



A statistical framework to track temporal dependence of chlorophyll–nutrient relationships with implications for lake eutrophication management



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ABSTRACT

A reliable chlorophyll–nutrient relationship (CNR) is essential for lake eutrophication management. Although the spatial variability of CNRs has been extensively explored, temporal variations of CNRs at the individual lake scale has rarely been discussed. The paucity of information about temporal dependence in CNRs may in part be due to the lack of a suitable statistical framework that helps guide such investigations. In order to reveal temporal dependence of CNR, this study develop a novel statistical framework. In the framework, we employ quantile regression to generate overall (the entire dataset), annual (subsets for each year), and accumulative (subsets collected before a certain year) CNRs. We aim to 1) show biases of annual relationships by comparing the overall and annual relationships and 2) determine whether or not data accumulation is enough to develop a reliable CNR. We use Lake Champlain and Lake Kasumigaura as case studies to illustrate the necessary steps needed to utilize this novel framework. Results show that large interannual variations exist for CNRs. Accumulative relationships tend to converge to the overall relationship, indicating that overall relationships are reliable for informing lake-specific eutrophication management in the two case study lakes. The novel statistical framework that we propose for a procedure to estimate reliable CNRs is important for informing lake-specific eutrophication control decision-making processes.

1. Introduction

Globally, anthropogenic nutrient loading from industrial and agricultural development has deteriorated water quality and impaired ecosystem structure and function in many lakes (Chislock and Doster, 2013; Ho et al., 2019; Huang et al., 2019; Le Moal et al., 2019; Smith and

Schindler, 2009). In most cases, nutrients, including phosphorus and nitrogen, are treated as two of the most manageable factors to curb lake eutrophication (Carvalho et al., 2013; Conley et al., 2009; Schindler et al., 2008). Therefore, quantitative relationships between chlorophyll (Chl) and nutrients are fundamental to guide lake eutrophication management (Filstrup et al., 2014; Huo et al., 2013; Huo et al., 2018; Jones

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et al., 2011; Liang et al., 2021b; Vinçon-Leite and Casenave, 2019).

The Chl-nutrient relationship (CNR) often varies substantially among lakes. The variations of CNRs across spatial scales complicate lake eutrophication management (Brown et al., 2000; Chen and Li, 2014; Phillips et al., 2008). For example, a “one-size-fits-all” approach – where a single CNR is used to manage eutrophication across a diverse set of lakes – is often not possible. This spatial variability in CNRs has been well established (Cha et al., 2016; Qian et al., 2019; Zou et al., 2020), and other studies have explored drivers of these spatially-varying relationships at multiple scales (Abell et al., 2012; Filstrup et al., 2014). Such studies have found that regional factors, such as land use and land cover, can explain some of the spatial variation observed in CNRs (Huo et al., 2014; Rohm et al., 2002; Wagner et al., 2011). In addition, recent research has shown that factors operating at the lake-scale, such as lake depth and watershed-scale (e.g., surrounding land use) can also influence CNRs (Qin et al., 2020; Read et al., 2015). Considering the spatial heterogeneity of lake-specific drivers of CNRs, lake-specific eutrophication management strategies (Xu et al., 2015a), such as the development of lakes-specific nutrient criteria (Liang et al., 2020; Olson and Hawkins 2013), has been recommended.

At the lake-scale, a reliable CNR is critical for informing successful management strategies. However, a lake-specific CNR might vary over time (Liang et al., 2019). Understanding potential temporal variations in CNRs could also be important for determining a reliable (i.e., temporally stable) lake-specific CNR. Despite some recent progress on developing lake-specific CNRs (Liang et al., 2020; Qian et al., 2019), we note that the temporal dependence of CNR has rarely been discussed or evaluated. Specifically, several questions exist, including 1) to what extent do CNRs vary annually and 2) if they do, how to determine whether or not a developed CNR is reliable enough to inform lake eutrophication management. Particularly, there lacks a statistical framework for revealing the temporal dependence of CNRs to answer the aforementioned questions.

