referred as COMSAT hereafter, includes 46 large southern Norwegian lakes sampled in the middle of the lakes by hydroplane in July and August 2011 (Thrane et al., 2014; Yang et al., 2015; Andersen et al., 2020). The selection of lakes was done with different constraints like size (>2 km²) or water pH (>5), as detailed previously (Thrane et al., 2014). The locations of the lakes sampled in both campaigns are shown in Figure 1.

2.2 Chemical analysis

Water temperature (T), pH, and electrical conductivity (EC) were measured at the CBA lakes immediately after sample collection. From each site, 50 ml of unfiltered water was collected and stored at 10°C in polypropylene tubes. Upon arrival at the laboratory, these samples were frozen at -20°C until total organic carbon (TOC), total nitrogen (TN), and total phosphorus (TP) determination at the University of Oslo. TOC was measured by infrared CO₂ detection after catalytic high temperature combustion (Shimadzu TOC-VWP analyzer). TN was measured by detecting nitrogen monoxide by chemiluminescence using a TNM-1 unit attached to the Shimadzu TOC-VWP analyzer. TP was measured on an auto-analyzer as phosphate after wet oxidation with peroxodisulfate. We here use TOC as a proxy of DOM since this is by far the dominant constituent of DOM. By mass DOM is equivalent of DOC, and DOC make typically up some 95% of TOC in these boreal, low-productivity lakes (Larsen et al., 2011a; 2011b).

Concentrations of dissolved Argon (Ar), O₂, N₂, CH₄, CO₂, and N₂O were determined in duplicate for each lake water sample using the acidified headspace technique (Åberg and Wallin, 2014). From the surface of each lake, a volume of 30 ml was carefully collected directly into a syringe instead of collecting from the bucket to avoid potential disturbances (e.g., outgassing). Subsequently, a 20 ml headspace was created with atmospheric air in each syringe, before 0.6 ml of 3% HCl (≈ 1 M) was added. Syringes were closed and equilibrium was reached in the headspace at field temperature by shaking them for 3 min. Finally, 15 ml of the headspace gas was transferred into 12 ml evacuated vials and kept at room temperature until further analysis. Gas concentrations were determined at the Norwegian University of Life Sciences by automated gas chromatography (GC) analysis following the methodology

outlined by Yang et al. (2015). In brief, 2 ml of headspace gas were sampled (autosampler GC-Pal, CTC, Switzerland) and injected into a GC with He back-flushing (Agilent 7890A, Santa Clara, CA, United States). The GC was equipped with a 20-m wide-bore (0.53 mm) Poraplot Q column operated at 38°C and with He as carrier gas for separation of CH₄, CO₂, and N₂O from bulk gases (i.e., Ar, N₂, and O₂). N₂O and CH₄ were measured with an electron capture detector (ECD) operating at 375°C, and a flame ionization detector (FID), respectively. All other gases were measured with a thermal conductivity detector (TCD). Certified standards of CO₂, N₂O, and CH₄ in He were used for calibration (AGA, Germany), whereas N₂, O₂, and Ar were calibrated against air. The analytical precision for all gases was better than 1%.

2.3 Biogenic GHG saturation

Saturation relative to atmospheric equilibrium was calculated for all gases. For this purpose, Henry's law constants for 25°C were temperature adjusted to *in situ* water temperatures using the Clausius–Clapeyron equation with gas-specific solution enthalpies (Sander, 2015). Concentrations of GHGs in equilibrium with the atmosphere were calculated using the temperature adjusted Henry's law constants. The concentrations of GHGs in the surface water of the lakes were normalized relative to the concentration of dissolved Argon (Ar). Unlike Ar, which in water is only controlled by physical processes, O₂, N₂, CH₄, CO₂ and N₂O concentrations in water governed by both physical and biogenic processes (Aeschbach-Hertig et al., 1999). For this reason, the relative saturation of GHGs normalized to Ar was used as a proxy for the saturation of the biogenically derived lake GHG turnover (Yang et al., 2015). Thus, unless otherwise specified, hereafter the variables CH₄, CO₂ and N₂O are mentioned to refer to the normalized relative saturations of each GHG. A similar approach was used for the gas data collected during the COMSAT campaign (Thrane et al., 2014).

2.4 Catchment characteristics: Hydrological and climate parameters

Lake Altitude (in meters above sea level, m a.s.l.), lake surface (LakeArea in km²), and catchment surface (CatchmentArea in km²) were obtained using the “Lake database” (<https://temakart.nve.no/tema/innsjodatabase>) from the Norwegian Water Resources and Energy Directorate (NVE, <https://www.nve.no>). Drainage ratio (Drainage) was calculated as the ratio between catchment area and lake surface area, both in km². Lake depth (LakeDepth in m) was measured from an Airbus AS350 helicopter with an echosounder held within the top 0.5 m of the lake water column above the expected deepest point

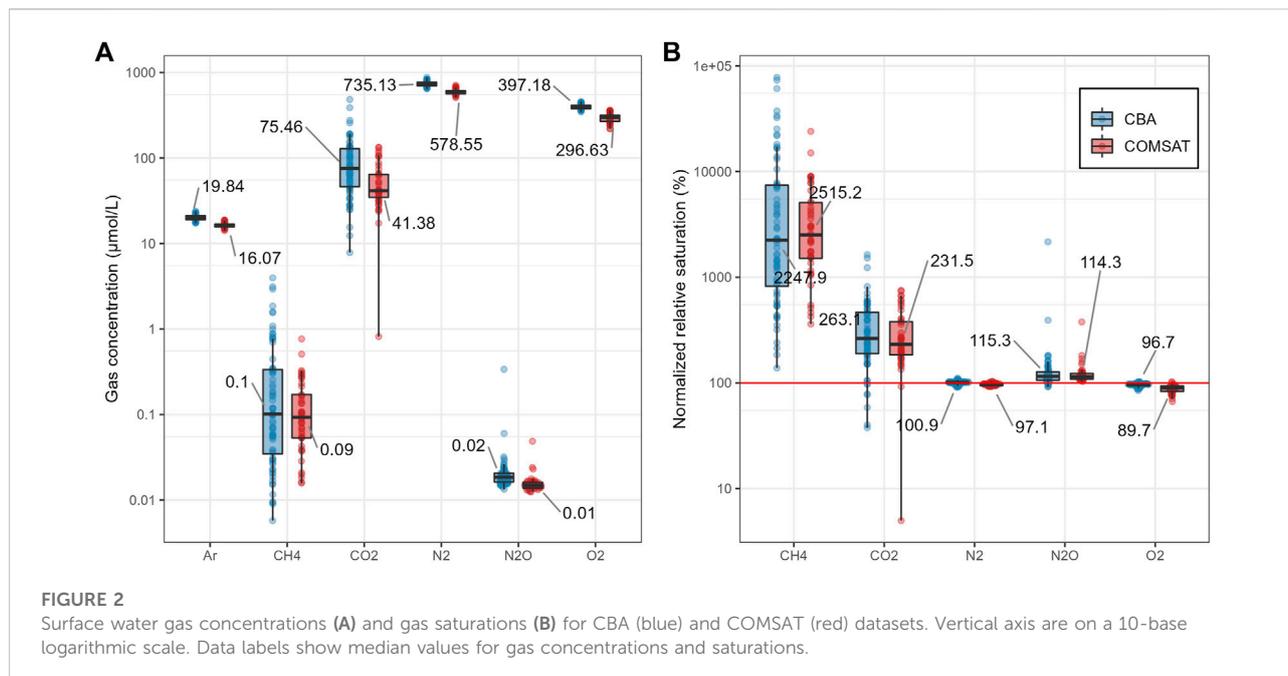
(Hindar et al., 2020). The remaining hydrological and climate parameters were compiled from the “NEVINA” database provided by the NVE (<http://nevina.nve.no/>). These were: specific runoff (Runoff in L s⁻¹ km⁻²); average inclination in the watershed (Slope in %); average annual air temperature (AnnualT in °C); average air temperature during the summer period (01/05–30/09) (SummerT in °C); average air temperature during the winter period (01/10–30/04) (WinterT in °C); average annual precipitation (AnnualP in mm); average precipitation during the summer period (SummerP in mm); and average precipitation during the winter period (WinterP in mm). Water residence time (Residence in yr) was estimated for each catchment from lake surface, lake depth, catchment surface, and specific runoff by assuming a cone-shaped morphometry for all lakes (Lindström et al., 2005).

