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Spatial predictors and temporal forecast of total organic carbon levels in boreal lakes

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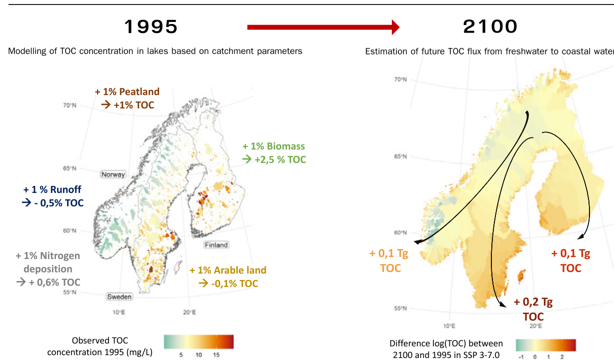
HIGHLIGHTS

- TOC concentration in boreal lakes depends on catchment characteristics, in particular on standing biomass.
- A space-for-time approach proves successful to predict future TOC concentration.
- Climate change > 4.5°C will lead to a strong increase in browning of freshwater and export of TOC to coastal waters.

GRAPHICAL ABSTRACT



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ABSTRACT

Browning of Fennoscandian boreal lakes is raising concerns for negative ecosystem impacts as well as reduced drinking water quality. Declined sulfur deposition and warmer climate, along with afforestation, other climate impacts and less outfield grazing, have resulted in increased fluxes of Total Organic Carbon (TOC) from catchments to freshwater, and subsequently to coastal waters. This study assesses the major governing factors for increased TOC levels among several catchment characteristics in almost 5000 Fennoscandian lakes and catchments.

Normalized Difference Vegetation Index (NDVI), a proxy for plant biomass, and the proportions of peatland in the catchment, along with surface runoff intensity and nitrogen deposition loading, were identified as the main spatial predictors for lake TOC concentrations. A multiple linear model, based on these explanatory variables, was used to simulate future TOC concentration in surface runoff from coastal drainage basins in 2050 and 2100, using the forecasts of climatic variables in two of the Shared Socio-economic Pathways (SSP): 1-2.6 (+2 °C) and 3-7.0 (+4,5 °C). These scenarios yield contrasting effects. SSP 1-2.6 predicts an overall decrease of TOC export to coastal waters, while SSP 3-7.0 in contrast leads to an increase in TOC export.

1. Introduction

In many boreal lakes, including Fennoscandian (Finland, Sweden, and Norway), a widespread browning of freshwater has been observed in the last decades. This phenomenon is caused by the increased flux of colored natural organic matter (NOM) from catchments to rivers and lakes (Monteith et al., 2007), and enhanced by a concurrent increase in iron

Abbreviations: TOC, Total Organic Carbon; NDVI, Normalized DIfference Vegetation Index.

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(Monteith et al., 2007, Xiao and Riise, 2021, Björnerås et al., 2017, Solberg, 2022). This upward trend in total organic matter (TOC) has been recorded since the 1980s when several monitoring programs were implemented to document the effect of acid deposition abatement policies. The changes in water quality in Norway were also assessed by regional synoptic surveys in 1986 (Henriksen et al., 1989), in 1995 (Northern European Lake Survey, (Henriksen et al., 1998), and in 2019 (Hindar et al., 2020, De Wit et al., 2023). The 1986-study covered 1000 lakes in Norway, while the 1995 survey included also almost 4000 lakes in Sweden, Finland, Denmark, as well as Scotland, Wales and Russian Kola and Karelia, while the 2019 survey again covered the 1000 Norwegian lakes. There was a 50 % decrease in sulfur deposition between the period 1976–85 and the period 1995–2001 (Skjelkvåle, 2003; Aas et al., 2002; Fagerli et al., 2022). This caused an increased solubility of NOM, thereby increasing the concentration of TOC in surface waters (De Wit et al., 2007; Monteith et al., 2007). Sulfur deposition is now back to preindustrial levels, meaning that any further reductions are considered to be marginal (Grennfelt et al., 2020). On the contrary, there are larger uncertainties regarding the reactive nitrogen deposition, which has not decreased as much as the sulfur. This nitrogen contributes both to soil and water acidification (Kanakidou et al., 2016), and promotes the net primary production of forests (Schulte-Uebbing and de Vries, 2018). A potential further increase in surface water NOM will likely not be driven by a further decrease in sulfur deposition, but by other factors, such as changes in nitrogen deposition, hydrology, and temperature, as well as re −/afforestation and other land use changes.

Increased surface runoff is expected to lead to increased flux of NOM from the forest floor to surface waters (De Wit et al., 2016). Higher temperatures provide longer growing seasons. This increases the primary production and thereby the amount of biomass in the catchments, as well as enhanced heterotrophic decomposition of the organic matter. Inherently, this leads to an increased flux of NOM to surface waters causing increased browning (Finstad et al., 2016; Kritzberg et al., 2020; Larsen et al., 2011a). Finstad et al. (2016) modelled the increase of TOC concentrations in 70 Norwegian lakes over 30 years. They found that a temporal increase in Normalized Difference Vegetation Index (NDVI), along with increasing temperature, were the main temporal explanatory parameters for the observed increase in freshwater TOC concentration. NDVI is used as a proxy for the density of vegetation (primarily forest) biomass (Beck et al., 2007). Accumulated reactive nitrogen from long-range atmospheric deposition is also expected to contribute to an increase in biomass (Vries and Schulte-Uebbing, 2019). The intensification of forest management in the 1950ies and 60ies increased the forest volume in Nordic countries. Moreover, abandonment of out-field resources as grazing pastures for husbandry, has resulted in increased standing forest biomass during the last centuries (Myrstener et al., 2021). In the coming decades, afforestation is expected to expand further, as the governments of Norway, Sweden, and Finland (collectively referred to here as Fennoscandia) consider forest as a necessary trade-off leverage for offsetting their carbon emissions and reaching their zero-emission goals (Nordic Council of Ministers, 2021; Vogt et al., 2022). Coniferous forests build up a thick layer of organic soil that increases the amount of NOM available for lateral transport (Škerlep et al., 2020). In addition, practices such as clear-cutting and peatland ditching are likely to promote the release of NOM towards the water system (Nieminen, 2004; Asmala et al., 2019; Finér et al., 2021).

The planned intensification of forest management and re-/afforestation has raised concern for possible effects on water quality (Norsk Vann, 2019). Raw water sources used for producing drinking water in Fennoscandia are predominantly surface waters. Browning has thus a potential impact on the water treatment to ensure potability of water (Eikebrokk et al., 2004). In addition, browning has a suite of ecosystem impacts in lakes, e.g., by increasing light absorbance, and thus reducing primary production (Karlsson, 2007; Thrane et al., 2014). Moreover, the increased influx of allochthonous NOM boosts the heterotrophic degradation, causing increased CO_2 and CH_4 emissions, especially in small lakes (Hessen et al., 1990; Tranvik et al., 2009; Wit et al., 2018; Yang et al., 2015). Increased browning also extends the duration of thermal stratification and affects fish, both via the reduced primary production, but also by the darkening affecting predators by hampering visual hunting (Craig et al., 2017; Finstad et al., 2014; Karlsson et al., 2009). Monitoring of the Secchi depth in the Baltic Sea have shown that the coastal water has steadily become less transparent since the beginning of the 20th century (Fleming-Lehtinen and Laamanen, 2012). Browning of freshwater lakes and rivers cascade along the aquatic continuum from rivers to the coast (Opdal et al., 2019). Increased NOM in surface freshwater thus inherently leads to an increased export of NOM to the coastal water, subsequently causing coastal darkening (Aksnes et al., 2009).

Identifying the governing factors for NOM concentration in space and time and understanding their role under changing anthropogenic and climatic pressures is therefore a prerequisite to forecasting NOMs potential detrimental effects to the water quality and ecosystem services.

Larsen et al. (2011a) developed an empirical model predicting the spatial distribution of TOC concentrations in the 1000 Norwegian lakes of the 1995 Northern European Lake Survey (Henriksen et al., 1998). The model was based on lake catchment characteristics, using catchment NDVI, area fractions of peat (Bog) and area specific surface runoff intensity (Runoff) as predictors. This study concluded that NDVI was a key predictor of surface water TOC concentration, as NDVI explained, together with Bog and Runoff, nearly 80 % of observed spatial variation in TOC.

