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Three Decades of Changing Nutrient Stoichiometry from Source to Sea on the Swedish West Coast

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Abstract

European ecosystems have been subject to extensive shifts in anthropogenic disturbance, primarily through atmospheric deposition, climate change, and land management. These changes have altered the macronutrient composition of aquatic systems, with widespread increases in organic carbon (C), and declines in nitrogen (N) and phosphorus (P). Less well known is how these disturbances have affected nutrient stoichiometry, which may be a more useful metric to evaluate the health of aquatic ecosystems than individual nutrient concentrations. The Swedish west coast has historically experienced moderate to high levels of atmospheric deposition of sulfate and N, and eutrophication. In addition, coastal waters have been darkening with damaging effects on marine flora and fauna. Here, we present three decades of macronutrient data from twenty lakes and watercourses along the Swedish west coast, extending from headwaters to river mouths, across a range of land covers, and with catchments ranging 0.037–40,000 km^2. We find a high degree of consistency between these diverse sites, with widespread increasing trends in organic C, and declines in inorganic N and total P. These trends in individual macronutrients translate into large stoichiometric changes, with a doubling in C:P, and increases in C:N and N:P by 50% and 30%, showing that freshwaters are moving further away from the Redfield Ratio, and becoming even more C rich, and depleted in N and P. Although recovery from atmospheric deposition is linked to some of these changes, land cover also appears to have an effect; lakes buffer against C increases, and decreases in inorganic N have been greatest under arable land cover. Our analysis also detects coherently declining P concentrations in small forest lakes; so called (and unexplained) ‘‘oligotrophication.’’ Taken together, our findings show that freshwater macronutrient concentrations and stoichiometry have undergone substantial shifts during the last three decades, and these shifts can potentially explain some of the detrimental changes that adjacent coastal ecosystems are undergoing. Our findings are relevant for all European and North American waters that have experienced historically high levels of atmospheric deposition, and provide

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a starting point for understanding and mitigating against the trajectories of long-term change in aquatic systems.

Key words: Organic carbon; Nitrogen; Phosphorus; Lakes; Streams; Eutrophication; Nutrients; Long-term trends; Sweden.

Highlights

- We analyzed three decades of nutrient data from lakes, streams and rivers in west Sweden
- We find widespread increases in carbon, and decreases in nitrogen and phosphorus
- These changes could affect the health of coastal ecosystems

Introduction

Research into terrestrial and aquatic biogeochemistry has often been partitioned by both ecosystem type (for example, forest, mire, lake, arable land, estuary, and so on) and process (for example, nutrient cycling, carbon (C) emission, hydrology, and so on). This is despite the self-evident fact that ecosystems and processes are dynamically linked via the transfer of water, solutes and particles from headwaters to the sea, and by interactions between different element cycles, particularly C, nitrogen (N) and phosphorus (P). Consequently, changes in the ecological status and management of terrestrial systems have the potential to alter the condition and function of downstream freshwater and marine ecosystems, and the goods and services they provide, in ways that cannot easily be predicted from the study of single systems or individual processes. A growing body of recent work now suggests that element ratios (that is, stoichiometry) may provide a more effective indicator of ecosystem status than individual nutrient concentrations (Taylor and Townsend 2010; Kopáček and others 2013; Burson and others 2016; Stutter and others 2018; Isles and others 2018; Deininger and others 2020; Graeber and others 2021), and that a landscape view, from source to sea, is needed to fully understand patterns and processes in biogeochemistry (Baker and Spencer 2004; Jones and others 2016; Palmer and others 2016; Xenopoulos and others 2017; Casas-Ruiz and others 2017; Maranger and others 2018).

There is overwhelming evidence that concentrations of macronutrients have been undergoing multidecadal changes in many European and North American aquatic systems. Principally, widespread increases in dissolved organic carbon (DOC) have been observed throughout the aquatic continuum (Worrall and others 2003; Evans and others 2005; Räike and others 2016; Eklöf and others 2021; Stetler and others 2021), although some waterbodies show no trends (Worrall and others 2004; Stackpoole and others 2017). Where increases occur, recovery from acid deposition and subsequent decreases in sulfate concentrations are thought to be the prime drivers (de Wit and others 2007; Monteith and others 2007; Evans and others 2012; Corman and others 2018), but climate and land-use potentially influence the trajectory of change in some areas (Kritzberg 2017; Lepistö and others 2021; de Wit and others 2021). Alongside this increase in DOC, concentrations of N and P have been declining in north European lakes, rivers and seas (Carstensen and others 2006; Radach and Pätsch 2007; Huser and others 2018; Lucas and others 2016). Mechanisms for this include decreasing atmospheric N deposition (Isles and others 2018) and the adoption of measures to reduce nutrient inputs from wastewater and agriculture (Carstensen and others 2006; Sharpley and others 2015). If C is increasing, while N and P are decreasing, it follows that C:N and C:P will be increasing at proportionally greater rates, with the result that ecosystems may be moving away from N and P excess. (Kopáček and others 2013).

The west coast of Sweden receives air masses from northwestern Europe, including large centers of industry, and therefore has been particularly affected by moderate to high levels of historical atmospheric sulfur (S, up to 25 kg ha^{-1} y^{-1}, Moldan and others 2004) and N deposition (up to 15 kg ha^{-1} y^{-1}, Moldan and others 1995). Subsequent reductions in S deposition in particular are believed to have driven large-scale recovery from acidification (Moldan and others 2013), resulting in fluvial DOC increases of about 2% per year since 1972 in the region (Kritzberg and Ekström 2012). In the coastal waters, significant decreases of inorganic N and P have been observed since 1990 (Frigstad and others 2013) but eutrophication remains an issue, and the coastal waters are classed as a ''problem area'' by the Convention for the Protection of the Marine Environment of the North-East Atlantic (Wesslander and others 2016). The consequences of these large-scale macronutrient changes are uncertain but potentially profound. Increasing DOC concentrations may fundamentally alter light and energy supplies, thus shifting the balance of autotrophic and heterotrophic processes, and change the entire aquatic food web structure (Båmstedt and Wikner 2016; Traving and others 2017; Creed and others 2018). Coastal water darkening has occurred in the region (Sandén and Håkansson 1996; Aksnes and others 2009; Frigstad and others 2013) resulting in suggested regime shifts from visual predators (that is, fish) to tactile predators (that is, jellyfish) (Eiane and others 1999; Sørnes and Aksnes 2006). Furthermore, the shift from nutrient limitation to light limitation has facilitated the widespread growth of filamentous algae since the 1940s (Johansson and others 1998; Eriksson and others 2002) at the expense of the previously extensive kelp beds (Moy and Christie 2012). There has also been a catastrophic loss of 60% of seagrass cover since the 1980s (Baden and others 2003; Nyqvist and others 2009) which in turn led to increased sediment suspension, thus further hindering seagrass recovery (Moksnes and others 2018). The exact cause of these seagrass declines is unclear, but it has been suggested that eutrophication and water clarity are both implicated (Rask and others 1999; Baden and others 2003). As well as the ecological implications, there are societal ramifications of these changes, because kelp and seagrasses are key habitats for commercially important fish (Heck and others 2003). Additionally, seagrasses can be greenhouse gas sinks (Ollivier and others 2022) and play a disproportionately large role in carbon sequestration (Pendleton and others 2012). Seagrass destruction results not only in the loss of C sequestration capacity but also in the release of C accumulated over centuries.