2.5 Land use coverage

Land use data for each catchment were obtained from the Copernicus Land Monitoring Service (CLMS), using CORINE Land Cover (CLC) databases 2018 and 2012, for CBA and COMSAT, respectively (see (Feranec et al., 2007) for further details). In the CLC database the land use is classified into seven categories: agricultural areas (Cultivated), forested areas (Forest), human-derived and artificial impervious areas (Artificial), open areas with little or no vegetation (Fell), areas covered by glaciers and perpetual snow (Glacier), inland wetland areas (Peatland), and inland waterbodies in the watershed, including lake's surface itself (Waterbodies). The CLC databases were clipped and intersected with the catchment areas using the open-source software QuantumGIS (QGIS, version 3.20.1). The area covered by the different land use types were calculated as percentage of the total surface using QGIS field calculator. In addition, the average Normalized Difference Vegetation Index (NDVI) value was calculated for each catchment and year (2019 and 2011, for CBA and COMSAT, respectively) using the average values of the summer months (June, July, and August) obtained from Copernicus data (<https://land.copernicus.eu/global/products/NDVI>).

2.6 Statistical analysis

Data analysis was performed using the open-source software R version 4.1.0 (R Core Team, 2021). The package *raster* (Hijmans et al., 2015) was used to obtain average NDVI values per catchment. To determine significant differences in dissolved gas concentrations between CBA and COMSAT datasets, the Mann-Whitney test was used at a significance level of $p < 0.05$. Variables were checked for normality using the *bestNormalize* package (Peterson, 2021) and transformed to logarithmic, exponential, or square root where needed to reduce



heteroscedasticity and obtain normally distributed residuals. However, variables were not standardized (i.e., mean centering and scaling) as comparisons of rescaled coefficients across datasets are problematic (King, 1986). A detailed list with the applied transformations is listed in the Supplementary Table S1. Once the transformations were performed, correlations were calculated using Spearman's correlation coefficients. In addition, principal component analysis (PCA) was performed for dimension reduction. This analysis was followed by K-means clustering. This non-hierarchical clustering method aims to partition the studied variables into k groups such that the sum of squares from points to the assigned cluster centres is minimized. Variation partitioning (function *varpart* in *vegan* package) (Oksanen et al., 2013) was used to evaluate the percentage of variance in biogenic GHG saturations that were in these clusters. Statistical modelling was carried out using the *mgcv* package (Wood and Wood, 2015) by fitting generalized additive models (GAM) to predict selected dependent variables (CH_4 , CO_2 , and N_2O). The selection of the predictive variable in the GAM models was performed by applying additional shrinkage on the null space of the penalty (*select = TRUE* argument in the *mgcv:gam* function) (Marra and Wood, 2011). To compare the contribution of each factor in explaining the total deviance of the model, we also formulated reduced models where we removed one of the terms at a time, as well as a null model. To be able to compare deviances, we fixed the smoothing parameters to the original model containing all terms (*sp* argument in the *mgcv:gam* function) and then calculated the contribution of each factor in the model as described previously (Ribic et al., 2010).

3 Results

3.1 Greenhouse gases

Across CBA lakes, the median surface concentrations of Ar, N_2 , and O_2 were 19.8, 735, and 397 $\mu\text{mol/L}$, respectively. For GHGs, median concentrations were 0.10, 75.5, and 0.02 $\mu\text{mol/L}$ for CH_4 , CO_2 , and N_2O , respectively (Figure 2A). GHG concentrations ranged widely across the CBA lakes, with CH_4 having a greater relative variability (CV = 183%) than N_2O (CV = 158%) and CO_2 (CV = 82.5%). On the contrary, Ar, N_2 , and O_2 showed smaller coefficients of relative variation (7.6, 6.7, and 7.0%, respectively). The biogenic gas saturations were low for O_2 (median = 96.7%) and N_2 (median = 101%), while they were high for the GHGs: N_2O (median = 115%), CO_2 (median = 263%), and CH_4 (median = 2248%) (Figure 2B).

There were significant differences between all gas concentrations in the CBA data and the COMSAT datasets ($p < 0.05$, Mann-Whitney), except for CH_4 ($p = 0.50$) (Figure 2A). However, by comparing the relative biogenic gas saturations between both datasets, significant differences were found only for N_2 and O_2 ($p < 0.05$) (Figure 2B). The saturations of the biogenically derived CH_4 ($p = 0.74$), CO_2 ($p = 0.42$), and N_2O ($p = 0.86$) did not differ statistically between the two datasets.

3.2 Chemical-, catchment- and land use data

Besides varying in pH (4.4–7.9) and conductivity (0.39–151 $\mu\text{S/cm}$), CBA lakes varied widely in TOC from