The 1000 lakes used to fit the model by Larsen et al. cover a very heterogeneous landscape, comprising large gradients in NOM, relative forest and bog coverage, and mean surface runoff, as well as length of growing season, with extensive subalpine and alpine areas. For this reason, the model predictions may not be representative of the more homogeneous boreal biome at large. To test the assumption that the key catchment properties NDVI, Bog and Runoff can serve as explanatory predictors for TOC concentrations at wider spatial scale, this study includes an additional 3735 lakes and their catchments properties from Sweden and Finland. These additional catchments represent more uniform topography and runoff, situated in low-altitude land of Sweden and Finland, with more productive forests, and with extensive areas of bogs. Other conceptually relevant predictors at the catchment level were also included in this study: i.e., mean temperature (Temp) and amount of rain (Precip), proportion of forest (Forest) and farmland (Arable), as well as total nitrogen (TNdep) and sulfur deposition (TSdep).

A Spatial Error Linear Model (SELM) is used as a modelling tool to account for the spatial autocorrelation of the response and predictor variables. The fitted TOC model, built on the optimum set of predictors, is validated on data from the recent re-sampling of the 1000 lakes in Norway (Hindar et al., 2020). Based on the model, the effect size (i.e., relative effect on TOC) of a relative increase of each of the predictor variables are estimated. The model is also used to forecast future average TOC concentration in an ensemble of Fenno-Scandinavian watersheds draining into the sea (here referred to as "coastal drainage basins"). Following the IPCC AR6 report (Masson-Delmotte et al., 2021), two global warming scenarios are considered: i.e., the SSP 1.2-6 scenario, with global warming limited to <2 °C, and the SSP 3-7.0 where global warming can reach up to 4 °C. Finally, these scenarios are compiled to estimate the future export of TOC from Fennoscandian peninsula into its coastal waters.

The aim of the study is to test two distinct hypotheses: 1) that a space-for-time approach, using a spatial "snap-shot" to predict temporal changes, can produce realistic forecast results; and 2) that global warming, including increased surface runoff, together with land-use changes, with increased standing biomass, is likely to exacerbate the ongoing browning of freshwaters and, subsequently, also coastal darkening.

2. Methods

2.1. Data preparation

The main water chemistry dataset assessed in this study (training dataset) comprised the data from 4735 lakes sampled during the Northern

European Lake Survey in 1995 (Henriksen et al., 1998). Except for one outlier, the smallest catchments areas started from 3500 m^2, while the largest watershed was over 5106 km^2. The average being 107 km^2, with a standard deviation of 416 km^2. TOC concentration in each lake, as well as catchment polygons defined with a digital elevation model, were downloaded from the NOFA database (Finstad, 2017). Nine explanatory parameters, describing each of the lake catchments, were compiled as predictor variables: i.e., NDVI; Forest, Arable and Bog; Runoff, Precip and Temp; TNdep and TSdep. These are briefly described below. Lake morphology parameters, such as lake depth, were not available for the Northern Lake Survey dataset and thus not included in the analysis. The complete procedure for preparation, extraction, and compilation, as well as the sources of data are described in Supplementary 1.

The test dataset, provided by the Norwegian Institute for Water Research (NIVA), comprised 1001 lakes sampled in 2019 (Hindar et al., 2020). The sampled lakes were essentially the same ones as for the Norwegian lakes in the training data set. In both surveys, the sampling was completed in the autumn, after the seasonal turnover, so that the surface sample represents the entire water column (De Wit et al., 2023). Each lake of the test dataset was matched with the corresponding lake of the 1995 training dataset, in order to use the same catchment polygons, from the NOFA database (see Supplementary 2).

Some of the variables were not strongly asymmetrical and were log-transformed using the natural logarithm, simply denoted “log” hereafter.

- Total organic carbon (TOC) concentration in mg C/L (Fig. 2) were extracted from the Northern European Lake Survey dataset from 1995 (Henriksen et al., 1998; Finstad, 2017) and from the 1000-lakes-survey in 2019 (Hindar et al., 2020). As the TOC concentration values were skewed, the data were log-transformed (logTOC) to normalize the distribution.
- Normalized difference vegetation index (NDVI) (Fig. 1a) for the summer months (June, July, and August) were downloaded from the GIMMS database (The National Center for Atmospheric Research (2018)) and averaged for 1994, i.e., the year preceding sampling of the Northern European Lake Survey. For the 2019 dataset, data from 2015 are used, as it was the latest available year in the GIMMS database.
- Mean area specific surface runoff (Runoff) values in mm/y (Fig. 1b) were derived from the CORDEX historical model. The period 1970–2000 was used for the 1995 data, and the period 1985–2015 was used for the 2019 data (CORDEX, 2021; Kreienkamp et al., 2012). As the surface runoff intensities were skewed the data were log-transformed (logRunoff).
- Mean annual precipitation (Precip) in mm/y (Fig. 1c) and temperature (Temp) in °C (Fig. 1d) for the years 1970 to 2000 were downloaded from the WorldClim database (WorldClim, n.d.) to be used as representative for the 1995 data. As the precipitation distribution was not normally distributed the data were log transformed (logPrecip). As these climate data were not retained in the final model, they were not downloaded for the 2019 data.
- Proportions of peatland (Bog), arable land (Arable) and forested area (Forest) (Fig. 1e, f, and g, respectively) within each catchment are computed from the Corine Land Cover (CLC) database (Copernicus Land Monitoring Service, n.d.), and correspond to the data of 2000, as land use data prior to this were not available. Several CLC categories were merged to create the three categories used in this study (see Supplementary 1). For the 2019 dataset, Corine Land Cover data for 2018 were used. Although distributions of the different land use are not normally distributed, they were not log-transformed due to many zeros in the data. Other transformations (square, square roots) did not contribute to normalizing the distribution.
- Total reactive nitrogen deposition (TNdep) in $mg.N/m^2$ and total sulfur deposition (TSdep) data in $mg.\ S/m^2$ (Fig. 1h and i, respectively) were extracted from EMEP models as the sum of dry and wet deposition (EMEP, 2022; “The Unified EMEP Model-User Guide”, 2012). Data from 2000 were used as these were the earliest available data, resulting in a slight underestimation of TNdep and TSdep since deposition has decreased since the 90s. For the 2019 dataset, EMEP data from 2019 were used.

2.2. Spatial TOC models

Larsen et al. (2011b) used a Multiple Linear regression model (LM) to predict the effect of climate change on the spatial distribution of TOC concentration in 1000 Norwegian lakes. In the present study, including in addition almost 4000 lakes in Sweden and Finland, a similar linear model is reproduced to investigate the explanatory value of the three predictors selected by Larsen et al. (i.e., NDVI, Bog and Runoff), along with other conceptually relevant predictors, for explaining the spatial differences in TOC levels in Fennoscandian lakes. All the statistical analysis were performed in R, version 4.1.2. (R Core Team, 2021).

The spatial autocorrelation of all predictors were determined by computing Moran's I (see details in Supplementary 4). All variables were found to be spatially autocorrelated. To take this into account, a Spatial Error Linear Model (SELM) was fitted with the same predictors as for the LM and the results compared. SELM was fitted using the “errorsarlm” function from the “spatialreg” package, version 1.1-8 (Bivand, 2019). Akaike Information Criterion (AIC) was used to compare the performances of the models.

The SELM and LM models are fitted on the 1995 training dataset and their slope estimates (i.e., slope β as in the simple model $y = \beta x + c$) were evaluated both on scaled and unscaled variables. Scaled estimates β (i.e., computed with centered and standardized variables) expresses the response variable change in standard deviation units per unit standard deviation change in the predictor variable, while unscaled estimates were used to compute effect sizes (expressing the relative change in the response variable for a given change in the predictor). Equations used to calculate effect size for each pair of response/predictor variables are explained in Supplementary 4.

The 2019 dataset of the 1000 Norwegian lakes survey (Hindar et al., 2020) was used to perform a test of the space-for-time model. The model fitted on the Fennoscandic 1995 data was used to predict TOC concentration values in the 1001 lakes in 2019, and the results were compared with actual observations.

