LETTER

Acceleration of cyanobacterial dominance in north temperate-subarctic lakes during the Anthropocene

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Abstract

Increases in atmospheric temperature and nutrients from land are thought to be promoting the expansion of harmful cyanobacteria in lakes worldwide, yet to date there has been no quantitative synthesis of long-term trends. To test whether cyanobacteria have increased in abundance over the past ~ 200 years and evaluate the relative influence of potential causal mechanisms, we synthesised 108 highly resolved sedimentary time series and 18 decadal-scale monitoring records from north temperate-subarctic lakes. We demonstrate that: (1) cyanobacteria have increased significantly since *c*. 1800 CE, (2) they have increased disproportionately relative to other phytoplankton, and (3) cyanobacteria increased more rapidly post *c*. 1945 CE. Variation among lakes in the rates of increase was explained best by nutrient concentration (phosphorus and nitrogen), and temperature was of secondary importance. Although cyanobacterial biomass has declined in some managed lakes with reduced nutrient influx, the larger spatio-temporal scale of sedimentary records show continued increases in cyanobacteria throughout the north temperate-subarctic regions.

INTRODUCTION

Over the past two centuries, humans have altered the global climate, accelerated nutrient cycles and modified terrestrial landscapes to such an extent that many are calling this period a new geological era, the Anthropocene (Steffen *et al*. 2007). Long-term ecological records, including monitoring time series spanning decades, natural archives such as lake sediments and historical records have been instrumental in advancing our knowledge of how populations to ecosystems have responded to the increased rate of change over the past few centuries (Smol 2008). In particular, freshwater ecosystems provide society with numerous ecosystem services, yet human alterations of freshwaters have decreased the value of marketable aquatic benefits by ~ 16% globally, equivalent to ~ $900 billion (Dodds *et al*. 2013). To avert further losses, large-scale syntheses of long-term ecological records from freshwaters are needed. Indeed, several of these syntheses have been published (e.g. long-term nutrient trends, Keatley *et al*. 2011; carbon

burial rates, Anderson *et al*. 2014) and are providing robust parameters for global scale models to make predictions about future responses.

Intensified land-use, sewage discharge and climate change during the past two centuries should favour disproportionate development of harmful algae in freshwaters (Wolfe *et al*. 2013). For example, blooms of cyanobacteria pose a serious threat to drinking water sources worldwide because many taxa contain harmful hepato- and neurotoxins. Despite the detrimental effects of cyanobacterial blooms on human health and ecosystem function (Jonasson *et al*. 2010; Lévesque *et al*. 2014), it remains unclear whether cyanobacteria are more abundant now than they were at the onset of the Anthropocene ~ 200 years ago. Both relatively short-term laboratory and field studies (often less than a few years) have shown a significant increase in cyanobacteria biomass and dominance of the community across large gradients of nutrients and temperature (Kosten *et al*. 2012; Taranu *et al*. 2012; Beaulieu *et al*. 2013). Warm temperatures can promote cyanobacteria

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directly because of their relatively higher temperature growth optima, and indirectly because of effective buoyancy regulation and the ability to shade out photosynthetic competitors in thermally stratified waters (O'Neil *et al.* 2012). Furthermore, changes in nutrient supplies associated with anthropogenic activities may favour cyanobacteria either by reducing N : P ratios which promote heterocystous species or providing chemically reduced nitrogen which enhances growth of low-light adapted, but highly toxic filamentous taxa (Donald *et al.* 2011).

The relative importance of climate change and nutrient inputs to lake ecosystems is scale dependent, thus confounding extrapolations of findings from experiments and modern surveys to the level of decades or a century. Indeed, over the past two centuries we have witnessed pronounced and rapid changes in multiple environmental stressors (Carpenter *et al.* 2011), which in some cases have interacted to generate unexpected non-additive responses or 'ecological surprises' (e.g. synergistic effects of warming and eutrophication; Taranu *et al.* 2012). Whereas the cumulative impact of climate change on freshwater ecosystems has been relatively gradual, the onset of modern agricultural practices and human-mediated changes to atmospheric and hydraulic loads has certainly resulted in an accentuated nutrient signal in recent decades (McGowan *et al.* 2012). Consequently, monitoring records often do not capture the full magnitude of climate change, particularly when only the most recent decades are considered (Thies *et al.* 2012; Tolotti *et al.* 2012). To date, much of the focus in the cyanobacteria literature has been on contemporary data sets. Little is known of continental-scale patterns in timing and extent of the cyanobacterial blooms, whether cyanobacterial growth has been disproportionate to that of other algae, or if historical changes are associated mainly with surface-water fertilisation (Bunting *et al.* 2007), rising water temperatures associated with climate change (Beaulieu *et al.* 2013) or other mechanisms (Molot *et al.* 2014). The paleolimnological approach is effective in providing information on baseline conditions against which the cumulative impacts of multiple environmental stressors can be assessed. The opportunity to advance our understanding has now emerged because of the growing number of long-term records that have been developed (spanning decades to centuries) by regularly sampling the water column of inland waters, or by analysing fossil pigment (carotenoid) biomarkers preserved in lake sediments. By considering data at this spatio-temporal scale, larger gradients of climate and anthropogenic change can be examined, providing a broader generalisation of the response of lakes to multiple stressors.

