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Key Points:

- 32% of Fennoscandian lakes showed temporal Fe-TOC decoupling over 30 years: TOC concentrations increased while Fe concentrations declined
- The combined effect of higher temperature and increased precipitation (warmer and wetter conditions) was associated with lower Fe levels
- The Fe-TOC decoupling was linked to short-timescale (2–4 years) increases in precipitation

Supporting Information:

Supporting Information may be found in the online version of this article.

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Temporal Decoupling Between Total Organic Carbon and Iron in Lakes Linked to Interannual Changes in Precipitation

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Abstract Widespread increases in lake browning, which affects primary production, have been observed in northern lakes. While lake browning is attributed to increases in terrestrially derived total organic carbon (TOC) and total iron (Fe), Fe does not consistently correlate with increasing TOC over time. This temporal mismatch between TOC and Fe indicates that we still do not fully understand the causes of lake browning, especially in the context of gradually changing climatic conditions. In this study, we utilized Fennoscandian 30-year (1990–2020) time series data for 102 lakes to describe possible reasons for the temporal decoupling between TOC and Fe. Using Bayesian mixed-effects models and wavelet coherence analysis, we found evidence for differential responses of TOC and Fe concentrations to changes in precipitation, temperature, and sulfur deposition. While TOC appeared more sensitive to the effects of precipitation, temperature and sulfur deposition in individual lakes, Fe concentrations were impacted by complex interactions among these environmental variables. Although TOC and Fe increased in most lakes in response to increased temperature and precipitation, 41% of the lakes—typically with larger catchment-to-lake area ratios and shorter water residence times—exhibited a declining trend in Fe. This analysis encompasses lakes of both significant and non-significant changes over time. This decline in Fe was associated with short-timescale (2–4 years) increases in precipitation, leading to a temporal decoupling between Fe and TOC. Our findings suggest that Fe concentrations do not increase uniformly with rising temperatures and increased precipitation, especially in regions where sulfur deposition has declined due to atmospheric recovery policies.

1. Introduction

Iron (Fe) is an essential micronutrient that supports life in the biosphere (Street & Paytan, 2005; Stumm & Morgan, 1996). Specifically, Fe, along with phosphorus (P) and nitrogen (N), supports primary production in aquatic ecosystems (Heikkinen et al., 2022; Martin, 1992; Moore et al., 2013; Paltsev et al., 2024; Sorichetti et al., 2016; Vrede & Tranvik, 2006). The concentration and speciation of Fe vary greatly among lakes and are linked to the biogeochemical cycling of other elements such as carbon (C), sulfur (S), N and P (Heikkinen et al., 2022; Riise et al., 2023; Stumm & Morgan, 1996; Temnerud et al., 2013; Y. Zhao et al., 2021). Ferric iron [Fe(III)] can form Fe hydroxides in oxic waters, binding P and reducing its bioavailability, while in anoxic waters, ferrous iron [Fe(II)] binds S, forming insoluble minerals (Jones et al., 1988; J. Karlsson et al., 2001; Xiao et al., 2016). It can also form stable complexes with organic carbon (OC), both dissolved organic C [DOC-Fe] or particulate C–Fe complexes—that affect the bioavailability of Fe and other elements (T. Karlsson & Persson, 2012; Knorr, 2013; Xiao & Riise, 2021). In addition, Fe(III) can strongly enhance the attenuation of short-wave radiation in water (Poulin et al., 2014; Xiao et al., 2015). Hence, lake browning, which is widely ascribed to increased catchment inputs of colored DOC to lakes (Creed et al., 2018; Williamson et al., 2015), has also been linked to increased concentration of Fe in lakes, in particular in Fennoscandia (Björnerås et al., 2017; Kritzberg & Ekström, 2012; Weyhenmeyer et al., 2014; Xiao et al., 2015).

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Increased catchment inputs of DOC to lakes are related to climatic parameters such as changes in precipitation and rising temperatures (Arvola et al., 2025; Catalán et al., 2016; de Wit et al., 2016; Hongve et al., 2004; Imtiazy et al., 2020; Räike et al., 2024). Also, climate induced permafrost thaw in subarctic and Arctic regions has been linked to increased DOC release from thick organic soils (Dabrowski et al., 2020; Liebmann et al., 2024). The input of DOC is also influenced by chemical drivers, for example, decreased deposition of acid rain, especially S (de Wit et al., 2021, 2023; Meyer-Jacob et al., 2019; Monteith et al., 2007). However, while S deposition has declined to near pre-industrial levels (Aas et al., 2019; Eklöf et al., 2021; Laudon et al., 2021; Shao et al., 2020), DOC (and total organic carbon: TOC) concentrations continue to rise in many areas (Finstad et al., 2016; Lawrence & Roy, 2021; Meyer-Jacob et al., 2019; Paltsev et al., 2024; Räike et al., 2024), though not in all (cf. Eklöf et al., 2021). Consequently, there is growing evidence that climatic factors and catchment properties (e.g., hydrology and vegetation) play an increasing role for catchment export of DOC and Fe (Björnerås et al., 2017; Finstad et al., 2016; Imtiazy et al., 2025; Jansson et al., 2008; Laudon et al., 2011; Räike et al., 2024).

Iron enters lakes in different forms—for example, as OC-Fe complexes, Fe-oxyhydroxides (FeOOH), in suspended silicate mineral particles (e.g., clay) or through atmospheric deposition (e.g., with dust) (Björkvald et al., 2008; Björnerås et al., 2021). In addition, in-lake processes such as changes in redox potential and oxygen concentrations, as well as sediment resuspension, can lead to either an increase or a decline in Fe concentrations in the water column (Kritzberg & Ekström, 2012; Peter & Sobek, 2018). In-lake processes are often linked to water residence time, with Fe (and DOC) concentrations being higher in lakes with shorter residence times and rapidly replaced lake water, allowing less time for DOC to oxidize and for Fe to settle to lake bottom (Jones et al., 1988; Riise et al., 2023; Weyhenmeyer et al., 2014). In turn, water residence time depends on lake and catchment size and precipitation amounts (Håkanson, 2004), meaning that any changes in precipitation are likely to impact not only Fe (and DOC) export from catchments (Björnerås et al., 2017; Forsberg, 1992; Heikkinen et al., 2022; Hongve et al., 2004; Sarkkola et al., 2013) but also Fe settling rates in lakes (Björnerås et al., 2021, 2022). Fe export can also be affected by air temperature (Ekström et al., 2016), which influences the potential for accumulation of organic matter in boreal soils (Kirschbaum, 1995; Thurman, 1985), the duration of the ice cover period (Kortelainen et al., 2013), and lake stratification patterns (Butcher et al., 2015; Kraemer et al., 2015). Furthermore, extreme weather events—such as prolonged droughts, floods, and intense storms—can significantly affect Fe cycling by altering in-lake processing and sharply increasing or decreasing the flux of Fe from catchments (Jeppesen et al., 2021; Ryder et al., 2014). For instance, droughts may concentrate Fe in sediments and reduce outflow, while subsequent floods can mobilize stored Fe, leading to sudden pulses of Fe into lakes (Škerlep et al., 2023). On a regional scale, especially in Fennoscandia, climate and weather are strongly influenced by climate oscillations (teleconnections) such as the North Atlantic Oscillation (NAO) and more local patterns such as the East Atlantic–West Russia pattern (EAWR) and the Scandinavia pattern (SCA) (Chen & Hellström, 1999; Hurrell et al., 2003; Oleksy & Richardson, 2021). However, a knowledge gap remains regarding the extent to which DOC and Fe concentrations in lakes align with such periodic climate patterns (Wilkinson et al., 2020).

Kritzberg and Ekström (2012) noticed that TOC and Fe concentrations were influenced by similar—but not identical—processes in 30 river mouths in Sweden, but they did not observe opposing trends between TOC and Fe in most of their samples over the 1970–2010 period. On the other hand, Škerlep et al. (2023) found that, despite long-term increases in DOC, Fe levels declined in higher-order streams in northern Sweden. A recent study by Paltsev et al. (2024) provided evidence of lake browning in Fennoscandian lakes with no concomitant increase or even a decrease in Fe during the 1995–2019 period. The reason for this discrepancy might be that (a) lake browning occurs with different intensities in different lakes (e.g., clear vs. brown lakes) (Arvola et al., 2025; Lapierre et al., 2021), (b) lake browning is not a process where both DOC and Fe *simultaneously* increase over time (Paltsev et al., 2024; Stetler et al., 2021), and (c) Fe enters lakes in forms other than OC-Fe or FeOOH that are not related to DOC (Ekström et al., 2016). Given the high sensitivity of Fe to redox conditions, its ability to form complexes with OC, and its strong influence by catchment processes (e.g., hydrology, soil properties and vegetation) (Björnerås et al., 2021; Heikkinen et al., 2022), both its quantity and quality are likely to vary greatly not only spatially (among lakes) but also temporally (among years). It can be reasoned that the proportion of Fe variability explained by temporal drivers may exceed that explained by spatial drivers—though this does not diminish the importance of spatial influences. Temporal variation could outweigh spatial variation due to the steadily increasing impacts of temperature increase and recovery from acidification in northern boreal ecosystems, which are among the most affected regions in this context (Creed et al., 2018). Furthermore, recent studies (e.g., Isles et al., 2018; Paltsev et al., 2024; Stetler et al., 2021) suggest that using *space-for-time* substitution—

where long-term averages of lake variables are correlated across lakes—may not adequately explain the effects of global change on biogeochemical cycling in lakes. Proper time series analysis is therefore needed to disentangle these spatial and temporal couplings.

Time series analysis assumes that investigated data are long term (preferably decades) and continuous (every year and preferably without any missing values) (Hampton et al., 2019). Biogeochemical time series usually do not meet this criterion mostly due to extensive, labor-intensive, and costly field sampling and laboratory work required to obtain these data. This is especially true when the goal is to combine spatial (many lakes) and temporal (many years for the same lakes) analyses. Only a few long-term data sets, such as decadal lake monitoring programs from Fennoscandian countries, provide the opportunity to study lakes *simultaneously* and *continuously* over space and time. Furthermore, most traditional time series analyses examine non-stationary changes (i.e., trends) with the assumption that correlation exists only between variables that exhibit consistent directional changes—such as a continuous increase or decrease over time (Wilkinson et al., 2020). However, a variable may affect other variable(s) on a different time scale (i.e., the variables may have repeating oscillations—e.g., at seasonal, annual or *n*-year time scales).

Wavelet coherence—a method within the wavelet analysis family—is used to identify the direction and strength of correlation between two time series across different time scales (Grinsted et al., 2004) and therefore expands traditional "correlation" into "coherence" perspective (i.e., it measures if two time series oscillate together over time). Originating in signal processing, wavelet coherence has since been applied to the analysis of climate, hydrological, and ecological time series (Centeno et al., 2020; Hieronymus et al., 2018; Juez et al., 2022; Liu et al., 2019; Mengistu, Creed, et al., 2013; Schmidt et al., 2019; Walter et al., 2020). This method can be applied to both non-stationary and stationary (i.e., wave-like/oscillating) time series (Centeno et al., 2020; Grinsted et al., 2004) and can detect periods of coherence even when they are confined to relatively short time spans (Schmidt et al., 2019). One of the most known oscillating time series in nature is climate teleconnections, which exhibit periodic or quasi-periodic fluctuations (Chase et al., 2006). Hence, wavelet coherence is particularly useful in finding correlations between climate shaped by these teleconnections and the export and transformation of chemical elements within the catchment-lake continuum (Mengistu, Creed, et al., 2013; Mengistu, Quick, & Creed, 2013; Walter et al., 2023; Wilkinson et al., 2020; Winder & Cloern, 2010; R. Zhao et al., 2018).

