Persistent and widespread long-term phosphorus declines in Boreal lakes in Sweden

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H I G H L I G H T S

- Substantial, long-term declines in TP were detected in boreal lakes across Sweden.
- Lake trophic status has changed in many lakes to ultra-oligotrophic.
- Increased reliance on forest biomass and hydropower will likely exacerbate P declines.
- Legislation is poorly equipped to handle impairments caused by oligotrophication.

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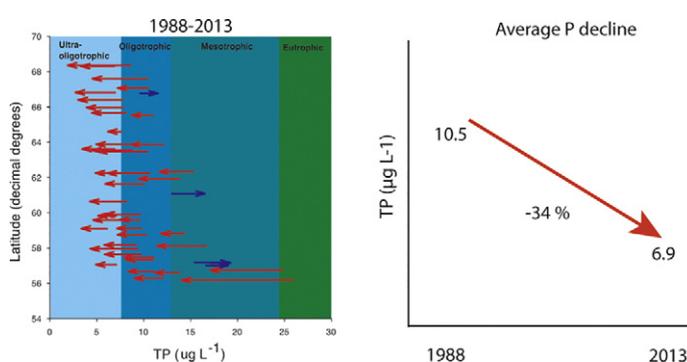
A B S T R A C T

We present an analysis of long-term (1988–2013; 26 years) total phosphorus (TP) concentration trends in 81 Swedish boreal lakes subject to minimal anthropogenic disturbance. Near universal increases in dissolved organic carbon (DOC) concentrations and a widespread but hitherto unexplained decline in TP were observed. Over 50% of the lakes (n = 42) had significant declining TP trends over the past quarter century (Sen's slope = 2.5% y^{-1}). These declines were linked to catchment processes related to changes in climate, recovery from acidification, and catchment soil properties, but were unrelated to trends in P deposition. Increasing DOC concentrations appear to be masking in-lake TP declines. When the effect of increasing DOC was removed, the small number of positive TP trends (N = 5) turned negative and the average decline in TP increased to 3.9% y^{-1}. The greatest relative TP declines occurred in already nutrient poor, oligotrophic systems and TP concentrations have reached the analytical detection limit (1 μg L^{-1}) in some lakes. In addition, ongoing oligotrophication may be exacerbated by increased reliance on renewable energy from forest biomass and hydropower. It is a cause of significant concern that potential impairments to lake ecosystem functioning associated with oligotrophication are not well handled by a management paradigm focused exclusively on the negative consequences of increasing phosphorus concentrations.

1. Introduction

Freshwater productivity can be limited by a number of factors including nutrients (nitrogen, phosphorous, carbon) and light. Phosphorus (P), often the limiting nutrient in lakes, is regulated by atmospheric inputs,

point and diffuse sources, and processes occurring in the lake and surrounding catchment. Human activity has altered freshwater P availability for centuries; however, most concern has focused on increasing anthropogenic P inputs and subsequent problems associated with eutrophication (Smith and Schindler, 2009). While the prevailing scientific and management paradigm has been that less P is better, studies have shown the deleterious effects of P declines (oligotrophication) on freshwater ecosystems, e.g. reservoirs (Stockner et al., 2000). This is a concern as widespread oligotrophication, which cannot be ascribed to direct anthropogenic pressures, is occurring in lakes and streams across the northern hemisphere (Arvola et al., 2011; Eimers et al., 2009; Hu and Huser, 2014; Stammler et al., 2017; Yan et al., 2008).

There are many anthropogenic and natural factors in the lake and surrounding catchment that can influence in-lake total phosphorus (TP) concentrations (Eq. (1)). These can be broken down into processes operating in the catchment that affect external P loads (L_{Ext}; g yr^{-1}), in lake processes regulating internal loading (L_{Int}: g yr^{-1}), in-lake retention (R_S; proportion as sedimentation), and climate-related and in-catchment processes controlling runoff (Q: m^3 yr^{-1}) (Nurnberg, 1984).

$$TP = \frac{L_{Ext} + L_{Int}}{Q}(1-R_S) \qquad (1)$$

As can be seen from Eq. (1), declines in P concentration consistent with oligotrophication can occur due to a decline in loading from internal and/or external sources, an increase in retention, an increase in runoff, or some combination of these factors. Oligotrophication may occur when external loads are reduced as a result of in-catchment processes including e.g. declining deposition, alterations in land management, increased uptake by terrestrial vegetation, or changes in P solubility. In-lake processes (e.g. thermal stratification) can alter sedimentation rates and geochemical factors controlling internal loading and retention. Changes in the timing and amount or runoff will also affect in-lake TP concentrations by influencing both delivery and flushing rates. These factors and the mechanisms by which they may lead to oligotrophication are summarized in Table 1.

Recovery from acidification can alter external and internal loading rates as well as retention, primarily through pH-mediated changes to fluxes of aluminum (Al) to lakes and sediments and dissolved organic matter (DOM) solubility (Huser and Rydin, 2005; Kopáček et al., 2008). Increased DOM fluxes may lead to increased external P loading as dissolved organic phosphorus (DOP) is a component of DOM. Declines in sulfate deposition since the 1980s may increase available soil P-binding sites due to less anionic competition (Geelhoed et al., 1997). However, recovery from acidification may also reduce external P loading because soils recovering from anthropogenic acidification may display increased P binding to (oxyhydroxide) metals (Gustafsson et al., 2012) and clay minerals (Gérard, 2016) that can limit P mobility in the watershed and subsequent transfer to lakes.

Climate change can affect external and internal loading, runoff, and potentially retention (Brantley et al., 2011). Warmer temperatures and a longer growing season can lead to increased P uptake by catchment vegetation (Jonard et al., 2015), changes in catchment hydrology caused by increased evapotranspiration (Crossman et al., 2016), and increases in internal loading (Jensen and Andersen, 1992). If warmer temperatures promote increased in-lake productivity, retention may also increase due to increased rates of sedimentation.

Finally, declining atmospheric P deposition may contribute to oligotrophication through a reduction in external loading (Zhai et al., 2009). Combustion-related sources of atmospheric P deposition related to combustion have decreased over Europe since the 1980s (Wang et al., 2015b); however, few long-term trends for total (natural plus anthropogenic) P deposition have been detected (Tipping et al., 2014) and none were detected at Canadian sites where surface water P declines have occurred (Eimers et al., 2009).

In boreal lakes, most water column P is present in organic forms, most of which are associated with DOM (Palviainen et al., 2016). Brownification, related to increasing surface water dissolved organic carbon (DOC) concentrations (a proxy for DOM), has been reported across much of the northern hemisphere and has been ascribed to recovery from acidification (Monteith et al., 2007) and a changing climate (Haaland et al., 2010; Hongve et al., 2004). If there are no changes in hydrology or DOM stoichiometry, increasing DOM fluxes from watersheds to lakes should also increase rates of external loading and in-lake P concentrations (Eq. (1)), but this has clearly not been the case in other studies (Arvola et al., 2011; Eimers et al., 2009; Hu and Huser, 2014; Yan et al., 2008).