2.3. Forecast of TOC prediction in coastal drainage basins

Changes in TOC concentrations in costal drainage basins by year 2100 are predicted using the validated SELM. Polygons representing the coastal drainage basins were used to cover the whole territory of Fennoscandia, not only the studied lakes catchments. These basins are the ensemble of the watercourses that drains into the sea within a coastal section (NVE, n. d.). The coastal drainage basins were defined using an elevation model delineated from a 10 m digital terrain model obtained from The Norwegian, Swedish and Finnish Mapping Authorities (Finstad, 2017). After cleaning, the dataset is composed of 1392 watershed polygons (see Supplementary 3). The smallest coastal drainage basin has an area of 27 km^2, the largest has an area close to 49,000 km^2, while the mean area is 751 km^2 with a standard deviation of 3723 km^2.

Future changes in climate parameters are based on predicted changes in climate drivers derived from the Coupled Model Intercomparison Project (Phase 6) (CMIP6), run by the World Climate Research Program (World Climate Research Program, n.d.) using two different SSPs (Riahi et al., 2017): SSP1-2.6 as the best scenario, assuming the average global warming will be limited to <2 °C, and SSP 3-7.0 as our worst-case scenario (CORDEX, 2021; Hausfather, 2019). Several institutions run the CMIP6 models to forecast future climate. Based on Raju and Kumar (2020), results from the model developed by the Centre National de Recherches Météorologiques (CRNM) were selected to extract future surface runoff intensity.

There is no forecast for NDVI in CMIP6. Summer NDVI values for 2050 and 2100 were instead predicted using a polynomial model based on forecast for temperature and precipitations in the period 2041–2060 and for the period 2081–2100. This model was fitted using summer months

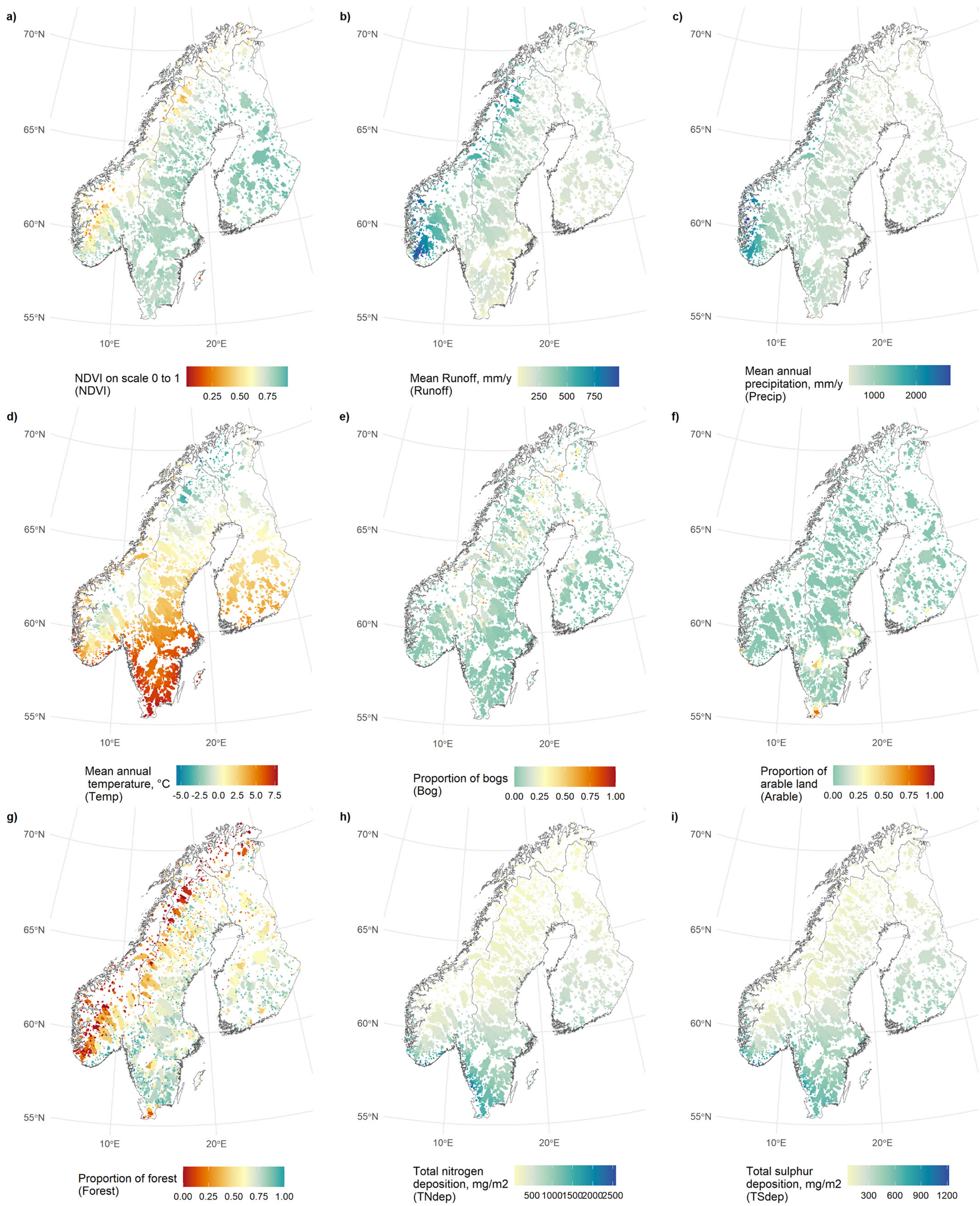


Fig. 1. Maps of the predictor variables: summer NDVI in 1994 (a); mean surface runoff intensity (Runoff, b), temperature (Temp, c), mean annual precipitation (Precip, d) from 1970 to 2000; proportion of peat (Bog, e), proportion of forest (Forest, f), proportion of arable land (Arable, g) for the year 2000; total nitrogen deposition (TNdep, h) and total sulfur deposition (TSdep, i) in the 4735 lake studied catchments in 2000.

NDVI, temperature and precipitations in 1995. The predicted future temperature and precipitation values were extracted from the CRNM model. Modelled summer NDVI and changes in this NDVI in the future are shown in Supplementary 3.

TNdep is also not modelled for the future and is estimated depending on the SSP. Between 2000 and 2020 the TNdep decreased due to reduced emission of both reduced (NH_3) and oxidized nitrogen (NOx), by respectively 8 and 42 % (European Environment Agency, 2021). Future trends in TNdep for the forecasts SSP 1-2.6 and SSP 3-7.0 were assessed based on the estimation for future N emissions described above. According to the Sixth Assessment of the IPCC report (Masson-Delmotte et al., 2021), the NOx emissions will decrease slightly for the SSP 1-2.6 (from 11 to 9 Mt/y globally) and increase for SSP 3-7.0 (from 11 to 21 Mt/y globally), between 1995 and 2100. There are no projections for reduced nitrogen, instead NH_3 is assumed to decrease by around 20 % between 2020 and 2100 in case of the SSP 1-2.6, and to remain constant for SSP 3-7.0.

2.4. Selection of predictor variables

2.4.1. Spatial distribution of predictors

Spatially distributed data of the mean values of summer NDVI, Runoff, Precip, Temp, Bog, Arable, Forest, TNdep and TSdep were assessed as possible predictors to be used in the SELM. Catchment means of these variables, describing each of the 4735 lake catchments in Fennoscandia, are shown in Fig. 1.

2.5. Pearson correlation coefficient, autocorrelation, and variance inflation factor

Predictor variables that are strongly correlated (multi-collinear) reduce the robustness of multiple linear models and the precision of their estimates (Vittinghoff et al., 2012). This intercorrelation is disclosed by investigating the Pearson correlation coefficients (r) for each pair of variables, shown in Table 1. In addition, the Moran's I of each variable was calculated (Fig. 2), estimating the spatial autocorrelation for each variable. Considering the substantial number of observations, the maximum Moran's I value for a non-autocorrelated variable would be $2.1 \cdot 10^{-4}$. All variables in this study had a Moran's I above this limit, indicative of a high degree of autocorrelation. Finally, the Variance Inflation Factor (VIF) (James et al., 2013), indicating the severity of multi-collinearity due to a single predictor, was computed, and presented in Fig. 2.

