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Key Points:

- Reduced air pollution is the primary driver of lake chemical change, while climate-induced changes on element cycling are of secondary importance
- Strong lake chemical recovery from acidification is demonstrated by reduced SO_4 and labile Al and increases in pH, alkalinity, and acid neutralizing capacity
- Lake display enhanced light- and nutrient-limitation (higher DOM and lower NO_3), while Ca and SiO_2 increased unexpectedly

Supporting Information:

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

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Changing Water Chemistry in One Thousand Norwegian Lakes During Three Decades of Cleaner Air and Climate Change

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Abstract We present long-term changes in Norwegian lake water quality across regional gradients in atmospheric pollution, air temperature, hydrology, and vegetation using (a) a national representative lake survey carried out in 1995 and 2019 (ThousandLakes), and (b) an annual lake survey from acid-sensitive catchments (78 lakes, TrendLakes) from 1990 to 2020. Our analysis encompasses all major chemical constituents, for example, anions and cations, dissolved organic matter (DOM), nutrients, iron (Fe), and silicate (SiO_2). During these decades, environmental changes included declines in sulfur (S) and nitrogen (N) deposition, climate warming, and increase in forest biomass. Strong chemical recovery from acidification is found, attributed to large reductions in atmospheric deposition, moderated by catchment processing from land use and climate change. Browning counteracted chemical recovery in some regions, while Ca increased unexpectedly. We suggest that increased weathering, from enhanced terrestrial productivity, is an important driver of increased Ca—substantiated by widespread, substantial increases in SiO_2. Light- and nutrient-limitation has become more prevalent, indicated by higher DOM, lower nitrate (NO_3), and lower $NO_3$to total phosphorous ratios. Declines in lake NO_3 occurred independently of N deposition, suggesting increased catchment N retention, possibly from increased terrestrial productivity. We conclude that decreased air pollution continues to be a dominant driver of long-term trends in lake chemistry, but climate-induced increase in terrestrial weathering processes, governed by increased biomass, is likely to have an increasing impact on future lake acidity, nutrient, and light status, that may cascade along the aquatic continuum from rivers to the coast.

Plain Language Summary Lakes in northern landscapes are sensitive to air pollution and climate change. Here, we investigate how lake water chemistry in a thousand Norwegian lakes has changed since the 1990s, using two large lake monitoring studies from the whole of Norway. Air pollution, climate change, catchment, and lake properties in these data sets are representative of large regions of Europe and North America. We found strong chemical recovery related to reduced atmospheric deposition of sulfur. We also found lower nutrient concentrations due to reduced deposition of nitrogen and increased catchment nutrient retention. The lakes have become less acidic and more nutrient- and light-limited. Unexpectedly, we saw widespread increases in calcium and silicate, which may be linked to increased terrestrial productivity under climate change. Overall, we conclude that air pollution continues to impact boreal lake water quality in addition to climate-induced changes in terrestrial biogeochemistry. We predict that changes in lake chemistry will continue in the future and will cascade downstream to rivers and coastal areas, with potentially large biological impacts.

1. Introduction

Lake monitoring programs from northern landscapes have, since the 1970s, provided invaluable evidence of large-scale environmental changes such as acidification (Schindler, 1988), chemical recovery from acidification (Stoddard et al., 1999), browning (Monteith et al., 2007), and nutrient and light-limitation (Creed et al., 2018). Monitoring data is also crucial for developing and testing hypotheses, for instance for the drivers of acidification, for example, Krug and Frink (1983), Schindler (1988), and VanBreemen et al. (1984) and browning (Evans et al., 2006; Monteith et al., 2007).

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Changes in water quality, first demonstrated in remote small headwater lakes, can cascade downstream along the entire aquatic continuum. For example, coastal darkening (Aksnes et al., 2009) is attributed to freshwater browning (Deininger et al., 2020; Raike et al., 2012) while reduced catchment nitrogen (N) leaching from declines in N deposition helped to alleviate coastal eutrophication (Burns et al., 2021).

Surface water acidification, related to anthropogenic emissions of sulfur (S) and N to the atmosphere for over a century (Grennfelt et al., 2020), has been extensively documented in the northern countries (Henriksen et al., 1998) and elsewhere in Europe and North America (Stoddard et al., 1999). The detrimental effects of acid deposition, especially S, are related to the mobilization of toxic inorganic aluminum (Al) in acidic soils (Van Breemen et al., 1984) with subsequent runoff to surface waters (Christophersen et al., 1990; Driscoll et al., 1980) and the precipitation of Al oxides on fish gills leading to hypoxia (Baker & Schofield, 1982). Regions with slowly weatherable bedrock and soils with low buffering capacity in northern Europe and North America were particularly sensitive (Hettelingh et al., 2007; McDonnell et al., 2014).

Currently, lakes and rivers in acid-sensitive regions show strong chemical recovery (Garmo et al., 2014), an expected response to reductions in S deposition because of the principle of electroneutrality and cation exchange (Reuss & Johnson, 1985); although chemical recovery is not always accompanied by biological recovery (Angeler & Johnson, 2012). Impacts of N deposition on lake water quality are less obvious, although it is accepted that N deposition promotes surface water acidification by enhanced leaching of nitrate (NO_3) (Henriksen & Brakke, 1988; Mitchell et al., 1996; Wright et al., 2001). Widespread browning of lakes, due to increased concentrations of Dissolved Organic Matter (DOM), has been an unexpected effect of cleaner air (Monteith et al., 2007).

Recent evidence suggests that in addition to atmospheric S and N deposition, climate and land use change are becoming increasingly important drivers of changes in boreal lake chemistry: lake browning (De Wit et al., 2016) and nutrient concentrations (N, phosphorus (P)) appear to be controlled by air pollution, land use, and climate (De Wit et al., 2020; Fölster et al., 2014; Vuorenmaa et al., 2018). Afforestation and increased terrestrial productivity have been suggested as a driver of browning in an 80-year record of DOC (Skerlep et al., 2020). Finstad et al. (2016) attributed DOM trends in Norwegian lakes to increased terrestrial productivity and reduced S deposition in equal weight. Increases in DOM are suggested as an explanation for the observed increases in surface water iron (Fe) in Europe and North America (Björnerås et al., 2017). Björnerås et al. (2017) used covariation between Fe and silicate (SiO_2) as support for the role of climate-driven hydrological change for DOM trends, and thus Fe trends.

Norway is exceptionally well-suited for investigating drivers of long-term change in water chemistry in northern regions because of its large latitudinal, longitudinal, and elevational gradients in climate, atmospheric deposition, land cover, and primary productivity and its regional variation in rates of warming, declines in anthropogenic deposition, and increases in forest growth which mirror environmental change elsewhere. Over the last few decades, Norway has experienced substantial environmental change. While S deposition in Norway has been reduced to 10%–30% of the deposition levels during the 1980s, the deposition of N has declined less (Aas et al., 2021; Colette et al., 2016). Most N deposition is retained in soil and vegetation (Campbell et al., 2004), possibly promoting forest productivity (Thomas et al., 2013). Large-scale increases in forest biomass found in Norway (De Wit et al., 2015) and elsewhere in Europe (Luyssaert et al., 2010) in turn may increase nutrient demand and reduce nutrient runoff. Atmospheric warming in Norway, estimated at 0.5°C decade^{-1} since the 1970s (Hanssen-Bauer et al., 2017), exceeds global warming for the same period by ca 1.5°C (IPCC, 2022). The growing season has become significantly longer in Norway (Hanssen-Bauer et al., 2017) and elsewhere in northern Europe (Aalto et al., 2022). Precipitation in Norway has increased by 18% since the late 1970s, leading to increased seasonal and annual discharge but with considerable regional and seasonal variation (Hanssen-Bauer et al., 2017).

To understand regional-to-national scale lake responses to environmental change, which typically occurs over decadal timescales, monitoring data is needed which covers both broad spatial scales and long time periods. Here, we assess the long-term changes in water chemistry based on two Norwegian lake surveys for the period 1995–2020, presenting a unique combination of a repeated, spatially extensive lake survey (i.e., "ThousandLakes") and a long-term annual lake survey (i.e., "TrendLakes") (78 lakes) in the context of large-scale environmental change. The annual lake survey is targeted to monitor responses to air pollution and includes therefore largely lakes from acid-sensitive regions in Norway (Dillon et al., 2003; Skjelkvale et al., 1998) with records starting during the 1980s. The nationally representative ThousandLake survey has provided the acid-base status of lakes in 1995 (Henriksen et al., 1998) and was repeated in 2019 (Hindar et al., 2020). Both surveys followed similar sampling and analytical procedures, resulting in two high-quality data sets which can be compared unhampered by methodological challenges.

We focus on changes and trends in acid-base chemistry and aluminum fractions, major nutrients, DOM, SiO_2, Fe, and major element ratios. We grouped the lakes into six ecoregions (Dillon et al., 2003) based on similarities in air pollution loading, acid-sensitivity, land cover, and climate and then tested the following hypotheses:

1. The changes between 1995 and 2019 in the ThousandLakes are consistent with the long-term trends in the TrendLakes and therefore represent wide-scale temporal trends rather than random interannual variation.
2. Regional variation in changes in lake chemistry is driven by changes in atmospheric deposition and by gradients of land cover and climate. In particular, we hypothesize that (a) chemical recovery is highest in regions where acidification in 1995 was the most distinct and reduction in S deposition are highest; (b) changes in lake NO_3 reflect the level and change in N deposition, while changes are moderated by forest cover; (c) changes in base cations follow changes in Strong Acid Anions (SAA); (d) changes in SAA, not in organic acidity, explain changes in recovery; (e) organically bound nutrients and complexed elements follow trends in DOM; (f) changes in SiO_2 are related to hydrology as groundwater is enriched in SiO_2 from weathering (Sutton et al., 2018), implying that SiO_2 covaries with hydrological flowpaths (Webster et al., 2021).