Furthermore, darker conditions associated with increased DOC concentrations can alter the energy budget of lakes. Increasing DOC concentrations limit light penetration, enhance thermal stability, and as a result can lead to cooler bottom-water temperatures during the summer (Tanentzap et al., 2008). Reduced light can potentially lower rates of primary productivity (Thrane et al., 2014) and shift aquatic ecosystems from nutrient to light limitation (Karlsson et al., 2009). Stronger and longer thermal stratification can effectively isolate bottom waters from the atmosphere. This may lead to increased bottom water anoxia and internal P loading (Spears et al., 2017). While there is a body of work documenting plausible drivers of temporal P trends (summarized in Table 1), none of the aforementioned mechanisms have been directly linked to the widespread surface water oligotrophication that has been observed across the northern hemisphere.

In this study we assessed long-term (1988–2013) changes to total P (TP) concentrations in a large (N = 81) dataset of Boreal lakes across Sweden. All lakes had minimal, direct anthropogenic disturbance in the watershed (e.g. point sources, urbanization or agriculture) or to the lake itself (e.g. liming). Multiple linear regression models were developed to relate TP trends to factors including land-use, catchment soil properties, climate, deposition and in-lake processes. Policy-related implications of declining P concentrations are discussed, and potential exacerbating factors e.g. hydropower and forest harvesting are examined as well.

2. Methods

2.1. Data collection and handling

To quantify temporal variation in lake water chemistry and assess possible correlates of declining TP in surface waters, we used long-term (1988–2013) Swedish national monitoring program data. The data set originally included 110 lakes (trend lakes covering latitudes 56–68 °N) with no point sources of pollution. All lake data were collected as part of the Swedish national monitoring program (Folster et al., 2014) and analyzed at the water chemistry laboratory (accredited by the Swedish Board for Accreditation and Conformity Assessment) at the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences. Epilimnetic samples (0.5 m water column depth) were collected four times per year (spring, summer, fall, winter) and analyzed for a suite of parameters (Table 2). Further details on methods, detection limits, and quality control can be found at the following website: http://www.slu.se/en/departments/aquatic-sciences-assessment/laboratories/geochemical-laboratory/water-chemical-analyses/. The dataset, at least to the authors' knowledge, is unprecedented for its temporal duration, spatial extent (covering nearly the entire latitude range for the Boreal ecosystem) and internal consistency of laboratory methods. Only lakes minimally affected by urbanization (<1% watershed area) or agriculture (< 5% watershed area) were included in the analysis, reducing the original dataset to 81 lakes. Outliers in water quality time series were identified by log transforming all data and excluding any values outside ± 3 standard deviations from subsequent analysis. DOC was determined as total organic carbon (TOC). This was considered acceptable because

Table 1

Summary of potential drivers of changing lake P concentrations and their effect on components of the equation for estimating steady state P concentrations: external loading (L_{Ext}), internal loading (L_{Int}), retention (R_S) and runoff (Q). Plus symbols (+) denote a positive relationship between the driver and the component of the equation; minus signs (−) a negative relationship and a blank is indicative of no causal relationship. Increases in loading will lead to an increase in P concentration while increases in either runoff or retention will result in lower concentrations.

Driver	Reference	Comments	L_{Ext}	L_{Int}	R_S	Q
Increasing TOC	Monteith et al. (2007) Spears et al. (2017) Thrane et al. (2014)	(i) TOC is a proxy for DOM. In northern lakes, a significant fraction of the TP is present in an organic form, which is derived from allochthonous (in catchment) production. If DOM stoichiometry remains constant, there should be a simultaneous increase in TP mediated via increased external loading.	+			
		(ii) TOC will associate with Al and Fe, reducing the capacity for formation of metal-(oxy)hydroxides in soils. If this mechanism is quantitatively important, there may be an increase in TP as there will be a potential for greater release of P from catchment soils and consequent increase in external loading, but a decrease in internal loading due to increased binding in the sediment by Al and Fe (depending on lake type and redox conditions) and an increase in retention associated with increased rates of organic matter (OM) precipitation.	+	−	+	
		(iii) Increasing TOC will intercept light more effectively, leading to warmer surface waters, greater thermal stratification, and a darker water column. More intense thermal stratification will increase the likelihood of bottom water anoxia and sediment P release from Fe and manganese. However, a darker water column may reduce rates of primary productivity and potentially oxygen depletion in bottom waters.		+	−	
Increasing iron	Kritzberg and Ekström (2012), Huser et al. (2011, 2012) Neal et al. (2008)	Widespread increases in surface water Fe concentrations have been reported, associated with a recovery from acidification and with wetter soils. Fe^{3+}-P complexes are generally insoluble. Wetter soils leading to greater redox-mediated reduction of Fe (Fe^{2+}) would be consistent with an increase in surface water TP as it would result in increased P mobilization from the catchment. Within the lake, an increase in soluble Fe would be consistent with an increase in internal loading. However, increased Fe concentrations in the water column could support higher rates of in-lake P precipitation and an increased sediment sorption capacity.	+	−	+	
Recovery from acidification	Kopáček et al. (2015) Moldan et al. (2013) Huser and Rydin (2005)	(i) P solubility is strongly pH dependent. Changes in soil pH associated with acidification may have profound effects on soil solution P concentrations. Depending on pre-existing conditions, recovery from acidification could lead to either a decrease on increase in surface water TP, if soils continue to acidify (as is happening in much of Sweden), Thus, there could be positive and negative effects on external loading.	±			
		(ii) Recovery from acidification is generally associated with a reduction in soil and surface water Al concentrations. Recovery could lead to greater P binding in watershed soils, leading to reductions in external P loads.	±	+	−	
		Less Al should lead to an increase in TP as there will be less formation of Al-P complexes, potentially increasing both external and internal loading. However, this is relatively unimportant because Al-PO_4 − P (direct) binding generally occurs at very high P concentrations. Precipitation of $AlOH_3$ at the sediment water interface (where pH is high enough) occurs in acidified lakes, but sorption of available P in sediment pore-water is not likely. Instead, Al probably scavenges P directly from Fe. This may explain the accelerated accumulation of Al-P in lake sediment after initiation of liming, because formation of Al(OH)3 could occur in the entire water column instead of a small zone at the sediment-water interface.				
		Declines in Al export from the catchment could contribute to an increase in internal loading and a decline in retention. This is corroborated by the declining accumulation of Al and Al-P observed in a southern Swedish lake recovering from acidification.				
		(iii) Recovery from acidification is a main driver of increasing TOC trends and has been implicated in the increase in Fe. (see above for potential mechanisms)	+	+	±	
Changing deposition	Stoddard et al. (2016), Wang et al. (2015a), Tipping et al. (2014)	Atmospheric P deposition is notoriously difficult to measure and most trend assessments rely on modeled results. However, atmospheric deposition (along with weathering) is the primary mechanism by which P enters a catchment. Thus, any large scale increases or decreases in P deposition should be expected to result in changes in external loading. Large-scale phenomena such as the North Atlantic Oscillation, which influences the degree of winter storminess, and pollution control technologies which reduce the emission of particulates may contribute to trends in P deposition	−			
Weathering	Norton et al. (2011), Landeweert et al. (2001)	Mineral weathering is the ultimate source of most P in the environment. Younger soils weather faster, releasing greater amounts of P. Fungally mediated weathering of P-containing minerals is known to be a relevant process in northern forests. Weathering rates can be influenced by temperature, and potentially site productivity. Any change in weathering rate should be reflected in changes in external loading and hence surface water TP.	+			
Reservoir creation	Stockner et al. (2000), Milbrink et al. (2011)	Reservoir creation is known to produce a "boom and bust" of short-term eutrophication followed by long-term oligotrophication. Eutrophication results from a short term increase in external loading caused by flushing of nutrients from newly flooded land while the oligotrophication results from a combination of factors including longer water residence times, which facilitate higher retention rates (and greater potential for P losses through sedimentation) and disconnection from riparian zones.	+	−	+	
Terrestrial plant growth and mortality	Akselsson et al. (2008), Kana et al. (2013), Crossman et al. (2016)	A significant fraction of the potentially mobilized P in some catchments is stored in terrestrial vegetation. Changes in terrestrial plant biomass can have profound consequences for catchment-scale P mass balance, both by altering P availability and runoff rates. Typically, increased vegetation growth leads to decreases in external loading and potentially decreases in runoff if there are large effects of transpiration.	−			−
		(i) Forest dieback can lead to increased external loading by releasing much of the P stored in vegetation to soil solution and surface waters, leading to potentially long-term increases in mobility. Widespread forest dieback will alter the hydrological cycle within the catchment leading to a greater fraction of precipitation contributing to runoff.	+			+
		(ii) Any increase in terrestrial plant growth, whether due to changes in climate, N deposition, or land management has the potential to sequester more P in vegetation, thereby reducing external loading rates. Long-term changes associated with a warmer climate and increased N deposition are likely leading to higher rates of terrestrial primary production and consequent reductions in surface water P.	−			−
		(iii) Biomass harvesting, whether for agriculture or forestry, will remove P from a catchment, reducing its	−			