Here, we provide the first continental-scale test of whether cyanobacteria have increased since the start of the Anthropocene and whether these changes have been disproportionate relative to other ubiquitous phytoplankton (in this study diatoms). We quantified cyanobacteria and diatom trends using regression-based analyses of two forms of data; highly resolved records of fossil pigments preserved in lake sediments, and decadal-scale time series of extant phytoplankton biomass. Our paleolimnological synthesis was based on available pigment time series from temperate and subarctic lake sediment cores of the Northern Hemisphere spanning the past

~ 200 years ($n = 108$ lakes). The long-term monitoring (LTM) programs spanned 8 to ~ 40 years of seasonal sampling (18 ± 9 years; $n = 18$ lakes). Most study lakes were located in North America and Europe, situated in either lowland ($n = 91$ lakes: 259 ± 220 m a.s.l.) or alpine ($n = 29$ lakes: 1831 ± 935 m a.s.l.) regions. We hypothesised that changes in nutrients (phosphorus and nitrogen) and atmospheric temperature would explain a significant amount of the variation in cyanobacteria trends and that cyanobacteria rates of increase would be accelerated relative to diatoms. Prior contemporary surveys have identified nutrients and temperature (Kosten *et al.* 2012; Beaulieu *et al.* 2013) as predictors of cyanobacterial biomass, but no studies have directly quantified the importance of these environmental variables at large spatio-temporal scales (i.e. continental and century time scales). Our synthesis of centennial-scale records unambiguously demonstrates the paramount importance of nutrients relative to temperature and identifies where and when cyanobacteria exhibited a disproportionate increase through time.

MATERIALS AND METHODS

Biological and physical data

Paleolimnological data

To acquire historical cyanobacteria records spanning the last ~ 200 years from lowland and alpine lakes located within the Temperate-Subarctic zone, we conducted a literature search of paleolimnological pigment records published before 2013 using the ISI Web of Science database and Google Scholar with different combinations of the following keywords: *Paleolimnology/Palaeolimnology*, *Pigment*, *Eutrophication*, *Land use*, *Trophic*, *Pristine*, *Lake* or *Freshwater*. We also included many of our own unpublished core records. In total, 191 studies were identified (sum of all potential studies). We first eliminated sites subjected to whole-lake manipulations (e.g. explicit additions of nutrients) because we were interested in the response of lakes to landscape-level changes. Fossil analyses were retained if core resolutions were sufficiently high (i.e. at least a 20-year resolution between *c.* 1800 CE and present day). Numerous Holocene-scale studies were identified by our search but most had an insufficient temporal resolution and thus were excluded ($n = 14$; Table S1). We further eliminated sites located outside of the Temperate-Subarctic zone (i.e. sites from the Tropics, Subtropics and Arctic; Table S1). Tropical to subtropical sites were excluded because too few sites had been analysed to appropriately characterise these lakes ($n = 7$ subtropical, $n = 2$ tropical). Arctic sites were excluded *a priori* because primary production in a vast proportion of these sites is benthic and would not track planktonic cyanobacterial changes (Vincent 2009). Published records were digitised using the GraphClick Version 3 (© 2008 Arizona Software, Phoenix, Arizona, USA). To control for among-lake differences in units and preservation (Leavitt & Hodgson 2001), concentrations of pigments per unit mass organic matter were standardised (*z*-transformed) in each core. In each study, pigments were extracted from sediments using oxygen-free organic solvents and then analysed using high-performance liquid chromatography according to

standard methods (Lami *et al.* 2000; Leavitt & Hodgson 2001; Buchaca & Catalan 2007).

We identified a total of 108 lakes that met these screening criteria and recorded historical changes in cyanobacterial pigments. For this study, we focused on myxoxanthophyll, a carotenoid which is found in most bloom-forming cyanobacterial taxa and which is characteristic of severe bloom events (Leavitt & Hodgson 2001). Furthermore, myxoxanthophyll had the largest representation across lakes identified in our search relative to other cyanobacterial pigments. Cases for which we could confirm that laboratories measured for myxoxanthophyll but found no samples above detection throughout the core were also included in our analyses. The average temporal resolution of each time series retained was 24 ± 17 samples per 100 years. Of the 108 studies where myxoxanthophyll was measured, 98 cores were ^{210}Pb-dated (Appleby 2001) and 95 of these cores also reported changes in the concentration of diatoxanthin, a pigment commonly used as an indicator of diatom abundance and which exhibits a similar chemical stability as that of myxoxanthophyll (Leavitt & Hodgson 2001). Once in the sediment, these pigments are stable for thousands of years and their use as biomarkers of past algal biomass has been validated in diverse environments (e.g. Leavitt & Findlay 1994). In particular, concentrations of diatom and cyanobacterial carotenoids have been demonstrated to vary as a linear function of changes in respective phytoplankton abundance in both multi-decade monitoring studies (Bunting *et al.* 2007) and whole-lake experiments (Leavitt & Findlay 1994). To assess the robustness of models, we conducted a complementary analysis on all lakes where diatoxanthin was reported, but not necessarily myxoxanthophyll (142 lakes in total, 125 of which were dated; Fig. S1).

Information on each lake and catchment (area, depth, water residence time, agricultural development), as well as water-column nutrient concentrations at the time of sampling, were obtained from original manuscripts or via correspondence with authors. Mean monthly air temperatures were obtained from sites within the Global Historical Climatology Network (Lawrimore *et al.* 2011) that were in close proximity to each study lake. Because sediment cores were obtained at different times across sites, the air temperature time series were truncated to the date of sample collection. For each time series, monthly air temperatures were averaged across April to October.