2. Materials and Methods

2.1. ThousandLakes Data Set

The survey in 2019 (1,001 lakes) included 983 of the lakes sampled in the 1995 survey (1,006 lakes), which is about 2.5% of all lakes in Norway with a surface area larger than 0.04 km^2 (Hindar et al., 2020) (Figure 1). The lakes in the 1995-survey, included in a Nordic lake survey (Henriksen et al., 1998), were selected using stratified random selection criteria designed to be a regionally representative selection of lakes in Norway. The 1995-survey had a distribution of 3:2:1 of lakes from southern, middle, and northern Norway, respectively, because environmental pressures (in particular acidification and eutrophication) decrease along a south to north gradient. Lakes that were limed, regulated, shallower than 1-m depth or an extension of a river were not selected. Eleven lakes from the 1995-survey were limed or regulated in 2019 and were replaced with nearby lakes with similar characteristics. The lakes were sampled from five size classes (0.04–0.1, 0.1–1, 1–10, 10–100, and >100 km^2), where 79% of the lakes were smaller than 1 km^2 and seven lakes were larger than 100 km^2. Five lakes that are strongly impacted by point nutrient sources (i.e., over 50% agriculture and/or PO_4 concentrations >30 $\mu g\ L^{-1}$ in 2019) were removed, leaving 978 lakes for further analysis.

Sampling of 975 lakes in 2019 was done from a helicopter and the remaining lakes were visited on foot. Sampling took place during or shortly after the autumn overturn, following the procedures from the 1995-survey (Hindar et al., 2020), and was done between day 260 and day 330 in 2019 and between day 252 and 340 in 1995 (Figure S1 in Supporting Information S1). Surface water temperature and lake depth were measured at each sampling point. Sampling started in the north and proceeded southward, where mountain lakes south of 64°N were visited earlier than lowland lakes. Lake temperatures in 2019 (measured) were on average 0.7°C lower than in 1995 (estimated) (Table S1 in Supporting Information S1), consistent with the comparison of 1-month antecedent air temperatures (Table S2 in Supporting Information S1). Lake temperature for the ThousandLakes in 1995 was estimated with Gaussian Process regression which was trained on lake temperature measured in 2019 ($n = 961$; $R^2 = 0.84$; RMSE = 0.89°C) in the MATLAB Regression Learner App with 5-fold cross-validation. Predictors included average air temperature at the lake outlet over the week prior to sampling, lake altitude and lake depth. Precipitation during the antecedent month was on average 16 mm higher for the entire country in 2019 than in 1995, except for inland lakes in the far west and south of Norway.

We tested whether sampling in each region was conducted shortly after or during autumnal lake turnover in both years by checking if average lake surface temperatures (Figure S2 in Supporting Information S1) were at or below 8°C. At this temperature, similar lakes in Europe and North America (Shatwell et al., 2019) as well as a well-studied lake in Norway (De Wit et al., 2018) have completed autumn turnover. In each region, averaged lake temperatures were indeed below 8°C.

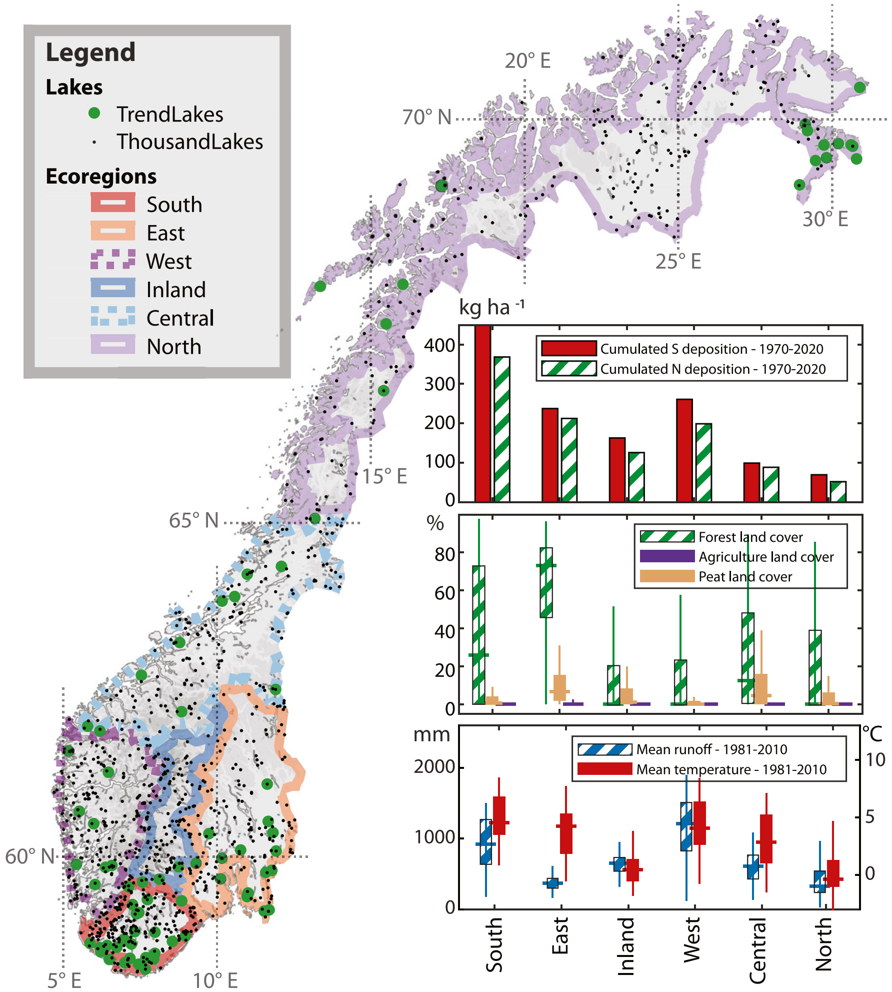

Figure 1. The location of the ThousandLakes (black dots) and TrendLakes (green dots) in six ecoregions in Norway, with cumulative S and N deposition for 1970–2020 as well as box- and whisker-plots of land cover (%forest, %peatlands, and %agriculture) and climate (ERA5 mean runoff and air temperature over 1981–2010) by region.

2.2. TrendLakes Data Set

The 78 lakes (TrendLakes), monitored annually to document responses to changes in the deposition of air pollution (Garmo & Skancke, 2020), are a subset of a survey of 1,005 lakes in 1986 (Henriksen et al., 1988) (Figure 1). All lakes are acid-sensitive headwaters in catchments dominated by granitic or gneissic bedrock, that is, poorly weatherable minerals low in base cations. Contrary to the ThousandLakes, low-elevation catchments with arable land on marine clays (i.e., below the marine limit, which is the highest Holocene shoreline which was elevated after the post-glacial rebound) are not included. The geographical distribution of the TrendLakes is skewed toward areas receiving relatively high loads of air pollution. The overlap between the TrendLakes and the ThousandLakes is limited to six lakes. Water sampling is done after the autumn circulation by local staff and sent to NIVA by mail and is received within 2–3 days after sampling.

2.3. Variables and Analytical Methods

The samples from both ThousandLake surveys were analyzed by NIVA's laboratory shortly after collection. There were some slight differences in methodology and limits of quantification (LOQs) (Table S3 in Supporting Information S1) between the 2 years but for most variables, the LOQs were well below the lake concentrations. Where this was not the case (SiO_2, 5 lakes; Fe, 345 lakes; and K, 15 lakes), we set any concentrations in 2019 that were below the 1995 LOQ at the LOQ for 1995. For the TrendLakes surveys, the same analytical methods were followed as for the ThousandLake surveys.

In 2019, the water samples were analyzed for pH (potentiometry), conductivity, major anions (SO_4, Cl) (ion chromatography), major cations (Ca, Mg, K, Na) (ion chromatography), alkalinity (Alk, acidimetric titration to a single endpoint at pH 4.5; the carbonate equivalence alkalinity (Alk-E), that is, the alkalinity corresponding to the pH where $[H^+]$ equals $[HCO_3^-]$, was estimated as proposed by (Henriksen, 1982)), TOC, DOC (filtered by 0.45 μm) (removal of DIC by acidifying and purging, post-combustion, IR-detection of CO_2), Al fractions (reactive Al, non-labile Al; labile Al calculated as reactive minus non-labile Al (ion exchange, spectrophotometry)), inorganic N species (NO_3 (nitrate + nitrite), NH_4), total N (totN) (spectrophotometry), total P (totP) (digestion with peroxydisulfate, spectrophotometry), Fe (ICP-MS), and SiO_2 (spectrophotometry). In 1995, the water chemical components listed above were analyzed using the same analytical procedures, except for major cations (ICP-OES), NH_4 (only 145 lakes in 1995), NO_3 (ion chromatography in 1995), and Fe (748 lakes only) (Hindar et al., 2020). All analytical work was carried out by NIVA laboratories with extensive Quality Assurance/Quality Control (QA/QC) routines (Hindar et al., 2020). Additional QA was done by flagging and reanalyzing samples from the 2019 survey that deviated markedly from the 1995 survey to a degree that was not compatible with temporal trends in TrendLakes.

Alkalinity is constituted by the "bases of weak acids," that is, HCO_3^-, NH_3, $Al(OH)_n^{3-n}$ (where *n* is 1–4), $Fe(OH)_n^{3-n}$ (where *n* is 1–3), as well as natural organic anions. Due to the limited pH range (4.4–8). In Norwegian surface waters, this is mainly HCO_3^-, $Al(OH)_n^{3-n}$, and natural organic anions. The following variables were calculated: Acid Neutralizing Capacity (ANC, sum of base cations minus sum of strong anions, in equivalents ($[Ca^{2+}] + [Mg^{2+}] + [Na^+] + [K^+]$) - ($[Cl^-] + [SO_4^{2-}] + [NO_3^-]$)) (Reuss & Johnson, 1985), organic acid adjusted ANC (ANC_{oaa}, by subtracting TOC*(10.2/3) from ANC, assuming a fixed charge density of 10.2 μeq^{-1}/mg C, and dividing by 3 assuming 1/3 of the charge density representing strong acid anions in line with a triprotic model (Lydersen et al., 2004)), total organic N (TON i.e., totN-NO_3), and CN of DOM (TON/TOC; w/w)). ANC is used as an indication of the acidification status of surface waters and is a key indicator in critical load calculations (Henriksen & Posch, 2001).