Table 1 (continued)

Driver	Reference	Comments	L_{Ext}	L_{Int}	R_S	Q
		availability to surface waters and potentially reduce external loading rates. Most forest harvesting in the boreal is not compensated for by P additions, suggesting a net decline in P availability in managed forests. Any increase in harvest intensity, i.e. whole-tree harvest, improved silviculture or shorter rotation periods, will all lead to a reduction in P available for export to surface waters.				
Liming	Huser and Rydin (2005) Hu and Huser (2014)	By changing lake water and sediment chemistry, liming can have profound effects on in-lake P concentrations. Following liming, internal loading often declines and retention increases due to more effective scaveiging of P in the water column and an increase in sediment sorption capacity by Al, Fe, and Ca. This occurs because transport of P binding metals from the still acidified watershed soils can precipitate through the entire water column (due to greater pH) instead of only in a small zone near the sediment water interface where internal processes can buffer pH decreases due to acidification. Lime also contains significant amounts of Al, Fe, and Ca.	−	+		
Increasing temperature	Moss (2012) Jensen and Andersen (1992) Jeppesen et al. (2009)	Increasing temperature is not likely to have a direct effect on lake P concentrations. Indirect effects include a potential decrease in surface water P concentrations associated with stronger sequestration by terrestrial vegetation as well as a potential increase associated with faster rates of mineral weathering. Warmer temperatures promoting higher rates of in-lake primary production may contribute to increases in internal loading Increased temperature may lead to greater rates of evapotranspiration and a reduction in runoff, which could also decrease external loading.	−	+		−
Changing precipitation patterns	Jeppesen et al. (2009)	Changes in precipitation can alter the mobilization, transport and loss of P. Wetter conditions would support increased reducing conditions in soils, which could result in greater P mobilization. Wetter conditions could also lead to greater runoff and higher flushing rates. Higher flushing rates, and shorter water residence times could be associated with a decrease in sedimentary P losses.	+			+

>95% of organic carbon in boreal lakes is typically present in a dissolved form (Gadmar et al., 2002).

Temperature, precipitation, and runoff data were obtained from the Swedish Meteorological and Hydrological Institute (www.smhi.se). Annual means and temporal trends for temperature, precipitation, and runoff were calculated using these data (Table 2). Watershed boundaries for each lake were delineated using elevation data in a 50-meter grid with a maximum standard error of 2.5 m. Land-use in the catchment was estimated using CORINE (Bossard et al., 2000) land cover data (250-m resolution) that were aggregated for further analyses (Table 3).

Limited watershed soil chemical data were available. The most complete coverage of soil P and metals was available from the Swedish Geological Survey (SGU) database of samples from the 0.5 m soil depth. This dataset included measurements for 55 of the 81 study lake catchments.

Table 2
Mean and standard deviation (SD) for in-lake and climatic variables. Symbols * and † indicate high bivariate correlation with TOC and conductivity, respectively. N = 81 except for P deposition measured (N = 4).

Variable	Unit	Mean ± SD
Absorbance (filtered)*	420 nm/5 cm	0.14 ± 0.12
Absorbance (unfiltered)*	420 nm/5 cm	0.17 ± 0.15
Cl†	meq L^{-1}	0.11 ± 0.1
Conductivity	25 mS/m25	4.4 ± 2.2
NH$_4$	µg L^{-1}	22 ± 16
NO$_3$	µg L^{-1}	49 ± 40
pH		6.3 ± 0.6
PO$_4$ _ P	µg L^{-1}	2.6 ± 1.4
Secchi	m	3.4 ± 1.9
SO$_4$†	meq L^{-1}	0.1 ± 0.06
TOC	mg L^{-1}	8.9 ± 4.2
TN	µg L^{-1}	409 ± 142
TP	µg L^{-1}	11 ± 6
Precipitation	mm	782 ± 146
Temperature	C	4.5 ± 2.9
Runoff	mm	32 ± 14
P deposition (modeled)	mg m^{-2} yr^{-1}	6.7 ± 2.7
P deposition (measured)	mg m^{-2} yr^{-1}	7.0 ± 4.9
TOC:TP (mass)		939 ± 302
TOC:TN (mass)		23 ± 6
TN:TP (mass)		45 ± 12

Samples were collected from this depth, freeze dried, and then sifted manually with nylon mesh. Analysis of metals and P was performed by exposing 2 g of dry sample (<2 mm grain size) to a 10 mL solution of 7 M HNO$_3$ at 200C for 30 min on a heating plate (Swedish standard SS 028183 for extraction of macro and trace elements and PAHs in soil samples). Extracts were measured using inductively coupled plasma mass spectrometry (ICP-MS).