This study sought to explain what causes *a temporal decoupling* between total organic carbon (TOC) and total iron (Fe). We refer to temporal decoupling as a situation where changes in Fe over time (whether an increase or decrease) do not correspond with changes in TOC, meaning the slopes of their trends move in opposite directions. In our previous study (Paltsev et al., 2024), we found evidence that the temporal decoupling between TOC and Fe might exist in some Fennoscandian lakes over a 25-year period. In this research, we asked the following questions: (a) Are TOC and Fe in Fennoscandian lakes correlated over space but not over time (over 30 years)? If so, is this correlation different for clear and brown lakes? (b) Are changes in TOC and Fe related to precipitation, temperatures, and S deposition? If so, do the *combined* effects of these environmental variables influence TOC and Fe differently, thereby driving their decoupling? (c) Is the variation in Fe primarily driven by *long-term trends* in precipitation, temperature and S deposition, or by *their interannual variability* and *periodic patterns*?

We aimed to test two hypotheses: (a) climate (precipitation and warming) and S deposition play major roles in controlling the temporal variability of TOC and Fe, and (b) due to its redox sensitivity (including in the catchment soils) and mineral interactions (e.g., with sulfides or hydroxides), Fe is more responsive to the interannual fluctuations in climate patterns than TOC, leading to greater variability in Fe trends and the overall temporal decoupling of Fe from TOC in Fennoscandian lakes. We used Sen's slopes to assess rates of change in TOC and Fe, the Bayesian models to evaluate the influence of multiple environmental variables on Fe and TOC concentrations, and wavelet coherence analysis to identify potential temporal coherence between Fe and TOC, and between each of these variables and environmental variables. These statistical techniques were applied on a time series from 102 Fennoscandian lakes, which were extensive in time (31 years, 1990–2020) and space (from 55°N to 68°N), with broad gradients in TOC (1.3–20.5 mg L^{-1}) and Fe (14.3–4,272.5 μg L^{-1}), enabling exploration of associations between climate, deposition, and water chemistry on a regional scale.

2. Methods

2.1. Data on Water Chemistry, Climate, and Deposition

Data on water chemistry parameters—TOC and Fe—were obtained from Swedish (Miljodata-MVM, 2021) and Finnish lake monitoring programs. Lakes in these countries have been sampled on an annual basis since 1990 (with some missing TOC and/or Fe for some years—especially in Finnish lakes). To minimize potential bias in the analysis caused by missing data, we selected lakes with at least 28 years (90%) of available data for both TOC and Fe. This resulted in annual data for 102 lakes (Sweden = 90 lakes, and Finland = 12 lakes) for the period of 1990–2020. All 102 lakes had information on lake and catchment size, while 96 lakes had information on wetland and forest cover in catchments (Table 1, Table S1 in Supporting Information S1). To check if the coupling/decoupling pattern between TOC and Fe is similar/stable across the lakes with various stages of browning, we divided our study lakes into three groups in accordance with TOC concentration, as follows: clear lakes (TOC < 5 mg L^{-1}; $n = 23$), medium brown lakes (TOC 5–10 mg L^{-1}; $n = 50$) and brown lakes (lakes having the highest TOC concentration, i.e., TOC > 10 mg L^{-1}; $n = 29$). TOC was calculated as mean TOC over all years for individual lakes. These groups also represent "typical" categories of lakes in Fennoscandia described by other authors (e.g., Ask et al., 2012; Bergström & Karlsson, 2019).

Water for analysis of lake water chemistry (including TOC and Fe) was collected at 0.5 m depth in Sweden and Finland yearly between three and four times over the open water season (between May and early October). For this study, we used water chemistry data from May to late September to cover approximately the same duration of the open water season in both the northern and southern regions of Fennoscandia. All analyses were performed at accredited laboratories: at the Department of Aquatic Sciences and Assessment of the Swedish University of Agricultural Sciences in Sweden, and at the Finnish Environmental Administration and consultant Eurofins Environment Finland (Fölster et al., 2014).

TOC concentration was used as a proxy for DOC concentration based on previous studies showing that >90% of TOC is DOC in Swedish and Finnish lakes (Köhler et al., 2002; Kortelainen et al., 2006). Iron was measured as total Fe concentration, including dissolved, colloidal and particulate Fe, of which colloidal Fe is expected to be the major part (Wetzel, 2001; Xiao & Riise, 2021). We used the lake-to-catchment ratio as a proxy for water retention time (Xiao & Riise, 2021).

Mean monthly precipitation (hereafter: PRE) and mean monthly maximum air temperature (hereafter: AT) and sulfur (S) deposition (hereafter: Sdep) were determined for the summer period for each lake and for each year for the 1990–2020 period. We considered May through September as summer months. The period from May to September was selected to align with the water chemistry sampling period and to include spring runoff in our analysis because spring runoff can be the largest contributor of TOC and Fe to lakes (Björnerås et al., 2021; Stetler et al., 2021). Maximum temperature was chosen over mean temperature, because maximum temperature is a better predictor of extreme weather events (e.g., droughts) and spatial distribution of water chemistry and plankton/biota (Bergström et al., 2024; Hallstan et al., 2013), and was also shown to have a correlation with atmospheric teleconnections—for example, the North Atlantic Oscillation (González-Hidalgo et al., 2022; Trigo & Osborn, 2002). Temperature and precipitation values were extracted from the "TerraClimate Data set" (high-spatial resolution ($1/24^0$, ~4 km)) monthly air temperature and monthly precipitation grids (see Abatzoglou et al., 2018). We downloaded gridded, interpolated estimates of wet Sdep from the European Monitoring and Evaluation Programme (EMEP) (Amann et al., 2018; Colette et al., 2016).

2.2. Statistical Analysis

2.2.1. Analysis of Trends

Sen's slopes (Sen, 1968) were calculated to estimate the direction of change (i.e., positive or negative), significance, and rates of change in TOC and Fe concentrations as well as PRE, AT and Sdep in individual lakes over the 1990–2020 period. The Theil–Sen estimator is robust to missing data and outliers, and it does not assume normally distributed residuals.

In this study, we included all lakes in the analysis, encompassing both those with significant ($p < 0.05$) and non-significant slopes. We included all lakes to be able to demonstrate (a) "general" changes (tendency) across all lakes in Fennoscandia, and (b) that some temporal changes are driven not by long-term trends in climate (e.g., AT)

Table 1
General Information on Fennoscandian Lakes

	Parameter	Quartile I	Mean	Median	Quartile III
All lakes, $n = 102$ (100%)	Lake surface area size (ha)	24.3	115.4	46.2	129.2
	Lake catchment size (ha)	199.4	4567.2	592.7	1592.6
	Catchment/lake ratio	5.4	33	9.3	20.3
	Wetland area in catchment (%)	–	4.5	–	2.5
	Forested area in catchment (%)	68.4	73.3	76.0	89.2
	TOC (mg L^{-1})	5.2	8.5	8.2	10.9
	Fe (μg L^{-1})	87.2	453.8	211.3	491.7
	PRE (mm)	64	71	69	78
	AT (°C)	16.5	17.2	17.9	18.5
	Sdep (mg m^2)	10.9	18.1	17.8	24.1
Lakes with positive slopes in Fe, $n = 60$ (59%)	Lake surface area size (ha)	20.2	84.8	40.4	113.5
	Lake catchment size (ha)	174.7	1077.6	482.5	990.4
	Catchment/lake ratio	5.5	13.1	8.6	16
	Wetland area in catchment (%)	–	2.2	–	1.5
	Forested area in catchment (%)	72.3	79.1	76.8	90.7
	TOC (mg L^{-1})	5.7	8.6	8.3	11.1
	Fe (μg L^{-1})	106.5	372.4	211.3	458.9
	PRE (mm)	64	72	70	80
	AT (°C)	17.1	17.6	18.1	18.6
	Sdep (mg m^2)	13.9	19.5	18.7	25
Lakes with negative slopes in Fe, $n = 42$ (41%)	Lake surface area size (ha)	31.7	159.1	73.7	166.9
	Lake catchment size (ha)	393	9552.2	853.9	4567.5
	Catchment/lake ratio	5.2	61.4	14.8	50
	Wetland area in catchment (%)	–	7.6	–	4.3
	Forested area in catchment (%)	57.5	65.5	70.9	87.5
	TOC (mg L^{-1})	4.9	8.4	8.2	10.5
	Fe (μg L^{-1})	44.6	570.1	209.2	643.9
	PRE (mm)	62	69	68	76
	AT (°C)	15.9	16.7	17.4	18.4
	Sdep (mg m^2)	8.1	16.2	13.7	20
Decoupled lakes with negative slopes in Fe and positive slopes in TOC $n = 33$ (32%)	Lake surface area size (ha)	23.7	118.7	44.9	134.5
	Lake catchment size (ha)	300.8	1888.1	493.9	1675.2
	Catchment/lake ratio	4.9	54.9	10.4	32.8
	Wetland area in catchment (%)	–	5.1	–	2.9
	Forested area in catchment (%)	68.7	76.1	76.9	89.7
	TOC (mg L^{-1})	5.5	9.4	9.5	11.9
	Fe (μg L^{-1})	42.4	694.8	347.4	866.5
	PRE (mm)	61	70	68	68
	AT (°C)	16.6	17.5	17.83	18.5
	Sdep (mg m^2)	10.9	18.9	16.7	25.7

Note. To avoid misinterpretation, we excluded medians and first quartile values for the wetland area in the catchment. This is because many catchments lack wetlands entirely (i.e., Wetland area = 0), resulting in medians and first quartile values being 0 as well.

or Sdep but by interannual fluctuations and cycles (e.g., in PRE). We aimed to deviate from traditional *p*-value thresholds and include as many lakes as possible by considering the ecological context (Wasserstein et al., 2019; Wasserstein & Lazar, 2016). Many recent studies have also used a similar approach (e.g., Bergström et al., 2024; Hrycik et al., 2021; Paltsev et al., 2024).

2.2.2. The Bayesian Mixed-Effects Model

To evaluate the influence of multiple environmental variables on lake Fe concentration, we employed a Bayesian mixed-effects model (Ellison, 2004; Lunn et al., 2012). This modeling approach is especially suited for ecological data sets that are nested, have imbalanced sampling across groups, and involve complex interactions among covariates (Bolker et al., 2009; Korner-Nievergelt et al., 2015). In these models, parameters such as slopes can vary across lakes, and these parameters are themselves treated as random variables. Unlike traditional regression, Bayesian models produce posterior distributions rather than point estimates and *p* values, with inference based on the proportion of the posterior above or below zero. These posteriors are estimated using Markov chain Monte Carlo methods, which generate representative samples via iterative simulations where uncertainty is expressed as credible intervals (Bolker et al., 2009; Ellison, 2004).

For the model, we used time series of Fe concentration as the response variable and times series of TOC, PRE, AT, and Sdep as predictor variables. The model was performed on all lakes ($n = 102$) as well as on lake types separately (i.e., on clear, medium-brown and brown lakes). All predictors were z-transformed to aid model convergence and enable the interpretation of interaction effects (Bürkner, 2017). The model included all *main effects* and *interactions* among the four predictors, allowing us to assess if the effect of one environmental variable on Fe depends on the level of others. Additionally, it included random intercepts and random slopes for TOC by lake, accounting for spatial heterogeneity in baseline Fe levels and in the strength of the TOC–Fe relationship. To account for within-lake temporal autocorrelation, a first-order autoregressive structure [AR(1)] was also added to the model (Berninger et al., 2022).