In this study, we investigated long-term changes in the concentrations and ratios of C, N and P, throughout the aquatic continuum on the west coast of Sweden. Our aim was to determine whether any trends were related to land cover within watersheds, and atmospheric deposition, and whether they were consistent from headwaters to the sea. To do this, we analyzed three decades of macronutrient data from thirteen lakes and seven watercourses, which varied in catchment size by seven orders of magnitude (0.037–40,000 km^2) and encompassed a range of land cover (forest, wetland, arable).

Methods

Study Region and Overview

Our study region was focused on the west coast in Västra Götaland county in western Sweden. It extends approximately 100 km inland (east from the west coast) and ranges approximately 170 km parallel to the coast (north–south) (Supplementary Figure 1). The region covers temperate and hemiboreal vegetation zones (Sjörs 1999), and the dominant land cover for the county is forest (61% of the terrestrial area) followed by arable land (20%) (Statistics Sweden 2015). Mean annual temperature is 7.4 °C and annual precipitation is 833 mm at Vänersborg, located approximately in the center of the study region. We used data from the Swedish national monitoring program (Miljödata-MVM 2021; Fölster and others 2014), and selected nineteen sites in the region that had the longest (> 30 y) water chemistry records for all nutrients of interest (see next section). We supplemented these with data from an additional long-term monitoring site maintained by the Swedish Environmental Research Institute (IVL) (site ''F1,'' Persson and Broberg 1985; Moldan and others 2018). Together, this gave thirteen lakes (ranging in area 0.16–5440 km^2) and seven watercourses (including a headwater stream, large rivers, and river mouths), with catchments ranging 0.037–40,000 km^2. Sites included the largest source of water to the coastal zone in this region (Göta Älv, Sweden's largest river, mean annual flow rate = 540 m^3/s, SMHI 2022), which was sampled about 70 km from the coast. Most sites were independent of each other, with the exception of the following: ''Dalbergsån Dalbergså'' and ''Upperudsälv. Köpmannebro'' are two rivers that both flow into ''Megrundet N,'' which is a sampling point within the great lake Vänern. The outflow of Vänern is the Göta Älv, which is also sampled as ''Göta Älv Trollhättan.'' The small lake ''Alsjön'' also contributes water to ''Göta Älv Trollhättan.'' Elsewhere, water from the small lake ''Rotehogstjärnen'' contributes to the river ''Enningdalsälv N.Bullaren.'' The dominant land cover of these twenty catchments is forest and also includes variable amounts of wetland, arable, and waterbodies (Table 1).

Water Chemistry Measurements

Water chemistry measurements covered the period 1987–2020, except for two sites (''Ejgdesjön'' and ''Stora Härsjön'') where records started in 1989. Data were available for every year except for the

Table 1. List of Study Sites

Site	Waterbody type	Catchment area (km^2)	Catchment land cover (%)				Lake area (km^2)	Average lake depth (m)
			Open wetland	Arable	Lakes and watercourses	Forest		
F1	Headwater stream	0.037	0	0	0	100	–	–
Granvattnet	Headwater lake	0.92	3	1	20	68	0.20	1.6
Stora Galten	Headwater lake	1.30	0.4	4	24	69	0.29	9.5
Torrgårdsvattnet	Lake	1.48	0.3	0	29	70	0.40	9.7
Alsjön	Lake	1.55	9	0	5	84	0.056	*No data*
Härsvatten	Lake	1.90	3	0	21	76	0.24	*No data*
Rotehogstjärnen	Lake	3.45	12	0	5	82	0.16	3.6
Ejgdesjön	Lake	4.33	5	0.3	26	62	0.86	7.0
Fräcksjön	Headwater lake	4.61	2	0	6	82	0.27	4.1
Stora Tresticklan	Lake	8.36	4	0	15	80	1.29	*No data*
Västra Solsjön	Lake	9.13	0.6	1	24	71	1.85	12.3
Bodasjön	Lake	9.9	1	0.1	14	82	1.23	*No data*
Stora Härsjön	Lake	25.1	3	0.6	23	66	2.57	15.7
Bäveån Udde-valla	River	286	5	11	6	68	–	–
Enningdalsälv N.Bullaren	River	572	4	7	10	73	–	–
Dalbergsån Dal-bergså	River	832	7	33	5	49	–	–
Örekilsälven Munkedal	River	1333	5	14	4	71	–	–
Upperudsälv. Köpmannebro	River	3055	2	3	17	74	–	–
Megrundet N	Lake	39,307	4	11	21	58	5440	27
Göta Älv Troll-hättan	River	39,508	4	11	21	58	–	–

Sites are arranged according to catchment area, and details of land cover, lake area and lake depth are given. Note that totals do not sum to 100% as not included are the land cover categories ''other open land'' and ''exploited land.'' For four sites, the catchments extend outside of the land cover layer, and land cover data for these portions of the catchments are missing. The missing portions represent the following % of total catchment area: Enningdalsälv N.Bullaren 4%, Göta Älv Trollhättan 1%, Upperudsälv. Köpmannebro 3%, Megrundet N 1%.

site ''Dalbergsån Dalbergså'' in 2013. Sampling frequency per year was mostly consistent for each site but varied between sites; mean number of samples collected per year ranged from 3.6 to 23.2. For the nineteen sites sampled as part of the Swedish national monitoring program, all water chemistry analyses were carried out at the Geochemical Laboratory at the Swedish University of Agricultural Sciences. The laboratory has been SWEDAC accredited since 1992 and all current and historical analytical methods are well-documented (https://www.slu.se/en/departments/aquatic-sciences-assessment/laboratories/vattenlabb2/). Samples from the site maintained by IVL were analyzed at their laboratory.