2.2. Modeled data

Because there were only four stations with P deposition measurements (Table 2), and the duration of monitoring was <10 years (precluding statistically meaningful trend analysis), robust modeled P deposition estimates were used in the analysis of in-lake P trends instead of measured data. Deposition of P was simulated using a global atmospheric transport model LMDZ-OR-INCA (Wang et al., 2015b). The model was run at a horizontal resolution of 1.27° latitude by 2.5° longitude with 19 vertical layers from the surface to a pressure of 4 hPa (approximately 38 km above sea level). To run this model, P emissions were derived for fossil-fuel/biofuel combustion (by month, varying from 1960 to 2013), wildfires (by month, varying from 1960 to 2013), dust (by month, averaged over 2000–2011), primary biogenic aerosol particles (constant, for 2000) and volcanoes (constant, for 1990). Averages of mean annual

Table 3
Mean and median values for watershed and lake characteristics. Land use is presented as a percent of the total watershed area.

Variable	Unit	Median	Mean
Wetland	% total area	6.1	9.2
Open water	% total area	11.7	12.9
Forest	% total area	67.4	62.5
Vegetated, other	% total area	0	5.6
Agriculture	% total area	0.4	1.5
Urban/exploited	% total area	0	0.2
Other	% total area	8.2	8.1
Total watershed	km^2	5.0	53.3
Lake area	km^2	0.5	1.3
Mean depth	m	4.2	4.5
Altitude	MASL	162	209
Soil Al:P (mass)		62.3	68.5

values for modeled (6.7 mg m^{-2} y^{-1}, std. dev. = 2.5 mg m^{-2} y^{-1}) and measured (7.1 mg m^{-2} y^{-1}, std. dev. = 5.0 mg m^{-2} y^{-1}) deposition data were similar for 2005–2012, the years in which measurement data were available. It should be noted that the inter-annual evolution of dust, biogenic aerosol and volcano P sources were not considered here, which may explain the somewhat lower modeled values. Due to the heavy computational load, snapshot simulations were run for 1980, 1990 and 1997–2013. A detailed description of these simulations has been provided elsewhere (Wang et al., 2015a; Wang et al., 2015b).

Several additional modeled and calculated lake variables were used in the analysis. Mean depth (Z_m) was modeled when measured data were not available (Sobek et al., 2011). The morphometric Osgood Index (Z_m/(lake area)$^{0.5}$) was used to determine lake mixing regime (Osgood, 1988). Nutrient ratios (C:N, C:P, and N:P, by weight) were calculated using TOC, TN, and TP from lake water samples. TP data were corrected following previous work (Hu and Huser, 2014) to account for calibration drift in the early 1990s.

2.3. Statistical analyses

Time series data were analyzed using the Mann-Kendall trend test (Mann, 1945; Kendall, 1975). Significant monotonic trends ($p \leq 0.05$) for epilimnetic water chemical, deposition, and climatic variables were determined and Sen's-slope (Sen, 1968) trend estimates (unit yr^{-1}) were calculated for each lake. Slopes were then used to estimate relative annual changes (trend/mean, % y^{-1}) for each site. Relative trends for different parameters were then compared using linear regression and by multiple linear regression (MLR) using JMP 11.0. Variables with high bivariate correlations (>0.7) were excluded from multiple linear regression analysis to reduce instability in the matrix inversion due to multicollinearity (Table 2). All r^2 values presented were adjusted for the number of degrees of freedom.

Two sets of MLR analyses were performed. The first used only lakes with significant increasing or decreasing TP trends. This analysis was then validated using both significant and non-significant TP trends as response variables. Both analyses were performed using in-lake median concentrations for all variables, statistically significant water chemical and deposition trends, climate, land use, and soils data, and interactions therein as potential predictors. Because not all potential driving variables had significant trends for each lake (Table 4), a standard, stepwise MLR approach where all potential variables are included from the start, was not possible. Thus, a manual MLR model development was necessary. For example, the variable with the greatest bivariate
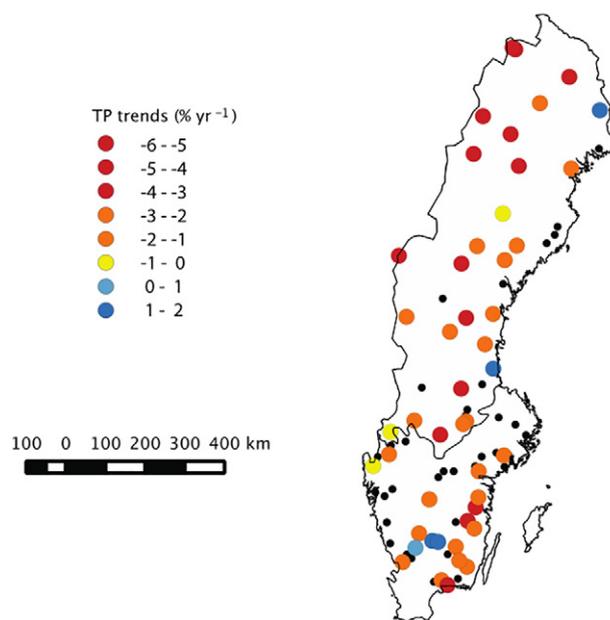

Fig. 1. Phosphorus trends across Sweden. Location and magnitude of total phosphorus (TP) trends for the study lakes across Sweden. Small, black circles represent lakes with no significant ($p > 0.05$) trend for TP as determined using the Mann-Kendall non-parametric test.

correlation with TP trend was the TOC concentration trend, and this was included as the first model parameter. All remaining explanatory variables were then tested at the second level, and the second significant variable explaining the greatest amount of variability was included in the model. The process was then repeated to determine a third significant variable, and so on, until a final model explaining variation in temporal lake TP trend was developed. Significance threshold for variable inclusion was $p < 0.05$ and model fitting ceased when an additional variable increased variance explained by <2%.

3. Results and discussion

More than half the studied lakes (n = 42) showed statistically significant ($p < 0.05$), declining epilimnetic TP concentration trends (Fig. 1) between 1988 and 2013, whereas 5 lakes had statistically significant increases in TP. The observed declines were not trivial, with relative Sen's
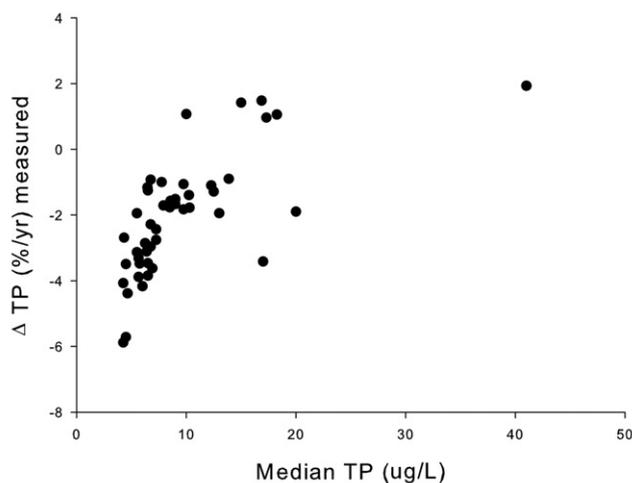
Table 4
Mean and standard deviation (SD) for significant trends (% yr^{-1}) and number of positive and negative trends for all study lakes (N = 81). Trends were determined using the non-parametric Mann-Kendall test.