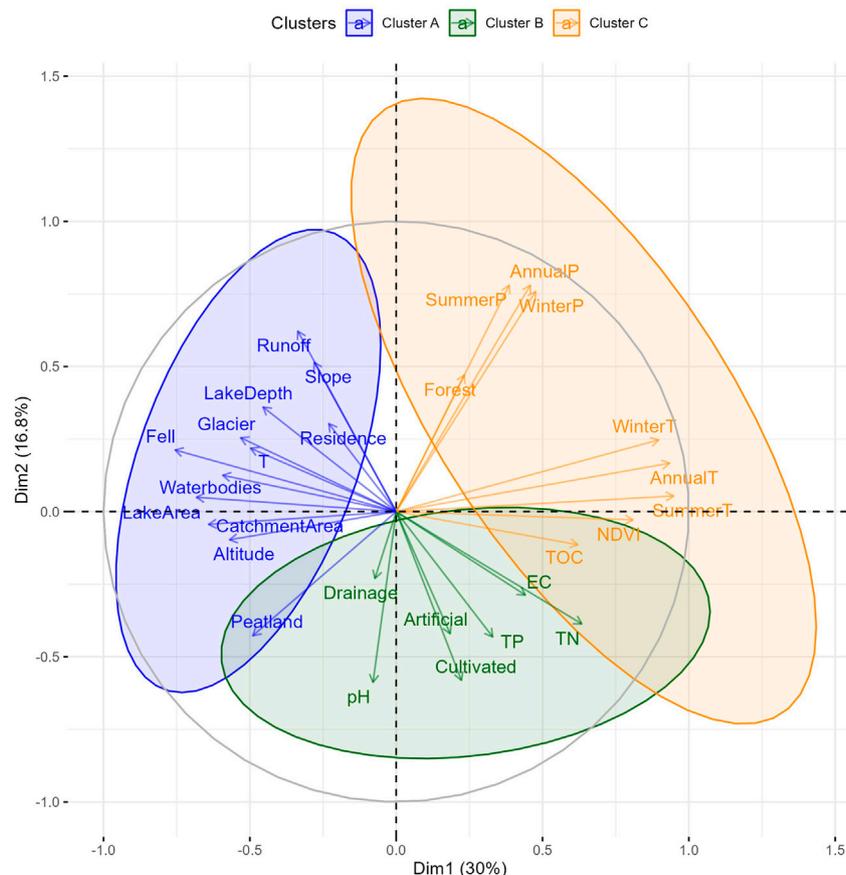


FIGURE 3

PCA analysis of the transformed variables depicting the two main principal components (PC) of parameter loading with the three defined clusters labeled in blue, green and orange (Clusters A, B and C, respectively). The two PCA axes explain 36.8% of total variability in the dataset.

2.54 to 116 mg/L (median = 13.3 mg/L), TN from 0.05 to 1.07 mg/L (median = 0.27 mg/L), and TP from 3.0 to 48.5 $\mu\text{g/L}$ (median = 8.90 $\mu\text{g/L}$). A complete description of the chemical data can be found in the [Supplementary Figure S1](#).

Catchment and Lake sizes varied from 0.02 to 369 km^2 (median = 0.93 km^2) and from 0.48 to 16,530 km^2 (median = 11.0 km^2), respectively. Drainage ratios varied from 2.76 to 405.8 (median = 24.6). The depths of the lakes ranged from 0.60 to 333 m (median = 12.0 m) and they are located at altitudes between 4 and 1,151 m a.s.l. (median = 190 m a.s.l.). Watershed slopes varied between 0.9 and 18.3% (median = 7.8%), while runoff values ranged from 8.0 to 41.8 $\text{L s}^{-1} \text{km}^{-2}$ (median = 18.1 $\text{L s}^{-1} \text{km}^{-2}$). Estimated water residence times varied widely from 0.01 to 7.4 years (median = 0.29 years). Detailed information about the catchment variables is provided in the [Supplementary Figure S2](#).

Most of the sampling sites in the CBA survey are first order lakes receiving drainage from pristine catchments. Forest was the main dominating land use type the watersheds (Forest, median =

83.7%), followed by inland waterbodies (Waterbodies; median = 3.7%) and agricultural land (Cultivated; median = 1.2%). Although generally comprising minor fractions of the catchment, peats and bogs (Peatland) and human-derived impervious surfaces (Artificial) reached coverages of up to 40.2 and 32.0%, respectively, in single catchments (see [Supplementary Figure S3](#)).

3.3 Explorative statistical analyses

Multivariate statistical analysis was performed on the transformed variables from the CBA dataset in order to explore whether the saturation of the biogenically derived GHGs significantly differed among explanatory variable groups. PCA of the transformed variables showed that 13 components were needed to explain 95% of the variance, with the first three components accounting for 59.1% of the total variance (30.0, 16.8, and 12.3%, respectively). Subsequent

K-means clustering analysis revealed three main groups of explanatory variables (Figure 3). The purpose of K-Means analysis is to group variables by minimizing data similarity between clusters, while maximizing data similarity within each cluster.

Cluster A (within the blue circle in Figure 3) includes water temperature measured during sampling (T) and non-forested land use (Waterbodies, Peatland, Fell, and Glacier), along with catchment physical properties (CatchmentArea, LakeArea and LakeDepth, Altitude, Residence, Slope, and Runoff). Cluster B (within the green circle in Figure 3) contains the water physicochemical parameters (pH and EC), nutrients (TN and TP), the coverage of Artificial and Cultivated land use types, as well as the Drainage ratio. Cluster C parameters (orange circle in Figure 3) are the biotic-related variables (Forest coverage, NDVI, and TOC) as well as climate factors in the watershed (temperature and precipitation).

Spearman rank-correlation analyses, using log transformed variables, provided information about internal correlation between the variables within the clusters, but also with the biogenic GHGs. Biogenic CH₄ normalized saturation was negatively correlated with lake size (LakeArea; $R = -0.78$; $p < 0.001$), watershed size (CatchmentArea; $R = -0.70$; $p < 0.001$), and lake depth (LakeDepth; $R = -0.50$; $p = 0.003$), whereas it was positively correlated with air temperature in the catchment (AnnualT; $R = 0.55$; $p < 0.001$) and NDVI ($R = 0.61$; $p < 0.001$). NDVI was positively correlated with average air temperature (AnnualT; $R = 0.69$; $p < 0.001$) (reflecting the altitude gradient) and thus Forest cover ($R = 0.53$; $p < 0.001$), whereas it was less negatively correlated with the percentage of water in the catchment (Waterbodies; $R = -0.54$; $p < 0.001$). The percentage of water in the watershed was also negatively correlated with the biogenic CO₂ normalized saturation ($R = -0.49$; $p = 0.004$), and as expected, positively correlated with lake size (LakeArea; $R = 0.75$; $p < 0.001$) and catchment size (CatchmentArea; $R = 0.64$; $p < 0.001$). Spearman's correlation plots are provided in Supplementary Figure S4.

Variation partitioning analyses (VPA) with transformed variables were carried out to assess relative contributions of the variables, grouped into the above defined clusters, to the biotic saturations of GHGs (Supplementary Figure S5). The combination of these variables explained 65.8% of the observed variation in biotic CH₄ saturation (Supplementary Figure S5A). The VPA showed that 38.1% of the total variation was shared between Cluster A, related mainly to catchment physical properties, and Cluster C, related to biotic and climate factors. Cluster A alone explained a large proportion of the variation (22.5%). The variance of biotic CO₂ saturation that could be explained by combining the whole set of variables was 39.7% (Supplementary Figure S5B). The largest explanatory value was shared by the three clusters (18.6%), explaining alone 4.4% (Cluster A) and 2.2% (Cluster B). Only 18.3% of the total variation of N₂O saturation was explained by the three clusters, with the variables within Cluster B providing most

of the explanatory value to the spatial variation in biogenic N₂O saturation (11.4%) (Supplementary Figure S5C). This approach seems to be useful for CH₄ saturation. However, the high residuals observed for CO₂ and N₂O saturation makes the VPA not so informative about the contribution of each cluster to the total variation.

3.4 Modeling biogenic CH₄ saturation

Based on the explorative analyses (i.e., PCA, correlation, and VPA analyses), a stepwise selection was done to fit a GAM model (Model 1) to the transformed (i.e., log) normalized relative CH₄ saturation by minimizing the residual deviance. Lake surface (LakeArea; log), NDVI (exp) and average summer precipitation (SummerP; sqrt) were selected as independent variables, explaining 71.1% of the total variance with a $R^2_{adj} = 0.691$ (Table 1). LakeArea was the factor that explained the greatest variance by itself (26.9%), followed by SummerP (4.12%) and NDVI (4.10%) (Table 2). The variable selection is in line with the results from VPA, showing that clusters A (i.e., LakeArea) and C (i.e., SummerP and NDVI) explained a large proportion of the variation in CH₄ saturation. All independent variables were statistically significant ($p < 0.005$). Effective degrees of freedom (edf) for the factors were not close to 1. Therefore, smoothers were applied in the model with the default number of knots ($k = 10$), as shown in Figure 4. Validation of Model 1 with the COMSAT dataset, by comparing the predicted values (Y-axis) with the measured values (X-axis), resulted in a R^2_{adj} of 0.18 ($p < 0.005$). Diagnostics for Model 1 are provided in the Supplementary Figure S6.