The Pearson correlation coefficient matrix in Table 1 highlights several groups of predictors that are strongly correlated. TNdep, TSdep and Temp are closely correlated, especially TNdep and TSdep with r = 0.97. The correlation coefficient between TNdep and Temp is 0.85, and the correlation coefficient between TSdep and Temp is 0.88. This is because both N and S deposition are mainly from long-range transported atmospheric pollution from sources in Europe located to the south of Fennoscandia. The deposition of nitrate and sulfate is thus higher in the southern regions of Fennoscandia, where also the temperature is higher. Additionally, the high correlation is partly due to that both S and N deposition are based on emission inventories that are calculated using the same meteorological model. TSdep also has the highest VIF (21.15), followed by TNdep (16.97). Likewise, Temp, TNdep and TSdep have a remarkably high Moran's I (all around 0.9), reflecting their similar spatial pattern. Because of the strong interaction between these three predictors, only one can be used in the model. Although the decrease in TSdep has been documented by several studies as the main driver for the temporal increase in TOC concentration (Monteith et al., 2007), we chose instead to keep TNdep as a predictor for atmospheric deposition in this study. TSdep has decreased substantially since the 80s, though soil acidification is still driven by the deposition of reduced and oxidized nitrogen (Lepori and Keck, 2012). In addition, reactive N is biologically relevant, being generally the limiting factor for terrestrial primary production, and has been shown to enhance C storage in biomass (Schulte-Uebbing and de Vries, 2018) and in soils (Janssens et al., 2010).

Table 1
Correlation plot for predictor variables.

	Arable	Temp	TNdep	TSdep	NDVI	Forest	Bog	logRunoff	logPrecip
Arable	1.00								
Temp	0.27	1.00							
TNdep	0.26	0.85	1.00						
TSdep	0.24	0.88	0.97	1.00					
NDVI	0.05	0.41	0.26	0.32	1.00				
Forest	-0.05	0.50	0.41	0.43	0.62	1.00			
Bog	-0.09	-0.24	-0.23	-0.25	[illegible]	-0.26	1.00		
logRunoff	-0.23	-0.45	-0.39	-0.41	-0.53	-0.58	0.06	1.00	
logPrecip	-0.04	0.19	0.24	0.24	-0.38	-0.36	-0.12	0.66	1.00

Forest and summer NDVI were also closely correlated (r = 0.58) although they do not match as closely as could perhaps be expected. Despite the finer resolution of the Corine Land Cover dataset for forest land cover, logTOC is slightly stronger correlated to summer NDVI (r = 0.68) than to Forest (r = 0.62). Moreover, NDVI reflects the density of the vegetation, and thus likely represents a better indicator for total standing biomass. Likewise, summer NDVI better reflects the photosynthetic activity of plants by representing greenness. Finally, the VIF of summer NDVI was lower than that of Forest (1.83 compared to 2.59), showing that it is less correlated to the other predictor variables. Therefore, only NDVI was kept as the vegetation biomass proxy in this study.

LogRunoff and logPrecip were also strongly positively correlated (r = 0.58). LogPrecip had a higher VIF than logRunoff (5.17 compared to 5.38), showing more general correlation to the other predictors, and a higher Moran's I (0.89 for logPrecip, compared to 0.79 for logRunoff). Conceptually, surface runoff is a more relevant predictor for TOC export from catchment to surface waters than precipitation, as the part of the precipitation that evapotranspires or infiltrates into ground reservoirs do not reach the surface water bodies. Moreover, logRunoff is stronger correlated to logTOC (r = −0.67) than logPrecip (r = −0.51). LogRunoff was therefore selected as the preferred NOM transport related predictor in the model.

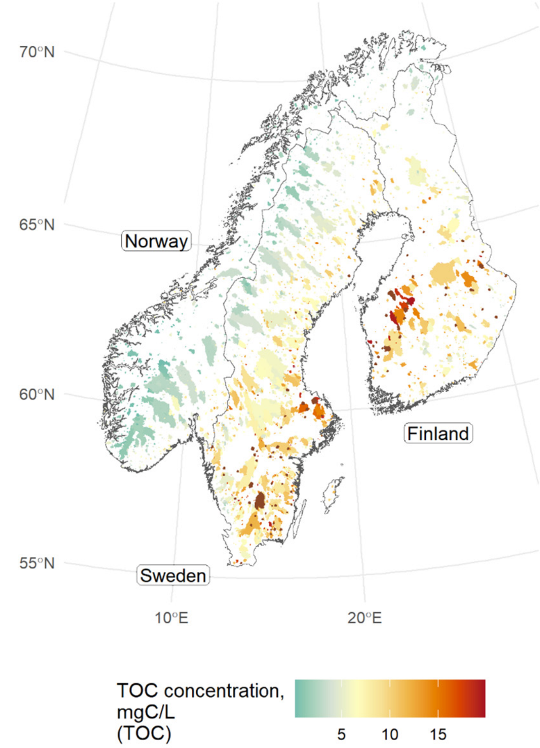
The proportions of Bog and Arable land were not strongly correlated to any other variable (Table 1), and both had low VIF and Moran's I (Fig. 2). Therefore, both were kept in the model as explanatory variables for TOC.

3. Results and discussion

3.1. Modelling of TOC concentration on the Northern Lake Survey dataset

A multiple Linear Model (LM) and a multiple Spatial Error Linear Model (SELM) were fitted for the log of TOC using the 5 predictors selected in the previous section. SELM takes into account the spatial autocorrelation of predictor variables in the error term of the regression. To compare the two models, the Akaike Information Criterion (AIC) was calculated. AIC for each model is presented in Supplementary 1.

Both models were fitted with and without scaled variables. This allows a comparison of scaled estimates with the actual effect size for each predictor a)



b)

Predictor	Moran's I	VIF	Pearson's r
Temp	**0.9**	**5.56**	0.47
logPrecip	**0.89**	**5.17**	**-0.51**
TNdep	**0.88**	**16.97**	0.32
TSdep	**0.86**	**21.15**	0.36
logRunoff	0.79	**5.38**	**-0.67**
Forest	0.48	2.59	**0.62**
NDVI	0.46	1.83	**0.68**
Bog	0.17	1.23	0.08
Arable	0.12	1.27	0.13

Fig. 2. a) Map of TOC concentration (mg C/L) in Fennoscandian catchments in 1995. b) Moran's I (i.e., spatial autocorrelation), Variance Inflation Factor (VIF) (i.e., multicollinearity) between predictor variables, and Pearson correlation coefficient with logTOC. Moran's I higher than 0.8, VIF higher than 5 and r higher than 0.5 are highlighted in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

variable (Fig. 3). Centered and scaled estimates (hereafter referred to as β), fitted on response and predictors variables, show the degree of contribution of each predictor to the response variable value. But this is in units of the local standard deviations of the variables, which complicates the generalization to other regions. The effect sizes are instead expressed as percent change in TOC concentration caused by a 1 % relative change, as explained in Supplementary 4: 1 % increase in summer NDVI (0.01 on the NDVI index); 1 % increase of surface runoff; 1 % increase in either Forest, Bog or Arable land coverage proportion; and a 25 mg/m^2 increase of nitrogen deposition (i.e., 1 % of the max TNdep). These relative changes of 1 % are arbitrarily chosen to represent the relative impact of each predictor variable on a comparable basis. This means that a 1% change of the predictor x, with an effect size δx in catchment A with initial TOC concentration TOC_A would result in a final TOC concentration of $TOC_A + \delta x \times TOC_A$.

Summer NDVI is the strongest predictor for logTOC in both LM and SELM. LM gives a higher impact to NDVI, with an estimate of 0.5 and an effect size of 4.02 %, while SELM gave a scaled estimate of 0.4 and an effect size of 3.27 % (Fig. 3).

LogRunoff had high scaled estimates, especially for the LM with β= -0.38, while SELM had β= -0.15. The effect size of a 1 % increase in logRunoff has a negative effect on TOC concentration (−0.68 % for LM and − 0.27 % for SELM) (Fig. 3). A plausible conceptual rationale for this negative effect is that the overall dilution effect is on average stronger than the increased episodic flushing of NOM. The spatial distribution of TOC concentration could also impact this result, as the highest surface runoff values are in steep mountain areas in the west coast of Norway, where soils are thin, and TOC concentrations are low.