Variable	Mean ± SD	Positive	Negative
Absorbance (filtered)	2.7 ± 1.2	58	0
Absorbance (unfiltered)	2.4 ± 1.4	57	2
Cl	−1.0 ± 0.7	3	48
Conductivity	−1.2 ± 0.8	3	72
NH$_4$−N	1.9 ± 2.2	24	5
NO$_3$−N	−2.5 ± 1.9	3	31
pH	0.2 ± 0.1	59	2
Secchi	−1.9 ± 1.2	2	47
SO$_4$	−3.5 ± 1.8	2	78
TOC	1.3 ± 1	58	2
TN	−0.01 ± 1.1	9	10
TP	−2.1 ± 1.7	5	42
Precipitation	0.61 ± 0.10	14	0
Temperature	0.37 ± 0.16	81	0
Runoff	0.23 ± 0.15	3	0
P deposition (modeled)	−2.4 ± 0.49	0	81
TOC:TP	2.5 ± 0.9	65	0
TOC:TN	1.4 ± 0.6	59	0
TN:TP	1.9 ± 1.2	32	2

Fig. 2. Median lake TP (1988–2013) versus trend for TP (% yr^{-1}) for all lakes with significant trends.
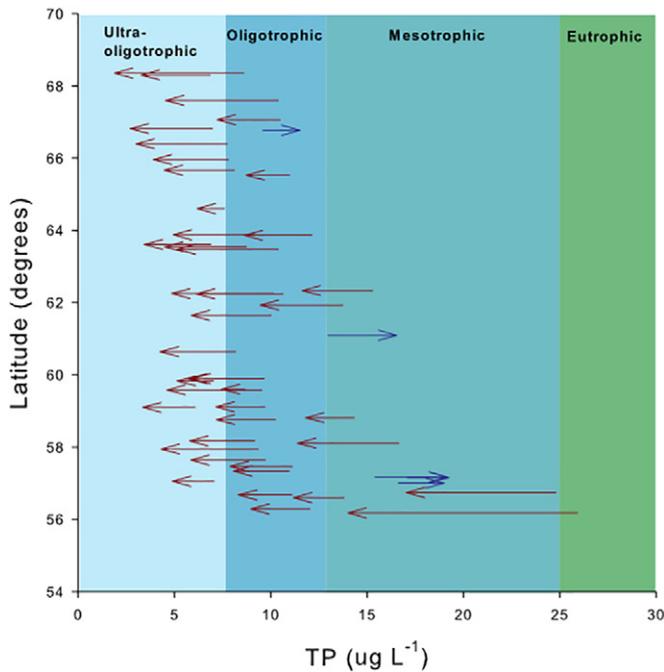
Fig. 3. Temporal changes in total phosphorus (TP) concentrations shown by differences in 2-year averages for mean, annual TP between 1988 and 89 and 2012–13. Trophic status, from ultra-oligotrophic (very nutrient poor) to eutrophic (nutrient rich), is based on standards for water quality in Sweden. Left and right arrow directions indicate decreasing and increasing trends, respectively.

slopes averaging >2.5% y^{-1} (2.1% y^{-1} when all significant trends, both positive and negative are included) and with declines ranging from 5.9 to 0.9% y^{-1}. Lakes that were already nutrient poor when monitoring began generally had the largest, relative P declines (Fig. 2). Mean TP concentration for lakes with significant declining trends decreased from 10.5 to 6.9 µg L^{-1} (based on mean values for the 1988–1989 and 2012–2013 periods, respectively). Using the Swedish lake classification system, 24 of the 81 study lakes moved to a lower trophic class and 18 became ultra-oligotrophic, tripling the number of lakes with very low nutrient, ultra-oligotrophic conditions (Fig. 3). No lakes moved to a higher trophic state. Increases in TN:TP ratios (from 26 to 35, mean by weight) suggest the potential for shifts from co-limitation by N and P to P limitation alone. However, further investigation of nutrient forms and ratios would be needed to adequately assess the status of nutrient limitation (Bergström, 2010).
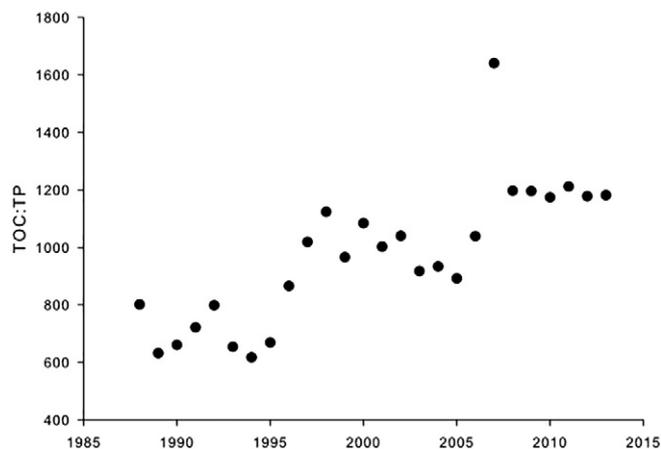
3.1. General trends in water chemistry and climate

The number of lakes with significant, increasing epilimnetic pH and TOC trends was also high, whereas water clarity (as Secchi depth) and P deposition (modeled) decreased for most lakes (Table 4). Unsurprisingly, TOC trends correlated strongly and positively with TP trends (r^2 = 0.63, p < 0.001) and were driven mainly by declines in sulfate and increasing temperature (MLR model r^2 = 0.72, p < 0.0001). The 80% increase in TOC:TP mass ratios (698 to 1254, Fig. 4), however, was not expected and explains why the substantial increase in organic matter transport to the lakes (expressed as TOC) did not entirely overwhelm the downward pressures on in-lake P concentrations. Because most organic matter in northern lakes is catchment-derived and DOM is the main vector for P transport (Baldwin, 2013), this indicates a change in terrestrial organic matter stoichiometry that may be correlated with the widespread decline in forest P status documented across Europe (Jonard et al., 2015). The increase in TOC alone, although substantial, was not enough to explain the increase in TOC:TP (i.e. a decline in P has occurred). Other variables with a substantial number of long-term trends included temperature, sulfate, chloride, and conductivity (Table 3).

