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# Characterization of lacustrine harmful algal blooms using multiple biomarkers: Historical processes, driving synergy, and ecological shifts

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## ABSTRACT

Harmful algal blooms (HABs) producing toxic metabolites are increasingly threatening environmental and human health worldwide. Unfortunately, long-term process and mechanism triggering HABs remain largely unclear due to the scarcity of temporal monitoring. Retrospective analysis of sedimentary biomarkers using up-to-date chromatography and mass spectrometry techniques provide a potential means to reconstruct the past occurrence of HABs. By combining aliphatic hydrocarbons, photosynthetic pigments, and cyanotoxins, we quantified herein century-long changes in abundance, composition, and variability of phototrophs, particularly toxigenic algal blooms, in China's third largest freshwater Lake Taihu. Our multi-proxy limnological reconstruction revealed an abrupt ecological shift in the 1980s characterized by elevated primary production, *Microcystis*-dominated cyanobacterial blooms, and exponential microcystin production, in response to nutrient enrichment, climate change, and trophic cascades. The empirical results from ordination analysis and generalized additive models support climate warming and eutrophication synergy through nutrient recycling and their feedback through buoyant cyanobacterial proliferation, which sustain bloom-forming potential and further promote the occurrence of increasingly-toxic cyanotoxins (e.g., microcystin-LR) in Lake Taihu. Moreover, temporal variability of the lake ecosystem quantified using variance and rate of change metrics rose continuously after state change, indicating increased ecological vulnerability and declined resilience following blooms and warming. With the persistent legacy effects of lake eutrophication, nutrient reduction efforts mitigating toxic HABs probably be overwhelmed by climate change effects, emphasizing the need for more aggressive and integrated environmental strategies.

## 1. Introduction

Cultural eutrophication has resulted in a worldwide proliferation of harmful (toxic, food-web altering, hypoxia-generating) algal blooms (HABs) in freshwaters (Ho et al., 2019), threatening global water security and biodiversity (Huisman et al., 2018). In particular, the coupling of nutrient enrichment and climate warming favors cyanobacterial dominance and toxin-producing blooms which have risen through the 20th and 21st centuries (Kosten et al., 2012; Taranu et al., 2015; Kakouei et al., 2021; Meerhoff et al., 2022), with a significant occurrence of cyanotoxins in water environments (Taranu et al., 2019; Hayes et al., 2020). Management and mitigation of HABs are of tremendous

concern due to their detrimental effects on ecosystem and human health (Havens and Paerl, 2015; Taranu et al., 2017). Systematic monitoring programs are typically initiated once environmental problems have emerged and they rarely capture the full spectrum of aquatic ecosystem deterioration (Battarbee et al., 2005; Qin et al., 2019). One key challenge we face, therefore, is to determine the historical baseline conditions and ecological processes (particularly decadal to centennial) of toxic cyanobacterial blooms in freshwater lakes (Huisman et al., 2018; Ryo et al., 2019; Cao et al., 2020), and what might be the underlying mechanisms of multiple interacting drivers through time (Leavitt et al., 2009; Bruel et al., 2021; Huo et al., 2022).

Retrospective assessment using well-dated sediment archives,

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promisingly, provide past information on ecosystem features prior to monitoring, thus allowing reconstruction of high-resolution eco-environmental trajectories (Smol, 2010; Perga et al., 2015; Zeng et al., 2023). Traditional paleolimnological techniques for micro-algae are mostly based on microscopic identification, and use morphological remains to infer long-term changes of diatoms and a few cyanobacteria (Battarbee et al., 2005; Bunting et al., 2016). Recent advances in molecular techniques enable the analysis of sedimentary DNA to provide a broader taxonomic specificity (Monchamp et al., 2018; Cao et al., 2020; Ibrahim et al., 2020; Huo et al., 2022). Meanwhile, these techniques can be either time/labor-consuming, inadequate, or limited by undefined taphonomic and analytical issues (e.g., cumulative degradation, insufficiency of targeted reference library) (Domaizon et al., 2017). Alternatively, the rapid developments of high-performance chromatography and mass spectrometry technologies have laid a solid foundation for highly efficient and reliable approaches for analyzing multiple molecular biomarkers of photoautotrophs. In particular, a combination of aliphatic hydrocarbons (Meyers, 2003), photosynthetic pigments (Leavitt, 1993; McGowan, 2013), and cyanotoxins (Henao et al., 2019) applied to lake sediments can achieve unprecedented information on past dynamics of primary producer communities (e.g. phytoplankton versus macrophyte dominance), HABs, and toxin production (Waters et al., 2021).

Impacts of multiple stressors on freshwater ecosystems, such as climate change, eutrophication, hydrological alterations, and trophic cascades, may (i) complicate the identification of drivers and their interactions that are responsible for key changes (Leavitt et al., 2009; Deng et al., 2014), (ii) increase the risk of catastrophic shifts (e.g., macrophyte extinction, HABs) (Scheffer et al., 2001), and (iii) compromise the potency of restoration measures by changing the baseline conditions (Battarbee et al., 2005; Bruel et al., 2021). In lake ecosystems, temporal variability in primary production can rise due to persistent increases in nutrient influx (e.g., the paradox of enrichment) (Cottingham et al., 2000; Bunting et al., 2016), after which the state shift to prolific cyanobacteria is triggered by relative minor forcing (e.g., warming) that push lakes beyond critical thresholds (Dakos et al., 2015). The complexity and interactivity of stressors on ecosystems further hamper our understanding of how reducing local pressures may increase ecosystem resilience to climate change (Monchamp et al., 2018; Bruel et al., 2021; Carpenter et al., 2022). Retrospective examination of past ecological shifts and variability characteristics using long-term records, therefore, may provide critical insights on how nonlinear and abrupt HABs in response to environmental change may improve lake management and mitigation strategies (Bunting et al., 2016; Ryo et al., 2019; Bjorndahl et al., 2022).

In this study, we quantified one-century long changes in the production, composition, variance, and rate of change of phototrophic assemblages, particularly toxigenic cyanobacteria and their toxic metabolites, in a typical eutrophic lake (Lake Taihu, China) suffering from increasingly frequent cyanobacterial HABs. High-resolution time series of sedimentary biomarkers (*n*-alkanes, chlorophyll and carotenoid pigments, microcystins) and geochemistry (organic carbon, nutrients, metals) were quantitatively integrated with monitoring data and historical archives using ordination analysis and generalized additive models. We address two key questions: (i) what are the dynamic trajectories of photoautotrophic succession (macrophyte versus phytoplankton), HABs, and cyanotoxin production, and how were catastrophic shifts shaped by multiple stressors interacting through time? (ii) whether current HABs and historical changes in ecological variability were consistent with the establishment of an alternative stable state in Lake Taihu, and what are the implications for ecosystem resilience and baselines? Our methodology has considerable potential application globally to provide evolutionary insights into the mechanisms of freshwater ecosystem degradation and to open new management perspectives for ecological adaptation (Perga et al., 2015) to climate warming.

## 2. Materials and methods

### 2.1. Study area and sampling

Lake Taihu, China's third largest freshwater lake (~2338 km<sup>2</sup>), is located in the rapidly developing and highly urbanized Yangtze River Delta (Fig. S1). Currently, the catchment area is dominated by agricultural land (47.9%), urban land (24.3%), and water bodies (13.6%) that supports 4.8% (~65 million) of China's population and produces 11.6% of national GDP, despite occupying only 0.4% (~36,500 km<sup>2</sup>) of China's land area. There are approximately 219 rivers, with inflows going to the northern and western lake sides, and outflows occurring on the eastern sides. Due to nutrient supply and climate warming, this shallow (<3 m) lake has experienced accelerated eutrophication accompanied by potentially toxic cyanobacterial blooms (*Microcystis* spp.) (Guo et al., 2019). Coupled with southerly or southeasterly winds, annual cyanobacterial HABs occur earlier and persist longer in the northern and western lake (Shi et al., 2019), threatening the water security of approximately 30 millions of inhabitants (Qin et al., 2019). However, continuous water quality monitoring and remote sensing observations are limited to recent ~30 years.

In order to reconstruct longer-term history of lake eutrophication and HABs for assessing ecosystem state dynamics and optimizing management strategies, two duplicate sediment cores (~40 cm) with intact sediment-water interfaces were retrieved (Smol, 2010) from one typical, relatively sedimentation-stable area of Lake Taihu (central west, N31.11623°, E120.06152°) in March 2017 and July 2020 (Fig. S1) (Lin et al., 2020). The cores were cut into 0.5-cm contiguous intervals, and subsamples were refrigerated at -4 °C. Modern samples including cultured cyanobacterium *Microcystis* (*n* = 5), predominant macrophytes (6 species), and surface forest soil (*n* = 5) in and around the lake were also collected (Table S1). In the laboratory, all the samples were freeze-dried, and core sediment subsamples were processed for radiochronology, physicochemical parameters, and biomarker analyses. The sediment core collected in 2020 was specifically for the measurement of microcystins, and other proxy analyses were conducted on the previous core. These cores from the increasingly blooming area were taken as an analytical test for multi-biomarker reconstruction in this study.

### 2.2. Radiochronology

Isotopes of <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>137</sup>Cs for sediment dating were measured using a gamma spectrometer (ORTEC., USA), which provides an absolute detection efficiency (≥95%) according to calibrated sources and standard samples. The age model of the sediment core collected in 2017 was computed with *serac* R package (Bruel and Sabatier, 2020). We have previously reported detailed protocols of geochronology and reliable results that suggested mean sedimentation rates of 0.38 cm yr<sup>-1</sup> for the lake area over the past hundred years (Lin et al., 2020). The age for another core was thus determined through core matching based on lithology and geochemical parameters (Cheng et al., 2023).

### 2.3. Sediment physicochemical analysis

The analytical determination of major sediment physicochemical parameters, including grain size, mass magnetic susceptibility, metal elements (e.g., Al, Ti, Fe, Mn), total phosphorus (P), total nitrogen (N), organic carbon (OC), and nutrient stoichiometry (molar ratios of C/N and N/P) have been well described in our previous studies (Lin et al., 2020, 2021). The temporal changes of these paleovariabiles are shown in detail in Fig. S2.

### 2.4. Multi-biomarker analysis

Three kinds of sedimentary molecular biomarkers were analyzed comprehensively in Lake Taihu cores to characterize the integral

historical processes of HABs. First, aliphatic hydrocarbons (particularly *n*-alkanes) were used to distinguish the relative contributions of organic matter from algae, macrophytes, and terrestrial plants, reflecting major succession of photoautotrophic communities (Meyers, 2003). Second, photosynthetic subfossil pigments are reliable indicators to track the past changes in lake primary productivity and algal community (McGowan, 2013), particularly the abundance and composition of bloom-forming algae (Bjordahl et al., 2022). On this basis, concentrations of cyanotoxins were measured in the sediments to infer the magnitude of past HABs and their toxin production through time (Waters et al., 2021).