Quantile regression (QR), as an alternative to traditional ordinary least-squares regression (OLSR), has been introduced to quantify cause-effect relationships between ecological variables. QR is used to explore the effect of one predictor on different quantiles of a response variable (Cade and Noon, 2003; Das et al., 2019), rather than the average of the response variable as in mean regression methods (e.g., OLSR; Altman and Krzywinski, 2015). The major advantage of applying QR to develop CNRs is that QR can estimate the upper boundary of Chl response to the nutrient (Chen and Li, 2014; Heiskary and Bouchard, 2015; Koenker and Bassett, 1978). Compared to OLSR-derived models, which fail to address the effect of unmeasured factors, QR-derived models can reflect the limiting effect of a nutrient on Chl by modeling the upper distribution of the Chl response to the nutrient (Cade et al., 1999; Xu et al., 2015a). Therefore, any unmeasured factors effecting CNRs including non-algal suspended solids, landscape factors, grazing by zooplankton, and other factors are considered in the QR analysis (Mazumder, 1994; Wagner et al., 2011). QR has been recently expanded in CNR estimations for lake-specific eutrophication assessment (Xu et al., 2015b), regional eutrophication management (Zou et al., 2020), and global nutrient limitation quantifications (Abell et al., 2012). However, there still lacks QR applications in revealing the temporal dependence of CNRs.

In this study, we propose a novel statistical framework to reveal lake-specific temporal dependence of CNRs with the application of QR. In the framework, we develop overall (the entire dataset), annual (subsets for each year), and accumulative (subsets collected before a certain year) CNRs. We attempt to 1) quantify any potential biases in annual relationships by comparing the overall and annual CNRs, and 2) determine whether or not data accumulation is appropriate for developing a reliable CNR by comparing the overall and accumulative relationships. We use data from Lake Champlain (LC; located in the United States) and Lake Kasumigaura (LK; located in Japan) as case studies to illustrate steps necessary to utilize this novel framework. We further discuss how our statistical framework could inform lake-specific eutrophication

management.

2. Materials and methods

2.1. Study area

We chose LC and LK as case studies based on two considerations, namely the availability of long-term monitoring data and lake-specific physical characteristics. Physical characteristics, measures of climate, and nutrient and Chl conditions for these two lakes are summarized in Table 1. LC is located in the northeastern United States. It is a deep lake, with an average depth of 20 m and with a surface area of 1127 km². The lake is well-known for its large watershed area to lake area ratio (19:1) (Isles et al., 2017). LK is located in Ibaraki Prefecture, approximately 50 km northeast of Tokyo, Japan. It is the second largest shallow lake in Japan with an average depth of 4.0 m. The lake has a surface area of 171 km². The watershed to lake area ratio is approximately 9:1 (Arai and Fukushima, 2014; Tsuchiya et al., 2019).

In this study, we included total nitrogen (TN) and total phosphorus (TP) in our analysis because they are the primary limiting nutrients of lake phytoplankton (Phillips et al., 2008; Stow and Cha, 2013). The concentrations of TN, TP, and Chl for estimating CNRs were obtained from the long-term monitoring datasets for LC and LK. The monitoring data for LC are freely available at <https://dec.vermont.gov/watershed/lakes-ponds/lake-champlain>. A total of 4413 paired TN, TP, and Chl concentrations for 12 sampling stations in LC from 1992 to 2018 were used in this study. The LK database was obtained from the National Institute for Environmental Studies 2016, and accessed via <https://db.cger.nies.go.jp/gem/moni-e/inter/GEMS/database/kasumi/index.html> 01-01-2020. Monthly time-series of 10 long-term sampling stations spanning the years 1985–2016, were used and resulted in 3807 paired Chl, TN and TP values for LK. All biweekly and monthly monitoring of TN, TP, and Chl concentrations were directly applied to develop CNRs.

Table 1
Comparisons of selected physical, chemical and climate characteristics between Lake Champlain and Lake Kasumigaura.

Parameter	Lake Champlain	Lake Kasumigaura
Location	New York, Vermont and Quebec, USA	Ibaraki Prefecture, Japan
Surface area (km ²)	1127	171
Maximum depth (m)	122	7.3
Average depth (m)	20	4
Watershed/ surface area	19:1	9:1
Average annual rainfall (mm)	845	Approximately 1250
Annual air temperature (°C)	−13.9–26.	Approximately 14.0
Water temperature (°C)	18.6 (2.7–28.8)	16.5 (2.3–32.4)
Sampling month	April–November	January–December
Sampling frequency	Biweekly	Monthly
Data available website	[a]	[b]
Chlorophyll (Chl, mg·m ^{−3})		
Mean	6.3	61.6
Std.D	6.7	37.8
Min	0.5	0.0
Max	116.4	466.0
Total nitrogen (TN, mg·m ^{−3})		
Mean	403.3	1511.2
Std.D	132.1	632.8
Min	110.0	475.0
Max	1710.0	4635.0
Total phosphorus (TP, mg·m ^{−3})		
Mean	24.2	115.8
Std.D	17.4	51.8
Min	5.0	13.0
Max	235.0	765.0