The model was run using four chains, each with 8,000 iterations (3,000 warm-up), for a total of 20,000 post-warmup samples (Bürkner, 2017; McElreath, 2020). The significance of model parameters was assessed by examining whether the 95% credible intervals (CI) excluded zero (Bürkner, 2017). We plotted 3D figures showing significant combined two-way interactions (effects) of predictor variables on Fe concentration. We also ran an additional Bayesian mixed-effects model to evaluate the influence of PRE, AT, and Sdep on TOC concentration in individual lakes. The model used parameters similar to those applied in the "Fe model."

2.2.3. Wavelet Coherence

Wavelet coherence analysis was used to investigate the temporal coherence between time series of: (a) TOC and Fe concentrations; (b) TOC concentration and PRE, AT, and Sdep; and (c) Fe concentration and PRE, AT, and Sdep for individual lakes. Prior to analysis, each time series was detrended and z-transformed to have mean = 0 and standard deviation = 1 (Sheppard et al., 2016; Walter et al., 2023).

The continuous wavelet transform was applied to transform the time series into a time-frequency space. Comprehensive reviews on theoretical aspects of the continuous wavelet transform (as well as the wavelet coherence) can be found elsewhere (e.g., Cazelles et al., 2008; Grinsted et al., 2004; Torrence & Compo, 1998). In brief, a wavelet is a function (Ψ_t) resembling a small wave that moves/oscillates along a time series and that can be applied as a bandpass filter to the time series (Grinsted et al., 2004). The wavelet function compresses or stretches along the time axis (x) with various frequencies (Schmidt et al., 2019). In this study, we used the Morlet wavelet because it provides a good balance between time and frequency (Labat, 2005; Sheppard et al., 2016; Si & Zeleke, 2005) and has been successfully used for the wavelet analysis of environmental time series in the past (e.g., Centeno et al., 2020; Juez et al., 2022; Mengistu, Creed, et al., 2013; Schmidt et al., 2019). The Morlet wavelet is defined as (Grinsted et al., 2004):

$$\Psi(t) = \pi^{-\frac{1}{4}} e^{\omega_0 t} e^{-\frac{t^2}{2}} \tag{1}$$

where t is time and ω_0 is the non-angular frequency, which by default is equal to 6 (Grinsted et al., 2004).

Continuous wavelet transform $W(f, \tau)$ at scale f and time τ is represented as Equation 2:

$$W(f,\tau) = \frac{1}{\sqrt{f}}\int_{-\infty}^{+\infty} X(t)\Psi^{*}\left(\frac{1-\tau}{f}\right)dt = \int_{-\infty}^{+\infty} X(t)\Psi^{*}_{f,\tau}dt \tag{2}$$

where *X(t)* is the time series, (*) indicates the complex conjugate form, and *dt* is the integration measure (Cazelles et al., 2008).

Wavelet coherence identifies regions in the time–space where two time-series (*X* and *Y*) co-move ("covariate") by comparing the wavelet spectra of these time-series (Juez et al., 2022), which are defined as (Grinsted et al., 2004):

$$R^2_{f,\tau} = \frac{\left|S\left(f^{-1}\ W^X_{f,\tau} W^{Y^*}_{f,\tau}\right)\right|^2}{S\left(f^{-1}\ \left|W^X_{f,\tau}\right|^2\right)\cdot S\left(f^{-1}\ \left|W^Y_{f,\tau}\right)\right|^2\right)} \tag{3}$$

where $R^2_{f,\tau}$ is a measure of square wavelet coherence between *Y* and *X*, $W^X_{f,\tau}$ is the first time series wavelet transform with the complex conjugation of the second ($W^Y_{f,\tau}$) normalized by the individual power spectra of each time series, and *S* is a smoothing operator for both scale and time domains (Si & Zeleke, 2005; Torrence & Compo, 1998).

The statistical significance of coherence was evaluated by comparing the observed values to a distribution of coherence values generated from surrogate data (Walter et al., 2020, 2023). These surrogates were generated under the null hypothesis of no true coherence. Because the surrogate generation procedure modifies only the phase of the oscillations while preserving the power spectrum of each time series, it is regarded as a conservative approach (Sheppard et al., 2016; Walter et al., 2020). Coherence was considered statistically significant at $p < 0.05$.

Wavelet coherence measures how strongly two time series are correlated in both the magnitude of their oscillations and the consistency of their phase relationship over time across different timescales. The coherence magnitude ranges from 0 (no relationship) to 1 (perfect coherence). Phase indicates whether the relationship is positive (in-phase, i.e., time series move/oscillate synchronously, where an increase in one time series is accompanied by an increase in the other time series at a given period), negative (out-of-phase, i.e., time series move in opposite directions), or partial (or time lagged). However, since we did not have within year data (i.e., monthly or weekly data, which are appropriate for time lagged relationship), the analysis of this type of relationship was beyond the scope of this paper. Phase relationships of individual lakes can be best presented on heatmaps (i.e., wavelet coherence density plots). The heatmaps do not show the magnitude of coherence, but rather where coherence occurred most frequently across lakes and time, with a specific phase relationship. On the heatmaps, phase values are typically visualized using a color scale, where phase differences are measured in radians and indicate the following: 0 radians = perfectly in-phase, $\pm\pi$ = perfectly out-of-phase and $\pm\pi/2$ = partial/lagged relationships. For simplicity, we classified phase differences between $-\pi/2$ and $+\pi/2$ radians as *in-phase*, and all other values as *out-of-phase*. Some phase values were located right on the boundary ($-\pi/2$ and $+\pi/2$; i.e., the coherence was not dominated by either an in-phase or out-of-phase pattern), hence we classified them as mixed.

Analyses were conducted in R software (v. 4.2.2; R Core Team, 2022) using the following packages: *trend* for the Sen's lopes estimation, *brms* (Bürkner, 2017) and *glmmTMB* for the Bayesian models and *wsyn* (Reuman et al., 2021) for wavelet coherence.

3. Results

3.1. Temporal Decoupling Between Fe and TOC and Long-Term Trends in Fe, TOC, and Environmental Variables

TOC and Fe were correlated with space ($R^2 = 0.58$, $p < 0.0001$; Figure 1a), but not over time (Figures 1b–1e). Median change in Fe concentration was 0.4 μg L^{-1} yr^{-1}, while median change in TOC concentration was 0.1 mg L^{-1} yr^{-1}. Among all study lakes, 59% had positive slopes in Fe, with 20% of lakes showing statistically significant positive slopes ($p < 0.05$). In comparison, 86% of the same lakes had positive slopes in TOC, and for 64% of lakes, these slopes were significant ($p < 0.05$) (Figures 1d and 1e, and Table S2 in Supporting

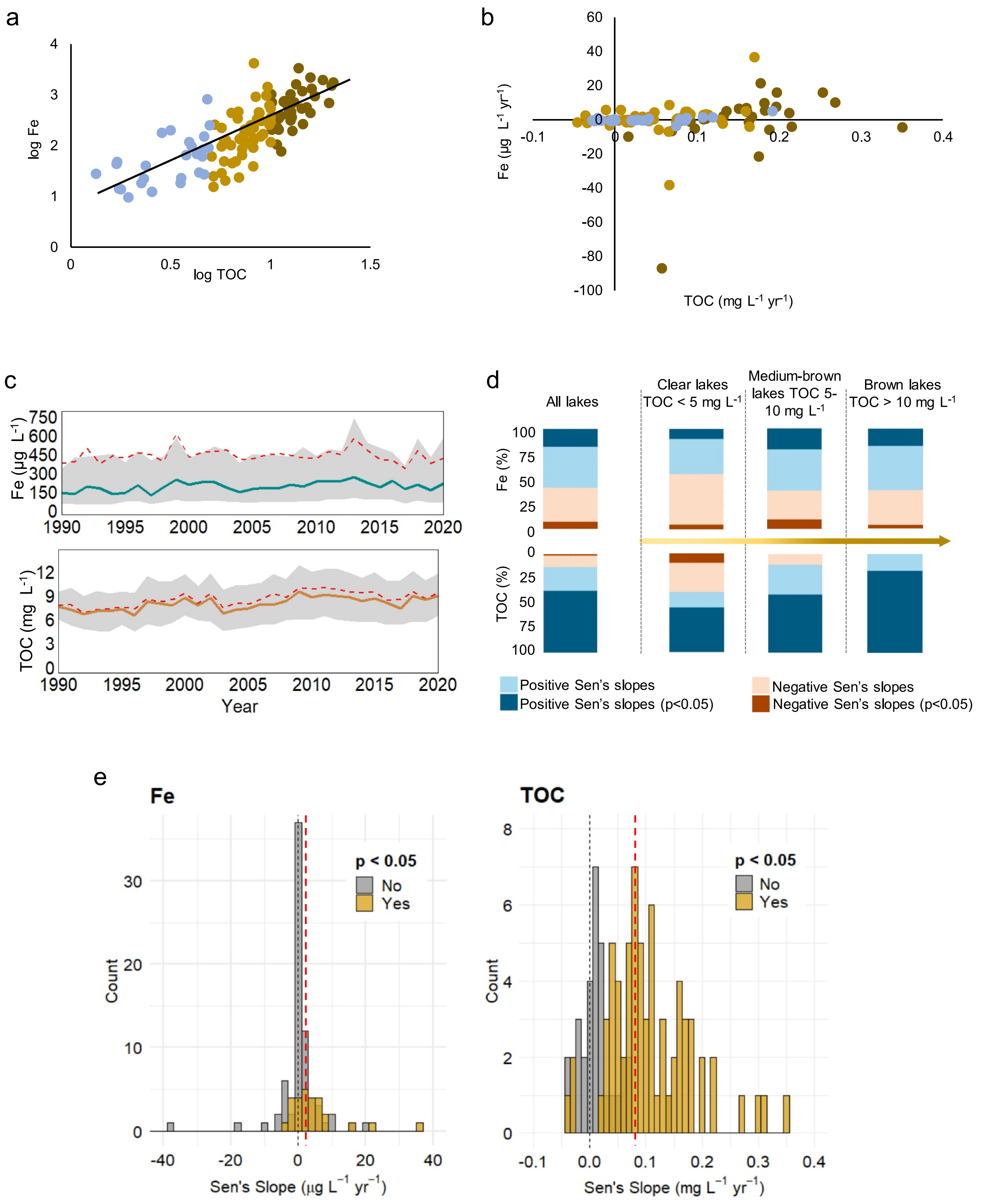
a
log Fe
log TOC
b
Fe (µg L-1 yr-1)
TOC (mg L-1 yr-1)
c
Fe (µg L-1)
TOC (mg L-1)
Year
d
All lakes
Clear lakes TOC < 5 mg L-1
Medium-brown lakes TOC 5-10 mg L-1
Brown lakes TOC > 10 mg L-1
Fe (%)
TOC (%)
Positive Sen's slopes
Positive Sen's slopes (p<0.05)
Negative Sen's slopes
Negative Sen's slopes (p<0.05)
e
Fe
TOC
p < 0.05
No
Yes
Count
Sen's Slope (µg L-1 yr-1)
Sen's Slope (mg L-1 yr-1)

Figure 1.