We collated measurements of ammonium (NH_4), nitrite + nitrate ($NO_2 + NO_3$), total phosphorus (TP), total nitrogen (TN), and total organic carbon (TOC). We adjusted TP concentrations for the period January 1991 to June 1996 by subtracting 1.2 µg/l, due to an error introduced in the sample analysis by an unstable baseline on an instrument (see Sonesten and Engblom 2001, for details concerning the error). Samples from site ''F1'' were filtered at 0.45 µm before TOC analysis, and therefore represent dissolved organic carbon (DOC). However, particulate organic carbon typically comprises less than 0.6% of TOC in streams draining small, forested catchments (Laudon and others 2011), and so we assume DOC concentrations in F1 ≈ TOC concentrations. Total inorganic nitrogen (TIN) was calculated as the sum of NH_4 and $NO_2 + NO_3$, and total organic nitrogen (TON) was calculated by subtracting TIN from TN. Concentrations were converted to moles per liter, and we calculated mean annual concentrations. The following long-term molar stoichiometric ratios were then calculated using the annual means of C:P, C:N, N:P, C:TON and C:TIN. Note that throughout the manuscript, ''C'' in all ratios refers to organic C only.

Statistics

There were considerable differences in the means and ranges of C, N and P, across the dataset over time and between sites as expected due to differences in land cover and waterbody position within the landscape. We therefore used Z scores which allow time series data to be standardized to visually compare underlying trends across multiple nutrients (for example, Evans and others 2010). Z scores were calculated for each site and year, for all determinands, by taking the mean annual concentration or stoichiometric ratio, subtracting the site mean, and dividing by the site standard deviation. We calculated the site mean and standard deviation using the data period 1987–1993 (see Supplementary Table 1), because this allows deviations relative to a starting Z score of zero to be evaluated (Evans and others 2017a). We used MAKESENS 1.0 (Salmi and others 2002) to test for the presence and magnitude of monotonic trends using Mann–Kendall tests (*Z* values) and Sen's slope estimates (*Q* values). To test for effects of land cover on driving changes in C, N and P, we used Spearman correlations between Sen's slopes and percentage land cover, catchment size and lake area, using SPSS 26. For Mann–Kendall tests and Spearman correlations, results were considered significant when $p < 0.05$, and nominal p values are used throughout. Land-cover was assumed to have remained stable over the study period; available statistics show that within the county productive forest land area has remained approximately constant during the study period (Riksskogstaxeringen 2022), pasture area has remained constant between 2003 and 2021, and arable area has decreased by 7% between 1998 and 2020 (Jordbruksverket 2022). To visually compare multi-decadal changes in C:N:P stoichiometry between the start and end of our monitoring period, we used ternary plots. We used the method presented in Smith and others (2017) whereby, after converting to moles, TN concentrations and TP concentrations are multiplied by 6.625 and 106, respectively. This conversion places the Redfield Ratio (C:N:P 106:16:1) in the center of the ternary plot as a reference point. Zones of relative nutrient depletion and co-depletion identified in the ternary plots were taken from Jarvie and others (2018). Note that Smith and others (2017) used inorganic plus organic C in their plots, while Jarvie and others (2018) used inorganic C alone. Here, we use organic C alone. Ternary plots were produced using TRI-PLOT (Graham and Midgley 2000).

Results

Long-Term Changes in Macronutrients

Z scores showed varying responses for different nutrients and stoichiometric ratios, but long-term increases were visually evident for TOC, C:P, C:N and C:TIN (Figure 1). The mean increase in TOC was 0.08 mg l^{-1} y^{-1}, but some sites showed large increases, with the greatest being 0.19 mg l^{-1} y^{-1} at the ''F1'' forest catchment (Supplementary Table 2). Mann–Kendall statistics demonstrated a high degree of coherency in site responses (Figure 2), with increases in C:P (18 significant), C:N (17 significant), and C:TIN (18 significant) at all sites, and TOC increases at all sites but one (15 significant) (Table 2, Supplementary Table 2). Additionally, all but two sites showed increases in C:TON (15 significant). Negative trends were visually evident for inorganic N and TP (Figure 1), and these were reflected in the Mann–Kendall results; mean decreases were 2.9 µg l^{-1} y^{-1} for TIN (14 sites showing significant declines) and 0.06 µg l^{-1} y^{-1} for TP (11 sites showing significant declines). There was no obvious trend in TON for all sites (Figure 1), and where significant changes were detected these were both positive and negative (Table 2). The direction of trends in macronutrient concentrations and stoichiometry was mostly consistent when sites were grouped into lakes and watercourses, with the exception of N:P which, on average, increased in lakes but decreased in rivers (Figure 2), although not all sites showed significant changes in N:P (Table 2).

At the start of the monitoring period (1987), all sites were P depleted relative to C, and five were P and N co-depleted relative to C (Figure 3). In 2020, relative to C, all sites remained P depleted, and eight were P and N co-depleted. Mean percent values from the ternary plots also reveal this movement away from the Redfield Ratio, toward P and N co-depletion, relative to C (Figure 3).