Water-column data

Using the ISI Web of Science database, Google Scholar, our unpublished data and online databases (e.g. the Estonian Environment Information Centre and the Long Term Ecological Research Network), we retrieved long-term records of phytoplankton biomass from the water column of 27 temperate to subarctic lakes. Time series with monthly resolution were available for 17 of these sites, whereas the remainder of lakes had annual estimates of phytoplankton biomass. To compare monthly and annually resolved data sets, monthly estimates of cyanobacterial biomass were averaged across the growing season (April–October). Sites were excluded from the following statistical analyses if lakes were sampled for fewer than 8 years (*n* = 5) or lacked concomitant time series of

nutrient data (*n* = 3 lakes). Consequently, a total of 18 lakes were selected for LTM analyses. These lakes were categorised according to whether (1) nutrient concentrations increased through time (eutrophication group), (2) nutrients did not significantly increase (no change group), or (3) re-oligotrophication efforts were cited and/or a decreasing nutrient trend was observed (restoration group).

Numerical analyses

Assessing paleolimnological trends

Nonparametric Mann–Kendall (M-K) tests for monotonic trends were used to quantify cyanobacterial trends during the past 200 years. This analysis is based on the Kendall rank correlation and was conducted using the *Kendall* library (McLeod 2011) of R version 3.0.2 (R Foundation for Statistical Computing). Briefly, pairs of measurements are rearranged to obtain an increasing rank of the first descriptor. The number of pairs of ranks that are also increasing on the second descriptor is then tabulated to provide a measure of association between both descriptors (Legendre & Legendre 2012). For each pair, a value of −1 is assigned if the former pair has a greater value for the second descriptor, or +1 if the value of the former is smaller. A positive summed score shows an increasing trend (i.e. the number of pairs in increasing order outnumber the number of pairs in decreasing order), whereas a negative value shows a decreasing trend. To identify the environmental drivers of centennial variation in cyanobacterial trends, the M-K trend coefficients for each stratigraphic time series were used as the response variable in a regression tree analysis using *rpart* and the wrapper function *MVPART-wrap* (Legendre & Legendre 2012) in R. This approach is particularly useful if time series exhibit threshold relationships with environmental drivers. To select the best tree and avoid overfitting, we plotted the cross-validation results and pruned the tree using the 1-SE rule (Legendre & Legendre 2012). Specifically, we selected the complexity parameter associated with the smallest tree where the estimated error rate was within 1 SE of the minimum error, and pruned the tree at this complexity parameter value. Several cross-validations were run to ensure the final tree was not atypical.

We compiled data for numerous potential explanatory variables that have previously been shown to explain significant variation in phytoplankton distribution or dynamics, but have yet to be considered for such a large spatio-temporal data set. Specifically, the variables we considered for our regression tree analysis included lake catchment area, percent agricultural land cover in the catchment, lake elevation, lake mixing regime (i.e. mono-, oligo-, di-, mero- or polymictic), water residence time, average and maximum lake depth, lake surface area, lake volume, mean seasonal air temperature and the concentrations of total phosphorus (TP) and total nitrogen (TN) in the water column measured near the time of coring. Nitrogen concentrations were only available for a subset of lakes (*n* = 63), thus the regression tree that included TN is provided as Supporting Information. Regression tree analysis also included three estimates for changes in air temperature: the M-K trend coefficient for temperature calculated across the entire time series, the post-1950 M-K trend and an

average of the 5 years leading to core sampling. We used a variation inflation factor (*VIF* < 5) to isolate independent (non-collinear) explanatory variables and eliminated collinear variables to improve interpretation of regression results. For example, TP concentration increased as a function of agricultural land cover (*GLM-R²* = 0.72, *P* < 0.0001) but only TP was retained in the final model because it was more widely available than were estimates of agricultural activity. Explanatory variables retained for the final regression tree included TP concentration, seasonal mean air temperature, lake surface area, maximum depth, water residence time and lake elevation. These variables were also evaluated in the supplemental regression tree analysis including TN (see Supporting Information).

An additive mixed-effect model (AMM) framework generated using the *mgcv* library in R (Wood 2006) was used to describe the general trends in cyanobacteria over the last two centuries. To test the significance of nonlinear relationships, we compared models with and without an AMM smoother function. Non-independence among data points from the same lake as well as the temporal autocorrelation were evaluated by comparing nested models with a random intercept or slope for lake and a continuous temporal autoregressive CAR (1) structure respectively. Because different geographic regions are characterised by different climates and land-use histories, we expected that the longer history of anthropogenic change in lowland Europe would result in a more pronounced cyanobacterial signal than in lowland North American or alpine lakes (Keatley *et al.* 2011). Thus, we evaluated the interaction between year and a categorical variable coding for the three major geographic regions of our study lakes (lowland Europe, lowland North America and Alpine). Further, given the expected relationship between cyanobacterial abundance and nutrient concentrations, an AMM with an interaction between year and lake trophic status was tested. Nested models were evaluated using the log-likelihood ratio test, with a maximum likelihood (ML) method for fixed effect comparisons (e.g. nutrient concentration), and a restricted ML method when comparing nested random structures (e.g. lake identity). Confidence intervals were derived using the *predict.gam* function in R, with type = 'response' specified in the model (*mgcv* library; Wood 2011). This produces robust prediction and SEs for a given fitted AMM object given a new set of values on the scale of the response. Finally, we conducted a resampling exercise of the time series data, where we created 10,000 bootstraps of 30 time series each from the initial pool of 60 lakes with reported catchment-scale anthropogenic disturbances to evaluate whether shrinking the data set down to the number of lakes reporting an absence of catchment-scale disturbances (i.e. 30 lakes) resulted in a substantially different AMM fit (see Supporting information).