**Aliphatic hydrocarbons.** Sediment core subsamples, samples of cultured cyanobacterium *Microcystis*, collected aquatic macrophytes, and surface forest soil were all analyzed for *n*-alkanes following standard procedures (Zhang et al., 2019). In brief, homogenized dry samples were ultrasonically extracted four times using the dichloromethane/methanol (9:1, v/v) mixture. Total lipids were concentrated to dry with pure nitrogen and saponified with 6% KOH in methanol at room temperature for 12 h. The neutral lipids containing *n*-alkanes were further extracted with *n*-hexane through silica gel column chromatography. Finally, *n*-alkanes were measured using an Agilent 7890 Gas Chromatograph (GC) equipped with an Agilent 5975 mass spectrometer (MS) with an external C16 standard for identification and quantification. A set of proxies (ACL, TAR, P<sub>aq</sub>, and S/M) were calculated based on odd-carbon *n*-alkanes to identify the composition and sources of sedimentary organic matter (Ficken et al., 2000; Meyers, 2003), which are explicitly described in the Supporting Information (Text S1).

**Photosynthetic subfossil pigments.** Sedimentary chlorophyll and carotenoid pigments were extracted and purified following the standard method with optimal mixtures of acetone, methanol, and deionized water at -20 °C for 12 h (McGowan, 2013). After filtering, nitrogen blowing, and re-dissolution, individual chlorophyll and carotenoid compounds were isolated and quantified using an Agilent 1200 series high-performance liquid chromatography unit (HPLC, Canada) with an ODS Hypersil column (250×4.6 mm; 5 μm particle size) and photo-diode array detector. Pigment concentrations were calibrated with authentic standards (DHI Lab, Denmark) and are expressed as nmol pigment g<sup>-1</sup> OC because the degradation of organic pool can partly compensate for bias of pigment diagenesis (Leavitt, 1993; McGowan, 2013). An ultraviolet radiation (UVR) index was calculated by dividing UVR-absorbing pigment (scytonemin derivative) relative to the sum of key carotenoids (diatoxanthin, lutein-zeaxanthin, and alloxanthin) and multiplying by 100 (Leavitt et al., 1997). Calibration in whole-lake experiments revealed that this index increases as a linear function of the depth of UVR penetration (Leavitt et al., 1997), such that higher index values indicate greater exposure to potentially damaging UVR, namely better light conditions and higher water clarity (McGowan et al., 2012).

**Cyanotoxins.** Sedimentary microcystins (MCs) were analyzed following previously published procedures (Cheng et al., 2023). Briefly, homogenized sediment subsamples were ultrasonically extracted three times using 0.1 mol EDTA/Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub> (1:1, v/v, pH=3) mixture. The extracts were loaded into hydrophilic-lipophilic balance cartridges previously activated with methanol and washed with deionized water. After a further washing step and elution with methanol, the toxin eluent was dried using pure nitrogen at 40 °C. Finally, three dominant MC congeners (MC-LR, MC-YR, MC-RR) (Henaio et al., 2019; Xue et al., 2023) were measured using a Waters H-class ultra HPLC tandem triple quadrupole mass spectrometer (USA) with an Acquity UPLC BEH C18 column (100×1 mm, 1.7 μm particle size). The calibration ranges were 0–200 μg L<sup>-1</sup> (R<sup>2</sup>≥0.99) using MC standards from Sigma-Aldrich (≥95%, Germany), and the relative standard deviations were less than 5% as quantified from external standards and blanks. The MCs were reported as ng toxin g<sup>-1</sup> OC to compensate for diagenetic degradation (Waters et al., 2021).

## 2.5. Historical data

Time series of major environmental variables from the study area were collected for the past ~120 years to quantify the statistical relationships between the paleorecords of lake ecosystem status (*n*-alkanes, pigments, cyanotoxins, OC, total nitrogen and phosphorus) and identify potential causal effects associated with regional changes in climate, agriculture, and urbanization (Fig. S3). Climate records including annual mean air temperature, wind speed, and total precipitation were obtained from a nearby meteorological station #58,358 of China (<https://data.cma.cn/>) with supplements from the global historical climatology network (<https://cdiac.ornl.gov/>). Estimates of agricultural intensity referring to total fertilizer and pesticide consumption and rice yield (Bunting et al., 2016) were compiled from official statistical data of nearby cities (<https://www.stats.gov.cn/>) and previous documents (Ellis and Wang, 1997). Historical records of urbanization (mainly urban sewage) as mirrored by total wastewater effluent, local population (dominated by urban inhabitants) and GDP (McGowan et al., 2012) were obtained from Taihu Basin Authority of Ministry of Water Resources and official statistical documents (Fig. S3). Although too short for our statistical analysis, limnological variables monitored during 2005–2020 were obtained from the Taihu Laboratory for Lake Ecosystem Research for data integration.

## 2.6. Numerical analysis

Temporal variations of historical document data, lake geochemical variables and biomarkers were visualized using Grapher 14 and R software. Sedimentary paleorecord time series (*n*-alkanes, pigments, total MCs, OC, nitrogen, and phosphorus) were log-transformed if needed, then centered (mean=0), standardized (variance=1.0), and harmonized to equivalent sampling intervals prior to statistical analyses (Bunting et al., 2016).

**Change phase analysis.** The timings of paleorecord changes were determined using stratigraphically constrained incremental sum of squares (CONISS) cluster analysis in the Tilia program. Sequential T-test algorithm based on variance analysis with prewhitening ( $p < 0.05$ , cut-off length=10) and calculation of cumulative sum of differences were also performed to identify significant change points and the main phases of past ecological dynamics (Andersen et al., 2009; Cao et al., 2020).

**Ordination analysis.** The gradient length of <1 total standard deviation unit for the paleorecords suggested that a linear model was suitable for unconstrained and constrained ordinations using Canoco 5 program (ter Braak and Šmilauer, 2012). Nonmetric multidimensional scaling (NMDS) analysis based on Bray–Curtis distances was performed to identify structural changes in paleorecords through time in a reduced two-dimensional space. For technical purposes, the scores of both NMDS axes (NMDS1, NMDS2) were taken as the main representative variation indices of ecological dynamics (Ibrahim et al., 2020). Up to ten environmental variables referring to regional climate, agriculture, and urbanization (Fig. S3) were included in the initial analysis to evaluate their significance and collinearity, via a series of forward selection and restricted Monte Carlo permutation tests ( $n = 999$ ) (ter Braak and Šmilauer, 2012). Four runs of redundancy analysis (RDA) were then conducted to explore potential relationships between paleorecords and individual categories of significant environmental variables of agriculture (fertilizer consumption), urbanization sewage (population), and climate (air temperature, wind speed), respectively. Variance partitioning analysis (VPA) was used to estimate the fractions of historical variances in the paleorecord time series explained by all categories of significant environmental variables and their interactions (Bunting et al., 2016).

**Generalized additive model analysis.** Generalized additive models (GAMs) analysis were performed to distinguish and quantify the impacts of local human activity and regional climate change on Lake Taihu ecosystem dynamics through time. The inferred human index (i.e.,



weighted averages of z-score normalized population and fertilizer consumption) and climate index (i.e., ratios of annual mean air temperature to wind speed) were chosen as representative predictor variables, while the response variables were scores of both NMDS axes calculated from the dataset collating multiple paleorecords (details in Text S2). The functions *gam* and *predict.gam* from “mgcv” package in R software (Wood, 2017) were used, and GAMs analyses were conducted following standard procedures (Simpson, 2018) and our previous descriptions (Lin et al., 2021).

**Temporal variability analysis.** Sedimentary pigments and other biomarkers can accurately record the variability of algal communities following fertilization through time (Cottingham et al., 2000; Meyers, 2003), and thus reliably reflect changes in ecosystem variability during state shifts (Carpenter et al., 2011; Bunting et al., 2016). Historical changes in the temporal variability of total autotrophic production (Chl *a* + echinenone), bloom-forming algal abundance (sum of canthaxanthin and lutein-zeaxanthin) (Bunting et al., 2016; Bjorndahl et al., 2022), cyanotoxins (total MCs) (Waters et al., 2021), and the scores representing paleorecord structure (NMDS1) were estimated by the numerical simulation of both variance and rate of change (RoC) following standard technical recommendations (Simpson, 2018;

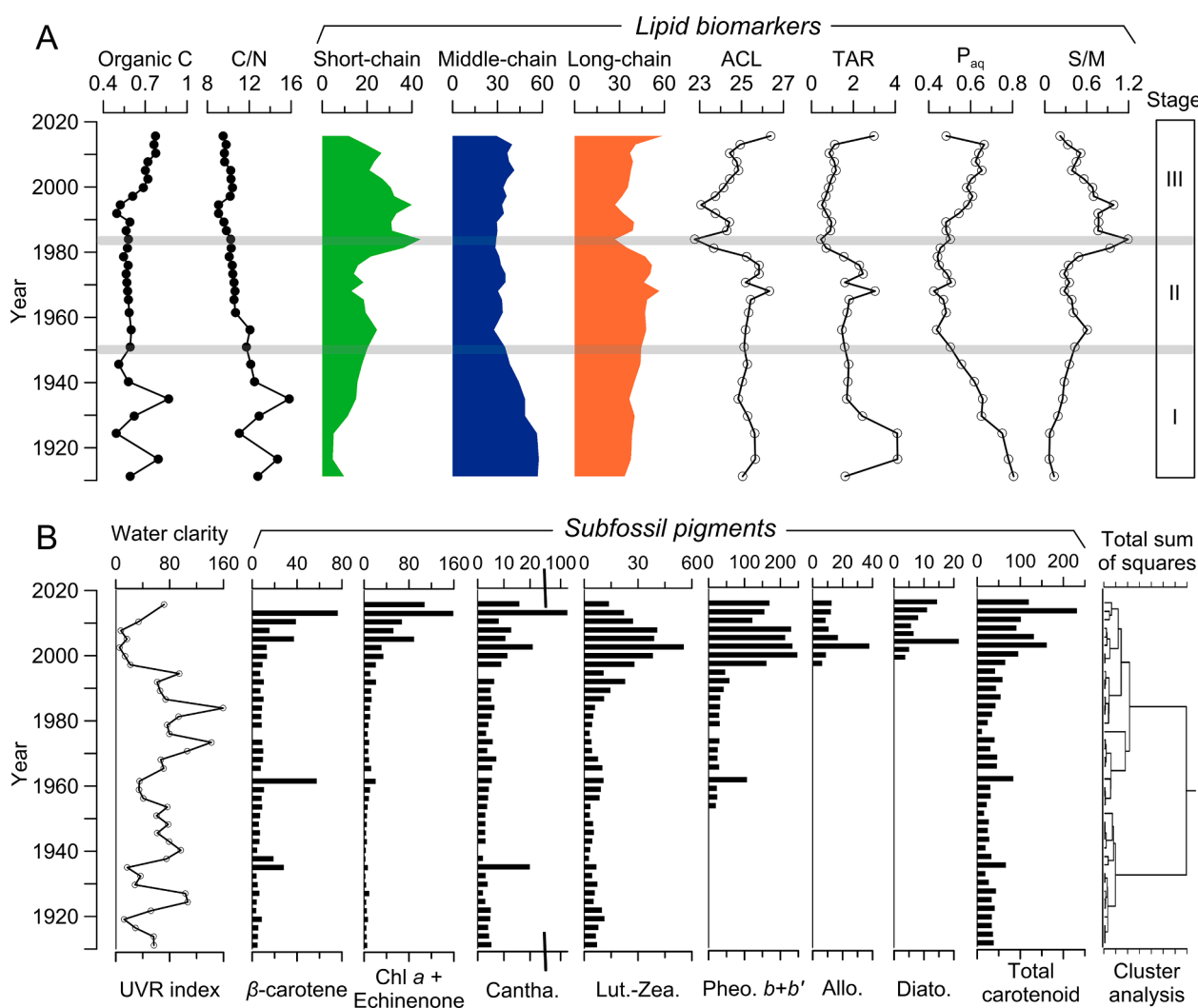
Bjorndahl et al., 2022). The variances were extracted by calculating the standard deviations performed with the “earlywarnings” packages in R (Dakos et al., 2015). The RoCs were generated from the posterior simulations of the fitted GAMs using the first derivative of a spline. All models fitting the selected time series contained a continuous-time first-order autoregressive [CAR(1)] process to account for temporal autocorrelation. These analyses were performed using R software with the “mgcv” and “vegan” packages (Wood, 2017; Simpson, 2018).