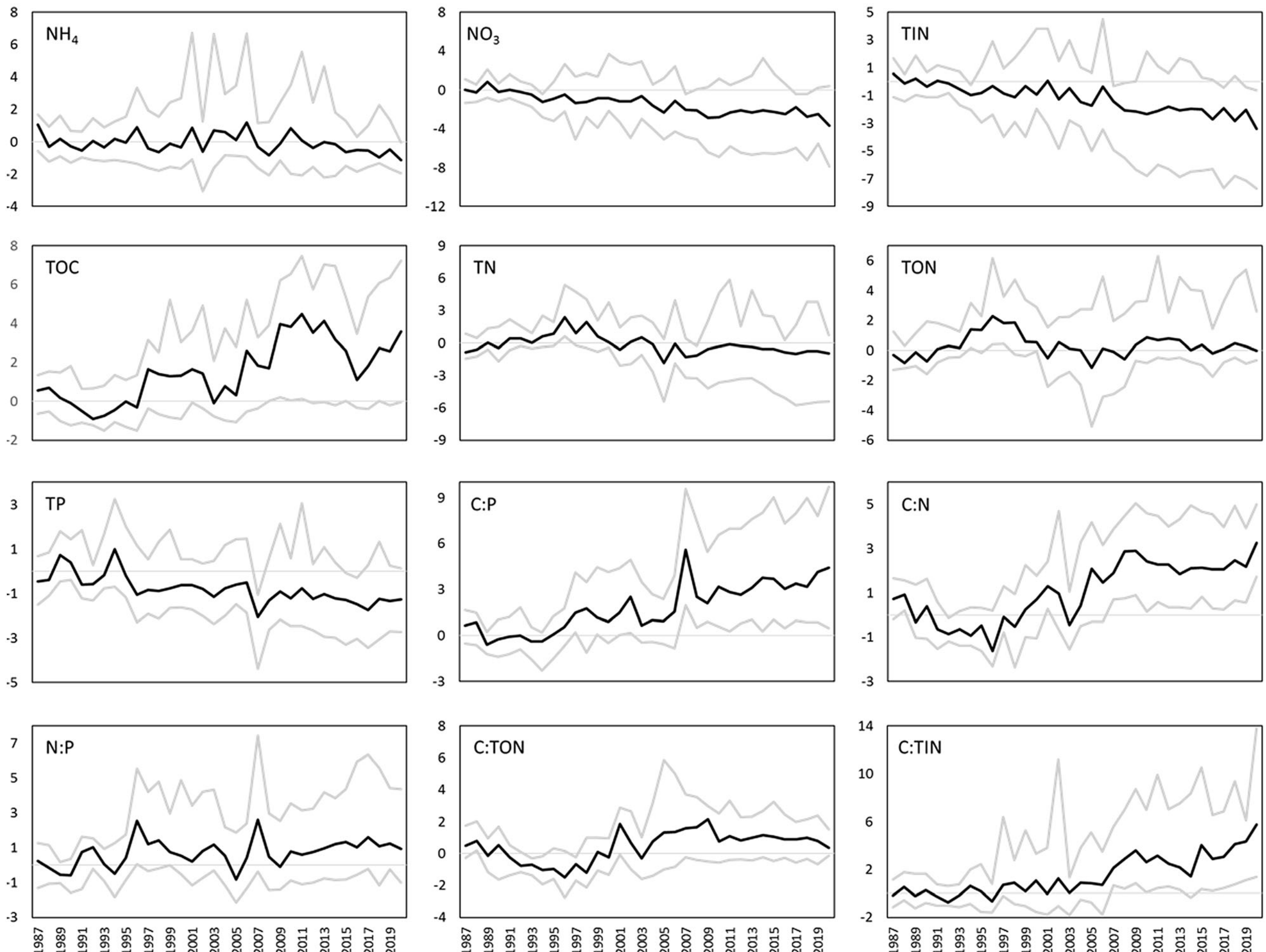

Figure 1. Time series of Z scores for all nutrients and stoichiometric ratios. In each plot, the central black line is the median, and the outlying grey lines are the 10th and 90th percentiles.

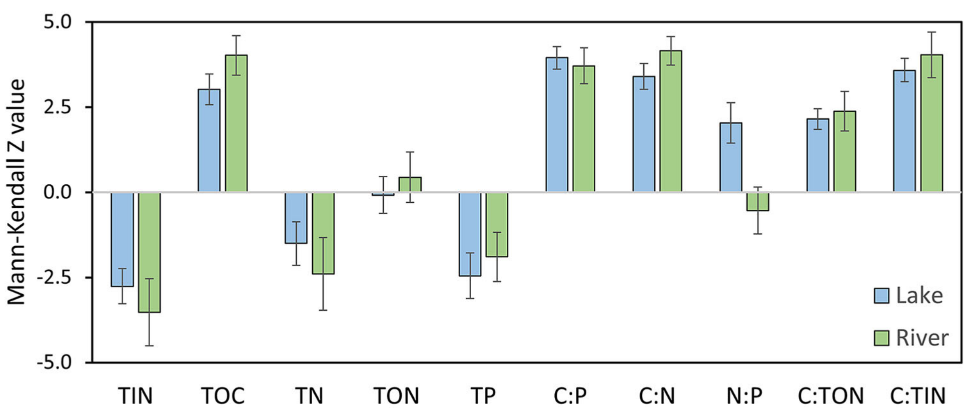

Figure 2. Mean (± standard errors) Mann–Kendall Z values for macronutrient concentrations and stoichiometry, grouped by lakes and rivers/streams. Positive and negative Z values correspond to increasing and decreasing trends, respectively.

Table 2. Mann–Kendall *Z* Values

Site	NH_4	NO_3	TIN	TOC	TN	TON	TP	C:P	C:N	N:P	C:TON	C:TIN
F1	0.7	1.7	1.7	**4.8**	**2.3**	**3.0**	-0.6	**2.7**	**4.5**	1.0	**5.2**	0.2
Granvattnet	-1.3	0.0	-1.4	**4.0**	-0.8	-0.1	0.6	1.5	**3.1**	-0.6	**2.7**	1.9
Stora Galten	-0.4	-1.5	**-2.1**	0.9	-1.3	0.7	**-3.8**	**3.5**	1.4	**2.5**	-0.3	**2.6**
Torrgårdsvattnet	**-4.3**	**-4.6**	**-4.8**	0.0	**-3.5**	-1.8	**-4.2**	**3.9**	**2.8**	**3.5**	**2.0**	**2.6**
Alsjön	0.3	-0.1	-0.3	**2.6**	1.5	**2.1**	-1.9	**3.6**	0.8	**3.0**	0.4	**2.3**
Härsvatten	2.0	**-4.2**	**-2.9**	**5.2**	0.9	**3.3**	1.1	**4.2**	**5.2**	-0.5	**3.3**	**5.4**
Rotehogstjärnen	**2.8**	**-4.3**	-0.5	**4.2**	**2.1**	**2.1**	1.6	**3.0**	**2.8**	1.0	1.2	**2.7**
Ejgdesjön	-1.2	**-4.9**	**-4.9**	**4.4**	**-3.9**	**-2.1**	**-5.9**	**5.9**	**5.3**	**5.1**	**3.8**	**5.3**
Fräcksjön	-1.8	0.3	-1.2	**5.4**	0.3	1.3	-0.7	**3.7**	**3.8**	0.8	**2.6**	**4.0**
Stora Tresticklan	-0.8	**-5.1**	**-4.3**	**4.5**	0.0	1.7	**-3.0**	**5.4**	**4.5**	**3.6**	**2.6**	**4.9**
Västra Solsjön	0.8	**-2.5**	**-2.2**	**2.2**	**-2.8**	**-2.3**	**-5.3**	**5.3**	**3.4**	**4.9**	**2.8**	**2.8**
Bodasjön	**-2.2**	-0.5	-1.8	2.0	**-2.6**	-1.5	**-4.2**	**4.2**	**2.3**	**2.4**	**2.0**	**3.3**
Stora Härsjön	**-2.8**	**-2.6**	**-2.7**	**2.4**	**-3.8**	**-2.5**	**-4.6**	**4.9**	**3.8**	**3.3**	**2.6**	**3.6**
Bäveån Uddevalla	-0.2	**-4.4**	**-3.3**	**4.8**	-1.5	0.0	0.1	**3.7**	**4.2**	-1.6	**2.1**	**5.0**
Enningdalsälv N.Bullaren	-0.4	**-5.1**	**-5.0**	**5.5**	**-3.4**	0.6	**-2.7**	**5.9**	**4.5**	-1.0	**3.0**	**5.2**
Dalbergsån Dalbergså	-0.6	-1.9	**-2.0**	**3.2**	0.2	1.5	0.4	1.6	1.6	-1.1	-0.1	**2.8**
Örekilsälven Munkedal	0.1	**-3.3**	**-3.5**	**5.3**	**-2.6**	-0.9	**-2.0**	**4.7**	**4.2**	-1.0	**2.7**	**5.1**
Upperudsälv. Köpmannebro	**-2.6**	**-6.1**	**-5.7**	**3.7**	**-5.2**	**2.1**	**-5.4**	**4.7**	**5.1**	**3.0**	1.1	**5.2**
Megrundet N	**-3.4**	**-6.9**	**-6.9**	1.6	**-5.6**	-1.9	-1.5	**2.4**	**5.1**	**-2.4**	**2.2**	**5.3**
Göta Älv Trollhättan	-0.9	**-6.9**	**-6.9**	0.8	**-6.5**	**-3.3**	**-3.2**	**2.6**	**5.0**	**-3.1**	**2.6**	**4.8**
	μg N l^{-1}	μg N l^{-1}	μg N l^{-1}	mg l^{-1}	μg N l^{-1}	μg N l^{-1}	μg l^{-1}					
Mean change	-1.8	-91	-96	2.7	-81	12.0	-1.9	1809	8.0	41	9.7	37
Median change	-3.7	-64	-69	2.9	-93	7.5	-2.7	1251	8.4	18	11.0	29