2.4. Catchment Characteristics

Catchments were delineated based on a 10 × 10 m digital elevation model (DEM) for Norway (https://www.geonorge.no/, DTM 10 Terrengmodell (UTM33)). Briefly, the coordinates of the sampling location were used to select lake polygons from the Norwegian Water Resources and Energy Directorate (NVE) lake database. The location of the outlet was located by intersecting these polygons with the NVE river network (https://nve.no/map-services/). The DEM was then processed with the TauDEM package (version 5) (Tarboton, 1997) to delineate the watershed of each lake outlet point. The resulting catchment polygons were used to extract catchment properties (area, elevation (mean, minimum, and maximum)) and land cover (Norwegian Mapping Authority, https://www.kartverket.no/en; AR50 data)). Lake area was available from the NVE lake database.

2.5. Climate, Discharge, Deposition, NDVI Data

Historical total atmospheric deposition of S and N for Norway until 2015 (Schopp et al., 2003) and projections under the Current Legislation scenario until 2020 (Amann et al., 2018) were obtained from the European Monitoring and Evaluation Programme (EMEP). The deposition is based on emission inventories and do not include S from sea salts. The data are given in steps of 5 years between 1880 and 2019 on a 0.50° × 0.25° longitude-latitude grid. We averaged the 1990–1994 and 1995–1999 intervals to calculate a representative value for the year 1995. For 2019, the interval 2015–2019 was used. Additionally, national data on total S (sea-salt corrected) and N deposition data for 1992–1996 (representative for 1995) and 2012–2016 (representative for 2019; 2017–2021 were not yet available) were obtained from the Norwegian Institute for Air Research (NILU) (Aas et al., 2017).

Table 1
Mean Annual Temperature (MAT), Precipitation (MAP), Discharge, and Total Deposition of N and S in 1995 and 2019 for the ThousandLakes

		Norway				Delta 1995–2019					
	Units	1995	2019	Delta	p	South	East	Inland	West	Central	North
MAT	°C	2.6	3.5	0.9	<0.0001	0.6	0.9	1.1	1.4	1.0	0.5
MAP	mm	1406	1479	73	<0.0001	214	239	162	−52	−41	−66
Q	mm	647	653	5	0.3763	123	70	−43	−104	−37	−21
N deposition[a]	kg/ha/yr	3.9	2.4	−1.5	<0.0001	−3.0	−1.9	−0.8	−2.0	−0.6	−0.4
S deposition[a]		3.8	1.2	−2.7	<0.0001	−5.5	−2.8	−1.5	−4.4	−0.9	−0.6
NDVI_JJA	%/decade			0.6	<0.0001	0.0	0.7	0.4	0.1	1.5	0.7

Note. Change in summer (June–August) NDVI_JJA for 1991–2015 (median sen-slope). Annual values presented for Norway and median delta values (1995–2019) presented for ecoregions. Statistical differences for Norway tested with pairwise comparison, except change in NDVI (significant if different from 0 (one-sample *t*-test at a significance level of 0.05)).
[a] Source: EMEP, 1995 based on averaged 5-year intervals 1990–1994 and 1995–1999; 2019 based on average 2015–2019.

Wet deposition is based on measured concentrations in precipitation and precipitation amounts, while dry deposition is estimated from measured air concentrations and seasonal deposition velocities, both interpolated to a 50 km× 50 km grid and further transferred to a 0.25° × 0.125° longitude-latitude grid.

Climate data (average daily air temperature and precipitation) were extracted from the Nordic Gridded Climate Data set (NGCD) type 2 (1 km^2) (Lussana, Saloranta, et al., 2018; Lussana, Tveito, et al., 2018), available through the MET Norway Thredds Service (https://thredds.met.no/thredds/catalog/ngcd/catalog.html) and area-averaged over each catchment. For each catchment, antecedent weather conditions (average temperature and summed precipitation) for time periods prior to each sampling date (1, 2, 7, 14, 28, 90, 180, and 365 days) were calculated for 1995 and 2019.

Annual runoff (based on ERA5 monthly averaged data) was downloaded for 1995 and 2019 from Copernicus on a 0.25° × 0.25° grid and values were extracted for the grid cell containing each sampling location (Hersbach et al., 2019).

We used NDVI (Normalized Difference Vegetation Index; GIMMS NDVI3g) (Pinzon & Tucker, 2014) as a proxy for catchment vegetation cover and productivity (Pettorelli et al., 2005) for the period 1991–2015. The NDVI3g data set has a pixel resolution of 8 × 8 km and is recorded as bimonthly maximum NDVI. The annual mean NDVI3g record for the main snow-free period (June–August) and for the growing season (May-October) was calculated for each catchment. The long-term averages of NDVI correlated well with land cover (pair-wise correlations between NDVI_June-August to %forest, and %forest + peat: 0.66 and 0.69, respectively) in the ThousandLakes, and the June-August average has been used earlier in trend analyses of lake DOC (Finstad et al., 2016). Other NDVI-composites (i.e., averages for other periods) were highly internally correlated with the variables mentioned above and therefore not included in further analysis. The change in NDVI was calculated as Sen slopes (% yr^{-1}) over 1991–2015 using the ktaub MATLAB function (Burkey, 2022). The trends in summer NDVI (Table 1) show an upward trend of average 0.6% $decade^{-1}$.

2.6. Eco-Regions

The ThousandLakes and TrendLakes are grouped into 10 ecoregions that comprise similar deposition, climate, and biogeographical conditions (Dillon et al., 2003). These regions were reduced to six in the current paper, as shown in Figure 1, that is, South (region 4 + 5), East (1 + 2), Inland (3), West (region 6 + 7), Central (region 8), and North (region 9 + 10) (Table 1, Figure 1). Lake chemistry variation in relation to air pollution, acid sensitivity, proximity to the coast, elevation, vegetation, and soil characteristics is described in Henriksen et al. (1998). In short, South Norway has historically received the highest loadings of atmospheric pollution, followed by east and inland Norway, while central and northern Norway were much less exposed being further away from the sources. The most acid-sensitive areas are also located in southern Norway (Austnes et al., 2018). Sources of SO_4 other than air pollution such as sea-salts, marine sediments (Maxe, 2001), and geological sources also contribute to elevated concentrations of SO_4, especially in Northern Norway (Wilander, 1994). Similarly, the regional contrasts in Cl-levels can be related to the presence of marine clays and sea-salt aerosols, explaining why especially lakes in inland Norway, located above the marine limit, are low in Cl. Most agriculture is found in East Norway, while forest and peatlands are most common in East and Central Norway. Inland Norway has mostly high-elevation lakes with sparse vegetation.

2.7. Statistical Methods

The Mann Kendall test (MKT) (Hirsch & Slack, 1984) was used to test for the presence of monotonic trends in water chemistry in the TrendLakes and of NDVI composites, and the Theil-Sen estimator was used to derive regressions. These methods are robust against outliers and missing data and do not require normally distributed residuals. To quantify the change in the TrendLakes per ecoregion over the period 1995–2019, the Theil-Sen estimator (fitted over the full period with available data, 1990–2020) was used to predict values in 1995 and 2019 and then absolute and percentage differences were calculated relative to 1995.

Water chemistry from the 1000 lakes in 1995 and 2019 were paired for each lake. The temporal change was compared using a “matched pairs” comparison, for Norway and per region (One-Way analysis), where we tested if the differences between 1995 and 2019 were significantly different from zero using a *t*-test. The *t*-test assumes a normal distribution of the values. A nonparametric test was also run (Wilcoxon signed rank), which gave similar results (not shown). The analyses were done in JMP 16.0.0 (512257). We tested for regional differences in the temporal change with a pair-wise one-way analysis of means by region; the regional means were compared with Student's *t*-test ($p < 0.05$).

We used a two-pronged approach to explore links between drivers and change in the ThousandLakes. First, we used a multivariate Principal Component Analysis (PCA) on changes in key components of lake chemistry, excluding element ratios to avoid internal correlation. After interpretation of the PCA loadings and clustering, and the available mechanistic understanding from the literature, we used Partial Least Squares (PLS) to test the explanatory power of variables. PLS is particularly useful when a large set of explanatory variables is given, and variables are collinear (Wold et al., 2001). The PLS produces linear models from factors, which are linear combinations of the explanatory variables (Xs) and are obtained by maximizing the covariance between the Xs and the response(s) (Ys). PLS exploits the correlations between the Xs and the Ys to reveal underlying latent structures, using the NIPALS algorithm (nonlinear iterative partial least squares) and the leave-one-out method of cross validation. The Variable Importance Plot (>1.0) is used to identify the key variables. We allow for a wide range of potential explanatory variables including static properties (%forest, %peatland, lake to catchment ratio) and changes in drivers, for example, in deposition, antecedent weather (temperature and precipitation, for 1 and 6 months, to avoid internally correlated variables), NVDI, and in water chemistry. We checked for collinearity (Figure SI 3) and only allowed variables that were not significantly correlated ($p < 0.05$) or make mechanistic sense (i.e., a change in labile Al is a response to changes in SO_4, not the other way around; S deposition, not N deposition, to explain SO_4). The variables were selected in two steps, first by including all with variable importance (VI) > 1.0 and then reanalyzing with only the selected variables and then again keeping only those with VI > 1.0. The analyses were done in JMP 16.0.0 (512257).

3. Results

3.1. Temporal Change in Lake Water Chemistry: Comparison ThousandLakes and TrendLakes

We hypothesized that the changes between 1995 and 2019 in the ThousandLakes exceed the interannual variation and reflect wide-scale temporal trends rather than random interannual variation. The largest and most consistent changes in water chemistry of the ThousandLakes between 1995 and 2019 (Figure 2, Table 2, Figure S4 in Supporting Information S1) were considerable declines in SO_4 (−39%), NO_3 (−32%), and Cl (−6%) and increases in DOM and SiO_2 (31% and 32%, respectively). Sea-salt-corrected SO_4 shows similar relative change compared with uncorrected SO_4 (Table 2, Table S4 in Supporting Information S1). TON (+9%), alkalinity (+13 $\mu eq\ L^{-1}$), and Ca (+14%) also showed overall increases. Note that Ca increased despite the overall decrease in SAA (−25%). The derived variable ANC was higher in 2019 in all regions, with an average change of +42 $\mu eq\ L^{-1}$. This change is consistent with, though higher, than the increase in alkalinity (+13 $\mu eq\ L^{-1}$). Finally, the ratios of NO_3 to totP, Fe to DOM and nonlabile Al to DOM almost halved compared with 1995.