Assessing water-column trends

To test for heterogeneity in cyanobacterial trends over the monitoring timeframe among the three nutrient trajectory groups (eutrophication, no change, restoration), cyanobacterial M-K trend coefficients were converted to within-study effect sizes (using the Fischer's *z* transformation) and a meta-analysis was conducted with a categorical moderator variable for the nutrient trajectory grouping (*MAc* library in R; Del Re & Hoyt 2010). A random-effect model was used to test for general patterns across north temperate-subarctic lakes. More specifically, a pooled variance was calculated for each trophic category, and the aggregate effect size across all studies within each category was obtained by summing individual effect sizes, weighted by the pooled variance. The average length of each time series was comparable among the three categories with 13 (± 3 SD), 16 (± 5 SD), and 21 (± 9 SD) years for the eutrophication, no change, and restoration groups respectively. Average timeframes did not differ significantly among the three categories (*F* = 1.3, *P* = 0.311). To identify additional sources of variability in M-K trend coefficients, we quantified the explanatory power of a suite of environmental variables and lake characteristics (i.e. TP, mean air temperature, water residence time, lake depth, lake surface area and lake elevation) using linear regression and analysis of variance. The relatively small sample size precluded the application of univariate regression trees.

RESULTS

Analysis of the sedimentary time series during the last ~ 200 years showed that 58% of lakes had significant increases in cyanobacterial pigment concentrations (myxoxanthophyll), as indicated by the M-K test, whereas lakes exhibiting significant decreases (3%) were far less prevalent (Fig. 1). Cyanobacteria increased in all regions over the past two centuries, but positive M-K trends were more common (χ^2 = 10.6, *P* = 0.009) in lowland lakes (61% in North America and 70% in Europe) than at alpine lakes (36%). Regression tree analysis showed that both differences in nutrient concentration among lakes and air temperature were significant predictors of the variation in the magnitude of change in cyanobacteria over the past ~ 200 years (R^2 = 0.35; *n* = 83), but nutrients explained three times more variation than temperature (Fig. 2; Fig. S2).