3.5 Modeling biogenic CO₂ saturation

A second GAM model (Model 2) was fitted to the normalized relative CO₂ saturation (in log) following the approach described above. A deviance of 54.2% ($R^2_{adj} = 0.498$) was explained by selecting lake size (LakeArea; log), NDVI (exp), cultivated area (Cultivated; sqrt) and water temperature (T) as independent variables by stepwise method. LakeArea explained the highest variance (13.2%), followed by NDVI (8.39%), Cultivated (6.88%) and T (3.75%). All the variables were statistically significant ($p < 0.05$) (Table 2) and belonged to the three different clusters obtained in the PCA, in agreement with the VPA results for CO₂ saturation. In all the factors, edf were not close to one and therefore, smoothers were applied in the model with the default number of knots ($k = 10$) (Figure 5). Using COMSAT dataset to test the model gave a coefficient of determination (R^2_{adj}) of only 0.0075 ($p = 0.59$). Diagnostics for the Model 2 are shown in Supplementary Figure S7.

TABLE 1 Results of the generalized additive models (GAMs) calculated for biogenic saturation of CH₄, CO₂, and N₂O using both linear terms and smoothers (indicated with *s* before the variable). Edf: effective degrees of freedom. R_{adj}^2 : R-squared adjusted for the number of predictors in the model. Dev: deviance explained by each model in %. AIC: Akaike information criterion.

| Model | Response | Formula | edf | R_{adj}^2 | Dev (%) | AIC | n |
|-------|------------------------|--|------|-------------|---------|---------|----|
| 1 | log (CH ₄) | <i>s</i> (log (LakeArea)) + <i>s</i> (exp (NDVI)) + <i>s</i> (sqrt (SummerP)) | 5.67 | 0.691 | 71.1 | 73.660 | 71 |
| 2 | log (CO ₂) | <i>s</i> (log (LakeArea)) + <i>s</i> (T) + <i>s</i> (exp (NDVI)) + <i>s</i> (sqrt (Cultivated)) | 6.89 | 0.498 | 54.2 | 7.058 | 68 |
| 3 | log (N ₂ O) | sqrt (Cultivated) + <i>s</i> (sqrt (SummerP)) + <i>s</i> (Forest) + <i>s</i> (sqrt (Artificial)) | 7.17 | 0.437 | 48.9 | -199.88 | 68 |

TABLE 2 Output of each generalized additive models (GAM). Edf: effective degrees of freedom. Dev: deviance explained by each factor of the model in %.

| Model | Variable | Estimate | Standard Error | <i>t</i> -Statistic | <i>p</i> -Value | Dev (%) |
|----------------------|------------------------------|------------|---------------------|---------------------|-----------------|----------------|
| 1 (CH ₄) | Intercept | 3.419 | 0.045 | 75.95 | <2e-16 | - |
| | Variable | Edf | df residuals | F-Statistic | p-Value | Dev (%) |
| | <i>s</i> (log (LakeArea)) | 2.336 | 9 | 7.476 | <2e-16 | 26.9 |
| | <i>s</i> (exp (NDVI)) | 0.879 | 9 | 0.809 | 0.00479 | 4.10 |
| | <i>s</i> (sqrt (SummerP)) | 1.459 | 9 | 0.938 | 0.00463 | 4.12 |
| 2 (CO ₂) | Intercept | 2.432 | 0.028 | 85.81 | <2e-16 | - |
| | Variable | edf | df residuals | F-Statistic | p-Value | Dev (%) |
| | <i>s</i> (log (LakeArea)) | 1.948 | 9 | 2.205 | 0.00003 | 13.2 |
| | <i>s</i> (exp (NDVI)) | 1.838 | 9 | 1.157 | 0.00282 | 8.39 |
| | <i>s</i> (sqrt (Cultivated)) | 0.883 | 9 | 0.837 | 0.00402 | 6.88 |
| | <i>s</i> (T) | 1.221 | 9 | 0.407 | 0.04804 | 3.75 |
| 3 (N ₂ O) | Intercept | 2.044 | 0.011 | 192.69 | <2e-16 | - |
| | sqrt (Cultivated) | 0.020 | 0.006 | 3.489 | 0.00091 | 28.8 |
| | Variable | edf | df residuals | F-Statistic | p-Value | Dev (%) |
| | <i>s</i> (sqrt (SummerP)) | 2.363 | 9 | 1.976 | 0.00027 | 15.6 |
| | <i>s</i> (Forest) | 2.084 | 9 | 1.287 | 0.00177 | 12.0 |
| | <i>s</i> (sqrt (Artificial)) | 0.723 | 9 | 0.290 | 0.05034 | 12.0 |