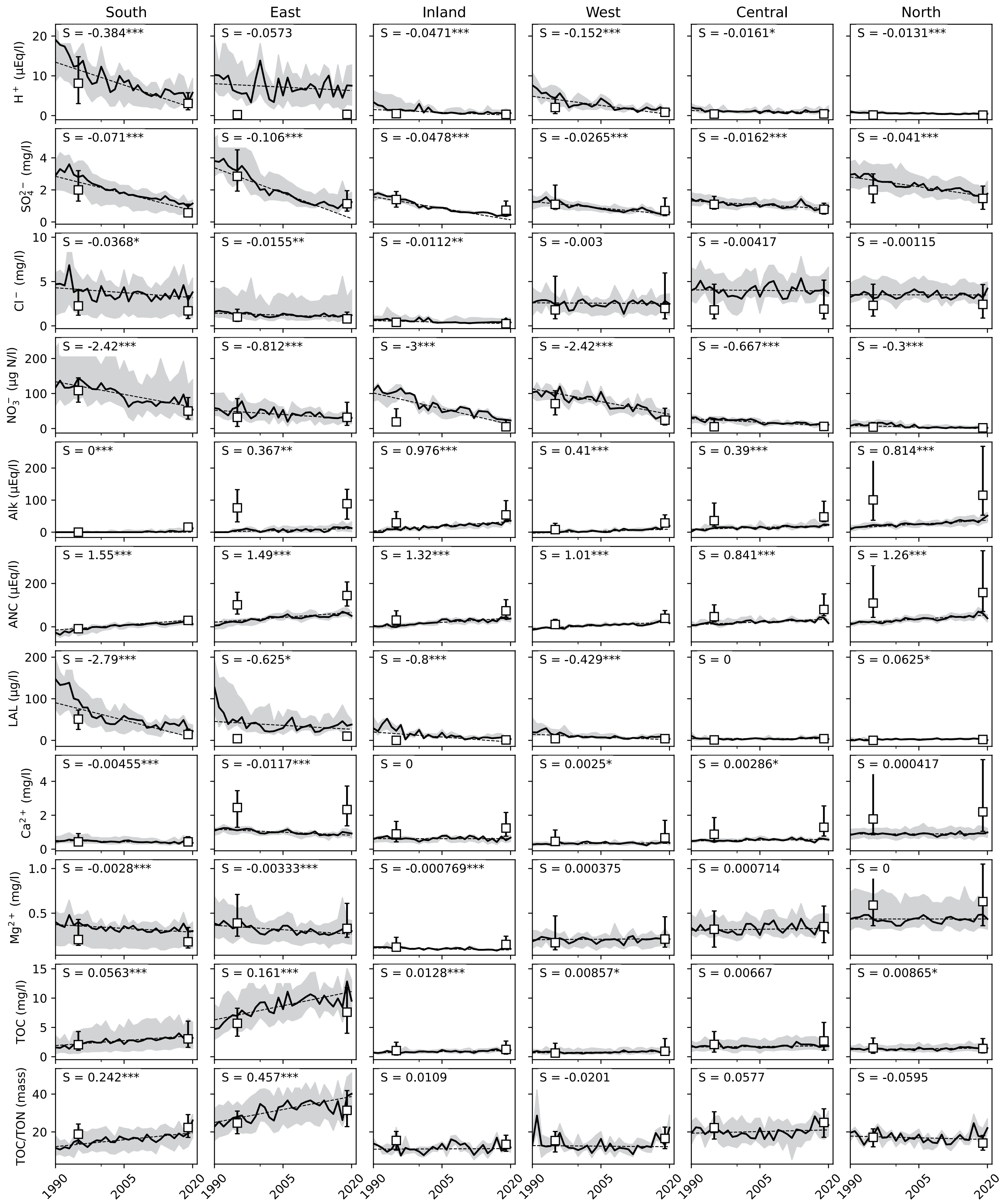

Figure 2. Time series of median (solid black line) and interquartile range (shaded gray area) for the 78 TrendLakes split into the six regions. Dashed lines show the Sen's slope of regional medians. The Sen slope, S, is given in the top-right corner of each plot, where asterisks indicate results of the Mann-Kendall trend test (*: $p < 0.05$, **: $p < 0.01$, and ***: $p < 0.001$). Median (square markers) and interquartile range (error bars) for the 1995 and 2019 1000 lakes are also shown. The *y*-axes are uniform across regions for a given variable.

Table 2
Results of Pairwise Comparison of Main Chemical Components ThousandLakes Showing Averaged Concentrations for 1995 and 2019 by Region and for all of Norway

	pH	SO$_4$ mg/L	Cl mg/L	NO$_3$ μg N/L	Alk μeq/L	ANC μeq/L	ANC$_{oaa}$ μeq/l	SAA μeq/l
Region	1995/2019/Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/Δ	1995/2019/Δ	1995/2019/Δ	1995/2019/Δ
South	5.02/5.38/0.36****	2.4/0.8/−65****	3.3/2.6/−21****	125/71/−43****	10/25/14****	2/41/39****	−8/27/35****	152/96/−56****
East	5.71/5.61/−0.10*	3.6/1.9/−47****	2.2/2.3/6n.s.	102/106/4n.s.	130/133/3n.s.	159/196/37****	139/166/27****	145/113/−32****
Inland	5.98/6.30/0.32****	1.5/1.0/−33****	0.5/0.3/−43****	37/17/−53****	57/81/24****	59/105/46****	53/99/45****	50/31/−19****
West	5.47/5.86/0.39****	1.9/1.2/−38****	4.2/4.0/-4n.s.	85/44/−47****	27/52/25****	27/70/42****	22/63/41****	164/141/−20***
Central	6.12/6.06/−0.06n.s.	1.5/1.1/−26****	3.2/3.5/9n.s.	23/22/-3n.s.	88/101/13***	100/141/41****	90/128/37****	119/119/0 n.s.
North	6.34/6.43/0.09n.s.	2.5/2.1/−17****	3.7/3.5/-6n.s.	16/8/−49****	203/206/3n.s.	217/265/48****	209/257/48****	157/141/−15****
Norway	5.51/5.77/0.26****	2.3/1.4/−39****	3.0/2.8/−6***	66/45/−32****	89/102/13****	97/139/42****	87/126/39****	135/110/-25****

	TOC mg/L	Labile Al μg/l	Non-labile Al	Fe μg/l	Ca mg/L	Mg mg/L	K mg/L	SiO$_2$ mg/l
Region	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ
South	2.9/4.2/44****	53/19/−64****	50/54/9*	130/186/43****	0.8/0.7/−12**	0.32/0.25/−20****	0.18/0.12/−32****	1.2/1.5/23****
East	6.0/8.9/49****	11/14/35***	52/58/12**	304/311/2n.s.	3.4/3.5/1n.s.	0.60/0.52/−12****	0.42/0.39/-6n.s.	2.3/3.5/48****
Inland	1.6/1.8/9*	4.2/3.0/−29*	15/10/−38****	76/52/−31***	1.3/1.8/36****	0.22/0.25/13**	0.18/0.15/−13n.s.	1.2/1.6/37****
West	1.6/2.0/22****	9.4/5.4/−43****	23/17/−27****	70/62/−11n.s.	1.0/1.3/35****	0.36/0.35/-2n.s.	0.25/0.20/−19****	0.6/0.8/37****
Central	2.9/3.8/31****	3.5/5.9/67**	29/25/−11**	120/91/−24**	1.9/2.4/26****	0.40/0.47/16****	0.23/0.22/-2n.s.	0.9/1.3/47****
North	2.4/2.4/0n.s.	3.5/4.6/30n.s.	14/7/−46****	114/45/−61****	3.6/4.2/15****	0.86/0.91/6*	0.42/0.45/6n.s.	1.3/1.4/7*
Norway	2.9/3.8/31****	16/9/−41****	31/29/−6**	143/132/−8*	2.0/2.3/14****	0.48/0.48/0n.s.	0.28/0.26/−8**	1.2/1.6/32****

	totP μg/l	totN μg/l	TON μg/l	NO$_3$:TotP g/g	CN DOM g/g	Fe:DOM mg/g	IlAl:DOM mg/g
Region	1995/2019/%Δ	1995/2019/%Δ	1995/2019/%Δ	1995/2019/Δ	1995/2019/Δ	1995/2019/Δ	1995/2019/Δ
South	3.1/3.1/1n.s.	261/230/−12****	135/159/17****	60.7/29.6/−31****	19.3/23.1/3.8****	38/33/−5*	23/13/−10****
East	6.5/7.8/20**	334/364/9*	232/257/11**	20.0/14.7/−5**	28.1/32.1/4.0n.s.	46/28/−18****	8/6/−2****
Inland	3.3/2.9/−12n.s.	125/123/-2n.s.	88/106/20****	21.5/10.3/−11****	16.0/14.8/−1.2*	59/22/−37****	21/8/−12****
West	3.3/3.2/-4n.s.	173/144/−17****	88/100/13****	43.1/18.8/−24****	17.0/17.0/0.0n.s.	84/22/−62****	27/11/−17****
Central	2.9/3.0/3n.s.	130/154/19****	107/132/24****	9.8/8.1/-2n.s.	23.6/25.5/1.9**	48/22/−26***	16/8/−7****
North	5.5/4.3/−22*	146/151/3n.s.	129/142/10n.s.	5.3/4.3/-1n.s.	18.9/14.8/−4.1***	84/32/−51****	14/7/−7****
Norway	4.1/4.0/-3n.s.	196/194/-1n.s.	127/138/9****	27.3/14.6/−13****	21.2/23.7/2.5**	60/28/−32****	18/9/−9****

Note. Change (Δ) is given in absolute units for element ratios, pH and Alkalinity and ANC and as %change relative to the 1995 values elsewhere. SAA, strong acid anions (equivalent sum of SO$_4$, Cl, NO$_3$); CN of DOM calculated as totN minus NO$_3$ divided by TOC. Significance levels indicate $p < 0.0001$ (****), with $p < 0.001$ (***), $p < 0.01$ (**), $p < 0.05$ (*), n.s. nonsignificant. pH was expressed in H$^+$ concentrations prior to testing. Note that comparison for Fe and Fe:DOM is based on fewer lakes[a].