Patterns of historical change in cyanobacteria were described better by nonlinear (additive mixed-effect) models than by linear regression and showed that the rate of cyanobacterial expansion increased during the mid-20th century (Fig. 3). Synthesis of piecewise regression analysis of each lake suggested that 25, 50 and 75% of lakes showed a break in the slope of cyanobacterial concentration vs. time at *c.* 1943, 1973 and 1985 CE respectively (Fig. 3a). In general, this pattern was driven by trends in lowland lakes which exhibited a significantly greater degree of acceleration of cyanobacterial growth than did alpine sites (χ^2 = 28.7, *P* < 0.0001), particularly during the last 25 years (Fig. 3b). Cyanobacterial trends also appeared to vary with land use, as there were more pronounced increases in lakes with reports of anthropogenic disturbances in their catchment (R^2 = 0.18; *n* = 60), compared to lakes situated in pristine catchments (R^2 = 0.06; *n* = 30), even after controlling for sample size (Fig. S3). In contrast to the difference between alpine and lowland lakes, rates of cyanobacterial increase did not vary among lowland lakes in Europe or North America. Instead, the maximum rates of cyanobacterial increase in these lowland sites varied as a function of trophic state in both regions, with the hypereutrophic
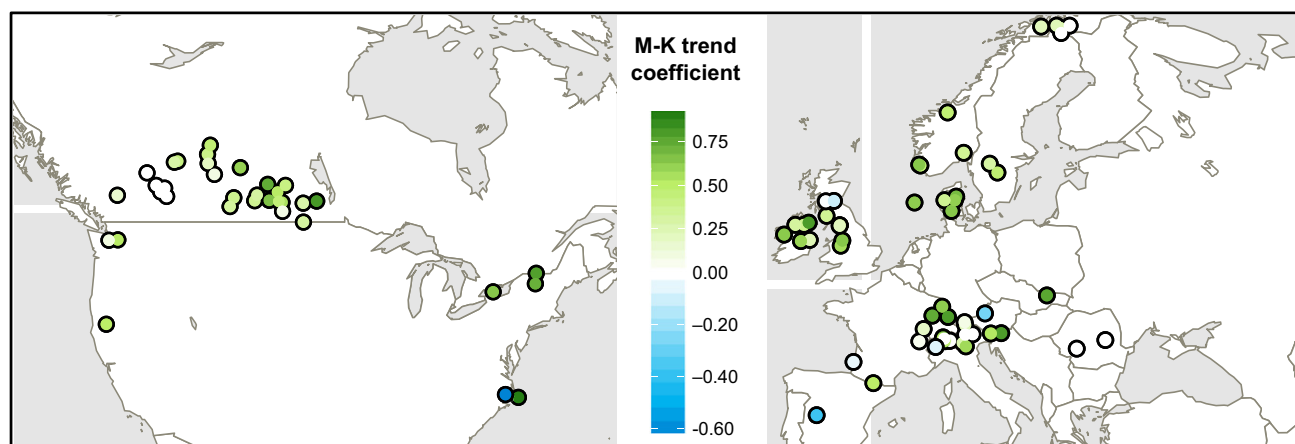
Figure 1 Map of Northern Hemisphere highlighting sediment core sites ($n = 108$) and their historical trends in sedimentary cyanobacterial pigment concentrations. Fill colours show the strength of cyanobacterial (myxoxanthophyll) trends through time determined by the Mann–Kendall (M-K) test, where green tones correspond to positive M-K trends and blue tones to negative ones. Not shown on the map is Pyramid Lake (Himalayas). See Table S2 for details on M-K trend coefficients and *P*-values.
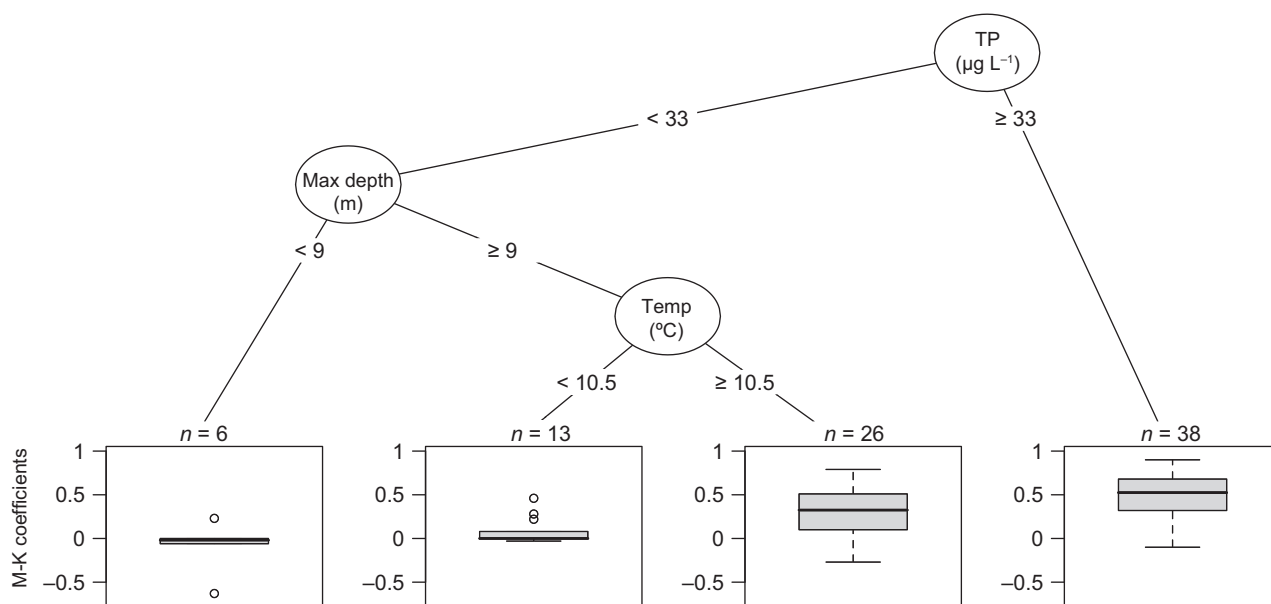