3.6 Modeling biogenic N₂O saturation

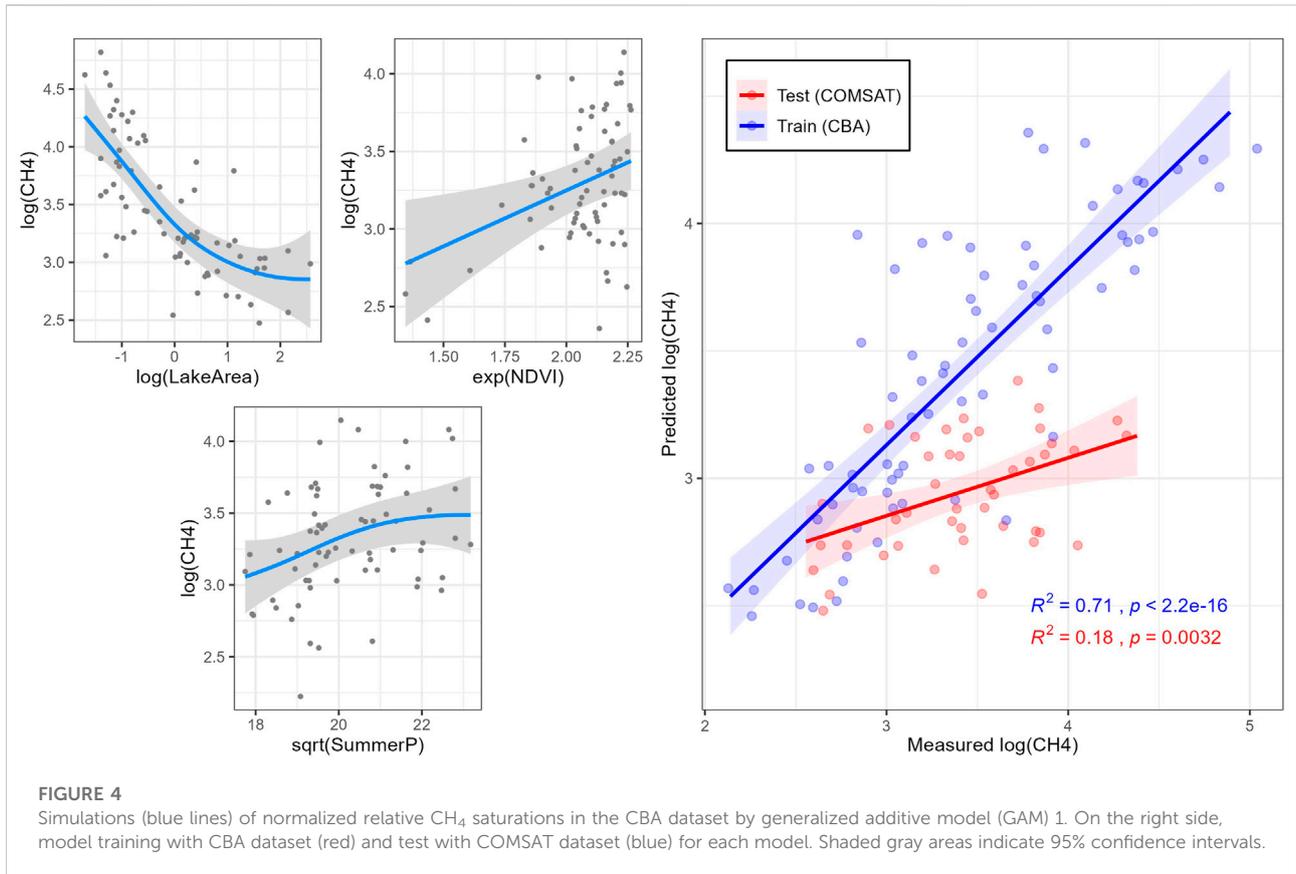
Differences in normalized N₂O saturation (48.9%) were predicted by fitting a GAM model (Model 3) including as independent variables agricultural area (Cultivated; sqrt), average summer Precipitation (SummerP; sqrt), Forest area (Forest) and impervious land (Artificial; sqrt) ($R_{adj}^2 = 0.437$). Variable selection was performed by stepwise method following the results from explorative analyses (i.e. PCA, correlation and VPA). In line with those, cultivated (Cluster C) was the factor explaining by itself the highest variance (28.8%), followed by SummerP (15.6%), Forest (12.0%) and Artificial (12.0%) (Table 2). Cultivated land showed an edf value close to 1, so

smoother was removed from this variable (Figure 6). COMSAT dataset was used to validate the model and provided a R_{adj}^2 of 0.49 ($p < 0.001$), which is similar to that obtained with the training dataset. Diagnostics for the Model 3 are provided in Supplementary Figure S8.

4 Discussion

4.1 Greenhouse gas saturation

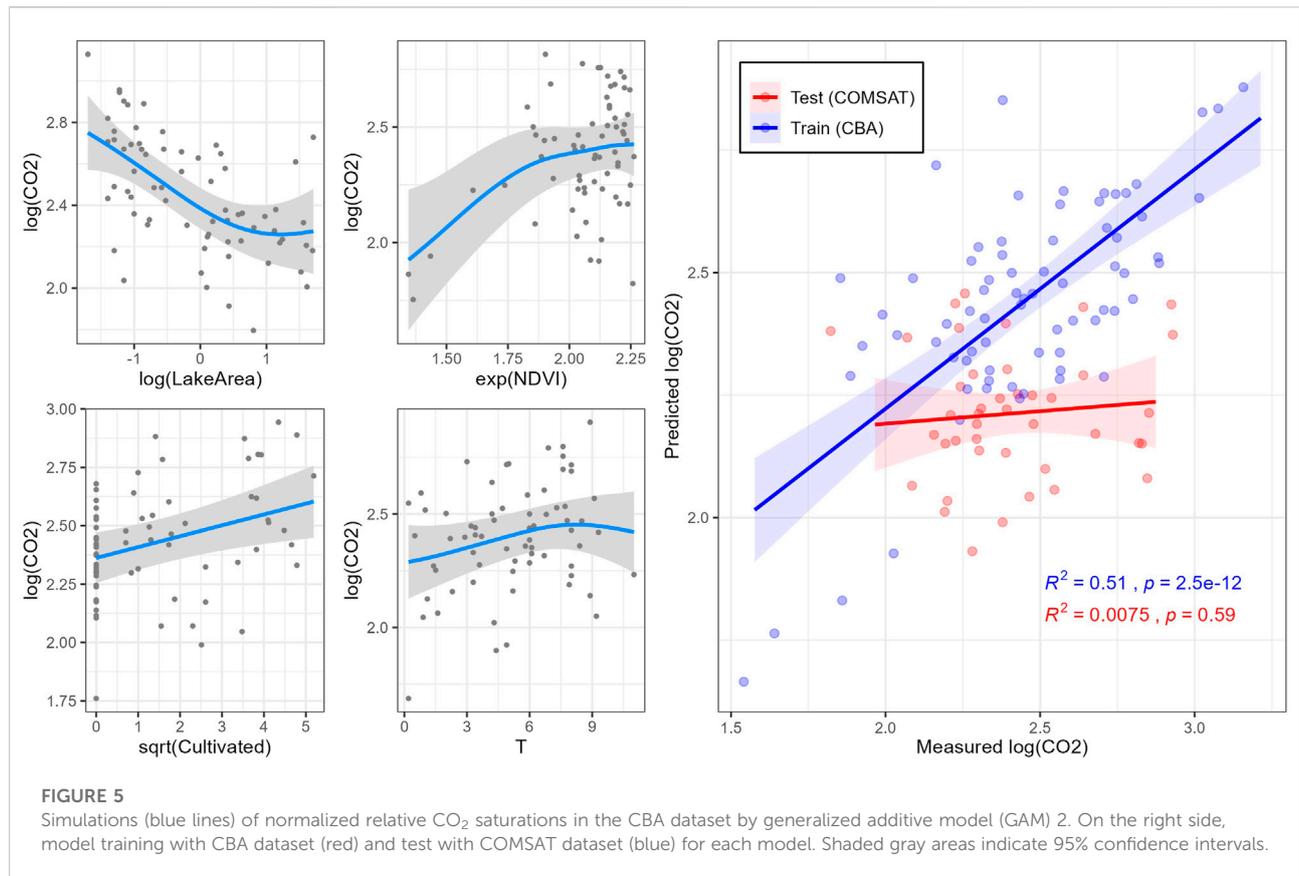
The biogenic GHG saturation levels were in close accordance to those reported for boreal lakes in COMSAT dataset (Yang



et al., 2015). The biogenic oversaturation of CH₄, CO₂, and N₂O demonstrated a substantial impact of biogenic processes on GHG concentrations (Figure 2B). Methanogenesis and denitrification are microbially-mediated processes, which are affected by physical (e.g., temperature, rainfall) and chemical (e.g., nutrients) levels in the lake (Roland et al., 2017; Peña Sanchez et al., 2022). The well oxygenated waters of the study lakes (median = 96.7%, Figure 2B) are likely to constrain methanogenesis and promote methanotrophy in most of the water column. Still a pronounced CH₄ oversaturation (median = 2248%) was found, reflecting a pronounced methanogenesis in deep waters and sediments. In small lakes (<1 km²), convective mixing in the epilimnion and deepening of the mixing layer may be the main mechanism transporting CH₄ to lake surface layers (Kankaala et al., 2013). Prior studies in small boreal lakes have noted the importance of convective cooling of water masses in autumn, where CH₄ effluxes exceed CH₄ oxidation in the water column (Kankaala et al., 2006). This is a possible explanation for the observed results, as our survey was conducted in autumn and 37 of the 73 lakes (51%) are classified as small. In large lakes (>1 km²), CH₄ can be originated from shallow epilimnetic sediments (Bastviken et al., 2008) and be transported laterally from the littoral/riparian zone and the catchment area (López Bellido et al., 2013). It is important to bear in mind the possible

bias in large lakes, as the sampling was performed from the littoral zone (Section 2.1). Nevertheless, the consistency between these datasets are noteworthy, even if the COMSAT lakes were sampled in the middle of the lakes in contrast to the littoral sampling of the current (CBA) dataset.