### 3. Results

Multiple analyses of sediment cores from Lake Taihu revealed distinct dynamics of lake primary producers following fertilization and climate change. Proxies for organic matter, nutrients, autotrophic production, and cyanobacterial HABs showed particularly prominent changes, which exhibited three similar phases since the 1900s.

#### 3.1. Sedimentary OC, nutrients and n-alkanes

Sediments were mainly composed of coarse silt (16–63  $\mu\text{m}$ ), accounting for >50% prior to 1950. Peaks of OC, C/N ratios (>12), total



**Fig. 1.** Temporal changes of lipid and subfossil pigment biomarkers in Lake Taihu sediment cores. (A) Profiles of sedimentary organic C (%), molar ratios of C/N, total *n*-alkanes within different length of chains (%), and selected *n*-alkane proxies (ACL, TAR,  $P_{aq}$ , and S/M) (Text S1). The gray horizontal bars partition three potential change phases for lipid biomarkers. (B) Profiles of sedimentary chlorophyll and carotenoid pigments ( $\text{nmol g}^{-1}$  OC) and UVR index, along with the CONISS cluster analysis. Cantha. = canthaxanthin (colonial cyanobacteria), Lut.-Zea. = lutein-zeaxanthin (chlorophytes and cyanobacteria pigments, i.e., “bloom-forming” taxa), Pheo. *b* + *b'* = pheophytin *b* + *b'* (chlorophytes), Allo. = alloxanthin (cryptophytes), Diato. = diatoxanthin (primarily diatoms), Chl *a* + Echinenone (all phototrophs and total cyanobacteria), and  $\beta$ -carotene or total carotenoid (total autotrophic production).

phosphorus and nitrogen in the mid-1910s and ~1930s, coeval with higher sand content and lithogenic elements (e.g., Al, Ti) (Fig. 1A and Fig. S2), probably showed enhanced inputs of terrigenous nutrients and organic matter (Meyers, 2003). Sedimentary *n*-alkanes before the 1950s were dominated by middle-chain (*n*-C<sub>21</sub>, *n*-C<sub>23</sub>, *n*-C<sub>25</sub>) and long-chain (*n*-C<sub>27</sub>, *n*-C<sub>29</sub>, *n*-C<sub>31</sub>, *n*-C<sub>33</sub>) components (averaged 87.5%) (Fig. 1A and Fig. S4) similar to that of macrophytes and surface soil (Fig. S5). These characteristics together with relatively high values of TAR and P<sub>aq</sub> (>0.4), and low S/M ratios suggested dominant contributions of organic matter from submerged/floating macrophytes and allochthonous origin (Ficken et al., 2000; Meyers, 2003).

During the 1950s–1980s, content of fine silt (4–16 μm) and lithogenic elements increased slightly while OC and nutrients remained stable (Fig. 1A and Fig. S2). The C/N ratios decreased continuously to <10 and proportions of short-chain *n*-alkanes (*n*-C<sub>17</sub>, *n*-C<sub>19</sub>) increased sharply since the mid-1970s, reflecting substantially higher algal organic matter contribution, as algal communities (particularly cyanobacteria) have been identified as main contributors of short-chain *n*-alkanes in our survey (Fig. S5) and previous analyses (Zhang et al., 2017). Middle-chain *n*-alkanes and P<sub>aq</sub> decreased and remained at low levels while S/M ratios fluctuated and increased to a high level, suggesting a potential transition toward an algae-dominated lake state. Meanwhile, long-chain *n*-alkanes, ACL, and TAR as indicators of allochthonous organic matter input (Meyers, 2003) showed coeval decreases and relatively low values from ~1970.

Since the 1990s, sediments were dominated by fine particles (<16 μm) enriched with magnetic mineral and lithogenic elements (Fig. S2) relating to well-weathered terrigenous detritus (Lin et al., 2020). Sedimentary phosphorus, nitrogen, and OC showed ~1.5-fold increases while C/N ratios fluctuated around 10. Short-chain *n*-alkanes from algae maintained a relatively high abundance followed by a decrease, whereas middle-chain *n*-alkanes and P<sub>aq</sub> values remained relatively low and stable (Fig. 1A). These results along with relatively high S/M ratios suggested an algae-dominated organic matter source and a lake ecological regime that has greatly deviated from the historical conditions prior to 1950.

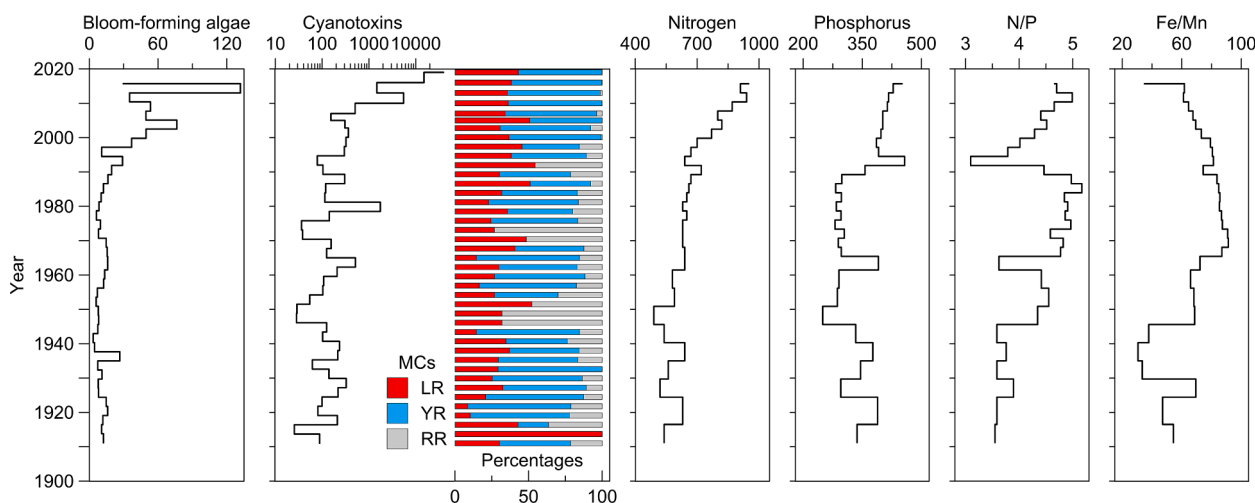
### 3.2. Subfossil pigments and cyanotoxins

Analyses of subfossil chlorophylls, carotenoids, and cyanotoxins revealed main shifts of algal communities are consistent with pronounced lake eutrophication (Figs. 1B and 2). An increase in the ratios of

labile to chemically-stable chlorophylls (Chl *a* : Pheophytin *a*) in the upper core (Fig. S6) reflected slightly improved pigment preservation when algal production increases as expected (Leavitt, 1993; Waters et al., 2021). Additionally, ratios of total carotenoids to total identified pigments did not change with depth, indicating a relatively constant preservation environment since the 1900s. This phenomenon coincided with generally stable redox conditions since the 1950s as inferred from Fe/Mn ratios (Fig. 2). Thus, the molecular biomarkers were well-preserved in recent sediments of Lake Taihu beyond similar human-impacted waters elsewhere (Bunting et al., 2016; Zastepa et al., 2017; Waters et al., 2021; Huang et al., 2022; Zeng et al., 2023).

Cluster analysis of pigment assemblages identified the most significant turning points around the 1950s and 1980s, and that algal communities showed two main patterns of change (Fig. 1B). First, concentrations of pigments from siliceous algae (diatoxanthin) and cryptophytes (alloxanthin) that commonly grow in the spring (Deng et al., 2014), were too low to be detected before ~1995, then generally increased in recent sediments. Second, subfossil biomarkers from summer bloom-forming colonial cyanobacteria and chlorophytes (canthaxanthin, lutein-zeaxanthin, pheophytin *b* and *b'*) (Guo et al., 2019), together with chemically-stable indicators of total algal abundance ( $\beta$ -carotene, total carotenoids), were relatively constant during most of the 20th century, increased 300–700% to high levels during the 1980s–1990s, then showed few fluctuations or lower trends in the recent decade. The echinenone (cyanobacterial pigment) that is often found to be extremely enriched in *Microcystis* (Hesse et al., 2001) was coeluted with labile Chl *a* (all phototrophs) in HPLC, which collectively showed a ~9-fold increase similar to  $\beta$ -carotene. Correspondingly, a sharp decrease of the UVR index occurred during the 1980s–2000s, indicating a marked decline in UV exposure and water clarity (Leavitt et al., 1997). Taken together, these results demonstrate that algal production increased ~3- to 7-fold since the 1980s when an ecosystem state shift was initiated. Recent modification of algal communities inferred from pigments is basically consistent with modern monitoring and molecular genetic results that showed a >50% phytoplankton biomass contribution from *Microcystis*-dominated cyanobacteria (Zhang et al., 2018a; Guo et al., 2019, 2023).