Bog, despite its low correlation with logTOC (0.08), has relatively high scaled estimates (0.13 for LM and 0.14 for SELM) and effect sizes (0.81 % for LM and 0.9 % for SELM) (Fig. 3). This is bolstering the role of bog coverage as an important spatial predictor for TOC concentration. Still, bogs evolve slowly, and their area-wise proportion is assumed to be constant over timescales relevant for this assessment. They can however experience short-term extreme events, such as droughts that are predicted to occur with increased frequency (Helbig et al., 2020). However, since we base our TOC forecast on 20-year averages of the main predictors (precipitation and temperature to predict NDVI, as well as surface runoff), the impact of extreme events on the TOC average is considered limited.

Arable land has negligible impact on TOC, along with a relatively low negative effect size (0.13 % for LM and 0.1 % for SELM) (Fig. 3). Agricultural soils store indeed less carbon than forest soils and bogs (FAO and ITPS, 2020), though the use of phosphate fertilizers may affect the primary production and thus the levels of autochthonous NOM in lakes through eutrophication. This effect does however not appear to be a preponderant factor governing NOM levels in the studied region.

TNdep has positive impact on TOC concentration (r = 0.32), with both relatively high positive scaled estimates (0.08 for LM and 0.1 for SELM) and effect sizes (0.42 % for LM and 0.52 % for SELM) (Fig. 3). The positive effect of TNdep on TOC may on the one hand be surprising as the deposition of acid rain, including the strongly correlated TSdep, is known to have a negative impact on TOC concentration in several studies (Monteith et al., 2007). The positive effect of TNdep may instead be due to a spatial covariation, as the TNdep is higher in the southern and warmer part of Fennoscandia, where high summer NDVI drives the high TOC concentrations. It can also be driven by the fertilizing effect of accumulating TNdep in the biomass. Future impact of reactive nitrogen deposition depends on public policies, as it is linked to combustion of fossil fuel and agricultural practices in central Europe. Here, the effect size is calculated for an increase of 25 mg/m^2 of TNdep, though TNdep might either decrease in the coming decades (SSP 1-2.6) or remain stable (SSP 3-7.0) compared to 1995.

The SELM model had a lower AIC than the LM model (see Supplementary 1). It also resulted in estimates being less extreme than the LM. Especially, summer NDVI and Runoff have lower estimates and effect sizes with SELM than with LM. On the other hand, TNdep had higher estimate and effect size with SELM compared to LM. This shows that the SELM model is more balanced and less likely to over- or underestimate the impact

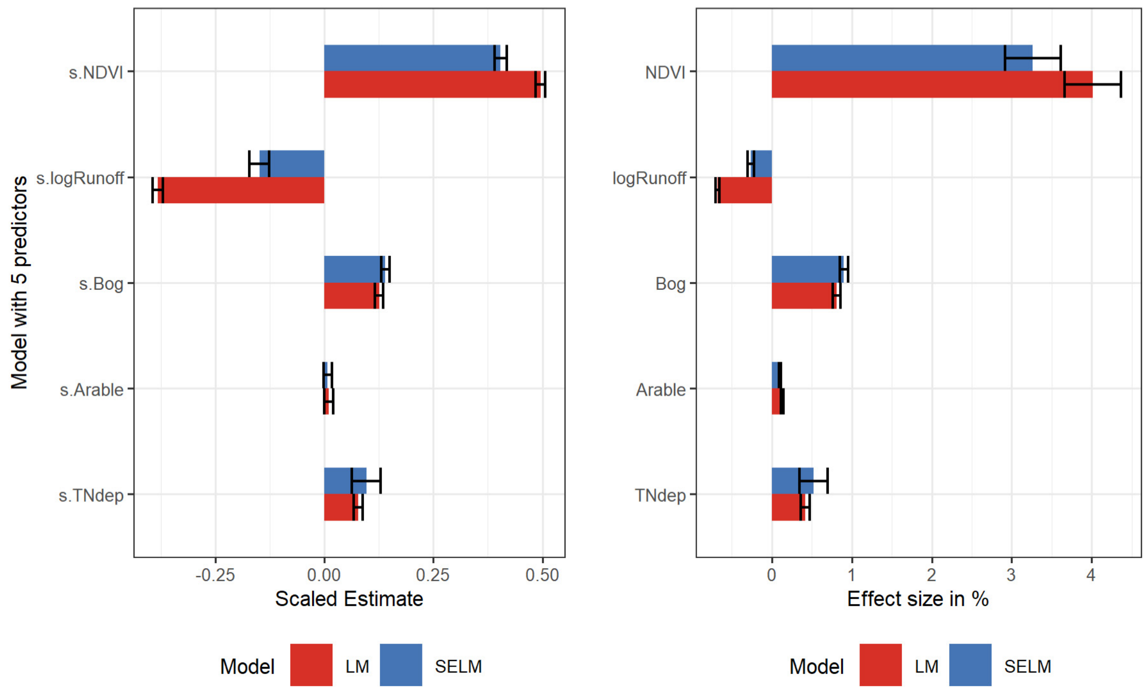


Fig. 3. Comparison of scaled estimates and effect sizes for a 1 % increase in the predictor variables using LM and SELM models, based on the Fennoscandian dataset with the 5 selected predictors. The s. prefix indicates that the variable is centered and scaled.

of each predictor. Moran's I of the residuals is higher for the LM than for the SELM, showing a stronger spatial pattern of the errors in LM. The SELM model performs therefore better in predicting TOC concentration in boreal lakes and is assessed further in the next section.

3.2. Validation against a new 1000-lakes-survey

The results of the validation test of the space-for-time model show that the SELM model fitted with the training data from 1995 of the five selected predictors gives satisfactory results. logTOC predictions were made from the 1995 model using input data from 2019. The Pearson correlation coefficient between these predictions and the actual observations from 2019 was 0.84. However, the model tends to over-estimate TOC concentrations in lakes with very low initial TOC concentrations (< 1 mgC/L), typically alpine oligotrophic lakes, and under-estimate the TOC concentrations in dystrophic lakes that had high initial TOC concentrations (>7,5 mgC/L), particularly in the south-eastern part of Norway (Fig. 4b).

This space-for-time approach employs a set of explanatory values taken at a snap-shot moment in time from a large variety of catchment types to model future changes. The relationships between explanatory parameters and the TOC response parameter at any single site is a result of biogeochemical processes that have evolved in the soils since the last glacial epoch (i.e., about 15,000 years), e.g., the long-term processes of generating the pool of soil organic matter. That catchments in the middle of the Norwegian mountains are predicted to have a higher TOC concentration in 2019 compared to the actual observations (Fig. 4b) is hence likely due to a recent increased NDVI in these regions (i.e., due to climate and land use change), not yet followed by the soil formation that a catchment with a similar NDVI value in 1995 would have had. On the contrary, the TOC concentration of lakes in south-east Norway are mostly under-estimated since in 1995 the TOC was still suppressed by S-deposition. High TNdep, being strongly correlated to TSdep, would have meant low TOC concentration, while in 2019 the role of TNdep as a predictor is more related to the positive fertilizing effect of accumulating reactive N in the watersheds than the negative acid rain effect.

Having in mind these potential discrepancies, this space for time model can be used to obtain an indication of future TOC concentration under various climate scenarios, as the prediction range and the trends predicted in this study nevertheless serves as a good indication in most catchments.

3.3. Forecast with SSP1-2.6 and SSP3-7.0

Future average TOC concentrations were modelled for 1392 coastal drainage basins in Norway, Sweden, and Finland.

As each coastal drainage basins comprise several lakes and catchments, there exists no empirical data for their respective average TOC concentration in their various water bodies. Therefore, the first step was to compute the TOC concentration in 1995 using the SELM model that was fitted and verified in the above section. In a second step, forecasts of the predictor variables Temp, Precip and Runoff were extracted from CMIP6 climate models based on two climate scenarios: i.e., SSP1-2.6 (global warming limited to <2 °C) and SSP 3-7.0 (global warming up to 4 °C). Summer NDVI was then modelled based on the Precip and Temp using a polynomial model, as described in Larsen et al. (2011b), though based on a beta distribution to constrain NDVI predictions between 0 and 1. The linear models were fitted with beta distributed response and logit link using the betareg package (Cribari-Neto and Zeileis, 2010), version 3.1-4, for R (see Supplementary 3).