Figure 2 Regression tree of cyanobacterial Mann–Kendall trend coefficients for the paleolimnological data. Data set comprises dated and non-dated cores with adequate environmental data (variance explained by model presented: $R^2 = 0.35$, $n = 83$). Temp refers to seasonal mean air temperature (April to October) averaged over the 5 years prior to sediment core sampling (the median core sampling date across lakes was 1997, and ranged from 1983 to 2011).

lakes showing the greatest relative increase (Fig. 3c). Likewise, the proportions of eutrophic to hypereutrophic lakes were comparable in North America (69%) and Europe (54%) ($\chi^2 = 1.9$, $P = 0.616$) respectively.

Rates of increase in cyanobacterial abundance over the past 200 years were greater than those recorded for diatoms, equally ubiquitous algae which have distinct environmental optima (Reynolds 2006). Analysis of sediment records containing biomarkers of both phytoplankton groups showed that nonlinear increases in pigment concentration after c. 1945 CE were significant and were disproportionately larger for cyanobacteria than for diatoms ($\chi^2_{\text{year-pigment}} = 14.3$, $P < 0.0001$). For example, the average increase in cyanobacte-

ria was 28% greater than that measured for diatoms in hypereutrophic lakes (Fig. 3c and e).

Quantitative evaluation of cyanobacterial M-K trends from LTM records spanning ten countries across three continents (Fig. S4) further suggests that variation was regulated mainly by changes in nutrient concentrations (Fig. 4). By separating LTM sites according to nutrient trajectories (eutrophication, no change and restoration), we demonstrated that cyanobacteria biomass increased significantly in nutrient-enriched lakes experiencing no to minimal nutrient loading restriction (effect size: M-K $\tau = 0.56$, $P < 0.01$, $SE = 0.23$), but declined in basins experiencing active management to reduce P and N inputs ($\tau = -0.65$, $P < 0.0001$). In addition, the effect of
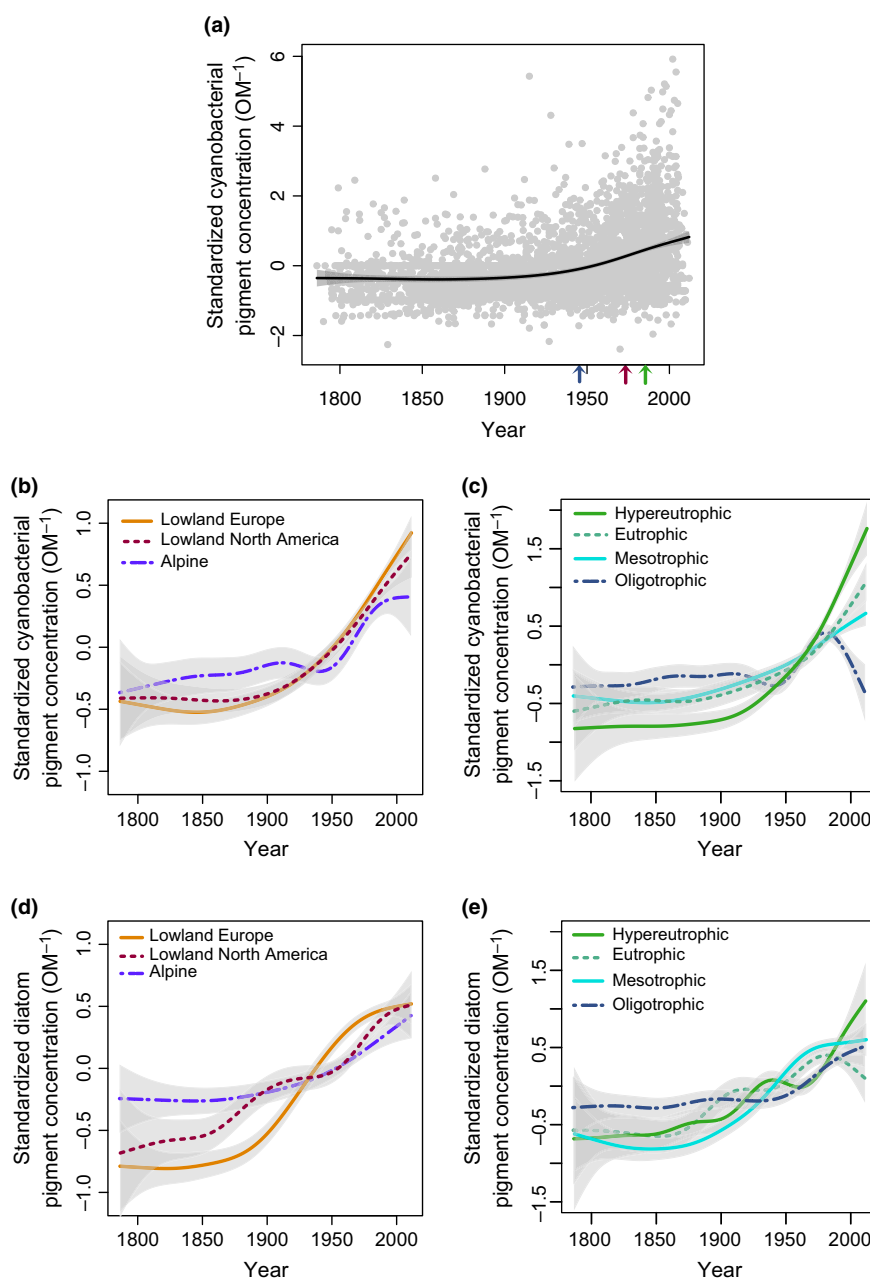
Figure 3 Additive mixed-effect model (AMM) response curves for the cyanobacteria and diatom paleolimnological data. Cyanobacteria concentration vs. (a) year ($R^2 = 0.14$, $n = 98$), (b) year × region ($R^2 = 0.15$, $n = 98$) and (c) year × trophic state ($R^2 = 0.17$, $n = 88$). Diatom concentration vs. (d) year × region ($R^2 = 0.15$, $n = 95$), and (e) year × trophic status ($R^2 = 0.16$, $n = 86$). Arrows in (a) provide the 1st quantile (blue: 1943), median (red: 1973) and 3rd quantile (green: 1985) of breakpoint regression dates. All models were significant, as was the temporal autocorrelation structure (range: $\phi = 0.36$–0.50, $P < 0.0001$). Grey bands indicate 95% confidence intervals of the predicted mean based on the AMM.

nutrients appeared to be greater for cyanobacteria than diatoms, as the ratio of cyanobacteria: diatom abundance (wet biomass per litre) decreased significantly when comparing eutrophication to restoration groups ($\beta = -0.28$, $P = 0.040$), while the effect size of nutrients was much smaller for diatoms than for cyanobacteria (ANCOVA: $F = 4.1$, $P = 0.03$; Fig. S5). Finally, the predominant role of nutrients was confirmed by considering alternative predictors of the LTM cyanobacterial trends. In this case, temperature alone accounted for 21% of the variation in observed M-K trends ($P = 0.030$), but the sig-

nificance of thermal effects was lost when the nutrient trajectory (i.e. increasing, decreasing or no change in nutrient status) was also included in the model. In contrast, nutrient trajectories alone accounted for 57% of the variance ($F = 13.0$, $P < 0.0001$).