Cyanotoxins (three microcystin congeners) were detected throughout the century-long sediment record, and their production showed historical variations corresponding to bloom-forming cyanobacterial proliferation (Fig. 2). In general, concentrations of total MCs fluctuated without clear directional trends before ~1950, peaked



**Fig. 2.** Historical reconstruction of harmful algal blooms and cyanotoxin production associated with sedimentary nutrient and redox variables in Lake Taihu. Bloom-forming algae is indicated by the sum of canthaxanthin and lutein-zeaxanthin pigments ( $\text{nmol g}^{-1} \text{OC}$ ). Cyanotoxins are reported as the sum of three dominant microcystin congeners (MC-LR, MC-YR, MC-RR) ( $\text{ng g}^{-1} \text{OC}$ ) and their relative percentages. Total sediment nitrogen ( $\text{mg kg}^{-1}$ ), phosphorus ( $\text{mg kg}^{-1}$ ), and molar ratios of N/P and Fe/Mn are also shown.

between the 1960s (514 ng g<sup>-1</sup> OC) and the 1980s (1752 ng g<sup>-1</sup> OC), then increased exponentially since the 1990s to the highest value (~40,000 ng g<sup>-1</sup> OC) in the surface sediments. Elevated production of total MCs was mainly sourced from increased contributions of highly toxic MC-YR (averaged 45%) and MC-LR (averaged 36%), whereas contribution of hypotoxic MC-RR (averaged 19%) generally decreased since the 1950s. Moreover, rapid growth of cyanotoxin production was in pace with enriched nitrogen, phosphorus, elevated N/P ratios, and decreased C/N ratios, indicating a combined effect of nutrient inputs and stoichiometry changes (Xue et al., 2023).

### 3.3. Driver-response relationships

NMDS analysis tracked three distinguishable groups from biomarker and geochemistry assemblages since the 1900s (Fig. S7), as also observed from the stratigraphic temporal cluster (Fig. 1). The main gradient of paleorecord changes was associated with increased levels of autotrophic production, bloom-forming cyanobacterial proliferation, cyanotoxin occurrence, and loss of macrophytes and water clarity as nutrient enrichment led to changes in trophic state. This ecological gradient was highly correlated with NMDS1 and explained 81.3% of variation compared to 18.7% from NMDS2. Further statistical analyses using sequential T-test and cumulative sum of differences for NMDS1 scores confirmed shifting points of the lake ecosystem around the 1950s and 1980s (Fig. S8).

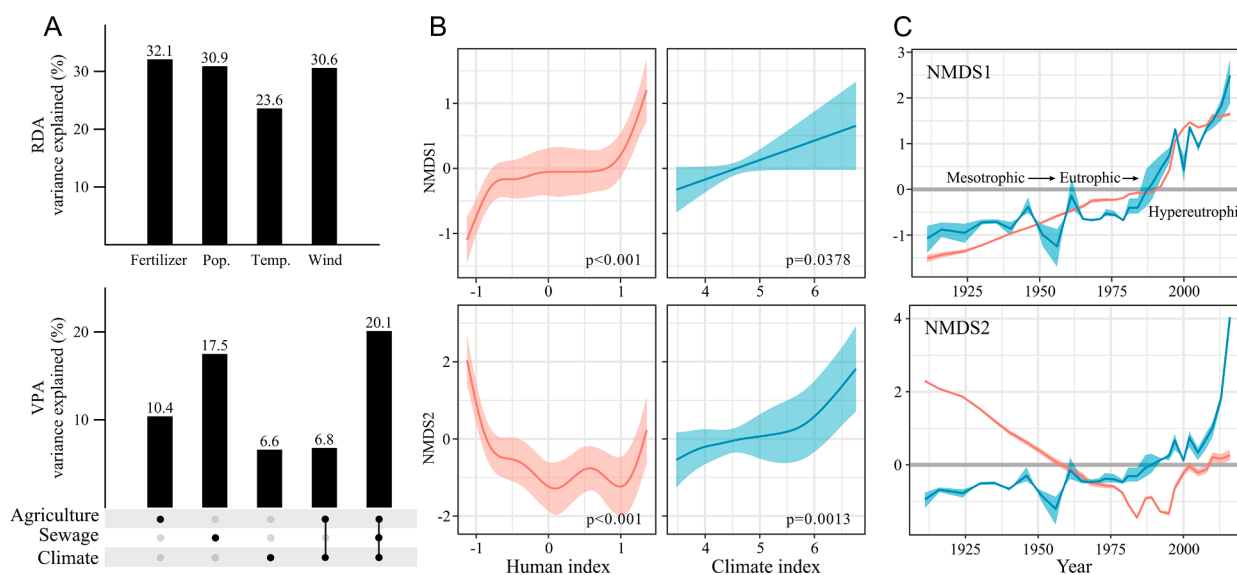
RDA with the agriculture predictor (fertilizer) alone explained 32.1% of variation in geochemistry and biomarkers during the past hundred years, similar to the variation explained by the sewage predictor (population, 30.9%) alone (Fig. 3A and Fig. S9). In contrast, RDA using climate variables either elevated air temperature (23.6%) or declined wind speed (30.6%) alone explained fewer changes in the paleorecords. VPA showed that environmental variation associated with agriculture, sewage, and climate totally explained 61.4% of past dynamics in lake ecosystem (Fig. S9). Comparison of individual and interactive categories suggested that most of the explained variance arose from total interactions between agriculture, sewage, and climate change (20.1%),

followed by that from sewage (17.5%) and that from agriculture (10.4%) alone (Fig. 3A). The remaining unexplained variance (38.6%) may be related to other environmental factors, particularly precipitation, hydrological fluctuations, and extreme weather events, which are important but usually hard to statistically quantify in long-term driver-response analysis (McGowan et al., 2012; Bunting et al., 2016).

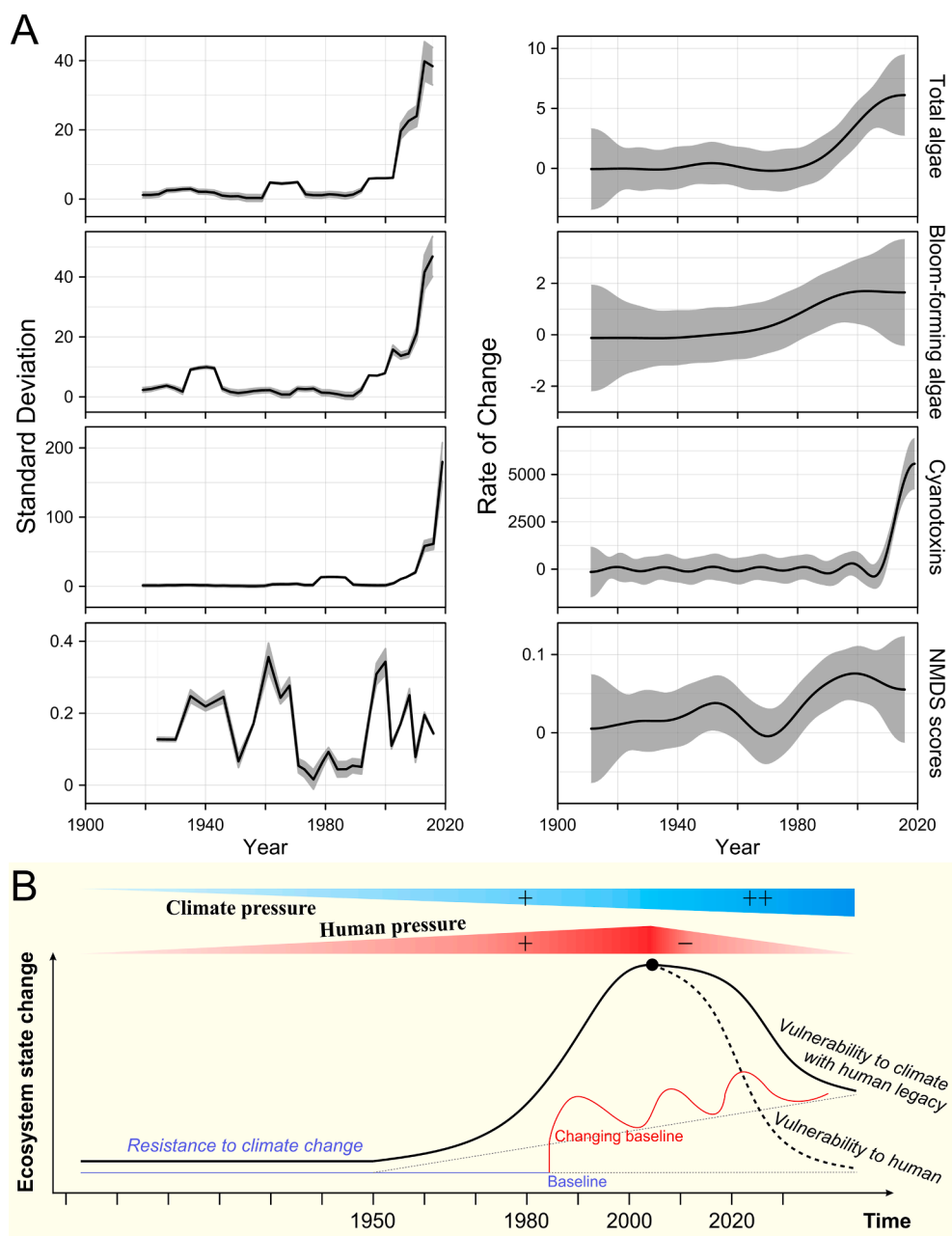
GAMs estimated when and to what extent human activities and climate change contributed to the past ecological dynamics of Lake Taihu, as indicated by the NMDS1 and NMDS2 scores of the paleorecords (Fig. 3). Documentary data-inferred human and climate indices (Text S2, Fig. S7) were considered as representative predictor variables according to our RDA results and previous investigations at Lake Taihu (Zhang et al., 2018b; Lin et al., 2021). Human and climate indices together explained 94.4% of variation in NMDS1 and 91.3% in NMDS2, respectively (Table S2). The smooth functions (Fig. 3B) illustrated a nonlinear response of NMDS1 to human index and a significantly linear response to climate. NMDS2 also responded significantly to human and climate indices with nonlinear trends. From a temporal perspective, the simultaneous effects of changing human activity and climate were detected for both NMDS axis scores since the 1980s when simultaneous positive effects on the NMDS1 prevailed (Fig. 3C). As lake trophic status deteriorated, the contribution of climate increased and even surpassed that of anthropogenic nutrient influx in recent decades.

### 3.4. Historical changes in ecological variability

The fitted GAMs were significant ( $p < 0.001$ ) for dominant biomarker time series of primary producers, community turnover, and cyanotoxins (Table S3), allowing estimation of historical patterns of lake ecosystem variability. Temporal trends in variance and RoC were similar for specific variables (Fig. 4A). Variance of time series in total and bloom-forming algae (particularly cyanobacteria) started to increase from the early 1990s, and the corresponding RoCs increased significantly since the mid-1980s. In contrast, cyanotoxins showed sharp increases in variance and RoC after ~2010, lagging behind the initial *Microcystis* blooms. Changes in variance and RoC of the scores of NMDS1 axis were



**Fig. 3.** Multivariate statistical analysis illustrating the dynamic relationships between paleorecords and forward selected environmental variables. (A) Century-long variation in geochemical variables and multi-biomarkers explained by categories of agriculture (fertilizer consumption), sewage (population), climate (air temperature, wind speed) via four individual runs of redundancy analysis (RDA) and one run of variance partitioning analysis (VPA) (Fig. S9). All categories of significant environmental variables and their interactions are included in VPA. (B) Generalized additive model (GAM) analysis of the fitted smooth functions between the inferred human index, climate index, and scores of both NMDS axes of paleorecords (NMDS1, NMDS2) (Fig. S7). (C) GAM results indicating the contributions (curves with 95% pointwise confidence intervals) of human (red) and climate (blue) variables to NMDS1 and NMDS2 through time. The relative importance (increase/decrease) is deduced from the centered-scaled values on y-axis scale except zero, and significant correlations with predictors are shown for each response variable in Table S2.