Future trends in TNdep were assessed based on estimations from IPCC as described above. According to their predictions following the SSP1-2.6 there will be a net decrease in TNdep through 2050 to 2100 and a net increase in the SSP 3-7.0 forecast. Finally, the average TOC concentration was modelled for two time periods: 2041–2060 and 2081–2100 (abbreviated as “2050” and “2100”). The details of predictor extraction and model fitting are presented in Supplementary 3.

Fig. 5 shows the difference between logTOC in 2050 and 2100, fitted for the different climate scenarios, relative to logTOC fitted for the coastal drainage basins based on the 1995 data. Coastal drainage basins are used in order to cover all of Norway, Sweden and Finland, and not only the lakes selected in the Northern Lakes Survey. This provides a better overview of future trends and enables a prediction of TOC export to coastal waters. Over- or under-estimation of TOC concentration, identified by the test on 2019-data, are assumed to be partially compensated by this double-fit. Moreover, the TOC is forecasted as an average over a 20-years

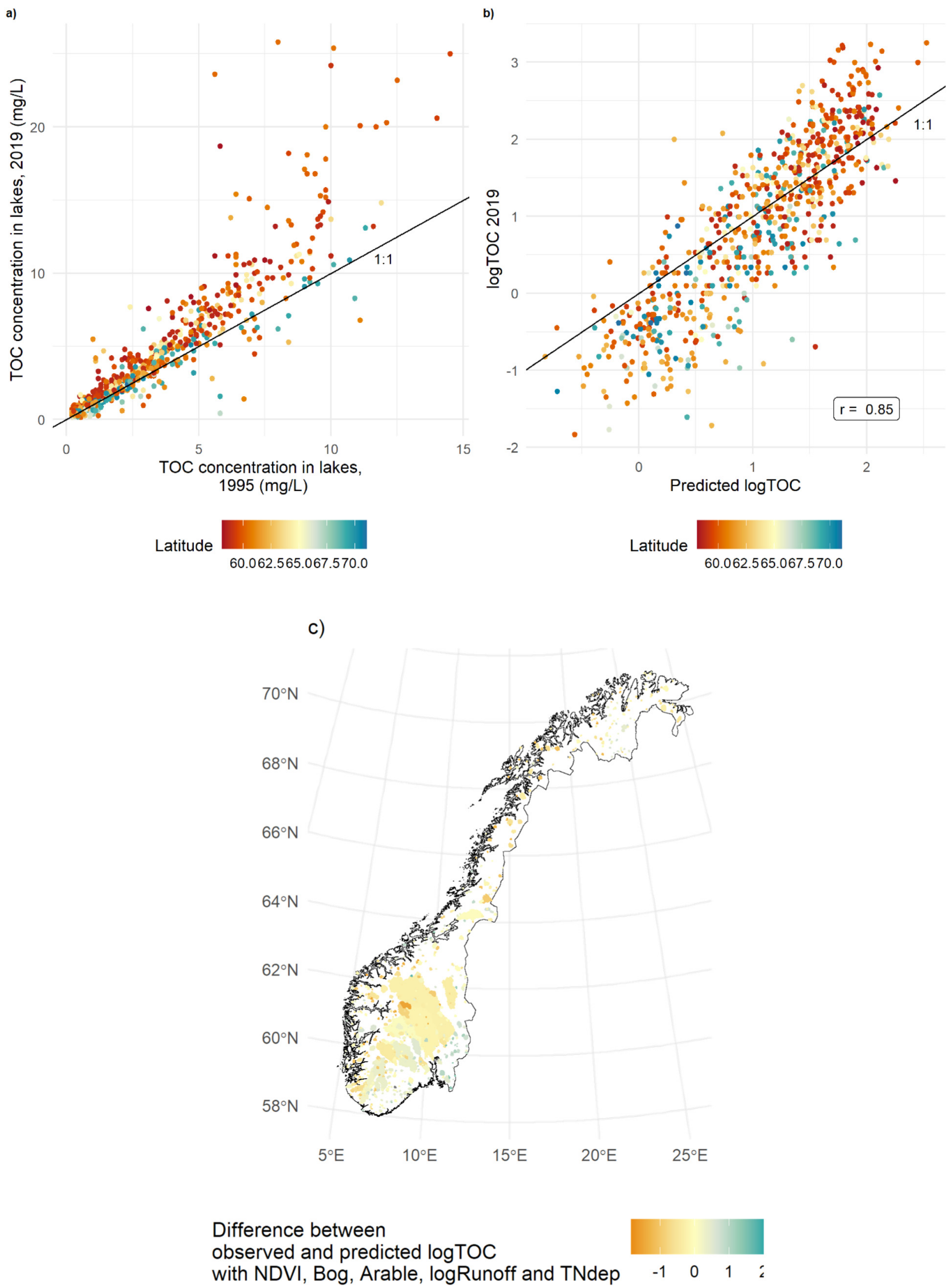


Fig. 4. Test of the SELM model, a) TOC concentration (in mg C/L) in 1995 and in 2019, b) predicted vs. measured logTOC; c) map of the spatial distribution of the difference between measured and fitted logTOC.

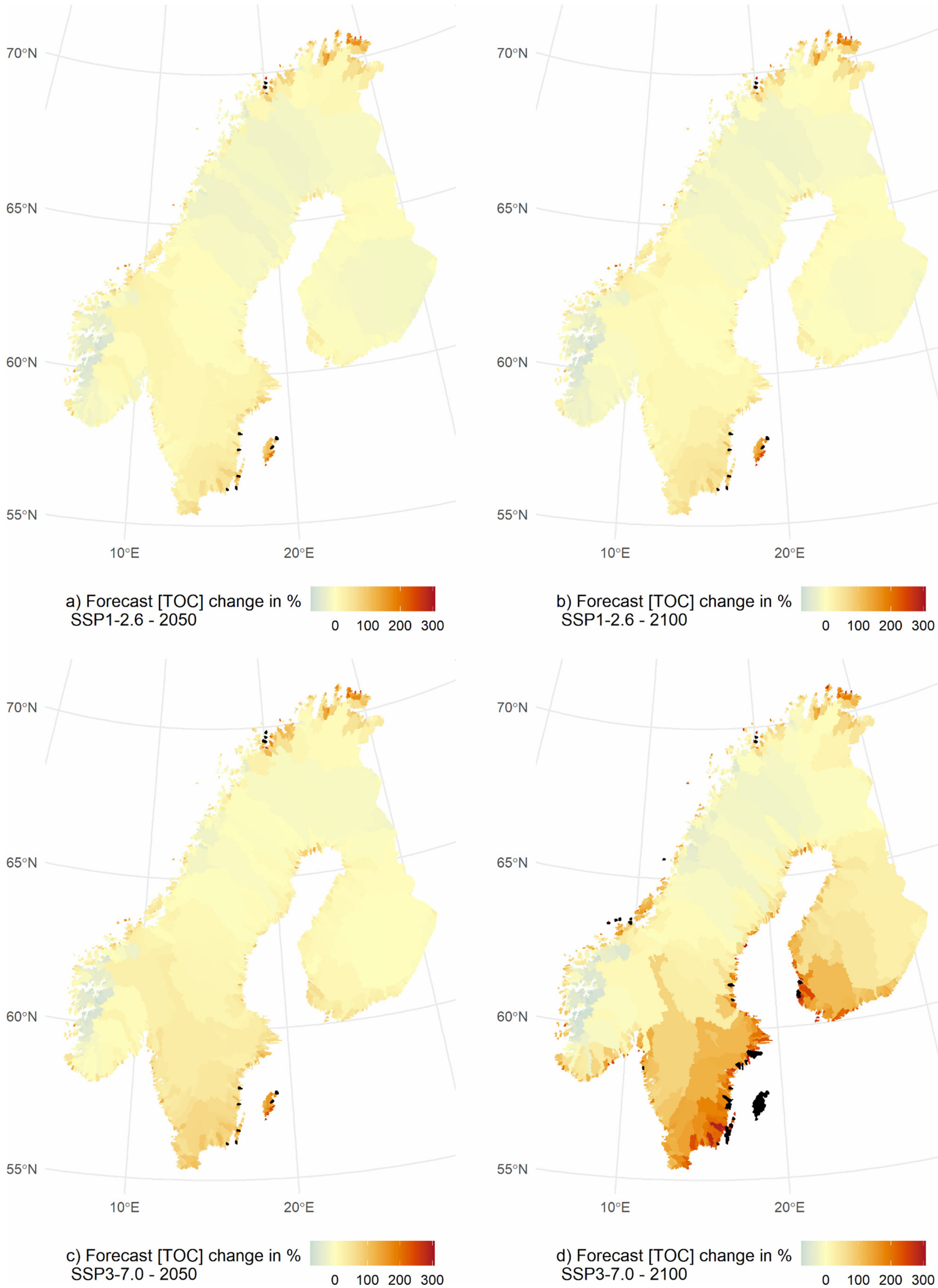


Fig. 5. Forecast of changes in TOC concentration (%) under SSP1-2.6 and SSP3-7.0 climate scenarios. The 5 % basins with the highest increase of TOC (> 307 % increase) are represented in black.

period, which reduce the "snap-shot" effect by considering a certain adaptation time of the system, although it does not account for centennial processes.