[a] Number of lakes per region for South, East, Inland, West, Central, and North are 198 (Fe: 143), 146 (Fe: 136), 130 (Fe: 82), 153 (Fe: 113), 147 (Fe: 113), and 205 (Fe: 160), respectively, which gives a total of 978 (Fe: 730).

Table 3

Changes in Main Chemical Components in TrendLakes by Ecoregion, Estimated by Comparing Predicted Values for 1995 and 2019

	pH	SO_4 mg/L	Cl mg/L	NO_3 mg N/L	TOC mg/L	Alk µeq/L	ANC µeq/L	Labile Al µg/l	Ca mg/L	Mg mg/L	CN DOM g/g
Region	Δ	%Δ	%Δ	%Δ	%Δ	Δ	Δ	%Δ	%Δ	%Δ	Δ
South	0.70 ***	−68 ***	−21 *	−48 ***	62 ***	0 ***[a]	37 ***	−88 ***	−22 ***	−19 ***	5.8 ***
East	0.09 n.s.	−89 ***	−27 **	−41 ***	54 ***	9 **	36 ***	−36 *	−25 ***	−23 ***	11.0 ***
Inland	0.92 ***	−86 ***	−50 **	−83 ***	41 ***	23 ***	32 ***	−120 ***	0 n.s.	−17 ***	0.3 n.s.
West	0.96 ***	−56 ***	−3 n.s.	−57 ***	29 *	10 ***	24 ***	−84 ***	19 *	4 n.s.	−0.5 n.s.
Central	0.17 *	−32 ***	−2 n.s.	−65 ***	10 n.s.	9 ***	20 ***	0 n.s.	14 *	5 n.s.	1.4 n.s.
North	0.26 ***	−38 ***	−1 n.s.	−103 ***	16 *	20 ***	30 ***	63 *	1 n.s.	0 n.s.	−1.4 n.s.
All	0.83 ***	−67 ***	−4 n.s.	−55 ***	32 ***	12 ***	28 ***	−85 ***	−5 n.s.	−11 **	2.4 *

Note. Predictions were derived using the Theil-Sen estimator fitted over the period 1990–2020. Change (Δ) is given in absolute units for element ratios, pH, alkalinity, and ANC and as %change relative to the 1995 values elsewhere. CN of DOM calculated as totN minus NO_3 divided by TOC. Asterisks summarize results of Mann-Kendall tests for monotonic trends, with $p < 0.001$ (***), $p < 0.01$ (**), and $p < 0.05$ (*) and n.s. is nonsignificant (no trend).

[a] Alkalinity was fairly constant throughout the period until high values in the last couple of years (see Figures S1–S3 in Supporting Information S1). This results in a Sen's slope of 0 and yet significant monotonic trend according to Mann-Kendall.

The concentration levels and trends in the TrendLakes (Figure 2, Table 3; Figure S4 in Supporting Information S1) show a high consistency with the differences between 1995 and 2019 in the ThousandLakes in each ecoregion (Table 2). This implies that the temporal change in the ThousandLakes can be understood as a long-term change, instead of interannual variation related to differences in weather and precipitation between the two sampling periods (e.g., Tables S1 and S2 in Supporting Information S1). Note that the TrendLakes monitoring program did not include Fe and SiO_2.

For pH, Ca, ANC, labile Al, and NO_3, the ThousandLakes data are outside the range found in the TrendLakes (Figure 2), consistent with the selection of TrendLakes from relatively acid-sensitive, base-cation poor areas of Norway. The TrendLakes also displayed higher recovery rates of pH and labile Al (TrendLakes: ΔpH and ΔLAl +0.83 and −85%, respectively (Table 3); Thousandlakes: ΔH and ΔLAl, +0.26 and −32%, respectively (Table 2)). We conclude that the direction of the region-wise change in both surveys is mostly consistent, supporting the hypothesis that the changes in water chemistry found in the ThousandLakes reflect a wide-scale temporal change and suggesting a commonality in underlying drivers in both surveys.

3.2. Comparison Between Ecoregions

The largest declines in SO_4 deposition are in South and West Norway, while North and Central Norway show little change (Figure 1, Table 1; Figure 3). Note that similar scores on the x-axis of Figure 3, that is, the same letter for two or more regions, indicate that changes for a component are not statistically different between these regions. The declines in lake SO_4 from south to north mirror the latitudinal pattern in S deposition change, whereas West and East Norway deviate, that is, West declined less in lake SO_4 than East (Table 2; Figure 3) despite a much larger long-term reduction in S deposition. Possibly, these deviations are related to interannual variation in precipitation (e.g., Table S2 in Supporting Information S1), leading to variations in the S deposition that are not well-represented by 5-year averages of deposition (e.g., Table 1). Considering the pH, labile Al, and alkalinity, South and West show the strongest signs of recovery: +0.4 pH unit in both regions and the largest relative declines in Labile Al. These regions were most acidified in 1995 (i.e., average pH < 5.5, ANC and alkalinity <30 µeq L^{-1}; Table 2). Alkalinity shows relatively little improvement in South Norway, possibly because of many lakes with zero alkalinity in the 1990s. The change in the composite indicator ANC is remarkably constant between the regions despite the considerable variation in regional acid sensitivity and acid deposition loads. ANC_{oaa} differentiates slightly more but both suggest significant, relatively constant recovery in all regions (Table 3).

Changes in pH and labile Al in East and Central Norway are not consistent with the decrease in S deposition. Instead, we see a slight reacidification, that is, decreases in pH and increase in labile Al (Table 2; also found in TrendLakes, Table 3). Note the large increases in TOC in these regions (East: +3.9 mg C l^{-1} (+49%); Central: +0.9 mg C l^{-1} (+31%); Table 2), which can be associated with increases in organic acidity. We attribute the relatively high TOC concentrations in East and Central Norway to high forest cover and a large contribution of peatlands (Figure 1).

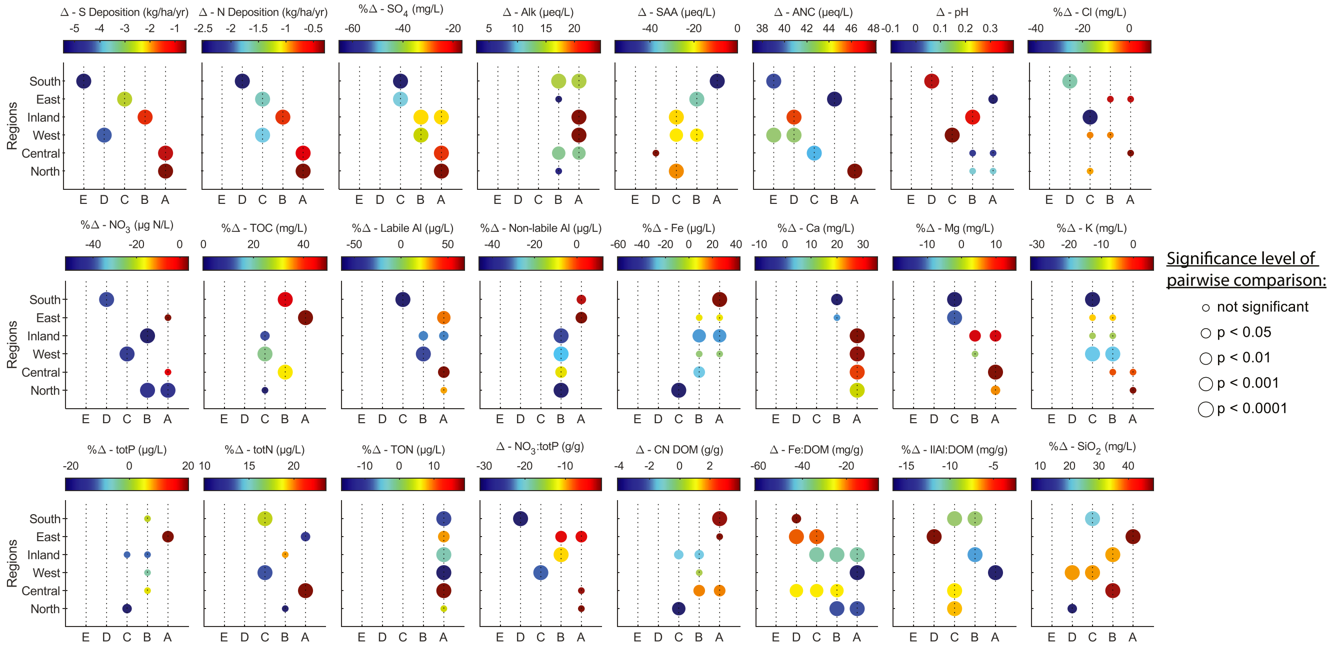

Figure 3. Joint presentation of (1) temporal change (between 1995 and 2019) of deposition and key water chemical components in the ThousandLakes in each region and (2) pair-wise comparison between regions of the magnitude of the temporal change. The regions are presented on the *y*-axis. The temporal change is color-coded (top bar) and its significance (difference from 0) is shown by the size of each circle. The pair-wise comparison between regions of the magnitude of the change is denoted by letters (on the *x*-axis), where different letters indicate that regions differ significantly ($p < 0.05$) in the magnitude of change for a given component. If circles for given regions have received the same letter, the change is uniform for those regions. If circles for each region receive different letters, the changes have regional contrasts. The letter A is awarded to the upper end of the change range and E to the lower end. The changes are presented as absolute or relative, as in Table 3.

Contrary to the expectation of simultaneous decreases in SAA and base cations, related to the principle of electroneutrality, Ca in the ThousandLakes increased in all regions except in South Norway (Table 2; Figure 2). The change in Mg is more consistent with ΔSAA than Ca but also shows increases in Inland, Central, and North Norway. The more acidic TrendLakes also show that declines in Mg are more pervasive than those in Ca (Table 3) and shows a positive Ca trend in West and Central Norway. Apparently, deviations from the expected declines for Ca and Mg are most distinct in the least acid-sensitive regions.