CO₂ exchange is mainly governed by biotic processes, specifically by the balance between photosynthesis and respiration. In boreal lakes, CO₂ oversaturation is to a large degree an effect of microbial respiration boosted by allochthonous C (Hessen et al., 1990), regulated by factors like catchment area or residence time (Larsen et al., 2011a). Our results showed CO₂ oversaturation in the CBA survey (median = 263.1%, Figure 2B) similar to those observed in the COMSAT survey (median = 231.5%). The lakes from the CBA survey were located in catchments dominated by coniferous forests, primarily spruce and pine. The positive correlation between CO₂ saturation and TOC concentrations ($R = 0.3$; $p < 0.001$) (Supplementary Figure S4) suggests in-lake CO₂ production by TOC mineralization. These results are in line with previous studies (Kortelainen et al., 2006; Whitfield et al., 2011). In addition, a positive correlation was found between CO₂ saturation and TN concentrations ($R = 0.36$; $p < 0.001$) (Supplementary Figure S4). Primary production in boreal lakes can be promoted by N availability (Elser et al., 2009). Nutrients also boost microbial activity, and our data demonstrate that boreal lakes



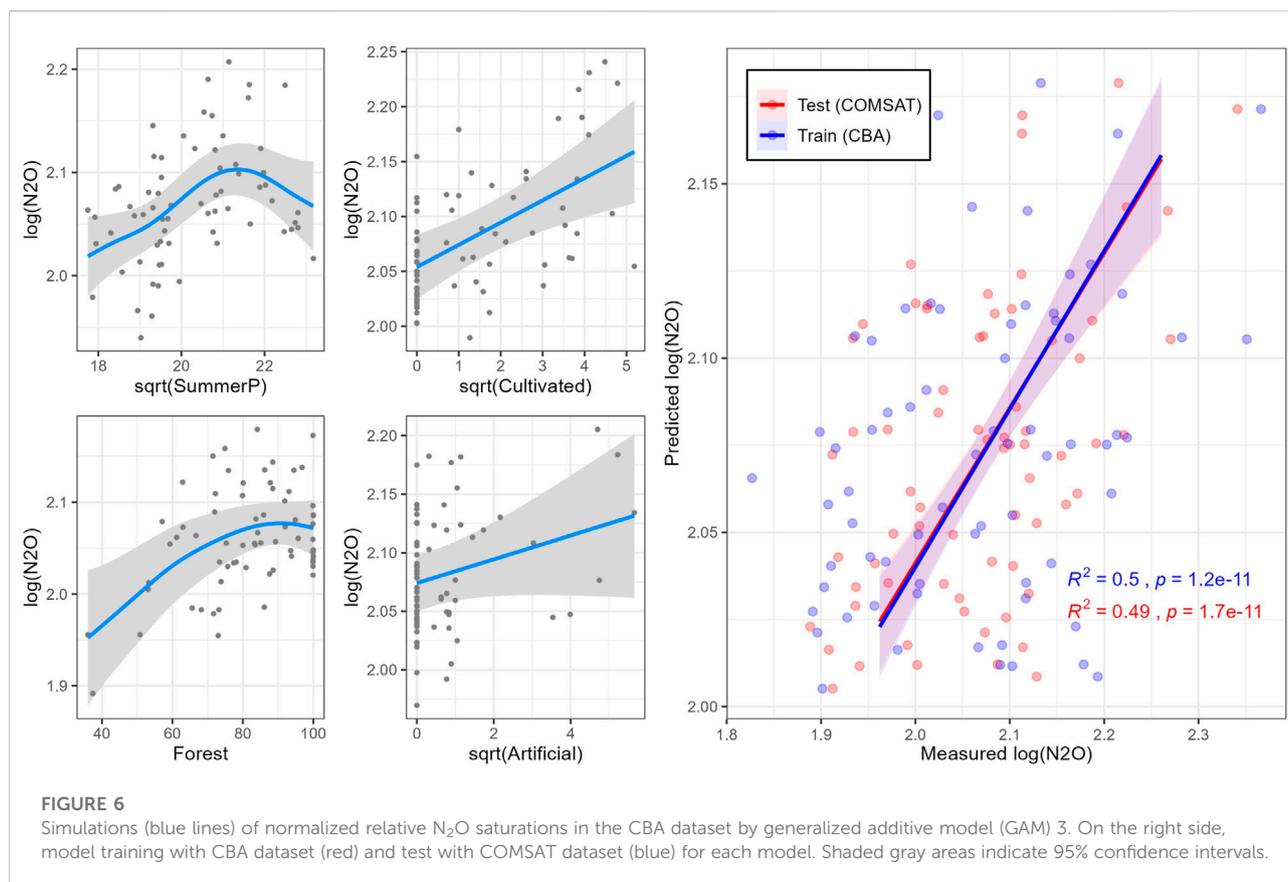
generally retain net heterotrophy in spite of nutrient loading and potentially high primary production, in support of Huttunen et al. (2003a). These positive correlations with TOC and TN suggest the transport of bioavailable OM from catchment soils to lakes and in-lake breakdown of DOM is a main driver for dissolved CO₂ (Sobek et al., 2003; Whitfield et al., 2011). Furthermore, lateral export of DIC from soils to aquatic environments may contribute to lake CO₂ (Öquist et al., 2009; Vachon et al., 2017). In boreal catchments, DIC may derive from the dissolution of soil CO₂ as well as from mineral weathering (Nydahl et al., 2020). Water pH may regulate the CO₂ concentration by keeping a large proportion of the DIC as free CO₂ at low pH values, which in turn depends on the input of humic acids (Nydahl et al., 2019). Our results followed this trend, with a weak negative correlation between CO₂ saturation and pH ($R = -0.21$; $p > 0.001$) (Supplementary Figure S4).

Consistent with the literature (Huttunen et al., 2003a; Whitfield et al., 2011; Yang et al., 2015; Kortelainen et al., 2020), our research found N₂O oversaturation, but lower than CH₄ and CO₂. While being relatively low, the oversaturation of N₂O (median = 115.3%, Figure 2B) reflects the potential of boreal lakes as to act a net source for N₂O due to nitrification and denitrification processes. A positive correlation was found between N₂O saturation and TOC concentrations ($R = 0.35$; $p < 0.001$) (Supplementary Figure S4). Since DOM (or TOC)

mainly derives from allochthonous sources, it suggests that N₂O concentration also depends on the transport of bioavailable OM from catchment soils. N₂O saturation in boreal lakes has been generally associated with agricultural land in the watershed (Kortelainen et al., 2020). Our results support this by a strong correlation between N₂O saturation and agricultural land cover (Cultivated) ($R = 0.32$; $p < 0.001$), but also with artificial impervious areas (Artificial) ($R = 0.3$; $p < 0.001$) (Supplementary Figure S4). In lakes, N₂O production and emissions is boosted by atmospheric N-deposition (Yang et al., 2015), as well as nitrate load from land to lakes (Huttunen et al., 2003b). This latter flux may be enhanced in the boreal landscape by rising temperatures leading to earlier floods due to snow melt (Blöschl et al., 2017) and consequent transport of nitrates to surface waters (Kortelainen et al., 2020).