The forecasts of average TOC concentration show similar pattern of changes under both the SSP1-2.6 and SSP3-7.0 scenarios, with the latter representing more extreme changes (Fig. 5). The strongest increase in TOC is predicted in the south-east of Sweden and south-west of Finland, while there is predicted a slight decrease in mountain areas in Norway and in some northernmost basins. The modelled changes of TOC are mainly governed by a combination of forecasted changes in NDVI, runoff and TNdep, with different relative effects varying between regions. A loss of TOC from oxidation and sedimentation occurs along the watercourse, inherently dependent on the distance from the headwater origin to the coast (Weyhenmeyer et al., 2012). It should thus be noted that this assessment neglects the potential effect of increased TOC retention due to longer water courses in the coastal drainage basins compared to the catchments used in the training data set.

In the western and northern basins changes in TOC are basically driven by the forecasted changes in NDVI (see Supplementary 3), reflecting the expected changes in biomass, which is mainly governed by the temperature increase. A main effect of this increase is the longer growing seasons and thereby a larger biomass production in forests, along with a faster biodegradation rate of soil organic matter. This increase in growth is supported by sustained amount of precipitation in most of Fennoscandia, if not an increase as for the Norwegian western coast. This limits the potential for soil dryness and water stress, even though water lost by evapotranspiration will increase (D'Orangeville et al., 2018; Gauthier et al., 2015). In mountain regions of the Norwegian west coast, where increase of precipitation is highest, NDVI decreases in both SSP pathways. Indeed, at high altitudes, negative winter temperatures allow for increased snow fall with increased precipitation. This in turn impedes vegetation growth at these high altitudes, which are scarcely vegetated areas that are sensitive to the number of snow cover days (Asam et al., 2018). This can partly explain the predicted decrease in mean TOC concentration in this region.

In this space-for-time approach, the chain of processes between biomass production and carbon storage in forest soils are not integrated. Actually, there will be a delay before the export of TOC from soils to surface starts to increase or decrease in turn. Nevertheless, these results can be used as a good trend indicator. It is worth noting that, in this model, the northernmost, small coastal drainage basins display a significant increase in TOC (up to 300 %), that matches a strong increase in temperature (up to 10 degrees). However, the NDVI model over-estimates the summer NDVI in these regions (see Supplementary 3). On the other hand, the effect of thawing permafrost, as a driver of increased export of TOC (cf. Abbott et al., 2014), is not accounted for in this study since permafrost cover very limited areas in the northernmost areas.

These changes in NDVI do not account for the forest policies implemented by Norway, Sweden, and Finland to capture and sequester carbon dioxide to fulfill their Paris agreement obligations (Vogt et al., 2022). The specific relationship between the proportion of forest in the catchment and TOC concentration are provided in Supplementary 5. Nonetheless, an increase in forest cover would lead to an increase of NDVI, so the results of the model runs may serve as an indication of the impact of this increased planting of climate forest on TOC concentration.

Forecasted trends in surface runoff intensity resemble changes in summer NDVI patterns and often match the TOC concentration evolution in the same manner. A strong decrease of surface runoff was modelled in small coastal drainage basins on the Norwegian western coast. This is likely caused by poor performance of the runoff model in this region due to its steep topography. Still, this has a limited impact on the overall TOC forecast. Besides these small basins, the regions with increased surface runoff (i.e., Norwegian mountains, and some basins in northern Sweden) will, due to stronger dilution, experience less increase or even a decrease in TOC concentration, compared to the regions with decreasing surface runoff, such as southern Sweden and Finland. The largest increases in TOC concentration are predicted to happen in these areas (Fig. 5). In some very small coastal basins, mostly in South-east Sweden, changes are predicted to exceed +400 %. These should be judged with care and are likely caused by the low resolution of the predicting variables in tiny catchments. However, our model also predicts the doubling or tripling of TOC levels in some of the larger basins in south-east Sweden. NDVI is predicted to increase only slightly in these areas, and even to decrease in some coastal basins. The significant decrease in surface runoff could therefore explain this forecast, by reducing the dilution of TOC exported from land to water.

Our model does not consider seasonal patterns of surface runoff, which will be more impacted by climate change than the average yearly surface runoff (Hanssen-Bauer et al., 2017). Summer runoff will decrease in most regions, due to earlier snowmelt and more evapotranspiration, while winter runoff is predicted to increase in most regions. These changes may have antagonistic effects on the yearly average TOC concentration: mobilization of soil organic carbon will increase during the winter and spring (Håland, 2017), while increased residence time of the TOC in lakes will allow for more enhanced photo- and biodegradation of the NOM during the summer. Whether higher fluxes and mineralization rate lead to a net higher or lower yearly average NOM concentration in surface water is subject to discussion (De Wit et al., 2016). The SELM shows that the dilution effect (i.e., more surface runoff means more surface water) prevails over the mobilization effect, though this requires more investigation.

Finally, accumulation of reactive nitrogen also affects regionally the predicted levels of TOC. The TNdep predictions are only based on emissions targets from the AR6 report (see Supplementary 3) and applied evenly on Fennoscandia: the following trend must therefore be interpreted with caution. In the SSP 1-2.6, final levels are reached in 2050. In the SSP 3-7.0, the trend for decrease nitrogen emissions between 1995 and 2015 is reversed during the rest of the century, leading to no overall change. In southern Norway and in the south-western part of Sweden no large changes of TOC levels are predicted, except in SSP 3-7.0 in 2100. A slight increase in NDVI and a decrease in runoff should have led to an increase in TOC concentration, but as TNdep is projected to decrease significantly under SSP1-2.6 scenario, the predicted TOC increase in these regions is limited. On the contrary, under SSP3-7.0, there is no significant change in the rate of TNdep in 2100 compared to 1995. This modelled effect is likely a consequence of the inherent weakness in the space-for-time approach as it likely reflects the spatial location of TOC-rich surface waters in southern catchments with higher mean temperatures, even though there is a potential fertilization effect of TNdep. In addition, chronic TNdep has been reported to have a positive effect on TOC concentration by that TNdep limit microbial respiration and root activities in soils, reducing soil organic matter mineralization rates and thus increasing the pool of TOC readily exportable to surface water (Bowden et al., 2004; Janssens et al., 2010; Ramirez et al., 2012). However, the projected future changes of TNdep were roughly estimated in this study and depend largely on regulations concerning fossil fuels burning and agricultural practices. A finer evaluation of the potential evolution of public policies would be necessary to refine the contribution of future TNdep to TOC concentration in surface waters.