**Fig. 4.** Temporal changes in ecological variability and conceptual diagram illustrating the response of Lake Taihu ecosystem to multiple changing stressors through time. (A) Lake ecosystem variability inferred from variance (standard deviation, left) and RoC (rate of change, right) metrics of time series of total autotrophic production (Chl *a* + echinenone), bloom-forming algal abundance (sum of canthaxanthin and luteinzeaxanthin), cyanotoxins (total MCs), and the scores of NMDS1 axis. The gray ribbons are 95% pointwise confidence intervals on the fitted models. Where the simulation value does not include zero, the models detect increase or decrease in the ecological variability. (B) Under multiple interactive stressors, the ecosystem changed through time to different configurations with shifting baseline beyond pre-disturbance reference (Battarbee et al., 2005; Ryo et al., 2019), and ecological vulnerability to climate change inherited the human legacy (e.g., anthropogenic eutrophication and alterations of food web) with lower resilience.

particularly coherent in recent three decades, manifesting as a significant increase followed by a decline. Notably, temporal variability of these key ecosystem components increased to varying degrees following the apparent ecosystem state change.

#### 4. Discussion

This work shows that integrated application of multi-biomarkers and geochemistry proxies on lake sediment sequences provides a powerful means to retrospectively assess long-term dynamics in HABs, cyanotoxin production, and ecosystem variability in response to multiple stressors and their interactions. Our results revealed critical processes of eutrophication and primary production culminating in an abrupt ecological shift, characterized by increasingly-toxic cyanobacterial blooms (*Microcystis*) from the 1980s in Lake Taihu. Thereafter increasing ecological variability suggested that Lake Taihu was probably subjected to elevated variation in the forcing mechanism (Dakos et al., 2015),

and/or exhibited a paradox of enrichment (Cottingham et al., 2000; Bunting et al., 2016) with nutrient cyclic loops (Scheffer et al., 2001; Xu et al., 2021), which failed to establish an alternative stable state (Carpenter et al., 2011). Our findings have demonstrated increased ecological vulnerability and reduced predictability of cyanobacterial HABs following state change and climate warming, which has profound environmental implications.

##### 4.1. Multiple stressors underlying HABs and cyanotoxin production

Freshwater ecosystems are widely experiencing the “allied attack” of climate change and eutrophication (Moss et al., 2011), particularly for degrading shallow lakes (Zhou et al., 2022), given their potential driving synergy (Paerl and Scott, 2010; Meerhoff et al., 2022). As shown in previous investigations (Kosten et al., 2012), experiments (Richardson et al., 2019), modeling (Taranu et al., 2017; Kakouei et al., 2021), and syntheses (Havens and Paerl, 2015; Taranu et al., 2015; Jeppesen et al.,



2020), eutrophic conditions and warmer climate contribute to higher phytoplankton biomass and cyanobacterial dominance, thus increase the risk of toxic HABs and water quality degradation (Huisman et al., 2018). This phenomenon is extremely true for large, shallow Lake Taihu, similar to lakes Okeechobee (USA), Winnipeg (Canada), Võrtsjärv (Estonia), Kasumigaura (Japan), Chaohu (China), and shallow parts of other large lakes worldwide (Qin et al., 2019; Zhou et al., 2022). Accelerated climate warming and nutrients influx (N, P) from the watershed due to rapid agricultural expansion and urbanization (Fig. S3) are widely recognized as the driving factors (Xu et al., 2021; Zeng et al., 2023). Our paleolimnological reconstruction and ordination results (Figs. 2 and 3A) offered empirical complement of lake eutrophication from long-term perspective, associating largely with agricultural practices and urban sewage.

Moreover, interactions between climate change (warming, wind stilling) and anthropogenic nutrients in VPA explained a large part of the historical dynamics in the Taihu ecosystem (Fig. 3A). This is consistent with theoretical and empirical expectations that environmental effects work on lakes via coupled influxes of energy (temperature, irradiance, wind) and mass (precipitation, nutrients, particles) (Leavitt et al., 2009). According to the GAM results (Fig. 3C), the majority of the forcing interactions through time were cumulative with simultaneous positive effects since intensified fertilization and cyanobacterial blooms in the 1980s. Thus, our empirical results highlight the synergistic effects of eutrophication and climate change on cyanobacterial temporal dynamics.

As previously and newly-loaded nutrients are effectively retained and recycled in Lake Taihu (Zhu et al., 2020; Xu et al., 2021), warming effects have produced a suite of favorable environmental conditions to sustain the HAB potential. First, phytoplankton universally optimize growth as temperatures increase to a certain extent, notably for cyanobacteria and other bloom-forming algae that prefer warming conditions (Carey et al., 2012; Havens and Paerl, 2015). Second, enhanced water column stability and thermal stratification following joint regional warming and atmospheric stilling (Fig. S3) modify phytoplankton communities by favoring surface bloom-forming cyanobacteria through buoyancy regulation (Kosten et al., 2012). The consequent accumulation of detrital organic matter from algae in summer and autumn usually enhances deoxygenation of subsurface water layers, stimulates the release of internal phosphorus (Yin et al., 2022), and inhibits nitrification-denitrification coupling in Lake Taihu (Zhu et al., 2020). This is also indicated by the shifts in the sediment nutrient stoichiometry, such as elevated N/P ratios and fluctuated C/N ratios following blooms (Fig. 2). Bloom-induced underwater light attenuation (Fig. 1B) further reinforce buoyant cyanobacteria dominance (Carey et al., 2012; Guo et al., 2019). Third, rising temperature can not only increase the mineralization rate of watershed soils, but also be accompanied by more extreme rainstorms, both of which could enhance diffuse and legacy nutrient delivery to waters. Tropical cyclones and extreme weather have been reported to play an important role in stimulating cyanobacterial bloom formation in Lake Taihu (Zhu et al., 2014; Yang et al., 2016). Furthermore, human and climate-driven hydrological alterations (e.g., ~20% rise of Taihu lake-level since 1992) (Zhang et al., 2018b), combined with projected warming and drought (Woolway et al., 2020), are expected to extend vertical stratification and water residence time, which will likely promote HABs and complicate eutrophication management (Havens and Paerl, 2015; Yang et al., 2017; Meerhoff et al., 2022).

In turn, the interactive effects also strongly stimulate the production of hazardous cyanotoxins, particularly microcystins, as found in microcosm experiments (Lüring et al., 2018). Our biomarker reconstruction revealed that significant increases in MCs lagged behind the initial blooms by over a decade in Lake Taihu (Fig. 2), until the shift to and formation of toxic *Microcystis* blooms (Hu et al., 2016; Guo et al., 2019) as climate change and eutrophication intensified in the 1990s. In particular, this process promoted the strains producing more toxic

MC-LR and MC-YR and exacerbated the ecological risk, as similarly observed in other degraded freshwaters (Zastepa et al., 2017; Henao et al., 2019). Increasing temperature and nutrient concentrations are corroborated as major predictors of large increases in regional MC maxima (e.g., North America) (Taranu et al., 2017; Hayes et al., 2020) and even global patterns of MC congeners (Taranu et al., 2019). Despite faster photodegradation in some environments (e.g., the tropics), surface freshwaters like Lake Taihu in the warming future might experience a predicted higher occurrence of highly-toxic MCs, with increasing risks to environmental and human health.

Multiple stressors can further provoke large changes in food webs with potential cascading effects, in particular weakening top-down control in regulating phytoplankton biomass and HABs (Meerhoff et al., 2022). In Lake Taihu, long-term loss of macrophytes (Fig. 1A) probably have caused less refuge available for phytoplankton competitors, such as grazer macroinvertebrates and zooplankton (Moss et al., 2011; Janssen et al., 2014). Over-fishing and extensive stocking have altered lake fish community diversity and structure by favoring large amounts of small zooplanktivorous species (notably anchovy) (Mao et al., 2020). These shifts in aquatic communities substantially reduced algal grazing by zooplankton in the lake. Furthermore, fewer piscivorous fish, more omnivorous and herbivorous species, and decreased body size of zooplankton are expected in warmer waters (Jeppesen et al., 2020). Such changes are likely to reinforce the positive interaction between climate change and eutrophication, given the projected algal proliferation, toxin-producing cyanobacterial dominance, and less intensive top-down effects.

#### 4.2. Ecosystem state shift and baseline implications

Studies on shallow lakes suggest that increased variations in phytoplankton parameters and regulatory mechanisms are reliable indicators of ecosystem state change from clear-water to turbid-water dominated by prolific, nuisance cyanobacteria (Scheffer et al., 2001; Janssen et al., 2014; Yang et al., 2017). This catastrophic shift may arise from abrupt but persistent changes in external forcings (e.g., climate change, nutrient enrichment) and/or gradual variation in intrinsic factors (e.g., trophic cascades) (Bunting et al., 2016; Carpenter et al., 2022). Ecological theory predicts that during the transitions among alternate stable states, lake ecosystems may exhibit transient (rising then decreasing) variability in focal parameters (Carpenter et al., 2011; Dakos et al., 2015) following the re-establishment of internal feedback mechanisms (e.g., internal nutrient cycling, loss of macrophytes, shading) (Scheffer and van Nes 2007; Janssen et al., 2014). However, both temporal variance and RoC analyses in Lake Taihu explicitly demonstrated increasing phytoplankton variability after the ecological shift (Fig. 4A and Fig. S8), suggesting that a stable state has not yet been established. In particular, the persistent rise in variability of total algae, bloom-forming taxa, and cyanotoxins is more consistent with the paradox of enrichment wherein ecological resilience and predictability declined (Cottingham et al., 2000; Bunting et al., 2016; Carpenter et al., 2022).