Information S1). The proportion of lakes with positive slopes in TOC and Fe generally increased with higher TOC concentration (i.e., brown lakes became browner, more so than clear lakes) (Figure 1d). However, even in the brown lakes (TOC > 10 mg L^{-1}), where all lakes exhibited positive slopes in TOC over time, only 66% of the lakes had positive slopes in Fe.

Among the study lakes, 41% exhibited negative slopes in Fe (with 11% showing negative slopes with $p < 0.05$), whereas 14% had negative slopes in TOC (with only 3% being significant). The proportion of negative slopes in Fe were consistently higher than the ones in TOC (Figure 1d). For example, 38% of clear lakes (TOC < 5 mg L^{-1}) exhibited negative slopes in TOC, but more than half (52%) exhibited negative slopes in Fe. This indicates that in 14% of these lakes, Fe concentrations were declining even though TOC did not. Clear lakes also had the highest proportion of negative slopes for both TOC and Fe compared to medium-brown and brown lakes. Interestingly, while median and mean TOC concentrations were closely aligned over the 1990–2020 period (thick solid and red dashed lines on Figure 1c), mean Fe was always higher than median Fe, suggesting higher variability in Fe.

In 21% ($n = 9$) of the lakes with negative slopes in Fe, TOC also showed negative slopes, indicating that these lakes remained temporally coupled to TOC. Overall, this means that in 32% ($n = 33$) of the study lakes, Fe and TOC were temporally decoupled, with Fe declining while TOC continued to increase (i.e., decoupled lakes = lakes with negative slopes in Fe that do *not* also have negative slopes in TOC).

Positive slopes in PRE were found in most lakes (62%), with only 3% of the lakes exhibiting significant ($p < 0.05$) increase in PRE (Figure 2). None of the lakes with negative slopes in PRE had a significant decline. All study lakes had positive slopes in AT with 48% having positive significant slopes. Sdep declined significantly in all lakes.

3.2. Spatial Patterns

The study lakes exhibited substantial spatial gradients in TOC, Fe, AT, PRE, and Sdep, alongside substantial variation in properties such as lake size, catchment area, and land cover (Table 1). The study lakes spanned the boreal zone of Sweden and Finland, extending northward into subarctic regions, where six lakes were located within areas of sporadic permafrost (Figure 3). Clear lakes were primarily concentrated in northwestern and western Sweden and Finland (Figure 3a). In Sweden, these areas correspond to mountainous and upland regions, giving clear lakes the highest mean elevation among all lake types (320 m a.s.l.; Figure 4). In Finland, however, the spatial pattern may partly reflect the smaller number of sampled lakes, which does not capture the full range of lake types across the country (Kortelainen & Mannio, 1990). Clear lakes had the smallest catchment-to-lake area ratio (mean = 18.8) and the lowest fraction of forest cover among lake types (mean forested area = 53.2%) (Figure 4; Table S1 in Supporting Information S1). Medium-brown lakes (5 < TOC < 10 mg L^{-1}) were mostly located in central, southern, and eastern Sweden. These lakes were the largest (mean lake size = 151.5 ha) and had the largest catchments (mean = 6861.6 ha) with extensive forest and wetland cover. Brown lakes were mostly located in southern Sweden and Finland. These lakes tended to have the highest Fe concentration (mean Fe = 817.6 μg L^{-1}), although some clear lakes in Finland also displayed high mean Fe levels (Figure 3). Brown lakes were small (mean lake size = 66.5 ha) with the highest catchment-to-lake area ratio (mean = 46.1), the highest proportion of forest cover in their catchments (mean forested area = 83%) and situated at the lowest elevations (mean = 155 m a.s.l.). For details, see Table S1 in Supporting Information S1.

TOC concentrations increased across most study lakes, except for a subset in mountainous northwestern Sweden, where changes in TOC were negative (with two lakes having statistically significant negative trends in TOC; Figure 3b). Fe trends tended to vary geographically, with negative slopes dominating in northern Fennoscandia and mixed slopes in southern Fennoscandia. Lakes with negative slopes in Fe and decoupled lakes were generally larger (mean area of lakes with negative slopes in Fe = 159.1 ha, and mean area of decoupled lakes = 118.7 ha

Figure 1. Spatial correlation between Fe and TOC concentrations (both $\log_2$-transformed) based on mean values across all years for each of 102 Fennoscandian lakes; (b) relationship between Sen's slopes (changes over time) in Fe and TOC concentrations for the same 102 lakes; (c) time series of median (thick solid lines), mean (red dashed lines) and interquartile range (gray shaded area) of TOC and Fe for the 1990–2020 period; (d) the proportion of positive and negative Sen's slopes in Fe and TOC for all lakes and lakes divided in three groups—that is, clear, medium-brown and brown; and (e) histograms showing distribution of Sen's slopes for Fe and TOC. In (a) and (b) different lake colors stand for lake types: blue—for clear lakes, light brown—for medium-brown lakes, and dark brown—for brown lakes. In (d), the brown arrow symbolizes the intensity of browning from clear to brown lakes; in (e) vertical red dotted lines indicate the mean Sen's slope of Fe and TOC.

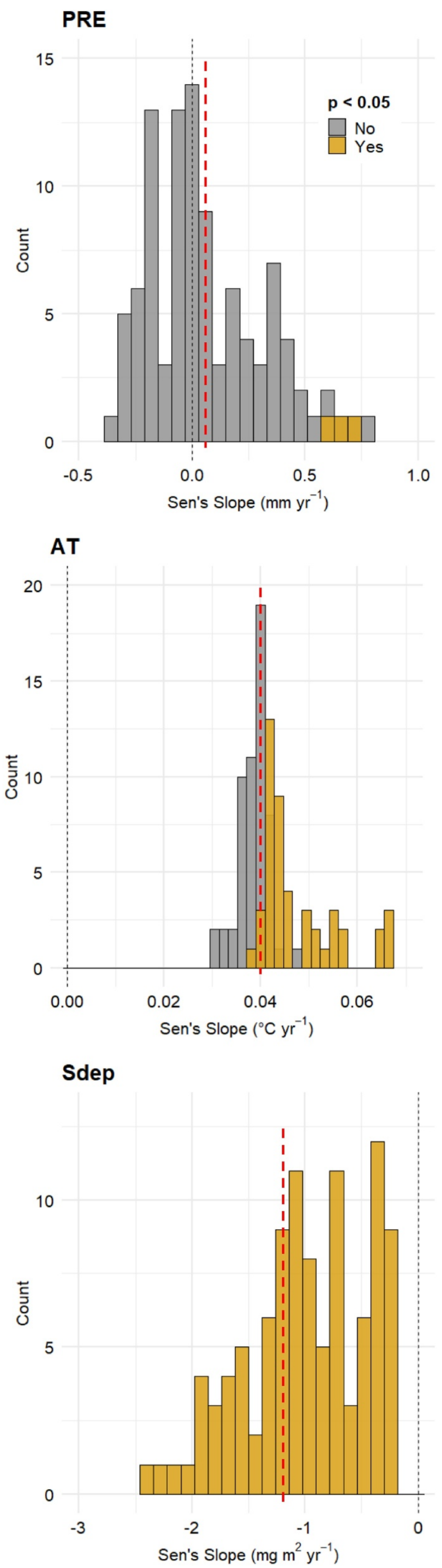

Figure 2. Histograms showing distribution of slopes for PRE, AT and Sdep, indicating slopes with a significant change ($p < 0.05$) (See legend on the upper panel). Vertical red dotted lines indicate the mean Sen's slope of each variable.

accordingly) than those with positive slopes in Fe (mean area = 84.8 ha). They also had larger catchments, higher catchment-to-lake area ratios, and greater forest and wetland coverage in their catchments (Table 1).

3.3. Influence of Environmental Variables on Fe and TOC

3.3.1. Fe Model

There were strong temporal associations between Fe concentrations and environmental variables (Table 2, Figure 5). We describe the results of the all-lake model ($n = 102$) in detail, while results for clear, medium-brown, and brown lakes are summarized more briefly; full results are presented in Table 2. TOC was the strongest predictor of Fe, with a positive effect (Estimate = 0.41, CI: 0.32 to 0.50), indicating that higher organic carbon content was associated with elevated Fe concentrations over time. AT had a small but significant positive effect (Estimate = 0.05, CI: 0.01 to 0.10), suggesting that warmer years were generally associated with higher Fe concentrations. PRE and Sdep did not show significant effects when considered individually.

The 3D response surfaces (Figure 5, also see Table 2) showed that lake Fe concentration is shaped by interactions between environmental variables rather than by simple additive effects (i.e., the plots showed that the influence of one variable depended on the level of the other). In an additive relationship, the effect of one variable (e.g., PRE) would be constant regardless of the level of another variable. There was a negative interaction between TOC and PRE (TOC × PRE), showing that high TOC and PRE were associated with a decrease in Fe concentration. However, this interaction was marginally significant and weak (Estimate = −0.02, CI: −0.04 to 0.00). Fe concentration was highest under conditions of low PRE combined with low AT (upper left corner, yellow peak). As PRE increases (moving right), Fe concentration generally decreases, especially at higher AT—that is, higher PRE and higher AT were associated with lower Fe concentration. This indicates a negative interaction (Estimate = −0.04, CI: −0.06 to −0.01), where the combined effects of wetter and warmer conditions led to the lowest Fe levels. The interaction between TOC and Sdep was positive (Estimate = 0.05, CI: 0.02 to 0.08), implying that Fe concentration tended to be higher in years when both organic carbon and S inputs were elevated. AT × Sdep showed a positive interaction (Estimate = 0.06, CI: 0.02 to 0.10): Fe concentration increased when both AT and Sdep were high. In contrast, low AT combined with low Sdep was associated with the lowest Fe levels, while intermediate conditions produced moderate Fe concentrations.

The three-way interaction among PRE, AT, and Sdep was negative (Estimate = −0.05, CI: −0.08 to −0.02; Table 2), indicating a suppressive effect on Fe concentration when all three factors were simultaneously elevated. Other two-way and higher-order interactions were not statistically significant. The model R^2 was 0.839 and the estimated error of prediction was 0.009 μg L^{-1}.

Across lake types, Fe concentrations showed various responses to TOC, PRE, AT, and Sdep (Table 2). In clear lakes, TOC, PRE, and Sdep had no significant effect, while AT was the strongest positive predictor. A negative TOC × PRE × AT interaction suggested that wet, warm conditions with high TOC reduced Fe levels, while a positive TOC × AT × Sdep interaction indicated that warm, high-TOC conditions with elevated Sdep increased Fe. The AT × Sdep effect was also positive but borderline significant. The model R^2 for clear lakes was 0.432 with a prediction error of 0.049 μg L^{-1}.