3.4. Coastal darkening

Modelling TOC concentration in coastal drainage basins provides an estimate of future TOC export to the Baltic Sea and the Norwegian coast from Fennoscandia. Most coastal drainage basins in Sweden and Finland drain into to the Baltic Sea (see Supplementary 2), while Norwegian coastal basins drain to Skagerrak, the North Sea, the Norwegian Sea and the Barents Sea (Sætre, 2007). All contribute to the Norwegian coastal current (NCC) freshwater with 50 % coming from the Baltic Sea and 40 % from Norway. The estimated exported TOC from each coastal drainage basin was computed as:

$$TOC_{exp} = [TOC]\left(mg.L^{-1}\right) \times Runoff\left(L.m^2.y^{-1}\right) \times Watershed\ \text{area}\left(m^2\right)$$

Past and future TOC concentrations (*mg C/L*) were obtained by taking the antilog of logTOC. The result of the above equation, giving the amount

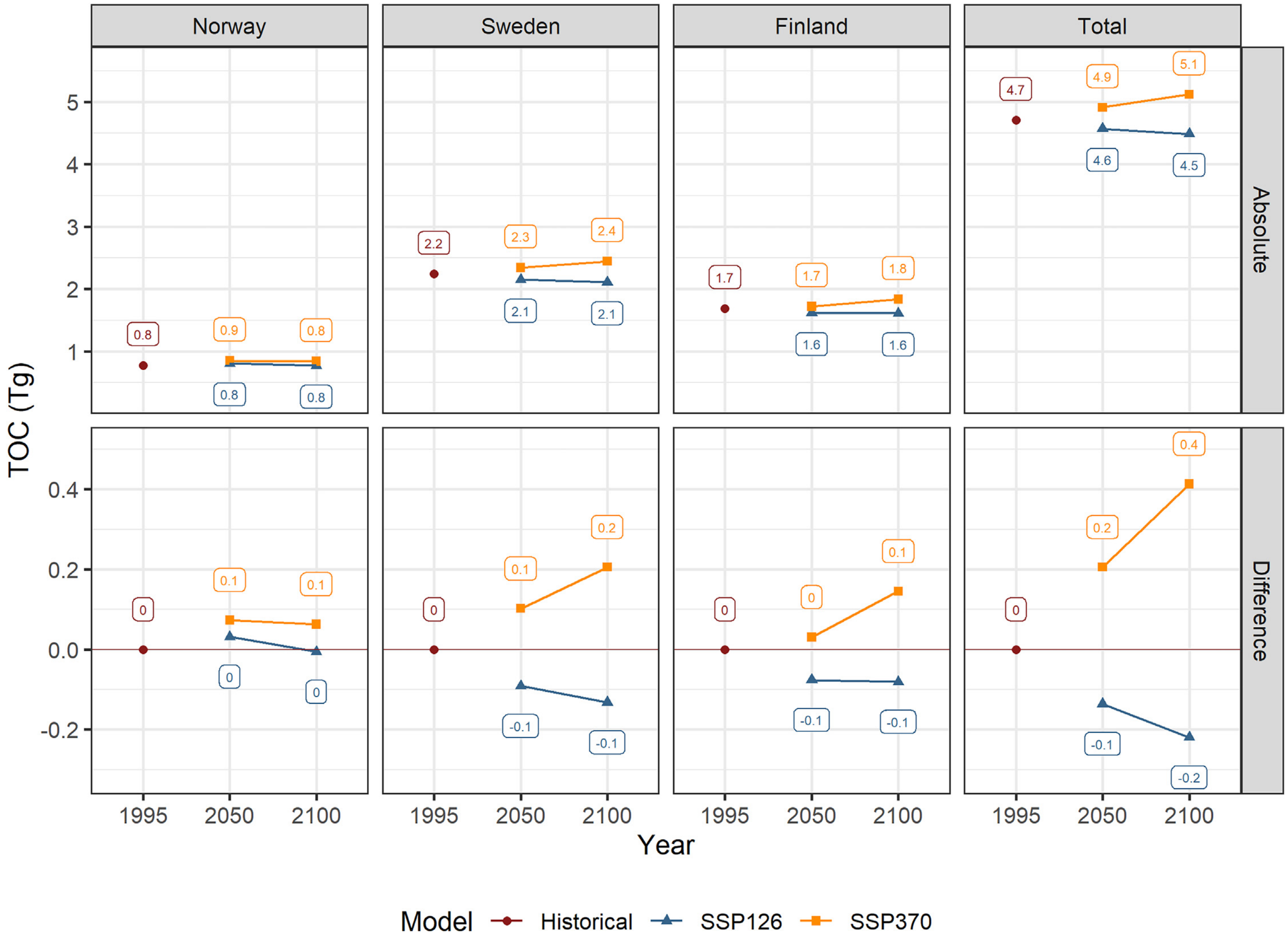


Fig. 6. Forecast of TOC export to coastal waters under SSP 1-2.6 and SSP 3.7-0 (Tg); above: absolute export, below: relative export compared to 1995.

of TOC exported in *mg C/y*, is then converted to *Tg*. Forecasts are presented in Fig. 6.

The amount of TOC exported is in the same order of magnitude as the estimations of De Wit et al. (2015) for Norway, who estimated an export of 0.96 Tg C/y in the period 1990 to 2008, for Norway alone. For Finland, our estimations are higher than those reported in Räike et al. (2016), who reported a yearly export of TOC from Finnish catchments to the Baltic Sea to be 0.92 TgC/y. Under the SSP 1-2.6 scenario, the export of TOC into the NCC will decrease by −0.1 Tg by 2050 compared to 1995, and by −0.2 Tg in 2100. On the contrary, under the SSP 3-7.0 scenario, the total TOC import into the NCC increases by 0.2 Tg in the forecast for 2041–2060, and by 0.4 Tg for the period 2081–2100. Swedish coastal drainage basins would contribute most to these changes. Associated with higher surface temperature, this browning of coastal water could lead to later spring bloom and spawning time.

These estimates do not account for mineralization and sedimentation of TOC along the water continuum, and hence we present a maximum estimate of coastal export where TOC entering the water system is transported all the way to the sea without losses. This "passive pipe" vision has been challenged by Cole et al. (2007), who highlighted the processes that organic matter undergoes along the water system. Photo-mineralization, respiration and sedimentation remove organic carbon from the water (Tranvik et al., 2018). Although the magnitude of this loss remains uncertain, it could be as high as 30 to 70 % (Algesten et al., 2004). In addition, this loss of organic C will, in turn, be impacted by changes in hydrology and temperature. Decreased retention time limits the degree of mineralization. An increase in runoff thus results in the export of less processed, more colored organic carbon to coastal waters (Weyhenmeyer et al., 2012). On the contrary, increased temperature favor higher mineralization rates in soils (Hicks Pries et al., 2017) and in water (Hanson et al., 2011). Moreover, microbial respiration rate also depends on the chemical composition of the freshwater (Crapart et al., 2021), which is impacted by land-use and atmospheric deposition. Sedimentation rates might also increase, without enhancing C-storage because of parallel increased decomposition rates (Velthuis et al., 2018). It should be remarked, however, that since these losses affect both current and future exports, the relative change in export should be less affected by "leaky pipe" processes than the absolute rate estimates.

4. Conclusion

This study demonstrates the relevance of using space-for-time models to forecast future regional environmental changes in TOC concentration in freshwater. The TOC concentration was best predicted by a linear spatial error model, with catchment characteristics as predictors. Long-term processes working on the catchment characteristics are not taken into account in the space-for-time approach. Nevertheless, the simulated changes from 1995 to 2019 were in good agreement with measured values ($r = 0.84$). NDVI, being a good proxy for the amount of biomass in the catchment, is a major predictor for TOC concentration. Surface runoff intensity was also a significant predictor with a negative effect on TOC concentration. The proportion of bogs in the catchment was a good spatial predictor for TOC concentration in lakes, while the proportion of arable land had almost no effect. Finally, a sustained atmospheric deposition of reactive nitrogen will, according to the model, have significant positive effects on the TOC concentration in lakes.

The forecast of TOC changes within this century indicates a slight decrease of total TOC export from Fennoscandia in the SSP1-2.6 scenario (−0.2 Tg in 2100) and an increase in the SSP3-7.0 scenario (+0.4 Tg). Under the SSP1-2.6 scenario, seasonal antagonistic effects of changes in predictors outweigh each other in several of the coastal drainage basins. In particular, the decrease of nitrogen deposition compensates the increase in biomass in southern Norway, limiting the increase of TOC amount in freshwater.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.161676.

CRediT authorship contribution statement

Camille Crapart: Conceptualization; Methodology; Software; Formal Analysis; Investigation; Data Curation; Writing original draft, review and editing; Visualization.
Tom Andersen: Conceptualization; Methodology; Resources; Writing: Review and Editing; Supervision.
Dag O. Hessen: Conceptualization; Resources, Writing: Review and Editing; Project Administration.
Rolf D. Vogt: Writing: Review and Edit.
Anders Finstad: Conceptualization; Resources; Data Curation.

Data availability

The data/code is available on https://camilmc.github.io/TOC_trend_1995/

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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