We hypothesized that regional changes in lake NO_3 are related to changes in N deposition and land cover. The decline in N deposition is again largest in South Norway with little change in the two northernmost regions, while East and West Norway show intermediate declines. The change in lake NO_3 roughly reflects the gradient of decline in N deposition, except for East (no decline) and Inland (a large relative decline) Norway. East Norway has most forest and inland Norway, dominated by high elevation catchments, has least forest (Figure 1), which suggests that a high catchment N retention capacity, represented by forest cover, reduces its sensitivity to change in atmospheric loads of N. In East Norway, the presence of agriculture also plays a role: East Norway has a small NO_3 decline, similar to North, when catchments with over 5% agriculture are excluded (data not shown). It is noteworthy that Northern Norway shows a significant decline in lake NO_3 even though N deposition has hardly changed.

The increases in DOM, represented by ΔTOC, are significant in all regions except in Northern Norway. Changes in East exceed changes in South and Central, which in turn exceed changes in Inland and West Norway (Figure 3). Again, this roughly coincides with changes in S deposition, except for West Norway, where the TOC increase is relatively modest. However, in the West, TOC concentrations are low, as a consequence of land cover, while the dilution from precipitation is considerable.

We hypothesized that nutrients in an organic form (totP and TON) and complexed elements (Fe, nonlabile Al) primarily follow the change in DOM, implying constant element ratios. The regional changes in TON, which are remarkably constant, mimic those of TOC, as they increase everywhere apart from North Norway. However, the CN of DOM shows contrasting change (Tables 2 and 3; Figure 2): increases in South and Central Norway, indicating N-depletion of DOM, while Inland and North Norway show decreases in the CN of DOM. TotP remains remarkably stable except for East Norway (+20%) and North Norway (−22%). If catchments with more than 5% agriculture in East Norway are excluded (data not shown), the increase in totP is less distinct but remains significant. It is puzzling that DOM in North Norway should be enriched in N while simultaneously depleted in P. The CN of DOM in North shows no change in the TrendLakes (Table 3), in contrast to the ThousandLakes. Such inconsistency is worrying and should caution overinterpretation of these trends.

Element to DOM (Fe:DOM, nonlabile Al:DOM) ratios do not remain constant but the direction of change is consistent in all regions of Norway (Tables 2 and 3; Figure 2). Fe and nonlabile Al on their own also show remarkably consistent changes across regions, that is, increasing in the South and decreasing elsewhere. This could point toward similar underlying drivers of change in both elements.

The decrease in the NO_3 to totP ratio is only significant in the four regions in the south, where the largest decrease in N deposition is found and also the most distinct increases in DOM.

The upward change in SiO_2 is almost as consistent and constant as the upward change in TOC, including the relative order of regional change, with largest increases found in East and the lowest in North.

We conclude that the regional variation in changes relating to chemical recovery appears to be largely related to atmospheric chemistry but that some changes are most likely moderated by land cover, especially for nutrients. Surprisingly, base cations, especially Ca, do not follow trends in SAA, while trends in element ratios of DOM do not follow DOM. This is possibly an indication of changes in DOM quality (discussed further in Section 4.3). Regional patterns of increases in DOM and SiO_2 are similar, which could indicate similar underling controls.

3.3. Controls of Temporal Change in Lake Water Chemistry

The assessment of region-wise differences suggests that air pollution is a key driver of temporal change in the ThousandLakes, where climatic factors and land cover are likely additional controls. We use changes in NDVI as a proxy for changes in land cover. A PCA loading plot (Figure 4) displays colocation of changes in key chemical components, land cover (forest, peatland, and agricultural land), deposition and discharge.

The first principal component (PC1) explains 19.8% of the variance in the data and can be interpreted as an axis of atmospheric deposition and chemical recovery. Here, the changes in S and N deposition, lake SO_4, base cations and alkalinity receive high scores, in addition to ΔNa and ΔCl, the main constituents of sea-salts (Wright et al., 1988). Note the colocation of ΔT_6m (positive since 2019 was warmer than 1995, Table S2 in Supporting Information S1) and change in deposition (negative), implying that warming was inversely correlated with a change in the deposition in all of Norway. The PC2, explaining an additional 13.5%, can be interpreted as an axis of land cover and climate. Here, forest, peat and changes in lake TOC score high, while discharge scores high, but negative. Components closely associated with DOM, that is, totP, TON, and nonlabile Al (i.e., organically complexed Al), also score high on this axis. Note that change in SiO_2 also has a high score on this axis. This axis could signify the importance of DOM for transporting of organically complexed Al, N, and P, and wetlands and organic carbon-rich mineral soils are known as significant sources of both TOC and SiO_2 (Struyf et al., 2010). Components with scores on both axes are changes in H, labile Al, and NO_3. Changes in pH are associated with chemical recovery but also with DOM, which contains weak organic acid, while labile Al highly depends on pH since Al speciation is strongly pH-dependent (Tipping, 1994). The change in NO_3 is likely to be a result of changes in the N deposition combined with catchment-processing, which could explain its intermediate position between PC1 and PC2. Overall, the PCA-analysis supports the key role of air pollution, land cover and climatic factors for change but does not identify the relative importance of variables, for which a PLS analysis is more suitable.

The variables in the PLS analysis that are explained best ($r^2 \approx 0.8$) are ΔCa and ΔNa and worst are ΔK and ΔNO_3 ($r^2 \approx 0.15$) (Table 4). The change in base cations is explained by ΔSO_4, ΔCl (the only variable selected for ΔNa), and ΔAlk (the most important for ΔCa). Land cover is not selected to explain the change in base cations, nor are the climate variables except for ΔT. However, ΔT is strongly internally correlated with ΔSO_4 (Figure S3 in Supporting Information S1) and is less likely to exert a direct effect on base cations than SO_4, so it is likely a case of correlation but not causation. Thus, the prominent increase in Ca appears to be primarily associated with increases in alkalinity and secondarily with the decrease in SAA, whereas the changes in Mg (mostly declines)

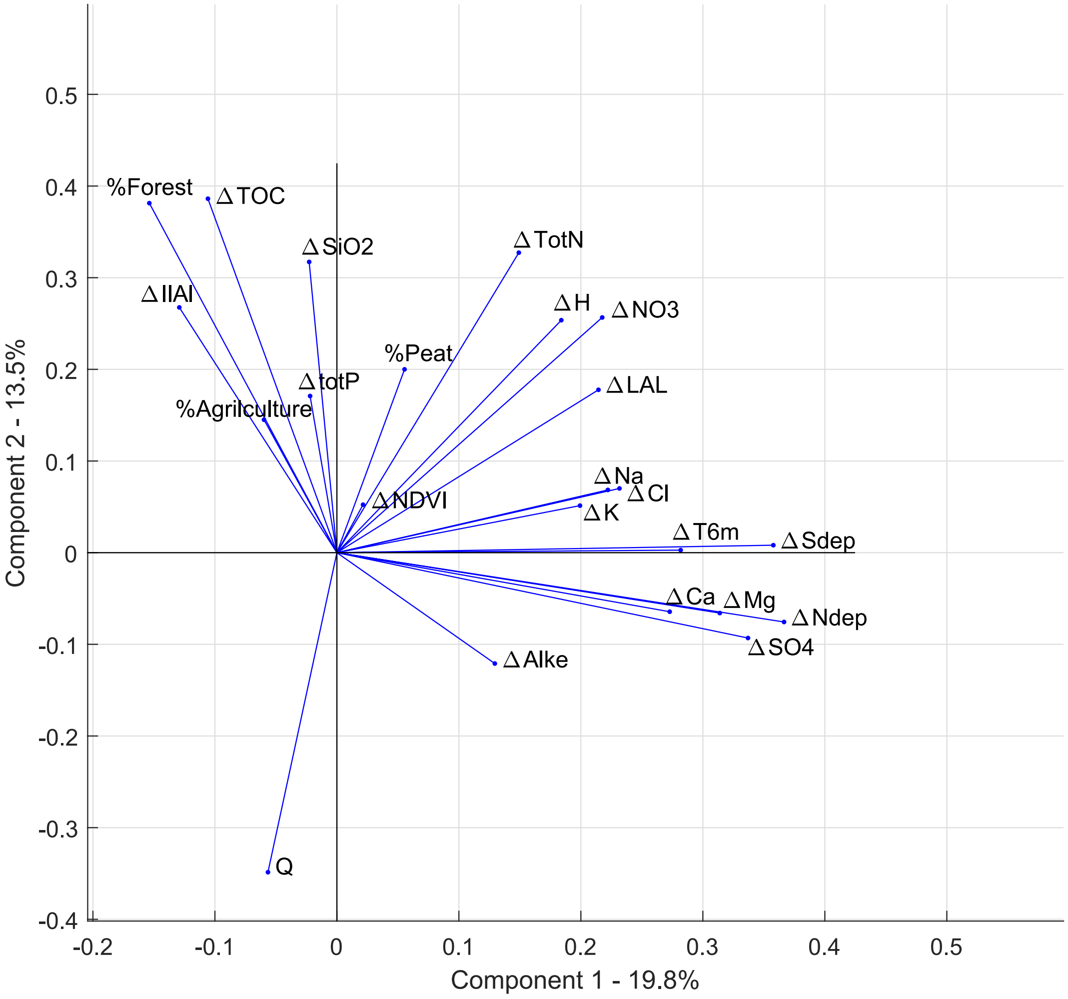

Figure 4. Principal Component Analysis on land cover (%peat, %forest, and %agriculture), long-term discharge (*Q*), changes in N and S deposition (1995–2019) (S dep, N dep), change in NDVI_JAS (NDVI) and temperature (T6m; 6 months average), and changes (1995–2019) in key water chemical components (Ca, Mg, K, Na, Cl, SO_4, NO_3, TOC, SiO_2, H, labile Al (Lal), nonlabile Al (IlAl), Alke, totP, and totN) in the ThousandLakes data set. All changes are indicated with prefix "Δ".

are more related to the declines in SO_4 and Cl. The PLS model suggests that the variation in Na is largely related to variation in Cl, which agrees with the PCA (Figure 4).