4.2 Hydrological and climate determinants of GHG saturation

The generalized additive models (GAMs) calculated for biogenic saturation of GHG included either or both hydrological (LakeArea) and climate (SummerP, T) parameters as explanatory variables (Table 1). Variation



partitioning analyses indicated that hydrological parameters had the greatest contribution to biogenic CH₄ saturation (22.5%, [Supplementary Figure S5A](#)), supporting earlier studies ([Kankaala et al., 2013](#); [Li et al., 2020](#)). In that sense, lake size (LakeArea) was the main explanatory factor for the spatial differences in biogenic CH₄ saturation (26.9% of the total deviance; [Table 2](#)), further supporting evidence from previous studies ([Bastviken et al., 2004](#); [Juutinen et al., 2009](#); [Kankaala et al., 2013](#); [Holgerson, 2015](#); [Denfeld et al., 2020](#); [Jahr, 2021](#)). Biogenic CH₄ saturation level was negatively correlated with lake surface (LakeArea; [Figure 4](#)). This result may be explained by the fact that small lakes have a high perimeter/surface area ratio, which means that they receive relatively higher loads of allochthonous DOM relative to water volume ([Holgerson and Raymond, 2016](#)). The increased contribution of allochthonous DOM promotes microbial metabolism ([Tranvik, 1998](#); [Forsström et al., 2013](#)). Therefore, littoral sediments can play a major role in increasing pelagic lacustrine CH₄ due to horizontal mixing in the surface layer ([Juutinen et al., 2003](#); [Rasilo et al., 2015](#); [Bartosiewicz et al., 2016](#)). This is also supported by the negative correlation found between log(CH₄) and log(LakeDepth) in ($R = -0.50$; [Supplementary Figure S4](#)). As stated, our sampling strategy may involve some bias in the results for large lakes. Lake size was also included as variable in the CO₂ saturation model (GAM model 2), showing a

negative correlation between both ([Figure 5](#)). LakeArea explained 13.2% of the total deviance ([Table 2](#)) and was the sole catchment parameter included in the model ([Figure 5](#)). Compared to the CH₄ model, this predictor explained less deviance (22.5 vs. 13.2%) but it was still the strongest predictor in the model ([Table 2](#)). Our CO₂ model 2 predicted higher CO₂ saturations in lakes with smaller surface areas (LakeArea), in line with previous studies showing significantly higher CO₂ emissions in small lakes ([Kortelainen et al., 2006](#)), most likely as a result of increasing lateral OM fluxes ([de Wit et al., 2018](#)).

As climate parameters, the average precipitation during the summer period (SummerP) was selected as independent variable in the CH₄ model (Model 1; [Table 1](#)). It explained 4.12% of the total deviance ([Table 2](#)), showing a positive correlation to the variation in biogenic CH₄ saturation ([Figure 4](#)). Summer precipitation was orthogonal to the loading vectors, reflecting lake size within Cluster A (i.e., LakeArea, CatchmentArea, and LakeDepth; [Figure 3](#)).

Precipitation boosted GHG-production (cf. Model 3; [Table 1](#)), and more so for CH₄ than for N₂O saturation ([Table 2](#) and [Figure 6](#)). These results are in line with previous studies showing that increased precipitation is associated with larger allochthonous DOM inputs to lakes ([Rantakari and Kortelainen, 2005](#); [Natchimuthu et al., 2014](#)).

For the CO₂ saturation model (Model 2; Table 1), water temperature (T) was included as explanatory variable showing a positive correlation (Figure 5). By itself, it explained 3.75% of the total deviance (Table 2), being the only model where T was selected. Water temperature reflects on the seasonal changes in air temperature. It also strongly affects microbial metabolism and promotes mineralization in lake sediments (Gudasz et al., 2010). However, this trend may be the opposite depending on the season of the year in which sampling is performed because of phytoplankton activity (Tadonléké et al., 2012). Since the CBA sampling was conducted in late fall, close to lake overturn, we can rule out CO₂ uptake by phytoplankton as a main driver.

The stepwise selection of variables for the GAM models highlighted the importance of catchment characteristics (hydrological and climate parameters) in predicting biogenic GHG saturation from boreal lakes. In fact, both LakeArea and SummerP may be understood as proxies for water residence time in the watershed. Previous studies have noted that mineralization capacity of lakes at the catchment scale was closely correlated to the mean residence time of surface water in the watershed (Algesten et al., 2004). An inverse relationship between the rate of OM decay (i.e., biogenic CH₄ production) and residence time have been reported (Catalán et al., 2016). In that sense, higher decomposition rates are found in systems with short residence times, as a result of the constant renewal of the labile organic pool (Jones et al., 2018). Headwaters can be considered as hotspots for OM processing as they receive fresh allochthonous OM. Thus, the lability of the allochthonous OM decreases along the aquatic continuum (Clayer et al., 2021). Our results are consistent with those previous studies. Although our estimated water residence times (Residence) were not included as explanatory variables in the GAM models, significant negative correlations were found between Residence and normalized CH₄ saturation ($R = -0.34$; $p < 0.001$), and normalized N₂O saturation ($R = -0.27$; $p < 0.001$) (Supplementary Figure S4). Using TOC as proxy for OM inputs, the negative correlation observed between TOC and Residence ($R = -0.25$; $p < 0.001$) (Supplementary Figure S4) points to residence time as a key driver. This also implies that changes in precipitation and runoff also will affect GHG-metabolism in lakes via this mechanism.

4.3 The influence of terrestrial primary production on GHG saturations

Variables related to primary production in the catchment were also included in the generalized additive models (GAMs) calculated for biogenic saturation of GHG. In general, higher GHG saturations were found coupled to higher values of the primary production proxies in the catchment (i.e., NDVI, Cultivation and Forest cover). NDVI was selected as

explanatory variable for CH₄ and CO₂ saturation models (Table 1). CH₄ model (Model 1) ranked NDVI as the third most explanatory variable (4.10% of the total deviance; Table 2), while it accounted for a higher deviance in the CO₂ model (8.39% of the total deviance; Table 2). NDVI is a proxy for terrestrial vegetation and primary production. NDVI was strongly correlated with TOC, AnnualT, and Forest cover (R of 0.43, 0.69, and 0.53, respectively; $p < 0.001$) (Supplementary Figure S4). Previous studies have shown a close link between NDVI in the watershed and levels of DOM/TOC in the runoff, suggesting that leachates from terrestrial primary production of litterfall are a major source of OM in boreal lakes (Larsen et al., 2011a; Finstad et al., 2016; Škerlep et al., 2020). The wide gradient of catchment NDVI and lake DOM is our studied lakes clearly reveals that the supply of allochthonous OM stimulates heterotrophic metabolism resulting in higher CH₄ and CO₂ saturations, which is observed in both models (Figures 4, 5).