Z values are given for all nutrients and stoichiometric ratios. Sites are arranged according to catchment size (smallest to largest). Significant trends are indicated by bold, black font. Colors indicate positive (green) and negative (blue) trends. Also shown at the bottom of the table are the average changes (as concentrations for nutrients, or values for ratios) for all sites between the start and end of the monitoring period (calculated as the average Sen's slope multiplied by 33 years).

Effects of Land Cover on Long-Term Macronutrient Changes

Fifteen significant correlations were identified between land cover, which was assumed fixed over the study period, and changes in macronutrient concentrations and stoichiometry (Table 3). Additionally, significant negative correlations were detected between forest and arable cover, and forest and lake cover (Table 3). The greatest declines in inorganic and organic N were found for sites with more arable land/less forest cover (Table 3, Figure 4F), but TON trends were also related to lake cover (Figure 4B): sites with greater lake cover within their catchment tended to show decreasing TON, while sites with less lake cover tended toward increasing TON. Increases in TOC were also correlated negatively with lake cover and positively with forest cover (Table 3, Figure 4A). Analyses of just the lake sites also revealed negative relationships between individual lake area and Sen's slopes for TOC and TON (Supplementary Table 3). The greatest increase in TP occurred in the catchment with greatest arable cover (33%, Dalbergsån Dalbergså), and there was a significant positive correlation between change in TP and arable cover (Figure 4D). However, that relationship was driven entirely by the 33% arable cover site; removing that site from the analysis produced no significant correlation. Thus, there was no overall relationship between long-term changes in TP concentration and any land cover type. However, significant decreases in TP were observed for forest lakes with zero (''Stora Tresticklan'' and ''Torrgårdsvattnet'') or minimal ($\leq$ 1%, ''Ejgdesjön,'' ''Bodasjön,'' ''Västra Solsjön,'' ''Stora Härsjön'') arable land cover within the catchment (Figure 5, Table 2). Three other forest lakes (''Alsjön,'' ''Härsvatten'' and ''Rotehogstjärnen'') had no significant trends in TP.

When considering drivers of stoichiometric change, arable cover was shown to be important, showing negative correlations with change in C:P (Figure 4E) and N:P (Table 3). A correlation between arable cover and change in C:TIN was only significant if the high arable cover site was included (Table 3), but there was a significant positive cor-

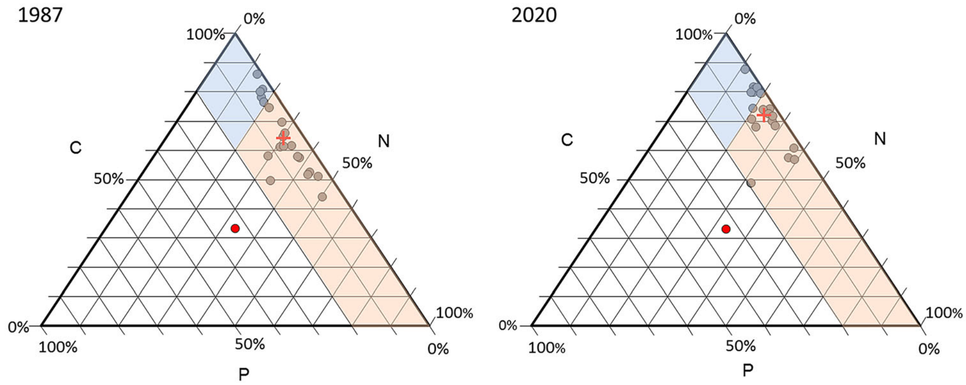

Figure 3. Ternary plots showing the relationships between C, N and P for all twenty sites (black circles) at the start (1987, left panel) and end (2020, right panel) of the monitoring period. The red cross indicates the mean of all twenty sites. The red point at the center of the plot marks the Redfield ratio (C:N:P 106:16:1) as a reference point. The pale orange trapezoid marks the region where P is depleted relative to N and/or C and the pale blue diamond marks where P and N are co-depleted relative to C (regions defined in Smith and others (2017) and Jarvie and others (2018).

relation between change in C:TIN and forest cover (Figure 4C). No significant correlations were found between macronutrient changes and wetland cover (Table 3).

Land use and catchment size were confounded, with a highly significant relationship between size and arable cover (rho = 0.69, $p = 0.001$). Significant correlations between Sen's slopes and catchment size were found for NO_3, TIN, TN, C:P, N:P (Supplementary Table 3), all of which also correlated with arable cover (Table 3), and we therefore assume any effect of catchment size was masked by this confounding.