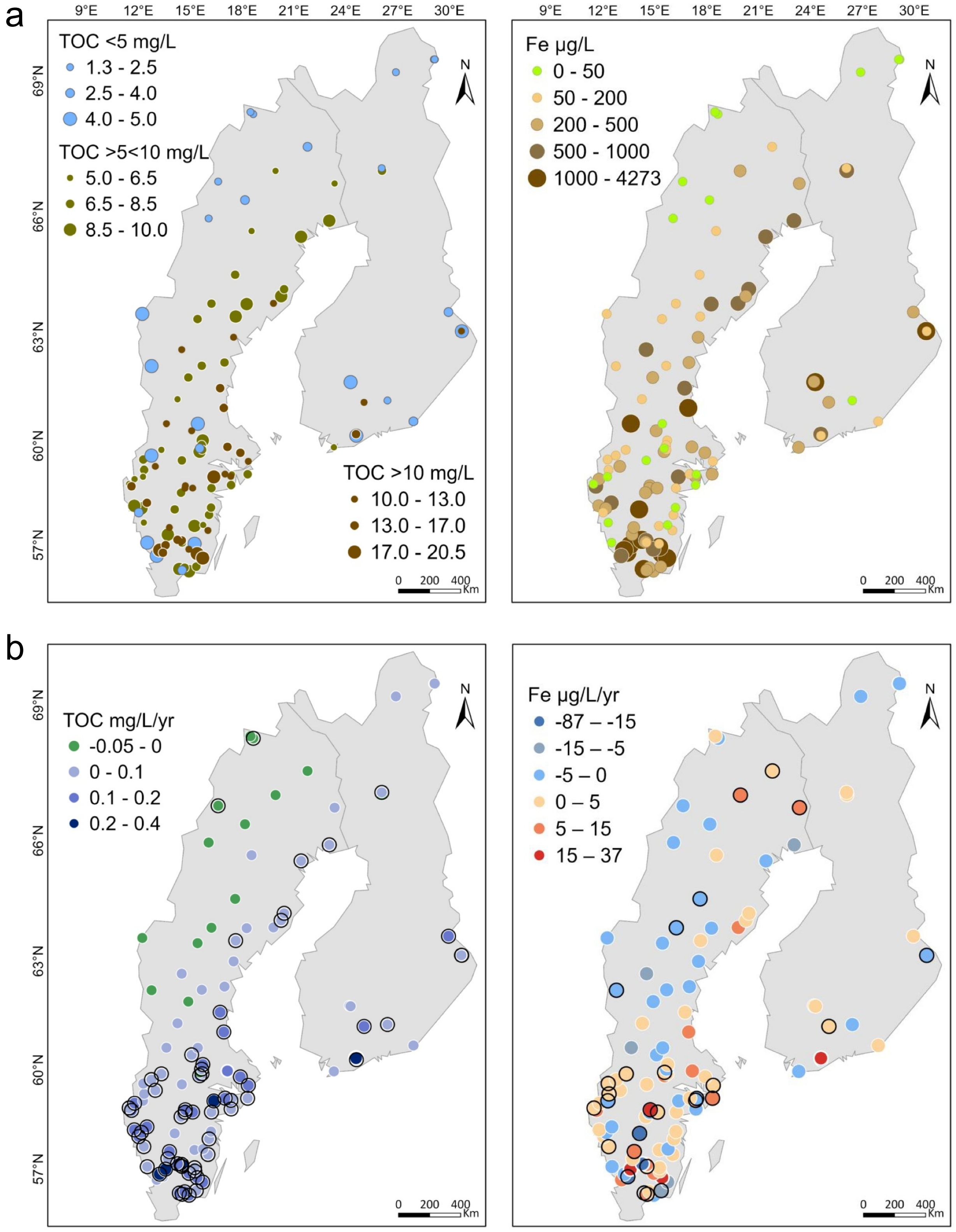

Figure 3. (a) Spatial distribution of TOC and Fe in Fennoscandian lakes (mean values across all years), and (b) spatial distribution of Sen's slopes for TOC and Fe. Lakes with Sen's slopes at $p < 0.05$ are outlined with black circles ($n = 102$).

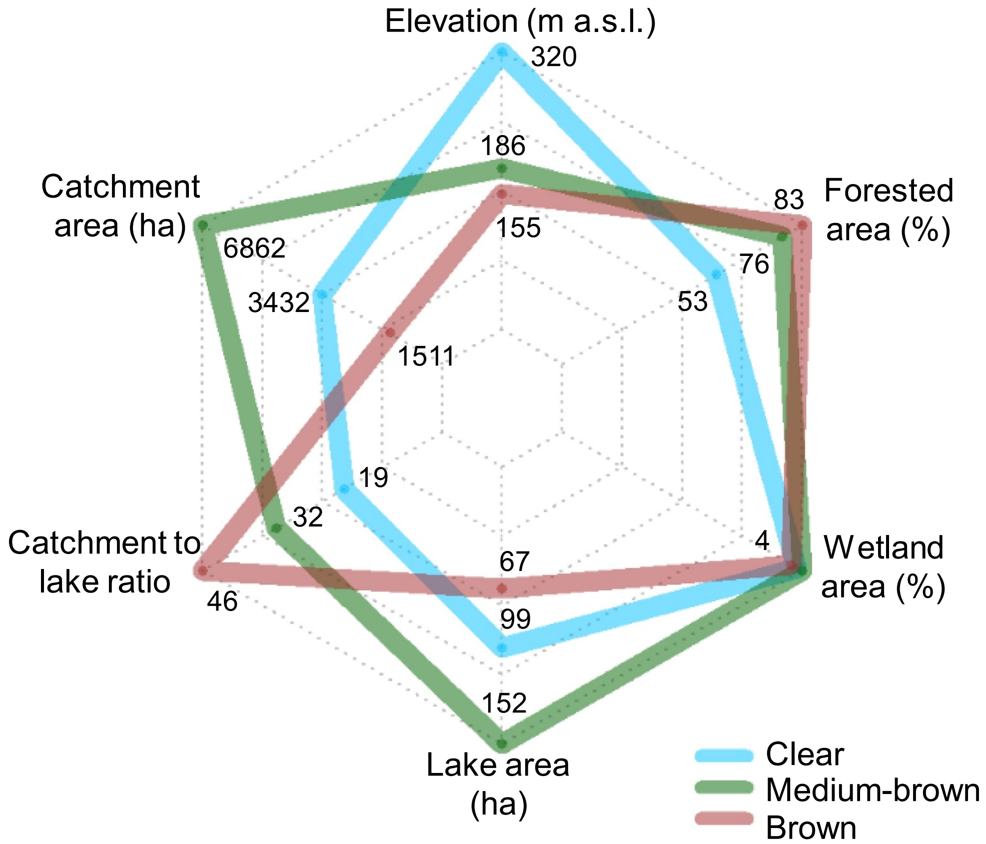

Figure 4. Radar chart comparing the main physical variables of Fennoscandian lakes by lake type. To ensure comparability, all variables were normalized to the range 0–1 before plotting, where 0, 0.25, 0.5, 0.75, and 1 correspond to the chart axes. Mean values of each variable were used (e.g., mean lake area) to create the chart, and these values are shown in units indicated beneath the variable names. For "Wetland area," the proportion is approximately the same for all lake types (i.e., ~4%). "Wetland area" and "Forested area" refer to the areas within lake catchments. For details, see Table S1 in Supporting Information S1.

In medium-brown lakes, TOC was the only strong positive individual predictor of Fe. The PRE × AT and PRE × AT × Sdep interactions both had significant negative effects, suggesting Fe declines under wet warm conditions with high Sdep. The AT × Sdep effect on Fe was positive but weak. The model R^2 was 0.845 with a prediction error of 0.017 μg L^{-1}.

In brown lakes, Fe was positively associated with TOC and Sdep. Increased PRE was linked to lower Fe. TOC × Sdep showed a positive effect on Fe levels, while TOC × AT × Sdep showed a weak negative effect, indicating that under warm, high-Sdep conditions, Fe does not track TOC increases. The model R^2 was 0.786 with a prediction error of 0.014 μg L^{-1}.

3.3.2. TOC Model

There were strong and significant temporal associations between TOC and each of the three environmental variables (PRE, AT, and Sdep; Table S3 in Supporting Information S1) in the all-lake model. Both PRE (Estimate = 0.08, CI: 0.06 to 0.09) and AT (Estimate = 0.08, CI: 0.06 to 0.10) had a positive effect on TOC (i.e., higher PRE and higher AT was associated with increased TOC), while Sdep had a negative effect on TOC (i.e., higher Sdep was associated with reduced TOC; Estimate = −0.07, CI: −0.08 to −0.06). Among the interaction terms, only the interaction between AT and Sdep was statistically significant (Estimate = −0.03, CI: −0.05 to −0.01). This interaction was also negative, implying that Sdep dampens the positive effect of AT on TOC.

Table 2
Summary of Fixed Effects From the Bayesian Model Predicting Fe Concentration

	Model				
Predictor	All lakes ($n = 102$)	Clear lakes ($n = 23$)	Medium-brown lakes ($n = 50$)	Brown lakes ($n = 29$)	Interpretation (for significant effects)
TOC	↑ 0.41 (0.32; 0.50)	x	↑ 0.23 (0.15; 0.31)	↑ 0.32 (0.24; 0.41)	Higher TOC is associated with increased Fe concentration
PRE	x	x	x	↓ −0.04 (−0.09; −0.01)	Fe is lower under wetter conditions
AT	↑ 0.05 (0.01; 0.10)	↑ 0.18 (0.04; 0.31)	x	x	Fe is higher under warmer conditions
Sdep	x	x	x	↑ 0.09 (0.05; 0.13)	Higher Sdep linked with higher Fe
TOC × PRE	↓ weak −0.02 (−0.04; 0.00)	x	x	x	Combined increases in TOC and PRE reduce Fe
TOC × AT	x	x	x	x	No effect
PRE × AT	↓ −0.04 (−0.06; −0.01)	x	↓ −0.05 (−0.08; −0.02)	x	Combined increases in PRE and AT (wetter and warmer conditions) reduce Fe
TOC × Sdep	↑ 0.05 (0.02; 0.08)	x	x	↑ 0.09 (0.05; 0.13)	Higher TOC and Sdep increase Fe
PRE × Sdep	x	x	x	x	No effect
AT × Sdep	↑ 0.06 (0.02; 0.10)	↑ weak 0.14 (0.00; 0.28)	↑ weak 0.05 (0.00; 0.10)	x	Combined increases in AT and Sdep increases Fe
TOC × PRE × AT	x	↓ −0.10 (−0.18; −0.01)	x	x	High TOC + wetter and warmer conditions reduce Fe
TOC × PRE × Sdep	x	x	x	x	No effect
TOC × AT × Sdep	x	↑ 0.18 (0.04; 0.31)[1]	x	↓ weak −0.05 (−0.10; 0.00)[2]	1. High TOC + warm conditions + Sdep increase Fe; 2. Higher AT + Sdep dampens Fe–TOC link
PRE × AT × Sdep	↓ −0.05 (−0.08; −0.02)	x	↓ −0.11 (−0.15; −0.06)	x	Combined wet, warm conditions and high Sdep reduce Fe
TOC × PRE × AT × Sdep	x	x	x	x	No effect

Note. Only significant results are shown. Significant effects are defined as those with credible intervals (CI) not overlapping zero. Values near arrows are estimates, which represent posterior means with 95% credible intervals. Estimates show the direction and magnitude of predictor effects. Arrows pointing up represent positive effects, while arrows pointing down represent negative effects. Credible intervals are shown in brackets (95% CI Lower and 95% CI Upper). “x” are non-significant results. Effects on the borderline of significance (marginally significant) are marked as weak.