In LC, average TN, TP, and Chl concentrations were $403.3 \text{ mg} \cdot \text{m}^{-3}$, $24.2 \text{ mg} \cdot \text{m}^{-3}$, and $6.3 \text{ mg} \cdot \text{m}^{-3}$, respectively (Table 1). Spatial variation of trophic status was observed in LC, with some areas being eutrophic (Xu et al., 2015b). In contrast, LK is hypereutrophic, largely due to the extremely high loads of organic matter and nutrients (Ishii et al., 2009; Masunaga and Komuro, 2020). The average TN, TP, and Chl concentrations were $1511.2 \text{ mg} \cdot \text{m}^{-3}$, $115.8 \text{ mg} \cdot \text{m}^{-3}$, and $61.6 \text{ mg} \cdot \text{m}^{-3}$, respectively (Table 1). In addition, concentration ranges for TN, TP, and Chl concentrations spanned a wider range in LK than those in LC.

2.2. Framework to reveal the temporal dependence of CNRs

The proposed novel statistical framework to quantify potential temporal dependence of CNRs is shown in Fig. 1. The framework includes three steps to estimate the overall, annual, and accumulative CNRs. Both Chl-TN and Chl-TP relationships were developed because both nutrients might influence Chl concentrations within lakes (Conley et al., 2009). In the framework, we want to explore two potential

temporal dependencies that may exist. On the one hand, we compare the overall and annual relationships to show if there exist biases in annual relationships. On the other hand, we compare the overall and accumulative relationships to determine whether data accumulation is sufficient to develop a reliable CNR.

In the proposed framework, the major difference among the three steps is the data used to develop the CNR. The first step is to develop the overall CNR using all available data (Fig. 1a). Next, we develop annual CNRs based on the data collected in every year (Fig. 1b). In the third step, we develop accumulative CNRs using the data collected before a specified year. For example, for the accumulative CNR for the year of 2010, we use all data collected before and including 2010 (Fig. 1c). Following these steps, we can determine if CNRs for the initial year are identical in step 2 and step 3. The CNR for the last year in the third step is the same as the overall CNR in the first step.

To explore the variation of annual CNRs, we compare the overall relationship with annual relationships. Here, the overall relationship could be treated as the baseline. If annual relationships all concentrate