Discussion

Long-Term Changes in Macronutrients

Our analysis revealed significant long-term changes in macronutrient concentrations in lakes and rivers of the Swedish west coast, with widespread increases in TOC and declines in TP and TIN. These changes have occurred throughout the aquatic continuum from small headwater lakes and streams to river mouths. Increasing TOC concentrations across Europe and North America during recent decades have historically been driven by recovery from atmospheric sulfate deposition (Monteith and others 2007), which results in increased organic matter solubility (Evans and others 2012). However, as sulfate concentrations further decline and begin to stabilize in aquatic ecosystems, climatic warming and changes in precipitation may begin to exert a greater control (de Wit and others 2021). Across much of Sweden concentrations of TOC are no longer rising, but the west coast received comparatively large amounts of sulfate deposition and other recent regional analyses have also detected continuing TOC increases (Isles and others 2018; Deininger and others 2020; Eklöf and others 2021). Within our dataset, greater catchment lake cover was linked to smaller changes in TOC concentrations, and a weaker, negative relationship was found between forest cover and TOC change (note that cover of these two land classes was negatively correlated). The proportion of catchment area occupied by lakes spanned a wide range (0–29%) and thus it is possible that these relationships simply reflect higher total production of TOC in catchments with a higher terrestrial surface area. It is also possible, however, that the generally oligotrophic lakes are acting as net DOC sinks (Evans and others 2017b) and that in-lake removal processes, particularly those with longer residence times, may be offsetting rising terrestrial DOC export (Köhler and others 2013). The data are consistent with either interpretation (Spearman correlation between % lake area and TOC concentration at the start of monitoring was -0.84, $p < 0.001$) but, when considering only the lake sites, the smallest lakes had the greatest TOC increases, which lends credence to the in-lake removal hypothesis. Increasing residence time throughout the aquatic continuum has also been linked to higher C:P, N:P, and C:N, possibly due to in-lake sedimentation of P-rich particles (Maranger and others 2018). Our data did not support this, with no relationship between catchment lake cover and stoichiometric ratios, but this may simply be an artefact of our small sample size ($n = 20$, compared to $n \sim 8500$ in Maranger and others 2018).

Table 3. Correlations Between Land Cover and Macronutrient Changes

		Wetland	Arable	Lake	Forest
NH_4	rho	0.29	-0.04	-0.32	0.33
	p value	0.21	0.85	0.17	0.15
NO_3	rho	-0.42	-0.69	-0.07	0.61
	p value	0.06	0.001	0.76	0.004
TIN	rho	-0.28	-0.75	-0.17	0.72
	p value	0.24	0.000	0.48	0.000
TOC	rho	0.39	-0.26	-0.79	0.59
	p value	0.09	0.27	0.000	0.006
TN	rho	0.1	-0.52	-0.4	0.54
	p value	0.68	0.018	0.08	0.014
TON	rho	0.3	-0.21	-0.58	0.47
	p value	0.2	0.38	0.007	0.038
TP	rho	0.22	0.48	-0.14	-0.26
	p value	0.36	0.031	0.55	0.27
C:P	rho	-0.42	-0.64	0.33	0.39
	p value	0.07	0.003	0.16	0.09
C:N	rho	-0.17	-0.35	-0.12	0.37
	p value	0.48	0.13	0.63	0.1
N:P	rho	-0.36	-0.56	0.35	0.24
	p value	0.12	0.010	0.13	0.31
C:TON	rho	-0.22	-0.32	0.14	0.12
	p value	0.35	0.17	0.56	0.61
C:TIN	rho	0.06	-0.48	-0.25	0.60
	p value	0.8	0.032	0.28	0.005
Wetland	rho		0.26	-0.42	-0.19
	p value		0.27	0.07	0.41
Arable	rho			-0.09	-0.68
	p value			0.72	0.001
Lakes	rho				-0.48
	p value				0.032

Spearman correlation coefficients (rho) and p values for correlations between percentage land cover and Sen's slopes for macronutrient changes. Green shading indicates where significant correlations were found for macronutrients, and blue shading where different land covers were correlated with one another. Significant correlations between arable and TP, and arable and C:TIN are driven by one site with the highest levels (33%) of arable land cover. Removing this site has the effect that these two correlations become non-significant (new values for arable vs TP are rho = − 0.31, p = 0.19, and for arable vs C:TIN are rho = − 0.43, p = 0.07), but no effect on the significance or direction of any other significant correlations.

Our analysis revealed widespread declines in NH_4, NO_3, and TIN, in agreement with findings from Swedish lakes (Isles and others 2018) and rivers draining into southern Norwegian coastal waters (Deininger and others 2020). On a national Swedish level, this region experienced high levels of atmospheric N deposition (> 10 kg ha^{-1} y^{-1}, Emmett and others 1998), which have subsequently declined (Isles and others 2018). For our sites, we found that the greatest declines were in catchments with the most arable land, where TIN concentrations were greatest, as observed in general (Boyer and others 2002) (Spearman correlation between % arable land and TIN concentration at the start of monitoring was 0.72, $p < 0.001$). This suggests that at the regional level declines in TIN are driven not only by changing atmospheric deposition, but also by changing management of agricultural lands. N:P was the only metric with an obvious difference in trends between lakes (increasing) and rivers (decreasing), and we infer this to be driven by the higher agricultural land use in the catchments of our rivers (mean arable cover = 11%, compared to 1% in lake catchments), rather than by any difference in N:P processing in different waterbody types. Contrary to changes in TIN, trends in TON were not coherent and included both significant increases and decreases. This is at odds with reports of increasing TON in Norwegian rivers in the same region which are suggested to be driven by enhanced leaching of organic matter (Deininger and others 2020), although we note

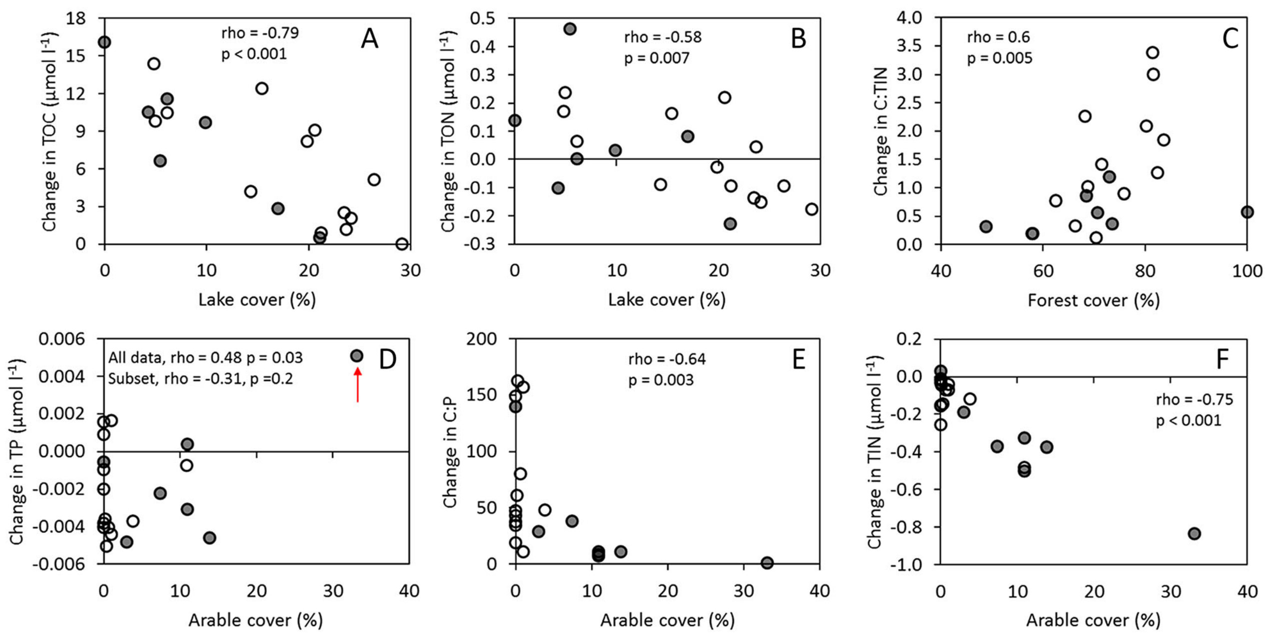

Figure 4. Scatter plots of significant correlations between long-term changes in macronutrient concentrations and stoichiometry (represented by Sen's slopes) and land cover. Open circles are lakes, filled circles are watercourses. Note that the significance of the TP vs arable cover correlation in panel D is driven by the site with 33% arable land (marked by the arrow); removing that site from the statistical analysis produces a non-significant negative correlation.