3.4. Temporal Coherence

Wavelet coherence analysis revealed distinct temporal patterns between Fe, TOC, and environmental variables (PRE, AT, and Sdep) across Fennoscandian lakes (Figures 6 and 7, Table 3). The coefficient of determination for significant coherence ranged from 0.44 to 0.56, depending on the environmental variable (Table 3). The number of lakes with identified coherence ranged from 47 to 57, indicating that TOC and/or Fe in approximately half of

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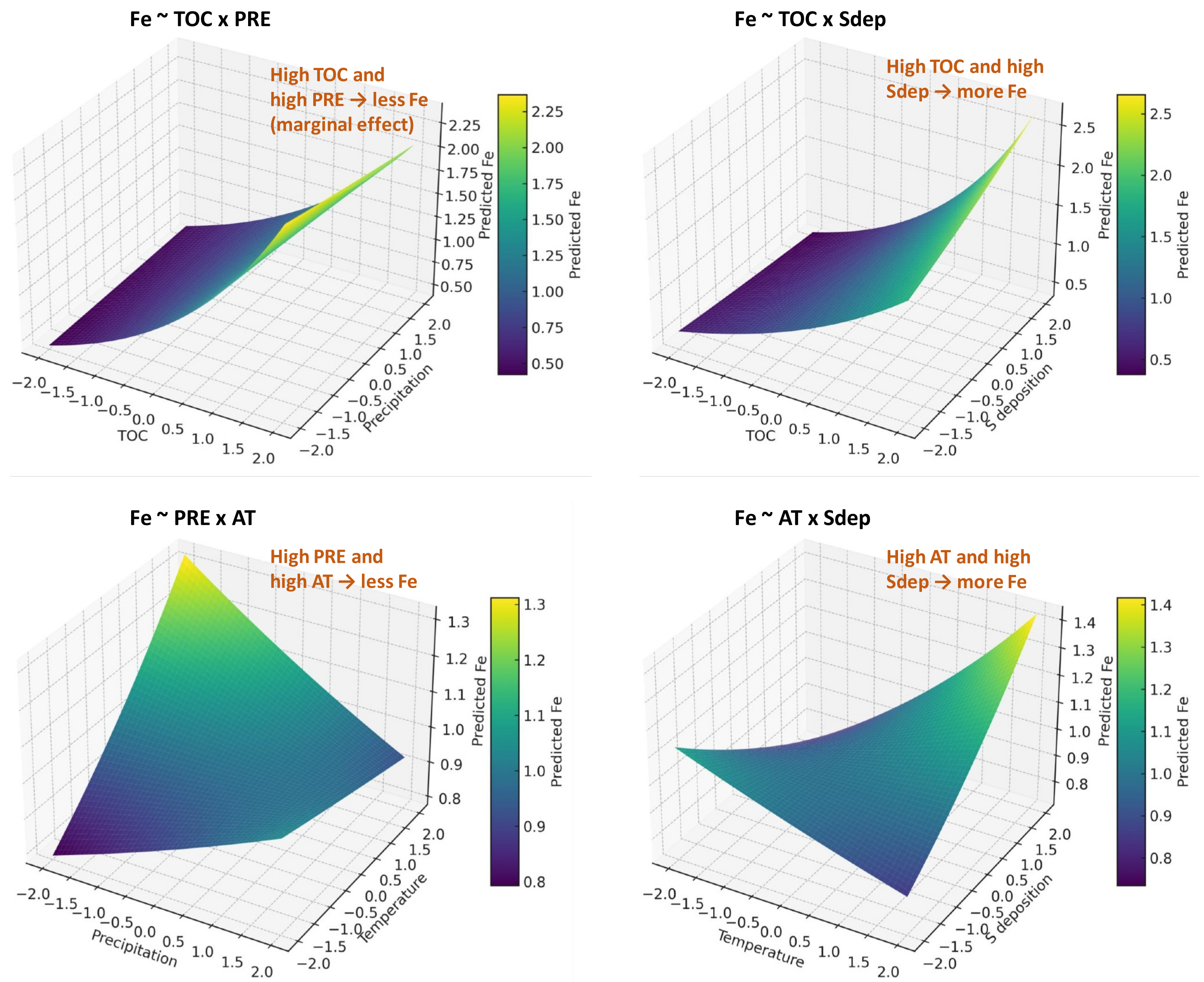

Figure 5. The 3D surfaces illustrating predicted Fe concentrations based on the significant interaction between TOC, PRE, AT and Sdep (in z-scores) for all lakes ($n = 102$), as estimated by the Bayesian model. Warmer colors indicate higher Fe concentrations. Note: TOC × PRE interaction is marginal (borderline of significance: See Table 2).

the study lakes responded to periodic patterns of environmental variables. Furthermore, coherence was most common over short timescales (2–4 years) for TOC versus Fe, as well as for Fe and TOC versus environmental variables (dense regions in the 2–4 years band in Figures 6 and 7), implying an overall sensitivity of TOC and Fe to interannual variability in the environment.

TOC and Fe were significantly coherent across multiple timescales, with dominant in-phase relationships clustering around 2–4 years, where the phase difference clustered around 0 radians (Figure 6). This indicates that changes in Fe and TOC were often synchronous, with mean coherence 0.49 (Table 3). However, a pattern of phase divergence was also observed—that is, while 25 lakes exhibited strong in-phase relationships (i.e., simultaneous changes in Fe and TOC), 16 lakes showed out-of-phase behavior. This indicates that TOC and Fe did not always co-vary synchronously over time.

Despite the overall synchrony of Fe and TOC (i.e., positive coherence), these parameters exhibited different coherence and phase relationships with environmental variables, notably with PRE (Figure 7). TOC showed consistently positive coherence (i.e., in-phase relationship) with PRE across all timescales, suggesting that increases in TOC levels were associated with increases in PRE. In contrast, Fe was frequently out-of-phase with

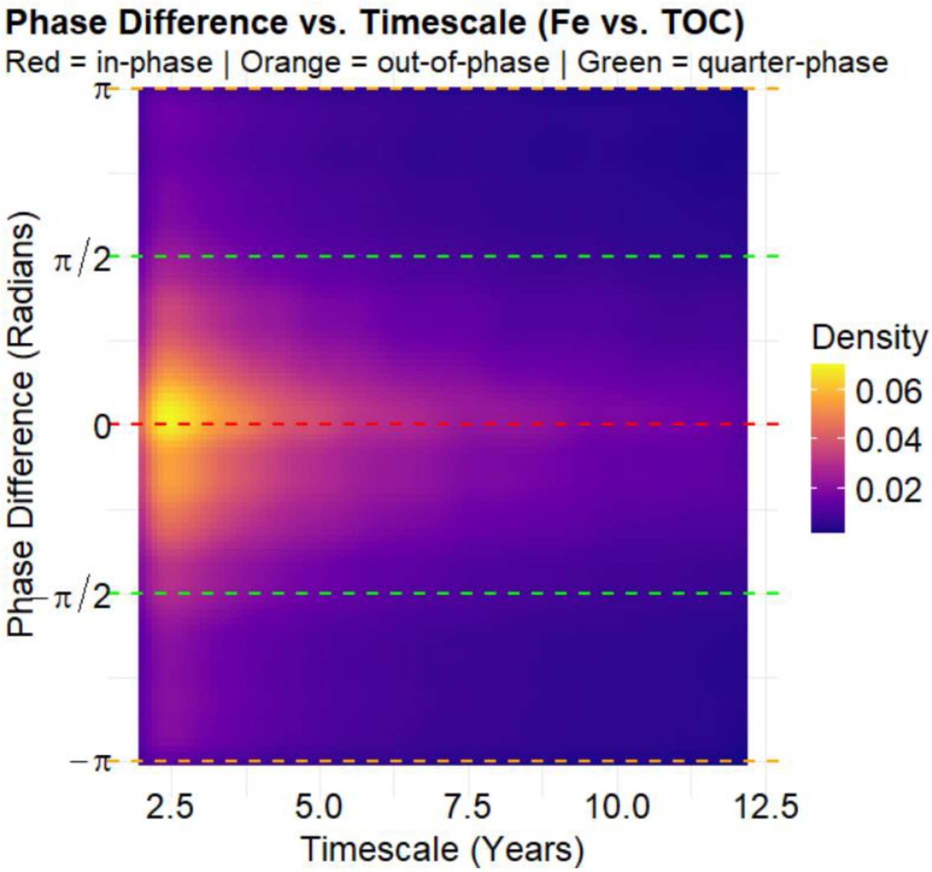

Figure 6. Wavelet coherence density plot showing the phase structure of TOC–Fe coherence across timescales for Fennoscandian lakes. Only significant coherence is shown ($p < 0.05$). The plot shows where coherence occurred most frequently across lakes and time. Density is the number of times a specific phase relationship occurred at a given timescale. The color gradient represents the density of phase angles, where warmer colors indicate higher density (i.e., more frequent phase relationships at a given timescale). A dense region (e.g., in the 2–4 years band) indicates that more lakes and more years had coherence there—not necessarily that the coherence was stronger. Horizontal dotted lines represent in-phase, out-of-phase and quarter phase relationships (see the title). 0 radians = perfectly in-phase; $\pm\pi$ ($\sim$3.14 or -3.14) = perfectly out-of-phase; and $\pm\pi/2$ ($\sim\pm1.57$) = 90° phase difference (quarter cycle lag). In this study we considered $\pm\pi/2$ (green dotted lines) as a boundary between in-phase and out-of-phase relationships. For details, see Section 2.

PRE over similar timescales (i.e., Fe had negative coherence with PRE). Out of 49 lakes with Fe–PRE coherence, 23 showed out-of-phase behavior, while 14 were in-phase. This suggests that increased PRE often resulted in lower Fe levels.

AT showed somewhat similar coherence patterns with both TOC and Fe (Figure 7), with more exhibiting in-phase relationships (19 for TOC, 22 for Fe) than out-of-phase (15 for TOC, 17 for Fe) relationships (Table 3). This indicates that warmer conditions tend to result in increased levels of TOC and Fe.

Fe and TOC responded differently to Sdep, showing coherence predominantly over 2–4 years. For TOC, this relationship was mostly out-of-phase (27 lakes with out-of-phase and 18 lakes with in-phase relationship), while for Fe, this relationship was mostly in-phase (16 lakes with out-of-phase and 30 lakes with in-phase relationship). This suggests that a temporal decrease in Sdep tended to result in an increase in TOC and a decrease in Fe levels.

Most of the lakes with mixed phase relationships were for TOC–AT and Fe-PRE coherence. This “mixed” category indicates a lack of a stable coupling pattern, which may reflect local lake-specific processes or nonlinear interactions not captured by dominant phase behavior.