3.2. Explanatory factors for in-lake P declines

To determine the main drivers of the TP trends (ΔTP) seen in the 47 study lakes with significant increasing (n = 5) or decreasing (n = 42) trends, stepwise MLR was performed. Three variables were included in the final model (N = 16, Fig. 5) predicting trends for P concentration in the study lakes (Eq. (2)), including:

$$\Delta TP = 1.7 * \Delta TOC - 0.024 * Al : P - 2.5 * \Delta pH - 1.9 \qquad (2)$$

Temporal trends in TOC (ΔTOC) explained 63% (p < 0.001) of the total variation in P trends in the final model above (Table 5). The positive relationship between TOC and TP has previously been reported by others (e.g. Nürnberg and Shaw, 1999). The positive relationship between ΔTOC and ΔTP is consistent with an increase in external P loading from the catchment. However, there is evidence of a long-term change in TOC:TP stoichiometry (Fig. 4).

The average catchment soil Al to P ratio (Al:P, by weight) for each lake was the next strongest predictor (7% variation explained, p < 0.05). Al concentration alone was not significantly correlated with TP trends, but the ratio of soil Al:P was, indicating that degree of binding-

Fig. 4. Temporal change in the annual mean ratio of total organic carbon (TOC) to total phosphorus (TP) for the study lakes (1988–2013).
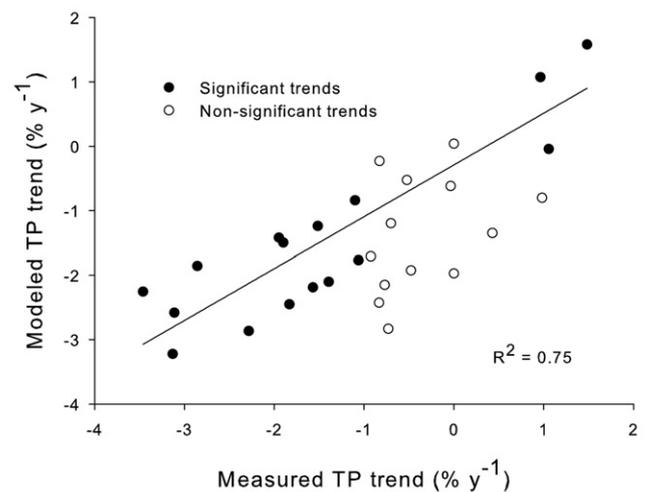

Fig. 5. Modeled versus predicted (see Eq. (1)) change in total phosphorus (TP) over the study period (ΔTP) for lakes with significant TP trends (filled circles) and non-significant trends (open circles). The r^2 value is for only the lakes with significant trends for TP.

Table 5
Parameters and estimates for models predicting TP trends in the study lakes. Model results are included for lakes with significant TP trends only (upper, N = 16) and both significant and non-significant TP trends (lower, N = 29).

Parameter	Estimate	Std error	t-Ratio	Prob > |t|
Intercept	−1.9	0.8	−2.3	<0.05
TOC trend	1.7	0.3	6.4	<0.0001
Al/P	−0.02	0.01	−2.2	<0.05
pH trend	−2.5	1.3	−1.9	<0.05
Intercept	−2.2	0.7	−3.0	<0.01
TOC trend	1.7	0.3	6.2	<0.0001
Al/P	−0.02	0.01	−1.8	<0.1
pH trend	−2.2	1.3	−1.7	<0.1

site saturation or Al mineral form (e.g. clays, oxides, hydroxides, etc.) may be potential explanatory factors for the observed P trends. Furthermore, TAl:TP ratios have been used to estimate binding P capacity in boreal lake sediments (Huser, unpublished data), but reactive Al forms and P bound to them would likely be a better predictor. Unfortunately these types of data were not available for the study lakes. Increasing P sorption has also been shown to positively correlate with DOC release from Al-rich humus soils typical of the boreal ecozone in Sweden (Giesler et al., 2005). Because DOC release from Al-rich soils may free up binding sites, an increase in available binding sites on Al mineral surfaces is a plausible mechanism for declining external P loads to the study lakes.

It should be noted here that soil metals and P data were only available for a limited number of catchments, which resulted in a decrease in the number of lakes that could be modeled when using these data. The soil samples, which were collected from a depth of 50 cm, can be assumed to provide a valid estimate of average catchment properties because previous studies have shown that the amount of reactive/exchangeable Al at this depth can be similar to (50–70%) or even equal (depending on extraction method used) to that found in overlying and surficial horizons (e.g. Walna et al., 2005). Thus, it seems likely that Al reactivity in deeper soil layers would generally be related to that of overlying soil layers.

Change in pH over time (ΔpH; 5% explained variation, p < 0.05) may have been indicative of decreased release of P from oxyhydroxide metals (or iron (Fe)/Al clay minerals) and transport to lakes caused by recovery from acidification. Because pH in surrounding catchment soils must generally increase in order for lake-water pH to rise, it is possible that transport of both P and P-binding metals to lakes is declining due to decreased mobility in the watershed. Increases in total Fe and Al, as well as P bound to Al and TP, have been detected in the sediment of acidified lakes (Huser and Rydin, 2005; Kopáček et al., 2000). But as pH has increased due to declines in acid deposition, accumulation of these metals and P in sediment has declined from peaks seen in the 1970–80s (Huser and Rydin, 2005), indicating a decrease in transport of these eleements to the lakes.

Previous work has suggested that nutrient availability in acidified lakes will increase as they recover from acidification, especially in lakes that were only slightly acidified during the industrial acidification period (Kopáček et al., 2015). Because most lakes used in this study are slightly acidic with mean pH increasing from 6.1 to 6.3 over the study period, the results presented herein may appear to contradict the work of Kopáček et al. (2015). It is often assumed that the solubility of phosphate-P ($PO_4 - P$) increases with increasing pH from the low (3.5) to medium pH (approximately 7–8) range in soils due to sorption and desorption processes involving Fe and Al oxides and hydroxides (Goldberg and Sposito, 1984). Previous work, however, shows that pH dependence of $PO_4 - P$ solubility in soils appears to be more complicated than this, and differences exist between sorption and solubility.

Recent work has shown that sorption maxima depend mainly on the type of binding site (clay mineral or metal oxide), amount of P, and soil

solution ionic strength (Gérard, 2016). Haynes (1982) cited a number of early soil studies with a minimum P solubility between pH 5 and 6. Similar results have been presented recently by Hartikainen and Simojoki (1997), Devau et al. (2011), and Weng et al. (2011), as well as a study including Swedish soils (Gustafsson et al., 2012). In some cases, especially at low $PO_4 - P$ pore-water concentrations, clay minerals appear to control P sorption instead of Fe and Al (hydr)oxides, with maximum sorption capacities between pH values of 4 and 7 (Gérard, 2016). A recent study of pH in soil water (50 cm soil depth) at nine sites in Sweden showed pH values generally increasing (slowly), ranging from 4.3 to 4.8 (Akselsson et al., 2013), indicating the potential for increased P binding and lower mobility in watershed soils. In addition, decreasing inorganic Al was also found in soil water for 5 of 9 Swedish sites (1986–2008, 50 cm soil depth) recovering from acidification (Akselsson et al., 2013), adding further support to the hypothesis that changes in atmospheric deposition chemistry are reflected in soil solution chemistry and may be affecting P mobility in these types of watershed soils.