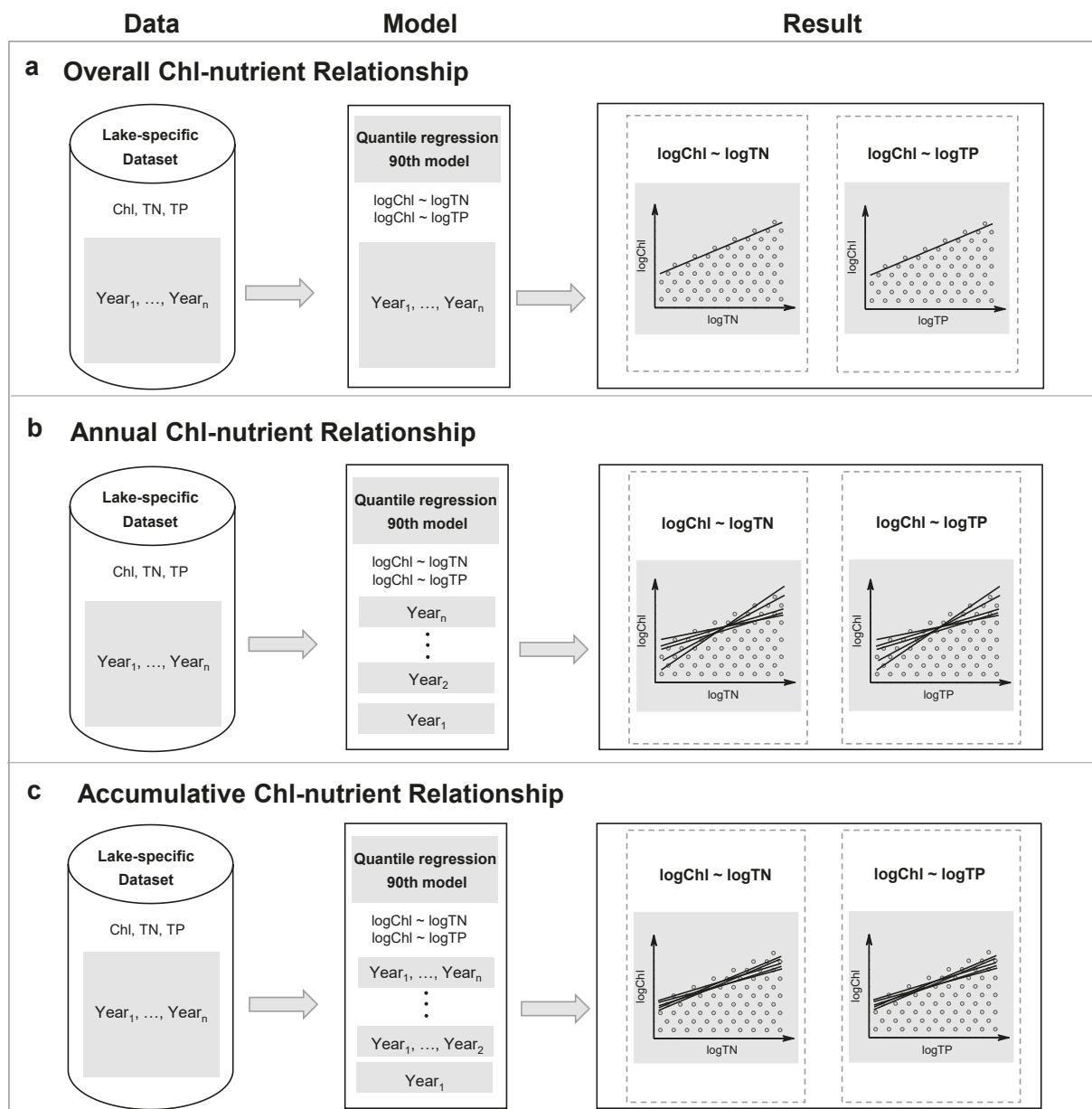


Fig. 1. The statistical framework to quantify potential temporal dependence of chlorophyll–nutrient relationships. The three steps include estimating (a) overall, (b) annual, and (c) accumulative chlorophyll–nutrient relationships.

near the overall relationship with small biases, we could determine that even an annual CNR is reliable to inform lake-specific eutrophication management. If the annual relationships deviate from the overall mean relationship substantially, it is worth exploring whether or not the overall relationship is reliable enough.

If there is large variation among annual CNRs, we can then compare the overall relationship with accumulative relationships to determine the reliability of the overall relationship. As aforementioned, the accumulative CNR for the last year is the same as the overall relationship. However, if the accumulative relationships are very similar to the overall relationship several years before the last year (bias less than 10%), we could conclude that the CNR is relatively stable over time. Therefore, the overall relationship may be reliable enough to inform lake eutrophication management. Otherwise, if the bias between the accumulative relationship and the overall relationship remains large until the year just before the last year, the overall relationship might not be adequate and more monitoring data may be required.

2.3. QR for CNR development

In the framework, we apply QR to develop CNRs in all three steps. The main function of QR is expressed as Eq. (1) (Koenker and Bassett, 1978):

$$y_i = b_0 + b_1 x_i + \varepsilon_i \quad (1)$$

where i represents the order of observations ($i = 1, 2, \dots, M$, M is the sample size), y represents the log transformed Chl concentration ($\log\text{Chl}$), and x represents the log transformed nutrient concentration ($\log\text{TP}$ or $\log\text{TN}$). b_0 and b_1 represent the regression intercept and slope. Since all the data are log transformed, the regression slope (b_1), is interpreted as the percent change in Chl concentration per 1% change in TN and TP concentration (Qian, 2017). ε is the error. The parameter estimation in linear QR is based on the minimum of absolute error (Eq. (2)) (Koenker and Bassett, 1978):

$$\min \left[\sum_{i \in \{i: y_i \geq (b_0 + b_1 x_i)\}} \tau |y_i - b_0 - b_1 x_i| + \sum_{i \in \{i: y_i < (b_0 + b_1 x_i)\}} (1 - \tau) |y_i - b_0 - b_1 x_i| \right] \quad (2)$$

where τ represents the quantile of the response. In our study, τ is chosen to be 0.9, following common practices when quantifying the limiting effect in ecological studies (Chassot et al., 2010).