The rising variability of primary producers probably reflects persistent increases in external forcings (Dakos et al., 2015; Huang et al., 2022) or the constraints of similar large lakes to establish alternative stable states (Scheffer and van Nes, 2007; Janssen et al., 2014; Bruel et al., 2021). Although a series of measures costing ~US\$30 billion have been implemented to address wastewater discharge and improve water quality in Lake Taihu watershed (Qin et al., 2019), recent warming climate and extreme weather continue to exacerbate non-point nutrient loading (Xu et al., 2021) and internal release from sediments (Yin et al., 2022). Meanwhile, growing evidence indicates that cyanobacterial dominance is exerting a positive feedback with climate warming (Huisman et al., 2018; Meerhoff et al., 2022), as blooms usually promote the release of nutrients (Xu et al., 2021; Yin et al., 2022) and greenhouse gasses (Yan et al., 2017) that increase the warming potential. Under

these circumstances, cyanobacterial dominance may thus be a self-perpetuating phenomenon (Havens and Paerl, 2015; Meerhoff et al., 2022), and toxic HABs are predicted to increase in Lake Taihu with climate change. Outside of the littoral habitats and east bays of Lake Taihu, it is arguable that the pelagic areas have always lacked enough macrophytes as a result of strong winds (Zhang et al., 2018b), therefore alternative stable states are unlikely to occur there.

It is particularly noteworthy that the synergistic effects of eutrophication and climate change can reduce ecosystem resilience and cause unexpected collapse (e.g., cyanobacterial HABs), even when individual stressors remain at levels that are considered to be safe (Ryo et al., 2019; Meerhoff et al., 2022). Since long-term eutrophication altered the nutrient cycling and reorganized the configurations of primary producers and food webs in Lake Taihu and beyond, as discussed above, the ecosystems are likely more responsive to warming than before state shift (Fig. 4B). Ecological resistance and vulnerability to climate change thus inherited the legacy of local human pressure similar to previous observations (Battarbee et al., 2005; Bruel et al., 2021). As a consequence, if local pressures (e.g., nutrients) must be reduced to build resilience and mitigate blooms, reduction strategies may be overwhelmed by climate change impacts on the ecosystems, unless more costly and aggressive approaches are employed. Furthermore, the current and future managers should acknowledge that the ecosystem changed permanently to different configurations with shifting baselines beyond the pre-disturbance reference (Fig. 4B) (Battarbee et al., 2005; Ryo et al., 2019). It is important to realize the contemporary process (e.g., re-oligotrophication), the hysteresis of ecological response (Scheffer et al., 2001), and the integral eutrophication history when setting appropriate eco-environmental baselines. Our study underlines the complexity and interactivity of multiple stressors on lake ecosystems and HABs, and the necessity of long-term perspectives to contextualize modern ecological conditions and inform integrated management.

#### 4.3. Integrated biomarker reconstruction: merits and prospects

In contrast to *in-situ* monitoring and remote sensing observations of lake surface, our novel analysis produces a longer-term, continuous record of lake primary productivity and HABs, which integrates phytoplankton from both surface and deeper waters (McGowan, 2013; Waters et al., 2021). This supplement allows to assess the complete trajectory, magnitude, and decadal variability of ecosystem deterioration, particularly covering a time span where abrupt ecological shifts and toxic HABs occur. The paleo-reconstruction can further shed light on the dynamics of algal community composition, toxin-producing taxa, and toxicity risk of cyanobacterial HABs through time, which may facilitate the development of effective control and restoration strategies.

However, specific taxa and biodiversity changes of harmful algae particularly cyanobacteria are still difficult to be determined using biogeochemical tools. The next-generation high-throughput sequencing of environmental DNA is increasingly recognized as an effective complementary means (Domaizon et al., 2017; Monchamp et al., 2018; Zhang et al., 2023). A combined application of molecular biomarkers and DNA techniques to well-dated sediment sequences is thus very promising in deepening our understanding of long-term lake ecosystem dynamics, biodiversity, functioning, and their linkages to environmental forcings. Meanwhile, the spatiotemporal heterogeneity of limnological characteristics, sedimentation, and anthropogenic disturbances in most large lakes like Taihu requires future research with multiple, well-placed cores (Lin et al., 2020, 2021) to improve the assessment of HABs and eco-environmental risk on the whole-lake scale.

## 5. Conclusions

We applied molecular biomarkers of aliphatic hydrocarbons, photosynthetic pigments, and cyanotoxins on lake sediment sequences to successfully reconstruct the long-term trajectories and variability of

cyanobacterial HABs, which provided empirical evidence on the abrupt regime shifts and causes underlying aquatic ecosystem degradation. The biomarker reconstruction in China's Lake Taihu revealed that significant increases in cyanotoxins lagged behind the initial blooms by over a decade, until the formation of toxic *Microcystis* blooms in the 1990s. This process was accompanied with the shift to and the occurrence of more toxic microcystins (MC-LR and MC-YR), leading to exacerbated harmful effects. Our ordination and numerical modeling results emphasize the synergy of climate warming and eutrophication through nutrient recycling and their feedback through buoyant cyanobacterial proliferation, which sustain bloom-forming potential and promote the occurrence of increasingly-toxic cyanotoxins. Cyanobacterial dominance particularly in shallow lakes may become a self-perpetuating phenomenon, and toxic HAB outbreaks are likely to occur continuously with climate change.

The increasing variance and rate of change metrics of phytoplankton (particularly bloom-forming taxa) in Lake Taihu probably indicated increased ecosystem vulnerability and resilience loss with potential risk to shift to another undesirable state. We deduce that an alternative stable state has not yet been established there. Current ecological resistance and vulnerability to climate warming inherited the legacy of local human pressure (e.g., lake eutrophication and alterations of food web), with shifting baseline beyond the pre-disturbance reference. To build resilience and reverse blooms, therefore, more aggressive and integrated strategies dealing with legacy nutrients and global warming are imperative. Taken together, our work provides novel approach and long-term evolutionary perspective for studying the underlying mechanisms triggering HABs with critical insights on water resource management.

#### CRedit authorship contribution statement

**Qi Lin:** Conceptualization, Investigation, Methodology, Formal analysis, Writing – original draft, Writing – review & editing, Funding acquisition. **Ke Zhang:** Conceptualization, Data curation, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Suzanne McGowan:** Methodology, Writing – review & editing. **Shixin Huang:** Formal analysis, Writing – review & editing. **Qingju Xue:** Investigation, Methodology, Writing – review & editing. **Eric Capo:** Formal analysis, Writing – review & editing. **Can Zhang:** Methodology, Writing – review & editing. **Cheng Zhao:** Methodology, Writing – review & editing. **Ji Shen:** Conceptualization, Supervision, Project administration.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.watres.2023.119916](https://doi.org/10.1016/j.watres.2023.119916).