The model predicting statistically significant TP trends (Fig. 5) was validated by developing a model including all available lakes (i.e. using significant and non-significant TP trends). Individual lake contributions to the final regression were first weighted by the absolute value of the Z-score derived from the Mann-Kendall trend test. Thus, lakes with more extreme positive or negative trends contributed more strongly to the resultant model. ΔTOC was the only significant predictor (at p < 0.05) when all trends for TP (significant and not significant) were included (Table 5). The next strongest variables for predicting variability in TP trends were pH trends and soil Al:P ratios (both p < 0.1). After including trends for TOC and pH and soil Al:P ratios, no other variables increased variance explained by >2%. Estimated coefficients were similar for both the models including only significant trends and both significant and non-significant trends (Eq. (3)), showing that similar drivers were also influential in lakes with smaller, non-significant trends. Based on the standard errors of the predictors, Eqs. (2) and (3) are statistically indistinguishable from each other (Table 5).

$$\Delta TP = 1.7 * \Delta TOC - 0.017 * Al : P - 2.2 * \Delta pH - 2.2 \qquad (3)$$

3.3. Other potential drivers of P declines

Somewhat surprisingly, even though trends in modeled P deposition were consistently negative (Table 4), there was no apparent relationship at the individual lake level (Fig. 6). The explanation for this may be related to catchment-to-lake area ratios. If this ratio is high, only a
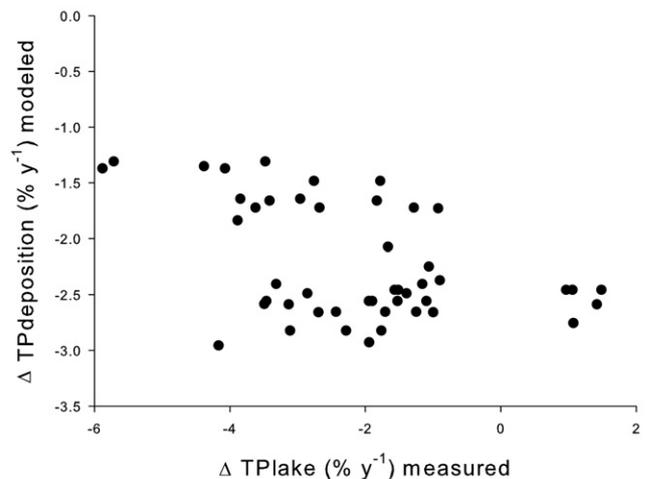

Fig. 6. Trends in TP deposition vs. in-lake TP concentration over the study period (1988–2013).

small portion of P deposited over the watershed will enter the lake via direct atmospheric deposition, suggesting this term is a relatively minor component of the external P load. The percentage of P directly deposited to the lake relative to total external P loading ranged from 3.4 to 16% for the dataset as whole. However, it should be noted that external loading (background loading unaffected by direct anthropogenic influence) was estimated using estimates for southern, middle, and northern Sweden on a regional basis (Uggla and Westling, 2003), precluding individual lake comparisons. No measured data for external loading were available for the lakes, at least to the authors' knowledge. Nonetheless, it is likely most P deposited to the catchment is retained in vegetation or soils, especially those soils containing greater amounts of Fe/Al oxides and hydroxides and/or clay minerals.

Inherent uncertainties due to limited data and the low spatial resolution of the deposition model (1.27° latitude by 2.5° longitude) may also explain the lack of relationship between deposition trends and lake water TP concentrations. In addition, only three positive and three negative trends were detected for deposition of P in a recent European study (Tipping et al., 2014). Other work has also shown no change in P deposition even as in-lake P concentrations have declined (Eimers et al., 2009). Of the four sites in our study with available P deposition data, no obvious trends were detected (Table 4).

Changes in precipitation and flow have also been suggested as drivers of P declines in surface waters, but the small number of trends for runoff detected in this study (N = 3) suggests otherwise, similar to a recent study of P declines in Canadian lakes (Crossman et al., 2016). Depletion of mineral sources of P in watershed soils is also unlikely, as soils in this region are relatively young, being formed after the last glaciation ca. 11,000 years ago (Walker and Syers, 1976). Regional climate cycles may also be responsible for the declining P concentrations reported here. For example, the Atlantic Multi-decadal Oscillation (AMO), which affects air temperatures and rainfall over much of the northern hemisphere, and in particular North America and Europe (McCarthy et al., 2015), appears to have been in an upward phase during the most recent ca. 15 years of the data range studied herein. An upward phase would generally lead to warmer and wetter conditions in these areas (van Oldenborgh et al., 2009). The few trends in precipitation and runoff suggest there have not been changes in runoff (Q) that could account for declining TP concentrations based on Eq. (1). All sites had significant increasing trends for air temperature; however, the potential effects of increasing temperature on P cycling are equivocal (Table 1). In addition, if the AMO enters a declining phase, our models suggest that P declines may accelerate, as TOC (the only positive term in our models) would be likely to stabilize or decline under cooler (Hongve et al., 2004) and drier (de Wit et al., 2016) conditions.

In-lake changes to P cycling may also be a driver of decreased in-lake P availability. Reductions in internal load and/or increased retention could both lead to oligotrophication. Increased retention associated with co-precipitation and increased sediment sorption and accumulation of P due to increased input of Al and Fe to acidic and limed lakes has been suggested as a driver of oligotrophication (Hu and Huser, 2014; Huser and Rydin, 2005; Kopáček et al., 2000; Kopáček et al., 2008; Norton et al., 2011). However none of the studied lakes was limed and the mean pH of the lakes in the dataset was 6.3 (N = 81, Table 2). In addition, the accumulation of P (and P binding metals like Al) in the sediment of anthropogenically-acidified lakes appears to have declined since peaking in the 1970s (Huser and Rydin, 2005).

Of the study systems with sediment data available, generally stable (N = 4) or declining (N = 2) accumulation of P (TP as well as different P fractions including Fe-, Ca-, and Al-bound-P) in sediment has been detected (unpublished data), suggesting little or no change in R_S (Eq. (1)). Although most of these lakes also had generally stable accumulation of P-binding metals like Fe and Al that strongly bind P, two lakes did have increasing accumulation of Fe in sediment during the past 30 years (data not shown), and this is likely due to the increase in TOC seen in the

lakes (Futter et al., 2014), streams, and rivers across Sweden (Huser et al., 2011; Kritzberg and Ekström, 2012). The number of lakes with sediment data was small, however, and further study would be needed to make any general conclusions. Declining P concentrations leading to reductions in external load to receiving lakes have been detected in unaffected headwater streams (Eimers et al., 2009), indicating that in-lake effects may not be the primary driving factor behind the observed TP declines.