Coefficients of determination (R^1) for 0.9 quantile were calculated to infer the goodness of fit to the upper boundary of data distributions for CNRs. The R^1 value is calculated as follows: $R^1 = 1 - (\text{SUM}_{(F)} / \text{SUM}_{(R)})$, where $\text{SUM}_{(F)}$ refers to the sum of weighted absolute deviations minimized to estimate each of the full parameter models, and $\text{SUM}_{(R)}$ refers to the sum of weighted absolute deviations minimized to estimate each of corresponding reduced parameter models of $\log\text{Chl} = b_0 + \varepsilon$ (Koenker and Machado, 1999). In order to be comparable to the OLSR-derived R^2 , the QR-derived R^1 for 0.9 quantile were converted to R^2 , which are calculated as follows: $R^2 = 1 - (1 - R^1)^2$ (McKean and Sievers, 1987; Schooley and Wiens, 2005; Xu et al., 2015a).

All models were fitted using the R software (R version 4.0.2; R Core Team 2020). The algorithm for parameter estimation of QR models was implemented in the *quantreg* package (Koenker, 2020). The code for the development of overall, annual, and accumulative CNRs is freely available at <https://doi.org/10.5281/zenodo.4087838>.

3. Results

3.1. Overall and annual CNR

Overall CNRs based on all available data are shown in Fig. 2. The overall CNR describes the upper response of Chl to each nutrient. In LC, the QR-estimated regression slopes of TN and TP are 1.33 and 0.87, respectively, indicating that a 1% increase/decrease for TN and TP concentration would lead to a 1.33% and 0.87% increase/decrease of Chl concentration. In LK, the QR-estimated regression slopes of TN and

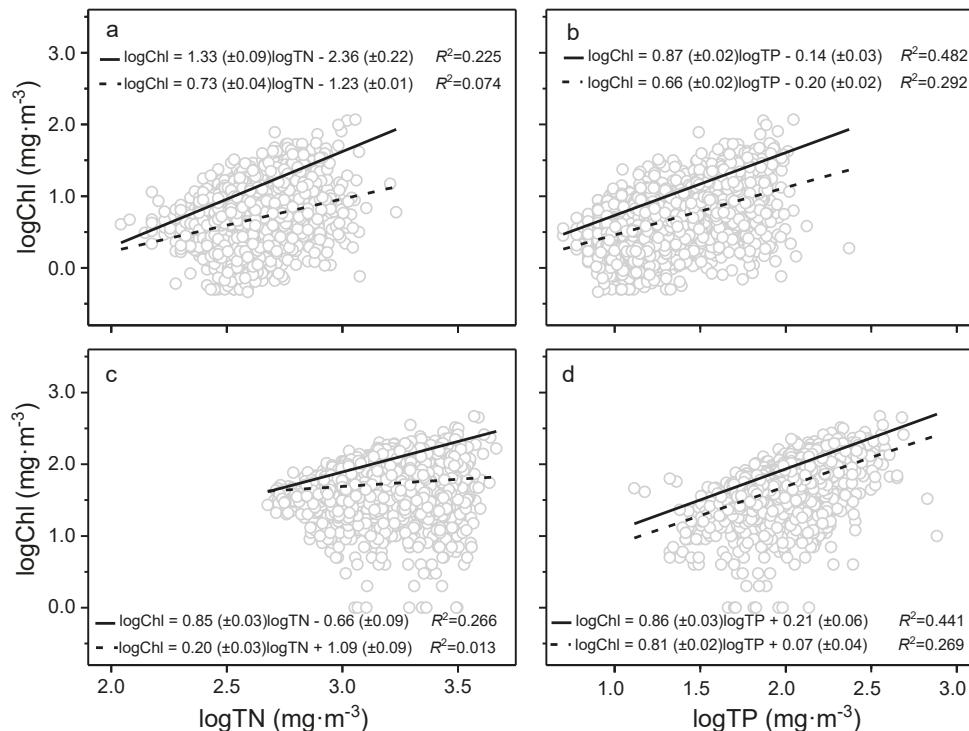


Fig. 2. Scatter plots of chlorophyll ($\log_{10}\text{Chl}$) against total nitrogen ($\log_{10}\text{TN}$, a & c) and total phosphorus ($\log_{10}\text{TP}$, b & d) for Lake Champlain (a & b) and Lake Kasumigaura (c & d). Black solid lines indicate the relationships for Chl-TN and Chl-TP at the 0.9 regression quantile. Black dashed lines are ordinary least-squares regression derived models for Chl-TN and Chl-TP relationship.