Cultivation coverage was included in both CO₂ and N₂O saturation models (Table 1). It explained 6.88 and 28.8% of the total deviance explained in CO₂ and N₂O models, respectively (Table 2). The positive correlation between the agricultural land cover (Cultivated) and CO₂ concentrations in boreal lakes has been pointed out previously (Kortelainen et al., 2006), likely reflecting inputs of easily degradable DOM (Crapart et al., 2021) as well as nutrient loads from croplands (Rantakari and Kortelainen, 2005). For N₂O saturation, a positive correlation between N₂O saturation and agricultural land cover has previously been reported for boreal lakes (Kortelainen et al., 2020). Our research shows that agricultural land cover and TN are positively correlated ($R = 0.54$; $p < 0.001$) (Supplementary Figure S4). N from fertilizers applied in the watershed are microbially processed in lakes, producing N₂O as by-product of both nitrification and denitrification. N₂O saturation model (Model 3; Table 1) also included Forest and Artificial, explaining respectively 12.0 and 12.0% of the total deviance (Table 2). Higher proportions of these uses were associated with higher N₂O saturations in lake waters (Figure 6). Likewise, Forest and/or Artificial land cover seems to be associated with DOM inputs into lakes (Mattsson et al., 2005; Pellerin et al., 2006; Finstad et al., 2016), promoting processes such as denitrification (Liu et al., 2015). In forest dominated areas from the boreal landscape, soil N seems to leach from catchment soils directly into the lakes in form of nitrate (Khalili et al., 2010), which can be denitrified giving N₂O as a by-product.

4.4 Evaluation of biogenic GHG saturation models

Given the major role of boreal lakes for GHG-emissions (Tranvik et al., 2009), accurate models are needed to better predict biogenic GHG saturations in lakes across the boreal

landscape. In that sense, our GAM models were tested on the independent COMSAT dataset. Despite COMSAT lakes were all sampled in the middle of the lakes, our results showed that the datasets are basically consistent. In fact, the COMSAT lakes tend to be a bit lower in GHGs, which can be attributed to a somewhat larger area. The major levels and patterns in saturation of the three GHGs was basically consistent between the two datasets. In decreasing order, the trained models had the highest testing success for $\text{N}_2\text{O} > \text{CH}_4 > \text{CO}_2$. The N_2O model (Model 3; Table 1) was validated using COMSAT dataset, obtaining similar results as those obtained when using the training dataset (CBA) (Figure 6). The relatively low deviance explained by Model 3 (48.9%) probably reflects the low variability in biogenic N_2O concentrations, as well as the lack of likely key governing factors for the in lake biogeochemical N_2O production, such as N-deposition (Pregitzer et al., 2008; McCrackin and Elser, 2010; Kortelainen et al., 2013).

Despite the consistency in patterns and levels of saturation between the two independent datasets, the CH_4 saturation model (Model 1; Table 1) showed a poor correlation between predicted and measured biogenic CH_4 saturations with the COMSAT dataset ($R_{\text{adj}}^2 = 0.18$) (Figure 4). This discrepancy could be attributed to the seasonality, the location, and the size of lakes sampled. The CBA lakes were sampled during fall turnover, while the COMSAT lakes were collected from the epilimnion during the summer stratification. The COMSAT survey covered a geographical gradient from western Norway (5.4°E) to the Norwegian-Swedish border (12.3°E). LakeArea distribution in the COMSAT dataset (median = 3.53 km²) was also significantly larger ($p < 0.001$) than the LakeArea from CBA dataset (median = 0.93 km²). Nevertheless, gas concentrations and especially biogenic saturations of GHGs were similar for both datasets (Figure 2). When the COMSAT dataset was restricted to lakes in central-eastern Norway ($n = 33$, longitude $> 7.7^\circ\text{E}$), the correlation between predicted and measured CH_4 saturations increased up to ($R_{\text{adj}}^2 = 0.39$). It should also be noted that the western part of Norway is characterized as one of the rainiest parts of Europe. The original COMSAT dataset has values from SummerP to 919 mm, which may definitely affect the adjustment of CH_4 saturation model.

Also the CO_2 saturation model (Model 2; Table 1) showed only weak correlation between predicted and measured biogenic CO_2 saturation in the COMSAT dataset ($R_{\text{adj}}^2 = 0.008$) (Figure 5). A feasible explanation for this discrepancy may be that samples for COMSAT were collected during the summer, when assimilation of CO_2 in the epilimnion is likely an important governing factor. The season of the year when sampling occurred may thus be a reason for the poor simulation of biogenic CO_2 saturation obtained when applying the model to the COMSAT dataset. In addition, LakeArea was the strongest predictor in the

model and, as stated above, lake size distribution differed significantly between the CBA and COMSAT surveys. When COMSAT dataset was downscaled to lakes with a longitude above 7.7°E ($n = 33$), the adjustment was slightly better ($R_{\text{adj}}^2 = 0.10$) but still far from that obtained with the training dataset ($R_{\text{adj}}^2 = 0.51$).

These findings show the difficulties in achieving good models for predicting GHG saturations in boreal lakes. When modeling, increasing the number of variables used in the models may increase the variance explained, but complicate the interpretation and application of the models. Within the CBA dataset, we achieved robust predictions. Despite the patterns and levels of GHGs saturations are consistent between the two datasets, the generation of robust models predicting GHGs demands multiple explanatory parameters, including size and seasonality.

5 Conclusion

The purpose of the current study was to model biotic saturation of GHGs in boreal lakes by using a set of chemical, hydrological, climate, and land use parameters. Our models were trained on a dataset of boreal lakes ($n = 73$) sampled in 2019 and then validated with an additional dataset of boreal lakes ($n = 46$) sampled in 2011. Both these datasets encompass an extraordinary gradient in altitude, precipitation, temperature and not the least their content of organic matter, and allowed us to tease apart various drivers. We find that: i) hydrological and climate parameters are key drivers for predicting GHG saturation; ii) the influence of external OM inputs (i.e., terrestrial) plays a key role in biogeochemical cycling. Most of the variation in biogenic CH_4 saturation was explained by lake size: the larger the lake, the lower the CH_4 saturation. On the contrary, summer precipitation and NDVI exhibited a positive correlation with CH_4 saturation. Biogenic CO_2 saturation was mainly explained by lake size (negative correlation) followed by NDVI, proportion of cultivated area, and water temperature (positive correlation in all of them). For biogenic N_2O saturation, the differences in land use (cultivated, forest, and artificial area) were the strongest predictors showing a positive correlation, as well as summer precipitation. The insights given by our extraordinary range of catchment and lakes variables provides new insights in drivers of GHG-production in boreal lakes, which again improve predictions of impacts of climate change and human activities. The comparison of two independent datasets, and using one as a training dataset for model prediction, nevertheless demonstrate that predicting GHGs demands multiple explanatory parameters, including seasonality, for generation robust models.

Data availability statement

The datasets presented in this study can be found in online repositories. The names of the repository/repositories and accession number(s) can be found below: https://osf.io/r39ng/?view_only=e9a3b3de84794bfc9883db481cb9a483.

Author contributions

NV, AE, and DH conceived the idea. All authors were involved in either fieldwork and/or sample analysis, in the analysis of data and final writing. All authors contributed to the article and approved the submitted version.

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Conflict of interest

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenvs.2022.880619/full#supplementary-material>

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