DISCUSSION

Our synthesis of available paleolimnological records and a comprehensive suite of LTM data provide clear evidence that
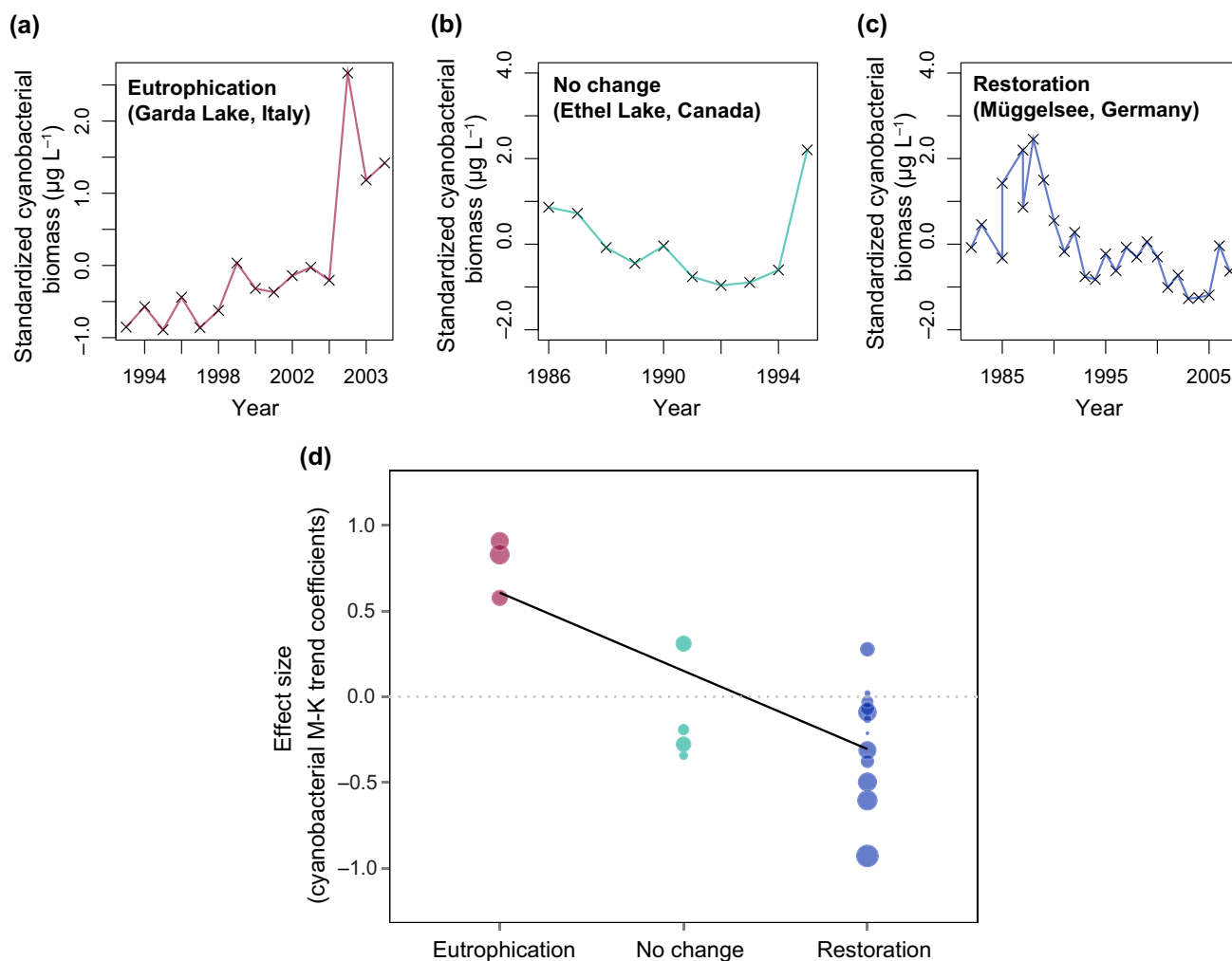
Figure 4 Meta-analysis of the long-term monitoring data (n = 18). Examples of cyanobacterial biomass time series from three individual lakes undergoing (a) eutrophication [reported increase in total nitrogen (TN) and total phosphorus (TP)], (b) no consistent nutrient change and (c) restoration lakes (reported decrease in TN and TP). (d) Meta-analysis of cyanobacterial trend coefficients with a random effect for nutrient trajectory. Circle size corresponds to pooled variance weight. Effect sizes decreased significantly (P = 0.0001) from the eutrophication to restoration groups: overall effect size of the eutrophication group (z = 0.64, P = 0.001, n = 3) was greater than the no change group (z = −0.69, P = 0.004, n = 4), which was greater than the restoration group (z = −0.78, P < 0.0001, n = 12).

cyanobacterial abundance has increased at the continental scale within the north temperate-subarctic zone. Our analyses further show that these potentially toxic primary producers have increased at rates greater than those of less harmful phytoplankton. Since the mid-1940s, the cyanobacterial rates of increase have accelerated, particularly in lowland nutrient-rich regions. This study adds to the growing body of evidence that human activities are now a widespread force in shaping the structure and functioning of inland waters (e.g. Keatley *et al.* 2011; Wolfe *et al.* 2013). Nutrients are being applied to the global landscape in excess of crop demands (MacDonald *et al.* 2011), leading to widespread eutrophication of lakes (Carpenter *et al.* 2011). Here, we show for the first time that temperate to subarctic lakes are responding to this enhancement with nonlinear increases in potentially toxic cyanobacteria.

Timing of the observed break points for many of the cyanobacterial pigment time series corresponded closely to that of

the proposed onset of the 'Great Acceleration' (*c.* 1945; *sensu* Steffen *et al.* 2007), in which exponential increases in fertiliser application and the introduction of phosphorus-containing detergents elevated the transport of nutrients to lakes via both lotic and atmospheric pathways (Holtgrieve *et al.* 2011; Mac-Donald *et al.* 2011). The fact that some previous long-term case studies note an increase in cyanobacteria in response to agriculture prior to the Anthropocene (Fritz 1989) lends support to our inference that global intensification of agricultural activities during the past 200 years underlies the observed cyanobacterial expansion (Steffen *et al.* 2007).

Residual variance in centennial cyanobacterial trends may reflect effects of other environmental forcing mechanisms. However, although warmer temperatures favour cyanobacterial development in numerous contemporary studies (Kosten *et al.* 2012; Taranu *et al.* 2012; Beaulieu *et al.* 2013), our continental-scale analyses demonstrate for the first time that the nonlinear expansion of historical blooms was primarily regu-