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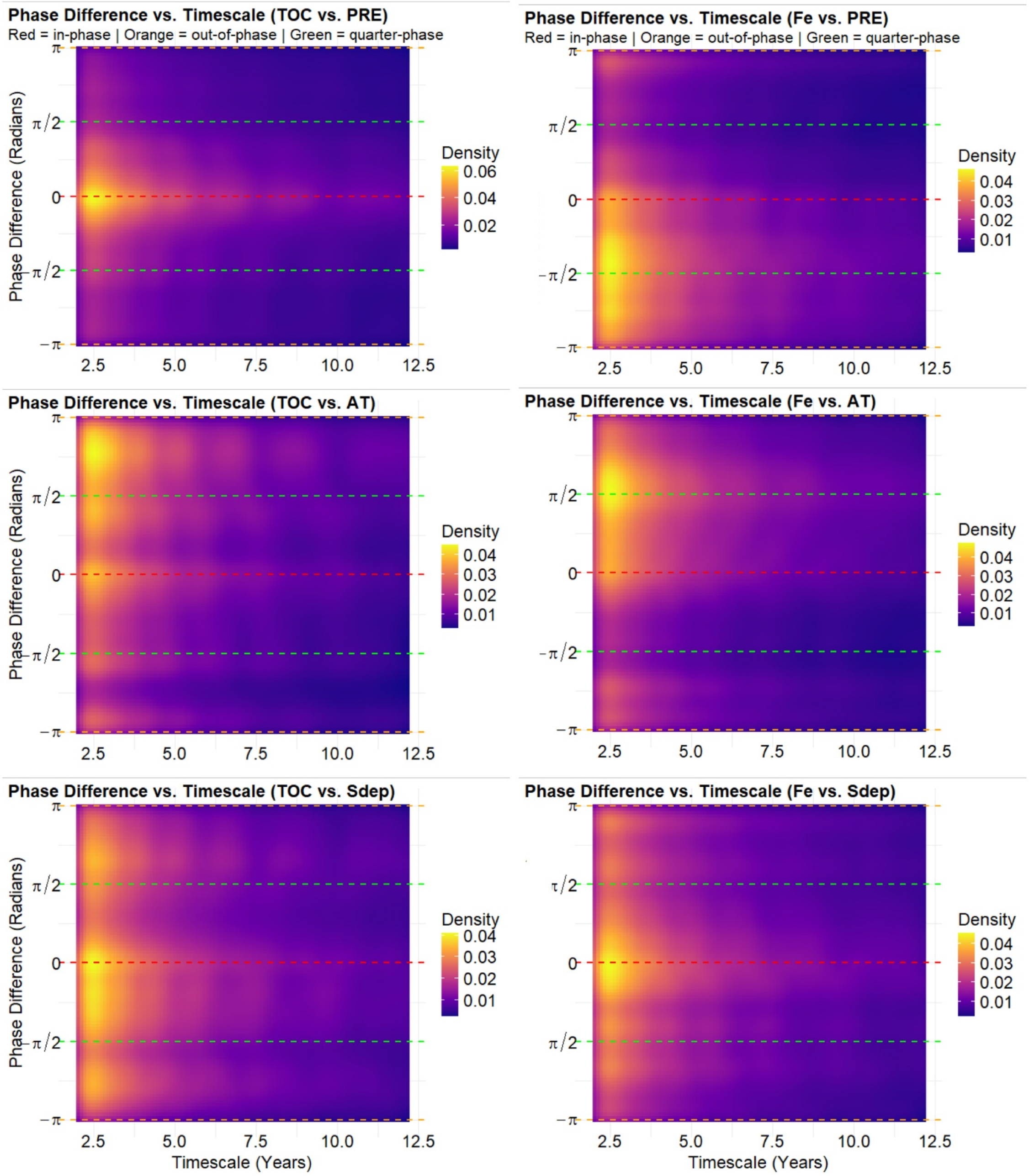

Figure 7. Wavelet coherence density plot showing phase structure of TOC and Fe coherence with environmental variables (PRE, AT, and Sdep) across timescales for Fennoscandian lakes. Only significant coherence is shown ($p < 0.05$). The plot shows where coherence occurred most frequently across lakes and time. Density is the number of times a specific phase relationship occurred at a given timescale. Guidance on interpreting the plot is provided in Figure 6 and in Section 2.

4. Discussion

In this study, we conducted comprehensive temporal analyses of TOC, Fe, climate variables and sulfur deposition (Sdep) across 102 Fennoscandian lakes with varying sizes and degrees of brownness. Our findings align with recent studies indicating that space-for-time substitution is inadequate for analyzing the effects of global change on aquatic ecosystems (Bergström et al., 2024; Paltsev et al., 2024; Stetler et al., 2021), particularly for long-term and cyclical (i.e., with periodic patterns) drivers such as rising air temperatures and changing precipitation patterns. We found that lake TOC and Fe concentrations in the study lakes are responsive to changes in temperature (AT), precipitation (PRE) and Sdep. This supports our first hypothesis that these environmental variables

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Table 3
Summary of the Results From the Wavelet Coherence Analysis Between TOC and Fe and Environmental Variables Across Fennoscandian Lakes

					Mean phase relationship (# lakes)		
	Parameter	Mean coherence	Mean timescale	# lakes with identified coherence	In phase	Out-of-phase	Mixed
TOC versus	PRE	0.48	4.0	51	30	10	11
	AT	0.56	4.2	47	19	15	13
	Sdep	0.44	3.5	54	18	27	9
Fe versus	TOC	0.49	4.0	50	25	16	9
	PRE	0.53	4.6	49	14	23	12
	AT	0.50	4.0	50	22	17	11
	Sdep	0.52	4.0	57	30	16	11

Note. Only lakes with coherence with $p < 0.05$ are shown. Means were calculated as mean values per lake (e.g., mean coherence is the average of all coherence magnitudes [between 0 and 1] per lake).

influence the temporal variability of TOC and Fe concentrations. The Bayesian model also revealed that AT and PRE had opposing effects on Fe when interacting, with negative PRE × AT and PRE × AT × Sdep interactions. This is consistent with the view that hydrological conditions can modify Fe export and emphasizes the need to consider interactions among environmental variables rather than individual effects. Additionally, the interaction of TOC with Sdep was negative, supporting the notion that TOC export is enhanced under conditions of declining Sdep (de Wit et al., 2021; Monteith et al., 2007), while Fe is more variable, depending on the combined hydrological and climatic context (Björnerås et al., 2017).

We further provided evidence that relatively short-timescale (between 2 and 4 years) variability in PRE led to the temporal decoupling of Fe from TOC in the study lakes, supporting our second hypothesis. This is the first study, to our knowledge, to analyze the nature of temporal correlations (trends vs. interannual fluctuations) between environmental variables and Fe cycling using wavelet coherence. While this approach does not per se reveal causality, it adds important temporal information not captured by other analyses and is very useful for generating and testing hypotheses on temporal versus spatial correlations.

4.1. Temporal Decoupling Between TOC and Fe

Previous studies on lake browning have often suggested a temporal correlation between TOC (or DOC/organic matter) and Fe concentrations in inland waters (e.g., Björnerås et al., 2017; Riise et al., 2023; Weyhenmeyer et al., 2014; Xiao & Riise, 2021). This is based on the observation that TOC and Fe generally correlate positively among boreal lakes, and the assumption that Fe entering lakes is primarily bound to OC-Fe complexes and FeOOH associated with OC (Heikkinen et al., 2022). Kritzberg and Ekström (2012) were among the first to observe that this relationship does not always hold true. Our study supports this observation, as lakes exhibiting positive slopes in TOC did not always show positive slopes in Fe, with this pattern persisting across lakes with different TOC concentrations (from clear with TOC < 5 mg L^{-1} to brown with TOC > 10 mg L^{-1}). This indicates that TOC and Fe do not always increase simultaneously, and in some lakes, they even exhibit opposite trends.

The proportion of lakes with positive slopes in Fe is lower than those with positive slopes in TOC, suggesting that lake browning in our study lakes may be driven mostly by TOC or by interactions between TOC and Fe. This interpretation aligns with findings from other studies (e.g., see review paper by Blanchet et al., 2022; Monteith et al., 2007). The intensity of browning also differed between lakes of different TOC concentrations (Figure 3). Brown lakes, for example, exhibit the highest Fe (median Fe = 542.8 μg L^{-1}) and TOC (median TOC = 12.8 mg L^{-1}) concentrations, and the highest rate of change in both Fe (median Sen's slope: 2.0 μg L^{-1} yr^{-1}; $p < 0.011$) and TOC (median Sen's slope: 0.16 mg L^{-1} yr^{-1}; $p < 0.0001$). Furthermore, 100% of brown lakes show positive TOC slopes, and they also have the highest proportion of lakes with positive Fe slopes (66%). These findings suggest that these brown lakes are becoming progressively browner.

TOC and Fe concentrations in the study lakes follow an elevation gradient, with clear lakes at the highest altitudes, brown lakes at the lowest, and medium-brown lakes in between, consistent with global patterns (Toming et al., 2020). TOC had a direct effect on Fe only in medium-brown and brown lakes (Table 2), suggesting that

TOC availability more strongly regulates Fe at lower elevations, where warmer, wetter climates, greater forest cover (Figure 4), and stronger catchment–lake connectivity enhance organic matter inputs. At higher elevations, clearer water and lower organic inputs may weaken this relationship statistically, while at lower elevations, the TOC signal is strong enough to directly influence Fe. However, this spatial gradient does not explain why many clear lakes occur at low altitudes or why temporal trends in Fe vary independently of lake type. This indicates that space-for-time substitution cannot always be used to predict temporal changes as complex interactions among variables may override even clear spatial climate patterns. Furthermore, internal Fe cycling could also be important—at moderate to high TOC concentrations, browning can enhance lake stratification, promote oxygen depletion, and trigger the release of Fe(II) from sediments, processes that are less likely in clear, frequently unstratified lakes in mountainous areas (Jansen et al., 2024; Lewis et al., 2023; Pilla et al., 2018; Puts et al., 2023; Solomon et al., 2015; Sorichetti et al., 2014).

4.2. Precipitation Drives Temporal Decoupling Between TOC and Fe

Climate factors (AT and PRE) have long been acknowledged for their role in controlling TOC and Fe levels in lakes and their catchments. While some studies have found no relationship between rising AT and changes in TOC and Fe concentrations in lakes (Erlandsson et al., 2008; Hall et al., 2021), our findings align with studies reporting such relationships (Asmala et al., 2019; Curtinrich et al., 2022; Kraemer et al., 2015; Räike et al., 2024; Williamson et al., 2015). Furthermore, we found that AT was the only variable in the "all-lake" and "clear lake" models that had individual effects on Fe levels, showing that an increase in Fe is associated with an increase in AT. Temperature changes may directly impact TOC exports from catchment soils by altering TOC and Fe leaching via increased organic matter decomposition and mineralization, which are both sensitive to variations in moisture and temperature (Feng et al., 2025; Räike et al., 2024). The response of Fe to AT in clear lakes may reflect that, in northwestern clear lakes, AT is a stronger driver of TOC and Fe export than in southern boreal areas, as cold soils limit microbial activity and decomposition. Even modest warming can enhance organic matter breakdown, while changes in snowpack and snowmelt timing intensify flushing of soil-derived TOC and Fe (Campbell & Hjalmar, 2019; Hirst et al., 2022; Jansson et al., 2008; Rawlins & Karmalkar, 2024). In contrast, southern catchments are more influenced by rainfall, making PRE the dominant control. Nevertheless, rainfall (PRE in this study) still influences clear lakes through its interaction with AT and TOC, where wet and warm conditions combined with high TOC reduce Fe concentrations. A positive relationship between AT and Fe would also be consistent with stronger and longer stratification, greater hypoxia, and enhanced internal Fe release (Jansen et al., 2024; Orihel et al., 2015; Sorichetti et al., 2014).

Since six study lakes were in permafrost regions, permafrost thaw may have influenced TOC and OC–Fe export from organic soils. However, because these areas are characterized by sporadic to isolated permafrost (Åkerman & Johansson, 2008), impacts on OC supply are likely weaker than in continuous permafrost landscapes (e.g., in Alaska or Siberia) (Puts et al., 2022; Vonk et al., 2015). This interpretation is supported by the fact that in all six lakes, TOC concentrations were declining, with significant DOC declines observed in two of them.