## References

- Andersen, T., Carstensen, J., Hernandez-Garcia, E., Duarte, C.M., 2009. Ecological thresholds and regime shifts: approaches to identification. *Trends Ecol. Evol.* 24, 49–57. <https://doi.org/10.1016/j.tree.2008.07.014>.
- Battarbee, R.W., Anderson, N.J., Jeppesen, E., Leavitt, P.R., 2005. Combining palaeolimnological and limnological approaches in assessing lake ecosystem response to nutrient reduction. *Freshw. Biol.* 50, 1772–1780. <https://doi.org/10.1111/j.1365-2427.2005.01427.x>.
- Björndahl, J.A., Gushulak, C.A.C., Mezzini, S., Simpson, G.L., Haig, H.A., Leavitt, P.R., Finlay, K., 2022. Abrupt changes in the physical and biological structure of endorheic upland lakes due to 8-m lake-level variation during the 20th century. *Limnol. Oceanogr.* 67, 1022–1039. <https://doi.org/10.1002/lno.12054>.
- Bruel, R., Sabatier, P., 2020. serac: a R package for ShortlivEd RADionuclide chronology of recent sediment cores. *J. Environ. Radioact.* 225, 106449. <https://doi.org/10.1016/j.jenvrad.2020.106449>.
- Bruel, R., Girardclos, S., Marchetto, A., Kremer, K., Crouzet, C., Reyss, J.-L., Sabatier, P., Perga, M.-E., 2021. Reframing Lake Geneva ecological trajectory in a context of multiple but asynchronous pressures. *J. Paleolimnol.* 65, 353–368. <https://doi.org/10.1007/s10933-021-00176-y>.
- Bunting, L., Leavitt, P.R., Simpson, G.L., Wissel, B., Laird, K.R., Cumming, B.F., St Amand, A., Engstrom, D.R., 2016. Increased variability and sudden ecosystem state change in Lake Winnipeg, Canada, caused by 20th century agriculture. *Limnol. Oceanogr.* 61, 2090–2107. <https://doi.org/10.1002/lno.10355>.
- Cao, X., Xu, X., Bian, R., Wang, Y., Yu, H., Xu, Y., Duan, G., Bi, L., Chen, P., Gao, S., Wang, J., Peng, J., Qu, J., 2020. Sedimentary ancient DNA metabarcoding delineates the contrastingly temporal change of lake cyanobacterial communities. *Water Res.* 183, 116077. <https://doi.org/10.1016/j.watres.2020.116077>.
- Carey, C.C., Ibelings, B.W., Hoffmann, E.P., Hamilton, D.P., Brookes, J.D., 2012. Ecophysiological adaptations that favour freshwater cyanobacteria in a changing climate. *Water Res.* 46, 1394–1407. <https://doi.org/10.1016/j.watres.2011.12.016>.
- Carpenter, S.R., Arani, B.M.S., Van Nes, E.H., Scheffer, M., Pace, M.L., 2022. Resilience of phytoplankton dynamics to trophic cascades and nutrient enrichment. *Limnol. Oceanogr.* 67, S258–S265. <https://doi.org/10.1002/lno.11913>.
- Carpenter, S.R., Cole, J.J., Pace, M.L., Batt, R., Brock, W.A., Cline, T., Coloso, J., Hodgson, J.R., Kitchell, J.F., Seekell, D.A., Smith, L., Weidel, B., 2011. Early warnings of regime shifts: a whole-ecosystem experiment. *Science* 332, 1079–1082. <https://doi.org/10.1126/science.1203672>.
- Cheng, C., Steinman, A.D., Zhang, K., Lin, Q., Xue, Q., Wang, X., Xie, L., 2023. Risk assessment and identification of factors influencing the historical concentrations of microcystin in Lake Taihu. *China. J. Environ. Sci.* 127, 1–14. <https://doi.org/10.1016/j.jes.2022.03.043>.
- Cottingham, K.L., Rusak, J.A., Leavitt, P.R., 2000. Increased ecosystem variability and reduced predictability following fertilization: evidence from palaeolimnology. *Ecol. Lett.* 3, 340–348. <https://doi.org/10.1046/j.1461-0248.2000.00158.x>.
- Dakos, V., Carpenter, S.R., van Nes, E.H., Scheffer, M., 2015. Resilience indicators: prospects and limitations for early warnings of regime shifts. *Philos. Trans. R. Soc. B Biol. Sci.* 370, 20130263. <https://doi.org/10.1098/rstb.2013.0263>.
- Deng, J., Qin, B., Paerl, H.W., Zhang, Y., Wu, P., Ma, J., Chen, Y., 2014. Effects of nutrients, temperature and their interactions on spring phytoplankton community succession in Lake Taihu. *China. PLoS One* 9, e113960. <https://doi.org/10.1371/journal.pone.0113960>.
- Domaizon, I., Winegardner, A., Capo, E., Gauthier, J., GregoryEaves, I., 2017. DNA-based methods in paleolimnology: new opportunities for investigating long-term dynamics of lacustrine biodiversity. *J. Paleolimnol.* 58, 1–21. <https://doi.org/10.1007/s10933-017-9958-y>.
- Ellis, E.C., Wang, S., 1997. Sustainable traditional agriculture in the Tai Lake region of China. *Agric. Ecosyst. Environ.* 61, 177–193. [https://doi.org/10.1016/S0167-8809\(96\)01099-7](https://doi.org/10.1016/S0167-8809(96)01099-7).
- Ficken, K.J., Li, B., Swain, D.L., Eglinton, G., 2000. An *n*-alkane proxy for the sedimentary input of submerged/floating freshwater aquatic macrophytes. *Org. Geochem.* 31, 745–749. [https://doi.org/10.1016/S0146-6380\(00\)00081-4](https://doi.org/10.1016/S0146-6380(00)00081-4).
- Guo, C., Zhu, G., Qin, B., Zhang, Y., Zhu, M., Xu, H., Chen, Y., Paerl, H.W., 2019. Climate exerts a greater modulating effect on the phytoplankton community after 2007 in eutrophic Lake Taihu, China: evidence from 25 years of recordings. *Ecol. Indic.* 105, 82–91. <https://doi.org/10.1016/j.ecolind.2019.05.034>.
- Havens, K.E., Paerl, H.W., 2015. Climate change at a crossroad for control of harmful algal blooms. *Environ. Sci. Technol.* 49, 12605–12606. <https://doi.org/10.1021/acs.est.5b03990>.
- Hayes, N.M., Haig, H.A., Simpson, G.L., Leavitt, P.R., 2020. Effects of lake warming on the seasonal risk of toxic cyanobacteria exposure. *Limnol. Oceanogr.* Lett. 5, 393–402. <https://doi.org/10.1002/lo2.10164>.
- Henaou, E., Rzymiski, P., Waters, M.N., 2019. A review of the study of cyanotoxins in paleolimnological research: current knowledge and future needs. *Toxins* 12, 6. <https://doi.org/10.3390/toxins12010006>.
- Hesse, K., Dittmann, E., Borner, T., 2001. Consequences of impaired microcystin production for light-dependent growth and pigmentation of *Microcystis aeruginosa* PCC 7806. *FEMS Microbiol. Ecol.* 37, 39–43. [https://doi.org/10.1016/S0168-6496\(01\)00142-8](https://doi.org/10.1016/S0168-6496(01)00142-8).
- Ho, J.C., Michalak, A.M., Pahlevan, N., 2019. Widespread global increase in intense lake phytoplankton blooms since the 1980s. *Nature* 574, 667–670. <https://doi.org/10.1038/s41586-019-1648-7>.
- Hu, L., Shan, K., Lin, L., Shen, W., Huang, L., Gan, N., Song, L., 2016. Multi-year assessment of toxic genotypes and microcystin concentration in northern Lake Taihu. *China. Toxins* 8, 23. <https://doi.org/10.3390/toxins8010023>.
- Huang, S., Zhang, K., Lin, Q., Kattel, G., Shen, J., 2022. Lake ecosystem regime shifts induced by agricultural intensification: a century scale paleolimnological investigation from the Huai River Basin (China). *Quat. Sci. Rev.* 285, 107522. <https://doi.org/10.1016/j.quascirev.2022.107522>.
- Huisman, J., Codd, G.A., Paerl, H.W., Ibelings, B.W., Verspagen, J., Visser, P.M., 2018. Cyanobacterial blooms. *Nat. Rev. Microbiol.* 16, 471–483. <https://doi.org/10.1038/s41579-018-0040-1>.
- Huo, S., Zhang, H., Monchamp, M.-E., Wang, R., Weng, N., Zhang, J.B., Zhang, H., Wu, F., 2022. Century-long homogenization of algal communities is accelerated by nutrient enrichment and climate warming in lakes and reservoirs of the North Temperate Zone. *Environ. Sci. Technol.* 56, 3780–3790. <https://doi.org/10.1021/acs.est.1c06958>.
- Ibrahim, A., Capo, E., Wessels, M., Martin, I., Meyer, A., Schleheck, D., Epp, L.S., 2020. Anthropogenic impact on the historical phytoplankton community of Lake Constance reconstructed by multimer analysis of sediment-core environmental DNA. *Mol. Ecol.* 30, 3040–3056. <https://doi.org/10.1111/mec.15696>.
- Janssen, A.B.G., Teurlincx, S., An, S., Janse, J.H., Paerl, H.W., Mooij, W.M., 2014. Alternative stable states in large shallow lakes? *J. Great Lakes Res.* 40, 813–826. <https://doi.org/10.1016/j.jglr.2014.09.019>.
- Jeppesen, E., Canfield, D.E., Bachmann, R.W., Søndergaard, M., Havens, K.E., Johansson, L.S., Lauridsen, T.L., Sh, T., Rutter, R.P., Warren, G., 2020. Toward predicting climate change effects on lakes: a comparison of 1656 shallow lakes from Florida and Denmark reveals substantial differences in nutrient dynamics, metabolism, trophic structure, and top-down control. *Inland Waters* 10, 197–211. <https://doi.org/10.1080/20442041.2020.1711681>.
- Kakouei, K., Kraemer, B.M., Anneville, O., Carvalho, L., Feuchtmayr, H., Graham, J.L., Higgins, S., Pomati, F., Rudstam, L.G., Stockwell, J.D., Thackeray, S.J., 2021. Phytoplankton and cyanobacteria abundances in mid-21st century lakes depend strongly on future land use and climate projections. *Glob. Chang. Biol.* 27, 6409–6422. <https://doi.org/10.1111/gcb.15866>.
- Kosten, S., Huszar, V.L.M., Becares, E., Coats, L.S., van Donk, E., Hansson, L.A., Jeppesen, E., Kruk, C., Lacerot, G., Mazzeo, N., De Meester, L., Moss, B., Lürling, M., Nöges, T., Romo, S., Scheffer, M., 2012. Warmer climates boost cyanobacterial dominance in shallow lakes. *Glob. Chang. Biol.* 18, 118–126. <https://doi.org/10.1111/j.1365-2486.2011.02488.x>.
- Leavitt, P.R., 1993. A review of factors that regulate carotenoid and chlorophyll deposition and fossil pigment abundance. *J. Paleolimnol.* 9, 109–127. <https://doi.org/10.1007/BF00677513>.
- Leavitt, P.R., Vinebrooke, R.D., Donald, D.B., Smol, J.P., Schindler, D.W., 1997. Past ultraviolet radiation environments in lakes derived from fossil pigments. *Nature* 388, 457–459. <https://doi.org/10.1038/41296>.
- Leavitt, P.R., Fritz, S.C., Anderson, N.J., Baker, P.A., Blenckner, T., Bunting, L., Catalan, J., Conley, D.J., Hobbs, W.O., Jeppesen, E., Korhola, A., McGowan, S., Ruhland, K., Rusak, J.A., Simpson, G.L., Solovieva, N., Werne, J., 2009. Paleolimnological evidence of the effects on lakes of energy and mass transfer from climate and humans. *Limnol. Oceanogr.* 54, 2330–2348. [https://doi.org/10.4319/lo.2009.54.6.part\\_2.2330](https://doi.org/10.4319/lo.2009.54.6.part_2.2330).
- Lin, Q., Zhang, K., Liu, E., Sabatier, P., Arnaud, F., Shen, J., 2020. Deciphering centennial anthropogenic pollution processes in large lakes dominated by socio-economic impacts. *Anthropocene* 32, 100269. <https://doi.org/10.1016/j.anucene.2020.100269>.
- Lin, Q., Zhang, K., McGowan, S., Capo, E., Shen, J., 2021. Synergistic impacts of nutrient enrichment and climate change on long-term water quality and ecological dynamics in contrasting shallow-lake zones. *Limnol. Oceanogr.* 66, 3271–3286. <https://doi.org/10.1002/lno.11878>.
- Lürling, M., van Oosterhout, F., de Senerpont Domis, L., Marinho, M.M., 2018. Response of natural cyanobacteria and algae assemblages to a nutrient pulse and elevated temperature. *Front. Microbiol.* 9, 1851. <https://doi.org/10.3389/fmicb.2018.01851>.
- Mao, Z., Gu, X., Cao, Y., Zhang, M., Zeng, Q., Chen, H., Shen, R., Jeppesen, E., 2020. The role of top-down and bottom-up control for phytoplankton in a subtropical shallow eutrophic lake: evidence based on long-term monitoring and modeling. *Ecosystems* 23, 1449–1463. <https://doi.org/10.1007/s10021-020-00480-0>.
- McGowan, S., 2013. Pigment studies. In: Elias, S. (Ed.), *Encyclopedia of Quaternary Sciences*. Elsevier, pp. 1–26. <https://doi.org/10.1016/b0-444-52747-8/00247-7>.
- McGowan, S., Barker, P., Haworth, E.Y., Leavitt, P.R., Maberly, S.C., Pates, J., 2012. Humans and climate as drivers of algal community change in Windermere since 1850. *Freshw. Biol.* 57, 260–277. <https://doi.org/10.1111/j.1365-2427.2011.02689.x>.
- Meerhoff, M., Audet, J., Davidson, T.A., De Meester, L., Hilt, S., Kosten, S., Liu, Z., Mazzeo, N., Paerl, H.W., Scheffer, M., Jeppesen, E., 2022. Feedback between climate change and eutrophication: revisiting the allied attack concept and how to strike back. *Inland Waters* 12, 187–204. <https://doi.org/10.1080/20442041.2022.2029317>.
- Meyers, P.A., 2003. Applications of organic geochemistry to paleolimnological reconstructions: a summary of examples from the Laurentian Great Lakes. *Org. Geochem.* 34, 261–289. [https://doi.org/10.1016/S0146-6380\(02\)00168-7](https://doi.org/10.1016/S0146-6380(02)00168-7).
- Monchamp, M.E., Spaak, P., Domaizon, I., Dubois, N., Bouffard, D., Pomati, F., 2018. Homogenization of lake cyanobacterial communities over a century of climate change and eutrophication. *Nat. Ecol. Evol.* 2, 317–324. <https://doi.org/10.1038/s41559-017-0407-0>.