lated by increases in the nutrient content of surface waters (Fig. 3). These results are consistent with the conclusions drawn by Leavitt *et al.* (2009), who noted that nutrient influxes can overwhelm the effects of energy inputs when stressors co-occur, particularly in the case of primary producers. Elevated atmospheric content of CO_2 could also have favoured increased phytoplankton primary production during the 20th century (O'Neil *et al.* 2012), but there are several lines of evidence which suggest that this mechanism did not produce the disproportionate increases in cyanobacteria observed at the scale of continents (Fig. 1). First, although dense cyanobacterial blooms can effectively deplete CO_2 in surface waters, these phytoplankton are frequently outcompeted by eukaryotic algae under CO_2-enriched conditions (Low-Decarie *et al.* 2011). Second, most inland waters are over-saturated with CO_2 (Raymond *et al.* 2013) such that the observed increase in atmospheric CO_2 concentrations should not significantly increase pCO_2 within the lake. Third, recent research reveals that riverine fluxes of dissolved inorganic carbon, rather than atmospheric CO_2 concentrations, are the primary controls of lentic CO_2 content (McDonald *et al.* 2013). Finally, comparison of 20th century trends in atmospheric CO_2 (Bruno & Joos 1997) and cyanobacterial abundance (Fig. 3) show substantial divergence, particularly prior to 1945, when CO_2 increased monotonically, but standardised pigment concentrations were relatively stable (mean breakpoint in the cyanobacterial trend *c.* 1960). Thus, while global increases in atmospheric CO_2 can have synergistic interactions with nutrients (Verspagen *et al.* 2014), we conclude that it is unlikely that greenhouse gas build-up is the principal driver of the trends reported in our study.

One could also argue that the lakes identified in our synthesis, although based on specific criteria that were defined prior to our search of the literature, reflect a bias that is present in the literature and thus might not be representative of lakes in temperate and subarctic regions. For example, previous research programs may have been explicitly targeting eutrophic lakes and thus we would capture a disproportionate abundance of nutrient-rich lakes. However, our synthesis of the paleolimnological literature included lakes from all trophic states (i.e. 52% oligo- to mesotrophic and 48% eu- to hypereutrophic) and this mirrors the distribution of trophic states found on the landscape of the conterminous USA based on a probability-based sampling of sites through the 2007 National Lake Assessment (50% oligo- to mesotrophic and 50% eu- to hypereutrophic; U.S. Environmental Protection Agency 2009). Furthermore, our focus on sites reporting myxoxanthophyll may have overestimated the cyanobacterial response due to intrinsic differences in studies lacking data on the myxoxanthophyll pigment. We quantified this potential bias by conducting a supplemental analysis of diatom trends in lakes where only diatoxanthin was reported (*n* = 39 lakes reporting trophic status; Fig. S6). In these sites, the proportion of pristine (41%) and impacted (38%) catchments were comparable to the full lake set (*n* = 95), however, oligo- to mesotrophic lakes were more common (67%) than eu- to hypereutrophic lakes (33%). The lack of myxoxanthophyll in at least the less productive sites can be due to the relatively low abundances of cyanobacteria, as has been

shown in lake surveys (Kosten *et al.* 2012). Finally, an important bias that remains to be addressed is the potential for unbalanced representation of pristine to impacted sites across all of our sites. We indeed found that lakes with direct anthropogenic disturbances dominated our lake set (*n* = 60), whereas lakes situated in pristine catchments were lower in number (*n* = 30). A random sub-sampling of 30 impacted lakes showed that the increase in cyanobacteria was always substantially greater in impacted lakes than in non-impacted lakes (Fig. S3). As such, our main findings do not appear to be biased.

Overall, an important consequence of the large-scale increase in cyanobacteria is the potential for a parallel increase in algal toxins in lakes. Although cyanobacteria are not uniformly toxic (Monchamp *et al.* 2014), lake surveys reveal that the best local predictor of algal toxin concentrations is the biomass of cyanobacteria within the water column (e.g. Dolman *et al.* 2012). Yet, modelling the concentration of cyanotoxins across broader spatial scales has been difficult to date (e.g. Orihel *et al.* 2012). Indeed, little is known about whether local-scale relationships are generalisable to larger networks of lakes (e.g. Beaulieu *et al.* 2014), or whether cross-scale interactions among environmental gradients (e.g. nutrient concentrations and stoichiometry, temperature and light availability) result in an increased variability at the landscape scale (e.g. Soranno *et al.* 2014). Ultimately, a better understanding of the drivers of cyanotoxin concentrations across spatio-temporal gradients is critical given the acute health effects resulting from direct and indirect human exposure (Lévesque *et al.* 2014) and the isolation of new, ubiquitous toxins potentially linked to an increased incidence of neurodegenerative diseases and cancer in humans (Jonasson *et al.* 2010). Although improved understanding of the environmental and stoichiometric controls of cellular toxicity in phytoplankton communities is needed (Finlay *et al.* 2010; Van de Waal *et al.* 2014), our study reveals that regulation and reversal of centennial-scale increases in nutrient influxes may be a prerequisite for improving continental water quality. Furthermore, there are important feedbacks to consider in light of the recent findings that cyanobacteria in oligotrophic lakes might facilitate a regime shift of the ecosystem towards a more eutrophic state (Cottingham *et al.* 2015).

The present study provides the first quantitative evidence across north temperate to subarctic latitudes that cyanobacterial expansion has been nonlinear over time, disproportionate relative to other algae and induced mainly by nutrient fertilisation. Given that agricultural fertilisation is expected to increase two-fold over the next 30 years to meet the nutritional demands of an additional billion humans, particularly in urban centres (Carpenter *et al.* 2011), we anticipate that water quality degradation associated with cyanobacterial outbreaks will continue to intensify. Fortunately, LTM studies demonstrate that nutrient-enriched ecosystems can recover within decades of nutrient diversion, even in the face of continued atmospheric warming (Jeppesen *et al.* 2007), although interactions with climate change and other factors may prevent re-establishment of the biotic composition that existed prior to fertilisation (Bennion *et al.* 2011).

AUTHORSHIP

ZET and IG-E developed the study design. ZET performed all statistical analyses. All authors provided unpublished and published sedimentary core data, were responsible for ^{210}Pb dating of sediment cores, and provided data on explanatory variables used in the analyses. ZET, IG-E, PRL and LB wrote the original draft and all authors provided valuable discussion points and revisions to the manuscript.

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