PRE influences the transport of organic matter and the elements bound to it into lakes (Mengistu, Quick, & Creed, 2013), as well as the fate of these elements within lakes (Creed et al., 2018; Fowler et al., 2020; Weyhenmeyer et al., 2016). The general consensus is that TOC export to lakes increases with rising PRE and associated runoff (Blanchet et al., 2022; de Wit et al., 2021; Imtiazy et al., 2025). Our study supports this, showing that both TOC and Fe responded to PRE in study lakes (Table 2 and Table S3 in Supporting Information S1; Figure 5). However, the relationship between Fe and PRE was more complex than that between TOC and PRE. While TOC responded to the individual effect of PRE indicating that increased TOC is associated with higher PRE, Fe responded to the interaction effect between PRE and AT and between PRE, AT, and Sdep. The PRE × AT interaction had a negative impact on Fe, showing that the Fe concentration decreased under conditions of simultaneously high PRE and elevated AT. This finding contradicts with studies that report a positive relationship between Fe and PRE (Björnerås et al., 2017; Hongve et al., 2004) but explains why Fe had negative slopes in nearly half of the study lakes. Furthermore, a recent study by Škerlep et al. (2023) observed that drought conditions led to a prolonged release of Fe from organic soils into Swedish streams. This suggests that the combined effects of AT and PRE, especially during extreme weather events, may substantially influence Fe cycling in unexpected ways.

In agreement with previous studies (Björnerås et al., 2017; de Wit et al., 2021; Monteith et al., 2007), our results also indicate a link between Sdep and Fe. However, Sdep is among the primary individual drivers of Fe change only in brown lakes. Many brown lakes are located in regions that historically received high Sdep (Andersson et al., 2018), but these areas have also experienced the strongest declines in Sdep in recent decades (Aas et al., 2019; Laudon et al., 2021). Long-term declines in Sdep may have contrasting effects on organic matter and Fe export from catchments to lakes. Recovery from acidification enhances the solubility and mobility of TOC, leading to increased TOC export (Meyer-Jacob et al., 2019; Monteith et al., 2007) and contributing to ongoing lake browning of brown lakes (hence, all brown lakes showed positive slopes in TOC; Figure 1d). In contrast, reduced acidity might lower Fe mobilization from soils, and in regions with historically high Fe exports where catchment Fe pools may already be relatively depleted. Increased PRE appears to also dilute Fe concentrations in brown lakes (Table 2). Thus, while TOC concentration often rises, Fe concentration can decline despite similar drivers, which is consistent with the 34% of brown lakes showing negative slopes in Fe. Our results show that the negative three-way interaction PRE × AT × Sdep on Fe in the "all-lake model" was similar in magnitude to the PRE × AT interaction. Additionally, other interaction terms involving Sdep (e.g., TOC × Sdep) were either not statistically significant or were associated with an increase in Fe levels. This suggests that while Sdep may influence Fe levels, its effect is likely secondary or modulated by PRE and AT.

Examination of Table 2 reveals that all negative interactions between Fe and environmental variables involve PRE. This suggests that PRE is the primary contributor to the observed decline in Fe levels, in contrast to AT, which was associated with increased Fe levels. The relationship between Fe and the PRE × AT interaction indicates a complex response of Fe to co-occurring environmental changes, potentially reflecting multiple underlying mechanisms (and at both temporal and spatial domains—e.g., catchment properties). For example, under warmer and wetter conditions, catchments may experience increased runoff and groundwater inflow, which can flush Fe from soils into lakes (Hongve et al., 2004; Weyhenmeyer et al., 2014). PRE can both mobilize (after dry periods) and dilute element concentrations (including substances such as FeOOH but also OC-Fe complexes) (Abhishek et al., 2024; Paltsev & Creed, 2022). Lakes with negative slopes in Fe are larger than lakes with positive slopes in Fe (Table 1). These lakes also have a higher catchment-to-lake ratio (which translates to shorter water residence time; Köhler et al., 2013; Riise et al., 2023) than lakes with positive slopes in Fe. Higher amounts of PRE, coupled with greater dilution and faster turnover rates, likely contribute to the declining Fe trends observed in the study lakes. On the other hand, shorter water residence times have been associated with higher Fe concentrations in the water column, likely because rapid flushing reduces the time available for Fe to settle into the sediments (Weyhenmeyer et al., 2014). However, this observation was made on spatial data (long-term means of Fe, TOC and climate variables) involving spatial correlations among lakes, not temporal analysis (long-term trends or interannual variability), indicating that the efficiency of standing waters as Fe sinks (Jonsson, 1997) may still hold true over time (i.e., when considering time series data). Moreover, the negative two-way interaction PRE × AT may also reflect nonlinear biogeochemical feedbacks on Fe, where warming-induced increases in microbial activity alter Fe cycling in sediments (e.g., enhancing reductive dissolution of Fe(III) and, where sufficient sulfate is available, subsequent burial of Fe as sulfides) (Kritzberg & Ekström, 2012; Melton et al., 2014).

Changes in forest and wetland cover have also been identified as important drivers of TOC and Fe export, which are generally linked to increased Fe levels (Björnerås et al., 2017; Blanchet et al., 2022; Ekström et al., 2016; Senar et al., 2018). Indeed, our results show that lakes with positive slopes in Fe had a relatively higher forest cover in their catchments (mean = 79.1%) than the lakes with negative slopes (mean = 65.5%; Table 1), and brown lakes had the highest forest cover across lake types (Figure 4). However, lakes with positive Fe slopes had a relatively lower proportion of wetlands in their catchments (mean = 2.2%) compared with lakes with negative Fe slopes (mean = 7.6%). Furthermore, wetland proportion did not vary across lake types (i.e., clear, medium-brown, and brown lakes all had ~4% wetlands on average). This pattern contradicts studies showing that wetlands generally act as major sources of TOC and Fe, with wetland-rich catchments contributing greater Fe influx to lakes (Creed et al., 2018; Senar et al., 2018). In our systems, however, Fe (and TOC) influx may be more strongly influenced by forested areas in the catchment (e.g., northern clear lakes with little forest vs. southern lakes with extensive forest cover) and by the catchment-to-lake area ratio, rather than by wetlands.

Clearcutting—more common in southern Sweden and Finland (south of 61°N)—can increase Fe and TOC through soil disturbance and altered water flow. Indeed, Fe concentrations are higher in the southern Sweden and Finland (506 vs. 370 μg L^{-1} in the north), and 77% of lakes with increasing Fe trends are located there. At the same time, other forestry-related processes, such as afforestation and forest regrowth, have also been linked to higher area-specific Fe export (Kritzberg, 2017; Sarkkola et al., 2013). Taken together, this highlights the complexity of Fe cycling, which does not always follow linear or unidirectional trends with climate or other anthropogenic disturbances.

4.3. Is the Decoupling Between TOC and Fe Primarily Driven by Long-Term Trends in Precipitation, or by Its Interannual Fluctuations?

In the previous section we discussed our finding that an interaction among PRE and AT (and Sdep) were associated with declines in Fe levels. Since AT increased significantly in nearly half of the study lakes ($n = 46$; mean rate of 0.04°C yr^{-1}, $p < 0.05$), and our Bayesian model showed that AT alone contributed to an increase in Fe concentration, we conclude that *long-term trends* in AT are likely contributing to rising Fe levels over time. However, compared to AT and Sdep, PRE exhibited both positive and negative rates of change (slopes) over the 30-year period, with significant increases observed in only three lakes. Hence, we cannot conclude that *long-term trends* in PRE are responsible for the decline in Fe levels and the temporal decoupling of Fe from TOC in study lakes. This raises an important question: if not long-term trends, what is the nature of the relationship between Fe and PRE—is it primarily driven by interannual fluctuations in PRE?

The wavelet coherence analysis showed that both TOC and Fe had interannual fluctuations that were coherent with fluctuations in PRE, AT and Sdep across various timescales, with a dominant oscillation pattern around 2–4 years. However, compared to TOC, which showed mostly positive coherence with climate variables, Fe exhibited more complex coherence patterns with these variables. This was especially evident for the Fe versus PRE relationship where Fe both increased and decreased with increasing PRE; however, a negative coherence—Fe decreased with increasing PRE—was dominant. This supports the idea that PRE plays an important role in decoupling Fe from TOC through short-timescale fluctuations rather than by a long-term unidirectional trend.

Increases in Fe levels were found to be frequently linked to increases in Sdep over the same 2–4-year period (in-phase relationship), suggesting that Sdep—similar to AT—contributes to Fe increase. Because our Sdep data are derived from wet deposition, they can coexist with PRE, making it difficult to disentangle their individual effects. However, the differing phase relationships between Fe versus PRE and Fe versus Sdep coherence demonstrate that PRE and Sdep each cause independent time-scale-specific influences on Fe concentration.

Strong coherence of TOC and Fe with environmental variables over short periods (2–4 years) likely reflects quasi-periodic fluctuations in climate and weather patterns. These fluctuations are governed by climate teleconnections, especially those with short periodicities described specifically for Fennoscandia such as EAWR and SCA (Ionita, 2014; Isles et al., 2023; Wang & Tan, 2020). The weaker coherence at longer timescales (e.g., 8–10 years) suggests that slow-moving drivers (e.g., long-term climate trends, or legacy effects from past deposition) are less synchronously aligned with changes in Fe and TOC. However, since the heatmaps do not show the magnitude of coherence—but rather indicate where coherence occurred most frequently across all lakes—it is possible that coherence is stronger at longer timescales than at shorter ones, even if this occurs in fewer lakes. In this study, our aim was to investigate the presence of interannual fluctuations in environmental variables on TOC and Fe levels. Therefore, quantifying the magnitude of coherence at different timescales was beyond the scope of this research.

5. Conclusions

Global change influences lake ecosystems directly and indirectly via changes in nutrient export to lakes and nutrient and mineral availability in lakes. Previous studies reported widespread increases in Fe trends for northern boreal and temperate lakes, which were associated with simultaneous increases in TOC. However, Fe in nearly a third of the Fennoscandian lakes in this study was found to be decoupling from TOC, with Fe concentrations declining while TOC continued to increase; both significant and non-significant patterns were included. This decoupling appeared to be driven by short-timescale interannual fluctuation (2–4 years) in precipitation, together with long-term temperature trends toward a warmer and wetter climate. Despite many previous studies showing a positive association between increasing precipitation and Fe, these short-timescale fluctuations indicate that an increase in precipitation can instead result in a decrease in Fe. Notably, the decoupled lakes were also large with high catchment-to-lake ratios and short water residence times, which may have made them more sensitive to these high-frequency precipitation changes. Thus, natural variability in lake types and their responses to climate forcing are key factors contributing to the decoupling of TOC and total Fe over time.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Gridded, interpolated estimates of wet sulfur deposition were downloaded from the European Monitoring and Evaluation Programme (Amann et al., 2018; Colette et al., 2016) and are available at https://www.emep.int. Data on air temperature and precipitation were extracted from the "TerraClimate data set" (Abatzoglou et al., 2018) available at https://www.climatologylab.org. Data on water chemistry and lake/catchment parameters were obtained from Swedish and Finnish lake monitoring programs (Fölster et al., 2014; Miljodata-MVM, 2021; Vuorenmaa et al., 2014). Data on water chemistry from the Swedish lake monitoring program are also available at https://miljodata.slu.se/mvm. The raw and processed data used for the statistical analysis and the preparation of figures in the study are available at Paltsev and Bergström (2025). Statistical analysis was conducted in R version 4.2.2 (R Core Team, 2022), available at https://www.r-project.org. Figures were also made in R. Maps were created using ArcGIS Pro version 3.0.0, licensed for Umeå University and available at https://www.arcgis.com.

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