that long-term trends in organic C and N can sometimes diverge (Rodríguez- Cardona and others 2022). In systems where DOC and DON trends are not coherent over time, we suspect that the DOM being exported during the recovery from acidification period is more N poor, as typical of large hydrophobic polyphenolic and aromatic compounds such as lignins and tannins and peat derived DOM which often has high DOC:DON (Yates and others 2019). The coupling or decoupling of DOC and DON suggests differences in DOM composition over time (or across sites). This can have consequences in terms of DOM being an energy source for the microbial community, particularly since DOM with high DON content often includes easily assimilable peptides and amino acids (Yu and others 2002).

For P, approximately half of the sites experienced significant declines, while no significant trends were found for the other half. This is in keeping with other findings showing widespread, but not universal, P declines in lakes in Sweden (Huser and others 2018; Isles and others 2018) and the Northeastern United States (Stetler and others 2021), sometimes referred to as ''oligotrophication.'' There was no relationship between any land cover and P trends, with significant declines observed both in river catchments featuring agricultural land and in forest lakes. Although reduced agricultural nutrient leaching may contribute to trends at some sites it cannot explain reductions at all sites, implying a broader shift in the nutrient demand and stoichiometry of forest and wetland ecosystems. This oligotrophication is not believed to be related to changes in P deposition, and possible causes include changes in climate, acidification recovery, and forest management (Crossman and others 2016; Huser and others 2018). One suggestion is that climate, forestry and oligotrophication are interlinked whereby rising temperatures lengthen the growing season, resulting in greater P (and N) uptake by vegetation, and therefore reduced hydrological nutrient losses (Lucas and others 2016; Huser and others 2018; Mosquera and others 2022). Conversely, Stetler and others (2021) raise the possibility that P concentrations may yet increase in future, following lags in soil pH recovery from acidification. Clearly, more work is needed to untangle the drivers of these P trends, and questions remain unanswered: how widespread is this oligotrophication, and will it continue in the future?

In addition to decreasing atmospheric deposition, rising temperatures, and changing land management, hydrological changes often explain some degree of shifting macronutrient concentrations and stoichiometry (Tranvik and Jansson 2002; Sebestyen and others 2009; de Wit and others 2016;

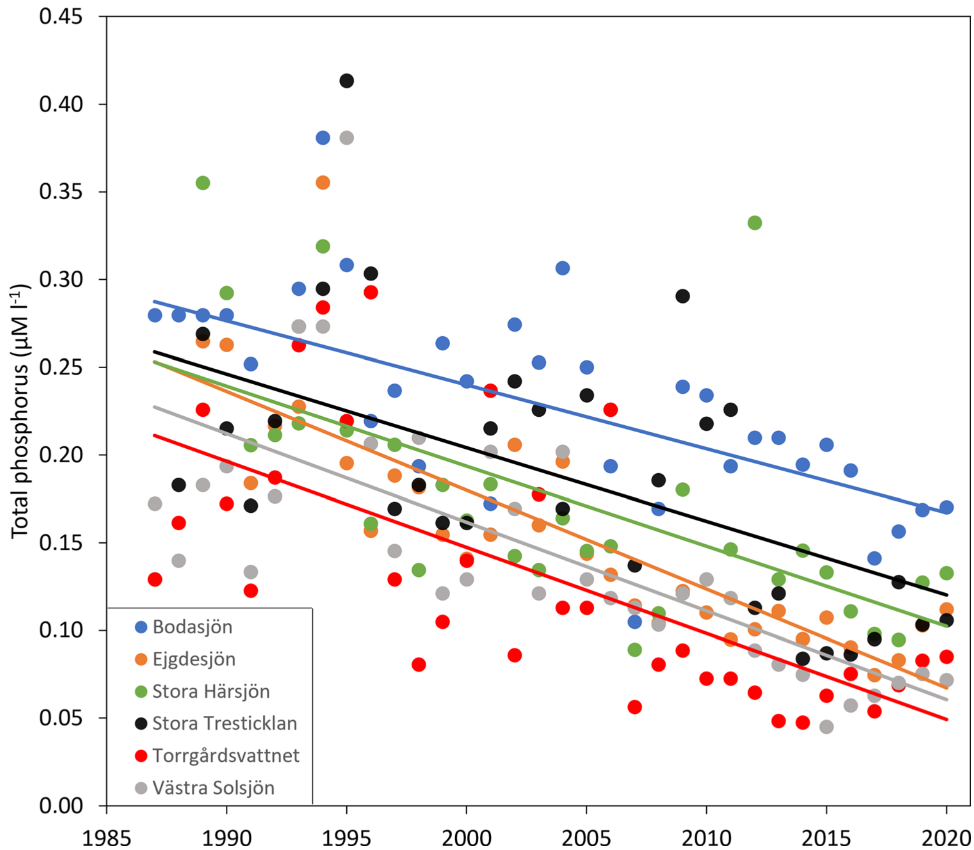

Figure 5. Scatter plots showing mean annual total phosphorus (TP) concentrations for six lakes against sampling year, with linear lines of best fit. All sites are forest lakes with zero/minimal arable land cover within their catchments, and Mann–Kendall tests show all trends are significant (Table 2, Supplementary Table 2).

Maranger and others 2018). Although catchments in south-west Sweden appear to have gotten wetter during the last sixty years, these changes in streamflow (Teutschbein and others 2022) and precipitation are not significant (Chen and others 2021), and our analyses of annual precipitation and flow data for our study region and study period show no significant trends (Supplementary Figure 2). Similarly, we find no change in seasonal flow patterns (Supplementary Figure 3). Although we cannot rule out that more nuanced temporal changes in precipitation or flow might be affecting macronutrient dynamics, these findings do suggest that hydrology has not been a dominant driver of C:N:P stoichiometry during the study period. In addition to hydrology, rising atmospheric CO_2 is known to change the macronutrient stoichiometry of terrestrial biomass, driving increases in plant C:P and C:N (Du and others 2019; Wang and others 2021). If decomposing plant litter, which leaches nutrients into drainage waters, becomes relatively more depleted in P and N, this could result in higher C:P and C:N in waterbodies. However, in our dataset we have no way to discriminate any effect of rising CO_2 from other drivers.