The PLS model for ΔSO_4 ($r^2 > 0.3$) (Table 4) confirms that the change in S deposition is its main driver in addition to forest cover and temperature. Again, it is possible that forest cover and temperature are selected because of their strong internal correlation with the gradient in the S deposition (Figure S3 in Supporting Information S1). The relatively low variation that is explained could be related to the presence of catchment-internal sources of S, SO_4 adsorption by soils, and to uncertainties in the modeled S deposition. The PLS model for ΔTOC ($r^2 > 0.3$) agrees with the dominant control of S deposition on browning. The PLS model for ΔSiO_2 ($r^2 > 0.3$) suggests a close association with changes in TOC and nonlabile Al, while climatic variables are not selected. Lastly, the PLS model for NO_3 hardly explains any variation ($r^2 > 0.1$). The temporal variation in NO_3 across large spatial gradients is typically difficult to explain (Burpee et al., 2022) and our study underlines this. Note that a change in NDVI is never selected as an explanatory variable in the PLS models.

4. Discussion

The ThousandLakes data document widespread chemical recovery, browning and changes in DOM elemental ratios and quality, SiO_2 enrichment in addition to reduced NO_3, and NO_3 to totP ratios. The statistical analysis

Table 4

PLS Models for Chemical Changes in Ca, Mg, Na, Non-Labile Al, NO_3, TOC and SiO_2 Between 1995 and 2019 in ThousandLakes

type	Name	Ca	Mg	Na	LAl	NO_3	TOC	SiO_2	SO_4
Static	%forest	-	-	-	-	-	2.1	1.3	1.7
	%peatland	-	-	-	-	1.0	-	-	-
	lake:catchment ratio	-	-	-	-	-	-	-	-
Delta	Temp_1 month	-	-	-	-	-	-	1.4	-
	T_6 month	-	1.1	-	1.4	1.3	-	-	1.2
	P_1 month	-	-	-	-	-	-	-	-
	P_6 month	-	-	-	1.4	-	-	-	-
	N deposition					1.7			
	S deposition	1.1	1.4	-	1.5		1.1	-	1.8
	Summer NDVI	-	-	-	-	-	-		
	SO_4	1.5	1.9	1.1	-				
	Cl	1.0	1.4	3.3	-		-		
	NO_3	-	-	-	-				
	Alkalinity	2.9	2.1	-	-				
	H	-	-	-	2.4				
	TOC	-	-	-	-	1.2		1.4	
	SiO_2	-	-	-	-				
	Non-labile Al	-	-	-				1.6	
	R^2	0.79	0.54	0.82	0.43	0.14	0.28	0.31	0.32
	R^2 (onlyVI > 1)	0.76	0.52	0.81	0.41	0.12	0.22	0.25	0.32

Note. All cells with content show selected explanatory variables; value refers to "variable importance" (VI), only shown for VI > 1.0. Static variables are land cover (% forest cover and peatland cover, lake to catchment ratio) and "delta" refers to difference between 1995 and 2019 (T, temperature; P, precip; 1 m, 6 m: averaged over 1 and 6 months prior to sampling date). R2 shows fraction of variation explained by PLS model with all variables initially selected, R2 (only VI > 1) shows fraction of varation explained with only selected variables from further refinement of PLS (in gray cells).

supports that the regional variation in chemical recovery is related to gradients and declines in air pollution in addition to land cover and possibly climate. However, the statistical analyses are limited by the information conveyed by the explanatory variables. Especially, climate-induced changes in catchment and lake processes and hydrology also could be responsible for some of the less well-understood change, for example, change in Fe, SiO_2, and element ratios of DOM and Ca. Below we place our results in the context of long-term environmental change and catchment biogeochemical studies.

4.1. Chemical Recovery From Acidification

Widespread decreases in lake SO_4 has been extensively documented in Europe and North America (De Wit et al., 2021; Garmo et al., 2014) and is related to reduced emissions of SO_2 to the atmosphere. Our results fit well with these changes but the overall SO_4 decrease of 39% in ThousandLakes (Table 2) is below the reported 70% decline in SO_4 deposition in Northern Europe 1990 (Aas et al., 2017; Colette et al., 2016). This is partly because geological and marine sources (Wilander, 1994) of SO_4 contribute to a baseline concentration and also because deposited SO_4 can be retained in soils and peatlands and slowly released leading to a delay in recovery (Prechtel et al., 2001; Reuss & Johnson, 1985), which has been observed in European forested catchments (Vuorenmaa et al., 2017).

With regard to the comparison between the relative change in lake chemistry and air pollution, note that estimations of levels and declines in S and N deposition vary considerably, especially for N (Table S5 in Supporting Information S1). Emission-derived deposition and interpolated estimates based on empirical measurements can give different results, as demonstrated by 15%–45% declines in tropospheric NO_2 over Europe estimated from emission inventories while satellite-observed trends indicated a decline of about 20% (Jiang et al., 2022).

Moreover, modeled wet bulk N and nonmarine S deposition were found to be 20%–40% lower than measured deposition for Norwegian stations (Marchetto et al., 2021). Inconsistencies in estimates of deposition pose a challenge for the identification of drivers of biogeochemical changes and calculations of critical load exceedances (Forsius et al., 2021).

The decreases in lake SO_4 concentrations have led to substantial chemical recovery of acidified lakes in acid-sensitive regions, as demonstrated by the rise in pH, alkalinity, and ANC (Grennfelt et al., 2020; Stoddard et al., 1999). We found the strongest recovery in South and West Norway (Table 2), the most acidified regions which have experienced the largest decline in S deposition (Table 1; Figure 1). Here, labile Al concentrations also decreased strongly, a variable that is not reported in international assessments because of the lack of harmonized methods (Hovind, 1998). Reduction of toxic Al is the best indication of improvement for sensitive aquatic life (Baker & Schofield, 1982; Havas & Rosseland, 1995). Increases in ANC were remarkably evenly, geographically distributed, suggesting a more general recovery from acid deposition than pH, alkalinity, and labile Al. Especially in the East, where the absolute increase in DOM was highest, alkalinity and pH did not increase, suggesting that organic acidity replaced mineral acidity in this region (e.g., Krug & Frink, 1983). We conclude that the regional patterns of chemical recovery from reduced S deposition have been modified by increases in DOM. Only South and West Norway still have ANC values around critical limits (Hesthagen et al., 2008), while the other regions have veered back to values not associated with problematic acidification for aquatic life. Note that there is considerable variation within each region, implying that proper assessment of lake water acidification status requires data evaluation on individual lake level.

Reduced anthropogenic emissions of Cl (Evans et al., 2011) are consistent with the reduction of Cl in the inland high elevation lakes (Table 2), whereas lower sea salt deposition is most likely the driver behind the decline in Cl in South Norway (Hindar et al., 2004).

4.2. DOM, Fe, and Browning

The strong increase in DOM found in the ThousandLakes (Table 2) is consistent with extensive documentation of lake browning in Europe and North America (De Wit et al., 2021), related to declines in SO_4 and possibly Cl (Monteith et al., 2007) and attributed to increased OM solubility (De Wit et al., 2007). We found no support for climate effects on changes in DOM (Table 4), in contrast to recent findings (De Wit et al., 2016, 2021). Similarly, no evidence for the effects of increased terrestrial productivity for browning (Finstad et al., 2016; Skerlep et al., 2020) was found (Table 4). Possibly, NDVI is not a sensitive indicator of increases in biomass since hardly any change in NDVI was found (Table 1) despite the marked increases in forest biomass in Norway in recent decades (De Wit et al., 2015). Experimental studies do not support a link between litter inputs, used as a proxy for increased terrestrial productivity, and DOM export (Froberg et al., 2007; Hagedorn & Machwitz, 2007). Evidence of the role of increased forest biomass as a driver of increasing DOM at decadal scales remains inconclusive.

A poorly understood pattern in the ThousandLakes is the stark regional contrast in trends of Fe (upward in South, decreases elsewhere; Table 2)—which also deviates from the increases in Fe found in 30 Swedish rivers (Kritzberg & Ekström, 2012). Since DOM is a vector of transport for Fe, increases in DOM would be expected to lead to increases in Fe but the only region that follows this pattern is South Norway. In the Swedish rivers, changes in Fe were higher than changes in DOM, in contrast to the ThousandLakes (Fe to DOM declined). In natural lakes in East Norway, positive trends in Fe exceeded positive trends in DOM (Xiao & Riise, 2021) consistent with Kritzberg and Ekström (2012). Similar to the decrease in Fe to DOM ratio, though not as distinct, the ratio of nonlabile Al to DOM declined in all regions. Kritzberg and Ekström (2012) proposed a relative increase in anoxic waters, for instance from peatlands in combination with changing hydrology, with high Fe as an explanation to the higher increases in Fe relative to DOM. Given the large hydrological gradients and variations in peatland cover in the catchment of the Swedish rivers and the ThousandLakes, a single hydrological explanation for changes in Fe stretches credulity.

The striking similarity of regional change in Fe and nonlabile Al and their ratios to DOM could point to common underlying drivers, although simple mechanistic explanations are lacking. Iron is redox-sensitive in contrast to aluminum. The oxidation rate of ferrous (Fe^{2+}) to ferric (Fe^{3+}) iron increases with pH in the pH range between 5.0 and 8.0 (Davison, 1993). Ferrous Fe, leached from wetlands and anoxic soils to oxic surface waters, may thus oxidize to ferric Fe and precipitate to $Fe(OH)_3$ at higher rates than previously. This mechanism, however, is not consistent with the regional contrasts in Fe trends (Table 2). Another explanation might be that the decline in SO_4 from deposition, largest in South Norway, has led to a reduced supply of sulfide, presumably limiting the fixation of Fe as pyrite (FeS_2) in anoxic conditions (Walker et al., 2021), which would lead to less Fe available for complexation with DOM. The lowering of nonlabile Al to DOM would be consistent with the lowered solubility of Al during chemical recovery (Van Breemen et al., 1984), while lower Al to soil organic matter (SOM) ratios are associated with higher SOM solubility (Berggren & Mulder, 1995; De Wit et al., 1999; Vogt et al., 1994).