Given the controlling variables in Eqs. (2) and (3), the predominant control of changing external loading on in-lake TP concentration trends seems plausible as the increased binding capacity of watershed soils, coupled to the increase in TOC:TP ratios seen in the lakes, both point to strong, in-watershed factors (climate, deposition chemistry, and watershed geology) that may overshadow any direct effect of declining P in deposition. Widespread declines in foliar P concentrations, indicative of declining in-catchment P availability (Jonard et al., 2015), as well as increased watershed soil binding and reduced transport of P to lakes as pH increases (Gérard, 2016 and references therein) add additional support to the results of our study.

3.4. Evaluation of hypothesized drivers of TP trends

Much of southern and central Sweden was acidified (Akselsson et al., 2013; Futter et al., 2014; Moldan et al., 2013) and most of the observed decline in TP discussed herein is consistent with recovery from acidification, e.g. increased retention in watershed soils; (Gérard, 2016; Crossman et al., 2016). The lakes included in this study, however, cover a broad latitudinal gradient representative of climatic conditions across the boreal ecozone as well as including areas in northern Sweden minimally affected by acid deposition and alpine areas where some of the largest, relative declines in TP (between 5 and 6%) were detected (Fig. 1). Increasing DOC concentrations are strongly linked to a recovery from acidification (Monteith et al., 2007) and potentially to a changing climate (Hongve et al., 2004), and ∆TOC was the only predictor in Eqs. (2) and (3) positively related to the TP trends found in this study. Increasing organic matter input, and thus increasing external loads of dissolved organic P input may mask the severity of the P declines detected here. To explore this, we set ∆TOC = 0 in Eq. (2), and the mean TP decline increased to 3.9% yr^{-1} with the few positive trends for lake TP in the dataset turning negative. The net impact of increasing temperatures on surface water organic matter concentration is still uncertain, however, and results from other studies show both increased concentrations from increased DOC production (Evans et al., 2005) and decreased concentrations due to decreased rates of C cycling (Allison and Treseder, 2008).

The trends observed herein were not related to trends in (modeled) P deposition. This suggests a disconnect between rates of atmospheric input and in-lake concentrations, presumably mediated by in-lake and in-catchment processes. Warmer conditions should lead to higher P weathering rates in the catchment, yet if this is occurring, the effect is being masked by other factors, potentially related to longer growing seasons and greater plant uptake. The controls exerted by terrestrial vegetation on in-lake P concentrations should be the subject of further research. Akselsson et al. (2008) have documented the risks associated with increased forest biomass harvesting while Jonard et al. (2015) have shown that there is a widespread decline in foliar P concentration across Europe. Standing forest biomass in Sweden has been increasing during recent decades, and this increase has been proposed as a mechanism to explain declines in riverine nitrate concentration (Lucas et al., 2016). This is consistent with a changing climate where warmer, wetter conditions lead to a longer growing season.

3.5. Potential effects of declining P in lakes

Some questions remain about the effects of ultra-oligotrophic conditions on in-lake productivity, but evidence from reservoirs has shown

that oligotrophication is linked to productivity declines (Milbrink et al., 2011; Rydin et al., 2008; Stockner et al., 2000). While there is an increasing awareness that water bodies can have too little nutrients (Anders and Ashley, 2007), the consequences of further oligotrophication are inadequately recognized in current legislation. Water quality management in Europe and elsewhere is based on a paradigm of "less nutrients are better" for assessing ecosystem health. In Europe, the Water Framework Directive mandates that all water bodies should achieve at least good ecological status by 2027. Somewhat alarmingly, the criteria to reach good status are based on upper nutrient concentration thresholds and do not consider the possibility of water quality impairment as a result of oligotrophication (EUWFD, 2000). Thus, increased reliance on renewable energy sources including hydroelectricity (Milbrink et al., 2011; Stockner et al., 2000) and forest biomass (Akselsson et al., 2008), common in Sweden and elsewhere, are likely to exacerbate the ongoing P declines with no management consequences. Further construction of reservoirs for hydroelectricity generation will increase retention while greater harvest rates of forest biomass will decrease total P mass in the tributary watershed (Table 1). In addition, reference condition TP concentrations are often estimated using proxies for DOM (Cardoso et al., 2007; Huser and Fölster, 2013). Continued increases in DOM will lead to higher estimated reference condition concentrations and greater apparent oligotrophication based on present-day TP when relying on these methods, suggesting the need for methods excluding DOM to determine reference conditions.

The methodology for TP analysis must now also be evaluated due to the low concentrations being detected in many of the study lakes. Analysis of TP in water samples is generally done using the ISO standard 6878:2005/Bran Luebbe Method G-175-96 method wherein the minimum detection limit is $1 \mu g L^{-1}$. Five lakes in this study, however, now have TP concentrations at the detection limit of $1 \mu g L^{-1}$. This means that a switch to low-level detection methods for P is needed, adding additional cost to monitoring programs. It is also likely trends for P in surface waters will be more difficult to detect simply due to the analytical bottom limit on P concentration. Given this limit, the trend results for TP in this study should be considered conservative.

With the available data it is not possible for us to determine if lakes in this study are returning to reference conditions (i.e. un- or minimally degraded state) or approaching hitherto unprecedented conditions with widespread organic matter concentration increases and TP declines. Paleolimnological evidence suggests lakes in the Boreal region generally become less productive as they age (Norton et al., 2011), but the rate of nutrient declines presented herein suggests additional pressure(s) may be accelerating this process. Recently published work showing increasing (instead of declining) trends for P in U.S. surface waters (Stoddard et al., 2016), in addition to declining trends in other regions (e.g. Canada and Finland; Yan et al., 2008; Eimers et al., 2009; Arvola et al., 2011, Stammler et al., 2017), indicates a potential, continental-scale shift in aquatic P availability may be responsible. Evidence relating these differing patterns to a singular disturbance mechanism, however, is currently lacking.

4. Conclusions

Although we cannot entirely discount the potential effects of other in-lake or catchment processes affecting P cycling, the results contained herein suggest that much of the variation in P declines seen in Swedish lakes, and presumably many of the lakes in northern boreal and arctic regions, can be attributed to changes in watershed processes related to terrestrial soil properties, recovery from acidification, and climate driven changes to organic matter input to lakes. Further empirical and modeling (e.g. Crossman et al., 2016) work is needed to determine if we are approaching a critical transition to a new, ultra-oligotrophic state in which increasing DOM and declining TP concentrations act in synchrony to suppress aquatic productivity.

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