Our findings, along with those of others (Gilbert 2010; Frigstad and others 2013; Isles and others 2018; Deininger and others 2020; McElarney and others 2021; Rodríguez- Cardona and others 2022), show that human activity, principally decreasing S and N deposition, has had a clear impact on the multi-decadal stoichiometric balance of aquatic ecosystems. On the Swedish west coast, the cumulative effect of three decades of changes in C, N and P concentrations has led to a doubling in mean C:P, and increases in C:N and N:P by 50% and 30%, respectively. Thus, the stoichiometry of these waterbodies is moving further away from the Redfield Ratio and becoming even more imbalanced (Elser and others 2022). The large rise in TOC concentrations has also led to increases in C:TIN and C:TON. Because land cover is forestry-dominated, organic C concentrations were already large at the start of the monitoring period, whilst P was low. Long-term changes have resulted in lakes and rivers becoming even more C-rich, and P-depleted. Declining N has resulted in more sites being co-depleted in both P and N, relative to C. It is important to note, however, that these nutrient changes are relative (for example, the ternary plots in Figure 2), and thus only suggest potential limitation; even if N:P is low, N may not be limiting if absolute concentrations are high enough (Bothwell 1985).

Effects of Long-Term Macronutrient Changes

Taken together, our results show that the Swedish west coast is clearly experiencing changes in both macronutrient concentrations and stoichiometry, which could be contributing to observed environmental problems. We found that these changes were not restricted to headwaters but were also evident in larger rivers and at river mouths; thus, it is evident that there is potential for a direct effect on the marine environment. Increases in TOC at the river mouths (sites ''Bäveån Uddevalla'' and ''Örekilsälven Munkedal'') was about 0.13 mg l^{-1} y^{-1}, translating to a total of 4.3 mg l^{-1} over the entire 1987–2020 period (representing an increase of 46% relative to our 1987–1993 baseline). The effect of organic C on aquatic food webs depends on other variables and can be unimodal (Finstad and others 2014). Initial increases in organic carbon can be a useful energy source (Cole and others 2011) but increasing concentrations can reduce light penetration and primary production (Jones 1992). As such, increasing TOC concentrations could be contributing to reported coastal darkening in the region (Sandén and Håkansson 1996; Aksnes and others 2009; Frigstad and others 2013) which is promoting the abundance of jellyfish at the expense of fish (Eiane and others 1999; Sørnes and Aksnes 2006), and has been implicated in the expansion of filamentous algae and the decline of kelp and seagrass beds (Johansson and others 1998; Eriksson and others 2002; Brun and others 2003; Moy and Christie 2012). It is also worth noting that relationships between TOC concentrations and light regimes may be non-linear, because the composition of organic carbon may also be undergoing long-term changes (Jane and others 2017). A detailed analysis of organic matter trends at the F1 catchment in our dataset (Evans and others in prep.) suggests that visible color may be rising faster than bulk DOC, implying an increased proportion of light-absorbing compounds, and the same finding has been reported for nearby lakes in south-east Norway (Xiao and Riise 2021), as well as more distant locations (for example, lakes in the Northeastern United States, Stetler and others 2021). Although we have no data on organic C composition across the wider dataset, the changes we see in organic C:TON stoichiometry are themselves indicative of fundamental shifts in the composition of organic matter (Wymore and others 2021; Rodríguez- Cardona and others 2022), and so a steady-state in dissolved organic matter composition cannot be assumed. Changes in N and P at the two river mouths were more equivocal than TOC changes: NO_3 and TIN decreased at both, and significant declines in TN and TP only occurred at ''Örekilsälven Munkedal.'' The largest river in the region (Göta Älv) (mean annual flow rate = 540 m^3/s, SMHI 2022), had a decline in TP, the largest decline in TN, and the second largest decline in TIN, but no trend in TOC. The lack of change in TOC is likely because this river drains the great lake Vänern (area = 5650 km^2) (refer to earlier discussion). Altogether, the results from these three sites suggest that coastal darkening may be localized to areas near the outflows of smaller rivers, while a reduction in TIN and TP is likely to be more widespread throughout the wider coastal zone, although further N and P declines are required for coastal waters to no longer be classified as eutrophic (Wesslander and others 2016).

In addition to affecting the coastal zone, these long-term stoichiometric changes will almost certainly have caused ecological changes within the sampled lakes and watercourses. For example, rising N:P and C:P in aquatic ecosystems (including in forested, oligotrophic lakes similar to those in our study) can disrupt food webs, because P-limitation in phytoplankton cascades down to P-limitation in zooplankton (Elser and Urabe 1999), and these effects can even translate into enhanced methane production within the water column (Elser and others 2022). Increasing N:P can also increase the prevalence of fungi that parasitize phytoplankton (Frenken and others 2017). Furthermore, the release of toxins by phytoplankton is partially regulated by C:N:P, whereby increasing N:P stimulates production of toxic N-rich secondary metabolites, while C-rich toxins become more abundant under N and P limitation (Van de Waal and others 2014). Thus, the long-term macronutrient changes we report are likely to be causing ecological changes throughout all components of the food web and across the aquatic continuum.

To conclude, it is evident that extensive changes have taken place in the biogeochemistry of waters draining from source to sea on the Swedish west coast, where long-term increases in TOC and decreases in TP and TIN, have resulted in widespread stoichiometric change: a doubling in mean C:P, respective increases in C:N and N:P of 50% and 30%, and significant increases in C:TIN and C:TON. Although not explicitly tested here, we assume that these changes have consequences for ecosystem functioning, although further studies are needed to test this. These are anthropogenic shifts, and changes in nutrient stoichiometry and organic matter composition have further implications for carbon burial, greenhouse gas emissions, food webs, metal transport and drinking water supplies (Xenopoulos and others 2021). As highlighted by Graeber and others (2021), aquatic nutrient stoichiometry has never been in stasis, regardless of human activities, and changes will continue to occur. By tracing past multi-decadal changes in C:N:P in the rivers and lakes of the Swedish west coast, our work provides a starting point to understand the trajectories of long-term change in European and North American waters that have been subjected to historical S and N atmospheric deposition. From this starting point arises the opportunity to mitigate and manage for upcoming changes to sustain the health of fresh and marine water ecosystems and safeguard their futures.

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OPEN ACCESS

DATA AVAILABILITY

Our data are from the Swedish National Monitoring program and the link to all of their data is: https://miljodata.slu.se/mvm/

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