4.3. Nitrogen Species, totP, and CN Ratios

The decline in NO_3 in TrendLakes exceeds the distinct decline in the ThousandLakes (Figure 2), while the relative lake NO_3 declines far exceed relative declines in N deposition in Europe, which is estimated to be approximately 20% since 1990 (Colette et al., 2016)). Declines in surface water NO_3 are well-documented (Garmo et al., 2014) and usually attributed to declines in N deposition, as well as interactions with catchment vegetation (Dise et al., 2009) and climate (De Wit et al., 2008; Kaste et al., 2020; Rogora et al., 2012). Experimental manipulations with NO_3 additions to plots and catchments have shown that removal of N deposition usually leads to a rapid decline in NO_3 leaching (Gundersen et al., 1998). The strong retention of deposited N by vegetation and soils (Vuorenmaa et al., 2017), however, implies that declines in atmospheric loading of N should not be expected to lead to immediate changes in surface water N under natural conditions. The relatively large NO_3 decline in Inland Norway, the region with high elevation lakes and sparse vegetation cover, suggests that high elevation catchments are most sensitive to changes in the N deposition. Similarly, rapid declines in N-species in alpine lakes were attributed to reduced N deposition during the COVID-19 pandemic (Rogora et al., 2022).

The increase in standing biomass in Norwegian forests (De Wit et al., 2015) and longer growing seasons (Hanssen-Bauer et al., 2017) point toward increased plant demand for nitrogen. The consistent and strong relative decline in NO_3 in East and North Norway, highly contrasting in N deposition, could be explained by increased catchment retention. In Northern Norway, increased grazing pressure from reindeer, reducing lichen density, is an important driver of increased forest cover (Tommervik et al., 2009) while in South Norway, reduced harvesting intensity drives increases in forest biomass (De Wit et al., 2015). Additionally, Northern Norway in 2019 was considerable warmer than 1995 (1.7°C in the 6 months mean antecedent temperature, Table S2 in Supporting Information S1), a warming signal that may have larger impacts on terrestrial productivity in cold than in warmer, regions. Climate warming and N deposition propel these changes forward (Tommervik et al., 2004), suggesting that catchment retention of nutrients in the future will increase.

Ratios of CN in DOM increased in South Norway in the ThousandLakes and the TrendLakes, indicative of N depletion of DOM, whereas little consistent change was shown elsewhere (Tables 2 and 3). Our findings are consistent with predominantly upward trends in the CN of DOM in northern streams across Europe and North America (Rodriguez-Cardona et al., 2022). Long-term enrichment of soils with atmospheric N would push CN ratios downward (MacDonald et al., 2002) and cannot logically explain these changes. The aforementioned changes in the Fe:DOM and Al:DOM ratios of DOM, along with the observed increases in water color which exceed DOM trends (Hongve et al., 2004), point toward changes in DOM character. Possibly, the increases in DOM and changing solubility has increased the mobilization of relatively more colored, hydrophobic (Vogt et al., 2004), N-depleted (Lajtha et al., 2005) humic acids.

TotP in the ThousandLakes showed little change despite the increase in DOM (Table 2) and is in contrast with the strong decline in TotP found in oligotrophic Swedish lakes (Huser et al., 2018). Huser et al. (2018) offer increased reliance on forest biomass for renewable energy, associated with increased P removal by tree harvesting, as one explanation for the reduction of P in the lakes. Such land use practices are not as common in Norway, which would potentially explain the lack of change in totP in Norwegian lakes. Reduced NO_3 and lower NO_3:totP ratios (Bergström, 2010), in combination with lower light availability as a consequence of browning, all point toward higher nutrient and light limitation, consistent with Creed et al. (2018).

4.4. Silicate

Significant increases in SiO_2 documented in the ThousandLakes are also found in several Norwegian rivers (Braaten et al., 2020), in Mjøsa, the largest lake in Norway (Hobæk et al., 2012), and in other lakes in Europe and North America (Björnerås et al., 2017; Vesely et al., 2005). We found that increases in SiO_2 were related to changes in TOC, forest cover, and nonlabile Al, while hydrological variables had no explaining power (Table 4). Thus, we find no support for our hypothesized explanatory effect of changes in hydrological flowpaths, that is, a greater contribution of SiO_2-enriched groundwater (Jin et al., 2010), to explain increases in SiO_2. Björnerås et al. (2017) linked widespread increases in Fe to increases in SiO_2, because both are derived from similar sources, that is, mineral soils. However, we found regional contrasts in Fe and SiO_2 changes, suggesting different controls for these elements. Mineral weathering of felsic rock is the primary source of SiO_2 in Norwegian soils derived from glacially eroded Precambrian igneous bedrock. This weathering is enhanced by root exudation of organic acids from vegetation and mycorrhiza (Drever & Stillings, 1997; van Scholl et al., 2008), which is consistent with positive correlations between forest cover and riverine SiO_2 export (Humborg et al., 2004) and lake SiO_2 (Hessen et al., 2009) and the strong correlation between ΔTOC and ΔSiO_2 in our study. The strong relation between primary productivity and root and soil respiration (Högberg et al., 2001), the increase in forest biomass in Norway (De Wit et al., 2015) and increased length of the growing season (Hanssen-Bauer et al., 2017) are consistent with amplified weathering rates by increased terrestrial productivity, resulting in temporal increased SiO_2 in the ThousandLakes and rivers, for example, an enhanced terrestrial silica pump (Carey & Fulweiler, 2012). An alternative explanation (Vesely et al., 2005) relates increases in SiO_2 in acidified lakes in central Europe to chemical recovery, for example, a decline in the inhibitory effect on silicate weathering from lower dissolved Al as well as lower precipitation of secondary minerals (aluminosilicates), cannot alone explain the widespread increase in SiO_2 in Norway, since some regions were not acidified.

4.5. Base Cations

A global analysis indicates that downward trends in Ca dominate in base-cation-poor regions of the world (Weyhenmeyer et al., 2019). This is attributed to the principle of electroneutrality, where cation charges must balance the decline in SAA equivalent concentrations. To our surprise, we saw an upward trend in Ca except for South Norway. The PLS analysis (Table 4) indicated that SAA was a downward driver of base cations while upward trends in alkalinity related especially well to Ca. Ca is the dominant base cation in the lakes (this study) and in soils (Steinnes et al., 1993). In acidic Norwegian soils, the primary source of Ca is weathering of silicate minerals, especially Ca-feldspars. Soils also contain large stores of exchangeable Ca, which have been depleted by leaching from soils to surface waters as a consequence of mobilization by acid deposition (Likens et al., 1998; Watmough et al., 2005). These pools are presently assumed to be slowly replenished by weathering (Reuss & Johnson, 1985). Above, we suggest that weathering from increased CO_2 pressure and organic acids is the best explanation for increased SiO_2, which could potentially also liberate base cations. However, the PLS model did not show any correlation between base cation change and SiO_2 (Table 4). Possibly, ion exchange confounds the relation between the change in base cations and SiO_2 since acidic soils have a higher affinity for base cations which will readsorb to the soil during chemical recovery (Blume et al., 2010). We interpret the strong relationship between alkalinity and Ca as a combined effect of chemical recovery and increased CO_2 pressure, where increased alkalinity is caused by chemical recovery (Reuss & Johnson, 1985) and increased CO_2 pressure in soils from enhanced terrestrial productivity. This interpretation implies that climate- and land use-induced increases in terrestrial productivity contribute to chemical recovery. Thus, a mechanistic connection between increased SiO_2 and Ca is possible, despite the lack of significant correlations, where the common driver would be climate-induced increases in terrestrial productivity and possibly chemical recovery. Further investigations into these relationships would benefit from mechanistic modeling of base cation exchange, chelation, and weathering (e.g., Gustafsson et al., 2018; Lawrence et al., 2014).

5. Conclusion

Significant change in chemistry in Norwegian lakes in the past three decades is attributed to large reductions in atmospheric deposition leading to recovery from acidification, whose effects are moderated by catchment processing impacted by land use and climate. In agreement with other lake trend assessments and established understanding of biogeochemical controls of acid-base chemistry, we found substantial chemical recovery from acid deposition and widescale browning. However, browning counteracted chemical recovery in some regions and should be accounted for in assessments of future trajectories of chemical recovery. Additionally, we found an unexpected increase in Ca which we attribute to increased terrestrial productivity, where the mechanistic connection is enhanced weathering from increase CO_2 pressure. This suggests that chemical recovery rates are enhanced by climate- and land use-induced acceleration of terrestrial carbon cycling rates.

We find that widespread increases in SiO_2 are consistent with explanations of increased weathering related to chemical recovery and increased terrestrial productivity, but for further elucidation of these relationships, physical-chemical modeling would be required. No support was found for hydrological drivers of changes in browning and SiO_2. Declines of NO_3, although poorly explained by statistical models, are consistent with interactions between declines in N deposition in areas lacking vegetation cover and increased N retention from enhanced terrestrial productivity.

We also found evidence of changes in DOM quality alongside well-documented increases in DOM, demonstrated by declines in elemental ratios (CN, Fe:DOM, nonlabile Al:DOM) of DOM and suggest that these are consistent with observed increases in color and evidence of chemical solubility controls of soil organic matter. The changes in DOM and nitrate further substantiate the enhanced light-limitation and reduced primary productivity in northern lakes.

Further understanding of biogeochemical controls would benefit from analysis of temporally intensive monitoring records and/or experimental catchment manipulations and use of physical-chemical models including improved descriptions linking the carbon cycle and acid-base chemistry.

The biogeochemical changes we have observed in these lakes can cascade downstream along the entire aquatic continuum. Based on the large-scale changes in lake water chemistry that we observe, and in the absence of hydrological change, we expect that rivers from natural catchments will become more nutrient- and light limited while northern coastal areas will receive higher freshwater inputs of DOM, alkalinity, Ca, Fe, and SiO_2.

Data Availability Statement

Land cover and water chemistry data from the ThousandLakes and TrendLakes are available at zenodo.org with the DOI: https://doi.org/10.5281/zenodo.7298477.

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