- Moss, B., Kosten, S., Meerhoff, M., Battarbee, R.W., Jeppesen, E., Mazzeo, N., Havens, K., Lacerot, G., Liu, Z., De Meester, L., Paerl, H.W., Scheffer, M., 2011. Allied attack: climate change and eutrophication. *Inland Waters* 1, 101–105. <https://doi.org/10.5268/IW-1.2.359>.
- Paerl, H.W., Scott, J.T., 2010. Throwing fuel on the fire: synergistic effects of excessive nitrogen inputs and global warming on harmful algal blooms. *Environ. Sci. Technol.* 44, 7756–7758. <https://doi.org/10.1021/es102665e>.
- Perga, M.-E., Frossard, V., Jenny, J.-P., Alric, B., Arnaud, F., Berthon, V., Black, J.L., Domaizon, I., Giguet-Covex, C., Kirkham, A., Magny, M., Manca, M., Marchetto, A., Millet, L., Pailles, C., Pignol, C., Poulencard, J., Reyss, J.-L., Rimet, F., Sabatier, P., Savichtcheva, O., Sylvestre, F., Verneaux, V., 2015. High-resolution paleolimnology opens new management perspectives for lakes adaptation to climate warming. *Front. Ecol. Environ.* 3, 1–17. <https://doi.org/10.3389/fevo.2015.00072>.
- Qin, B., Paerl, H.W., Brookes, J.D., Liu, J.Q., Jeppesen, E., Zhu, G., Zhang, Y., Xu, H., Shi, K., Deng, J., 2019. Why Lake Taihu continues to be plagued with cyanobacterial blooms through 10 years (2007–2017) efforts. *Sci. Bull.* 64, 354–356. <https://doi.org/10.1016/j.scib.2019.02.008>.
- Richardson, J., Feuchtmayr, L., Miller, C., Hunter, P.D., Maberly, S.C., Carvalho, L., 2019. Response of cyanobacteria and phytoplankton abundance to warming, extreme rainfall events and nutrient enrichment. *Glob. Chang. Biol.* 25, 3365–3380. <https://doi.org/10.1111/gcb.14701>.
- Ryo, M., Aguilar-Trigueros, C.A., Pinek, L., Muller, L.A.H., Rillig, M.C., 2019. Basic principles of temporal dynamics. *Trends Ecol. Evol.* 34, 723–733. <https://doi.org/10.1016/j.tree.2019.03.007>.
- Scheffer, M., Carpenter, S., Foley, J.A., Folke, C., Walker, B., 2001. Catastrophic shifts in ecosystems. *Nature* 413, 591–596. <https://doi.org/10.1038/35098000>.
- Scheffer, M., van Nes, E.H., 2007. Shallow lakes theory revisited: various alternative regimes driven by climate, nutrients, depth and lake size. *Hydrobiologia* 585, 455–466. <https://doi.org/10.1007/s10750-007-0616-7>.
- Shi, K., Zhang, Y., Zhang, Y., Li, N., Qin, B., Zhu, G., Zhou, Y., 2019. Phenology of phytoplankton blooms in a trophic lake observed from long-term MODIS data. *Environ. Sci. Technol.* 53, 2324–2331. <https://doi.org/10.1021/acs.est.8b06887>.
- Smol, J.P., 2010. The power of the past: using sediments to track the effects of multiple stressors on lake ecosystems. *Freshw. Biol.* 55, 43–59. <https://doi.org/10.1111/j.1365-2427.2009.02373.x>.
- Simpson, G.L., 2018. Modelling palaeoecological time series using generalised additive models. *Front. Ecol. Evol.* 6, 149. <https://doi.org/10.3389/fevo.2018.00149>.
- Taranu, Z.E., Gregory-Eaves, I., Leavitt, P.R., Bunting, L., Buchaca, T., Catalan, J., Domaizon, I., Guilizzoni, P., Lami, A., McGowan, S., Moorhouse, H., Morabito, G., Pick, F.R., Stevenson, M.A., Thompson, P.L., Vinebrooke, R.D., 2015. Acceleration of cyanobacterial dominance in north temperate-subarctic lakes during the Anthropocene. *Ecol. Lett.* 18, 375–384. <https://doi.org/10.1111/ele.12420>.
- Taranu, Z.E., Gregory-Eaves, I., Steele, R., Beaulieu, M., Legendre, P., 2017. Predicting microcystin occurrences in US lakes and reservoirs: a new framework for modeling the drivers of an important health risk factor. *Glob. Ecol. Biogeog.* 26, 625–637. <https://doi.org/10.1111/geb.12569>.
- Taranu, Z.E., Pick, F.R., Creed, I.F., Zastepa, A., Watson, S.B., 2019. Meteorological and nutrient conditions influence microcystin congeners in freshwaters. *Toxins (Basel)* 11, 620. <https://doi.org/10.3390/toxins11110620>.
- ter Braak, C.J.F., Šmilauer, P., 2012. *CANOCO Reference Manual and User's Guide: Software for Ordination (version 5.0)*. Microcomputer Power.
- Waters, M.N., Brenner, M., Curtis, J.H., Romero-Oliva, C.S., Dix, M., Cano, M., 2021. Harmful algal blooms and cyanotoxins in Lake Amatitlán, Guatemala, coincided with ancient Maya occupation in the watershed. *Proc. Natl. Acad. Sci. U. S. A.* 118, e2109919118. <https://doi.org/10.1073/pnas.2109919118>.
- Wood, S.N., 2017. *Generalized Additive models: An introduction With R (2)*. Chapman and Hall/CRC Press.
- Woolway, R.I., Kraemer, B.M., Lenters, J.D., Merchant, C.J., O'Reilly, C.M., Sharma, S., 2020. Global lake responses to climate change. *Nat. Rev. Earth & Environ.* 1, 388–403. <https://doi.org/10.1038/s43017-020-0067-5>.
- Xu, H., McCarthy, M.J., Paerl, H.W., Brookes, J.D., Zhu, G., Hall, N.S., Qin, B., Zhang, Y., Zhu, M., Hampel, J.J., Newell, S.E., Gardner, W.S., 2021. Contributions of external nutrient loading and internal cycling to cyanobacterial bloom dynamics in Lake Taihu, China: implications for nutrient management. *Limnol. Oceanogr.* 66, 1492–1509. <https://doi.org/10.1002/lno.11700>.
- Xue, Q., Xie, L., Cheng, C., Su, X., Zhao, Y., 2023. Different environmental factors drive the concentrations of microcystin in particulates, dissolved water, and sediments peaked at different times in a large shallow lake. *J. Environ. Manag.* 326, 116833. <https://doi.org/10.1016/j.jenvman.2022.116833>.
- Yan, X., Xu, X., Wang, M., Wang, G., Wu, S., Li, Z., Sun, H., Shi, A., Yang, Y., 2017. Climate warming and cyanobacteria blooms: looks at their relationships from a new perspective. *Water Res.* 125, 449–457. <https://doi.org/10.1016/j.watres.2017.09.008>.
- Yang, J., Lv, H., Isabwe, A., Liu, L., Yu, X., Chen, H., Yang, J., 2017. Disturbance-induced phytoplankton regime shifts and recovery of cyanobacteria dominance in two subtropical reservoirs. *Water Res.* 120, 52–63. <https://doi.org/10.1016/j.watres.2017.04.062>.
- Yang, Z., Zhang, M., Shi, X., Kong, F., Ma, R., Yu, Y., 2016. Nutrient reduction magnifies the impact of extreme weather on cyanobacterial bloom formation in large shallow Lake Taihu (China). *Water Res.* 103, 302–310. <https://doi.org/10.1016/j.watres.2016.07.047>.
- Yin, H., Zhang, M., Yin, P., Li, J., 2022. Characterization of internal phosphorus loading in the sediment of a large eutrophic lake (Lake Taihu, China). *Water Res.* 225, 119125. <https://doi.org/10.1016/j.watres.2022.119125>.
- Zastepa, A., Taranu, Z.E., Kimpfe, L.E., Blais, J.M., Gregory-Eaves, I., Zurawell, R.W., Pick, F.R., 2017. Reconstructing a long-term record of microcystins from the analysis of lake sediments. *Sci. Total Environ.* 579, 893–901. <https://doi.org/10.1016/j.scitotenv.2016.10.211>.
- Zeng, L., Swann, G.E.A., Leng, M.J., Chen, X., Ji, J., Huang, X., McGowan, S., 2023. Ecosystem deterioration in the middle Yangtze floodplain lakes over the last two centuries: evidence from sedimentary pigments. *Quat. Sci. Rev.* 302, 107954. <https://doi.org/10.1016/j.quascirev.2023.107954>.
- Zhang, C., Zhao, C., Zhou, A., Zhang, K., Wang, R., Shen, J., 2019. Late Holocene lacustrine environmental and ecological changes caused by anthropogenic activities in the Chinese Loess Plateau. *Quat. Sci. Rev.* 203, 266–277. <https://doi.org/10.1016/j.quascirev.2018.11.020>.
- Zhang, J., Shi, K., Paerl, H.W., Rühland, K.M., Yuan, Y., Wang, R., Chen, J., Ge, M., Zheng, L., Zhang, Z., Qin, B., Liu, J., Smol, J.P., 2023. Ancient DNA reveals potentially toxic cyanobacteria increasing with climate change. *Water Res.* 229, 119435. <https://doi.org/10.1016/j.watres.2022.119435>.
- Zhang, M., Shi, X., Yang, Z., Yu, Y., Shi, L., Qin, B., 2018a. Long-term dynamics and drivers of phytoplankton biomass in eutrophic Lake Taihu. *Sci. Total Environ.* 645, 876–886. <https://doi.org/10.1016/j.scitotenv.2018.07.220>.
- Zhang, Y., Qin, B., Zhu, G., Shi, K., Zhou, Y., 2018b. Profound changes in the physical environment of Lake Taihu from 25 years of long-term observations: implications for algal bloom outbreaks and aquatic macrophyte loss. *Water Resour. Res.* 54, 4319–4331. <https://doi.org/10.1029/2017WR022401>.
- Zhang, Y., Su, Y., Liu, Z., Yu, J., Jin, M., 2017. Lipid biomarker evidence for determining the origin and distribution of organic matter in surface sediments of Lake Taihu, eastern China. *Ecol. Indic.* 77, 397–408. <https://doi.org/10.1016/j.ecolind.2017.02.031>.
- Zhou, J., Leavitt, P.R., Zhang, Y.B., Qin, B.Q., 2022. Anthropogenic eutrophication of shallow lakes: is it occasional? *Water Res.* 221, 118728. <https://doi.org/10.1016/j.watres.2022.118728>.
- Zhu, L., Shi, W., Van Dam, B., Kong, L., Yu, J., Qin, B., 2020. Algal accumulation decreases sediment nitrogen removal by uncoupling nitrification-denitrification in shallow eutrophic lakes. *Environ. Sci. Technol.* 54, 6194–6201. <https://doi.org/10.1021/acs.est.9b05549>.
- Zhu, M., Paerl, H.W., Zhu, G., Wu, T., Li, W., Shi, K., Zhao, L., Zhang, Y., Qin, B., Caruso, A.M., 2014. The role of tropical cyclones in stimulating cyanobacterial (*Microcystis* spp.) blooms in hypertrophic Lake Taihu, China. *Harmful Algae* 39, 310–321. <https://doi.org/10.1016/j.hal.2014.09.003>.