TP are 0.85 and 0.86, respectively, indicating that a 1% increase/decrease of TN and TP concentration would lead to a 0.85% and 0.86% increase/decrease of Chl concentration. It is worth noting that the overall CNRs derived from the QR method fit better than those derived from the OLSR method with higher R^2 values for QR than for OLSR (Fig. 2).

Generally, there were no apparent patterns in the annual variation for the annual CNRs in both lakes. Regression parameters show large interannual variation (Figs. 3 and 4). Parameter estimation results of annual CNRs in LC are shown in Fig. 3 (hollow triangles). For the Chl-TN relationship, regression slopes range from 0.56 to 2.05, with an average of $1.35 (\pm 0.41)$, standard deviation. Regression intercepts range from -4.21 to -0.50 , with an average of $-2.46 (\pm 1.01)$. For the Chl-TP relationship, average regression slopes and intercepts were $0.87 (\pm 0.25)$ and $-0.16 (\pm 0.34)$, respectively. Corresponding ranges were from 0.34 to 1.54 and from -0.90 to 0.51.

Parameter estimation results of annual CNRs in LK are shown in Fig. 4 (hollow triangles). For the Chl-TN relationship, regression slopes range from -0.05 to 1.36, with an average of $0.79 (\pm 0.31)$. Regression intercepts range from -2.24 to 2.03, with an average of $-0.50 (\pm 0.92)$. For the Chl-TP relationship, average regression slopes and intercepts were $-0.96 (\pm 0.29)$ and $-0.002 (\pm 0.57)$, respectively. Corresponding ranges were from 0.25 to 1.56 and from -1.22 to 1.38. Despite of relatively larger ranges of TN, TP, and Chl concentrations in LK compared with LC, estimated regressions parameters for these two lakes did not show many differences.

3.2. Accumulative CNRs

Parameter estimation results for the accumulative CNRs are shown in Figs. 3 and 4 (solid dots). Unlike the annual CNRs, regression parameters of accumulative CNRs show an obvious trend with year and generally converge to stable values (i.e., the parameters values of overall relationships) over time. In LC, regression slopes for the Chl-TN and Chl-TP relationships both increase with increasing year, while

regressions intercepts both have decreasing trends (Fig. 3, solid dots). The regression slope for the Chl-TN relationship increases from 0.56 to 1.22 in 1995, and then become very similar to the 1.33 slope estimated for the overall relationship. Regression intercepts decrease from -0.58 to -2.23 for 1995, and then converge to be very close to the overall relationship. For the Chl-TP relationship, the slope increases from 0.49 to 0.80 for the year 2004, and then become very close to the 0.87 slope estimated for overall relationship. The intercept decreases from 0.36 to -0.05 for the year 2004, and then becomes very close to the overall relationship.

In LK, patterns reverse. Both regression slopes decrease and both intercepts increase over time (Fig. 4, solid dots). The regression slope for the Chl-TN relationship increases from 1.02 to 1.18, then decreases to 0.86 in the year 1993, and generally converges to the slope of overall relationships. Regression intercept decreases from -1.16 to -1.61 , and then increases to 0.68 in the year 1993, and converges to the overall relationships. For the Chl-TP relationship, the slope increases from 1.01 to 1.22, and then decrease to 0.93 in the year 2001, and generally converges to the slope of overall relationships. Regression intercept decreases from 0.08 to -0.31 , then increases to 0.10 in the year 2001 and tends to the overall relationships.

4. Discussion

4.1. The difference of nutrient limitations between two lakes

Difference in the upper boundary equations on the CNRs for LC and LK indicates that the nutrients limitation of chlorophyll varies between these two case study lakes. Nutrients limitation variation may be due to the difference in hydrodynamic disturbance of these lakes. Conjecturally, the geographic location and water depth of the two lakes are the main reason for different hydrodynamic conditions. LC is relatively stable in terms of hydrodynamic conditions due to its deep depth (Table 1). In contrast, the shallow LK is influenced by the mixing effect of storms, especially typhoons. The energy of these storms leads to the

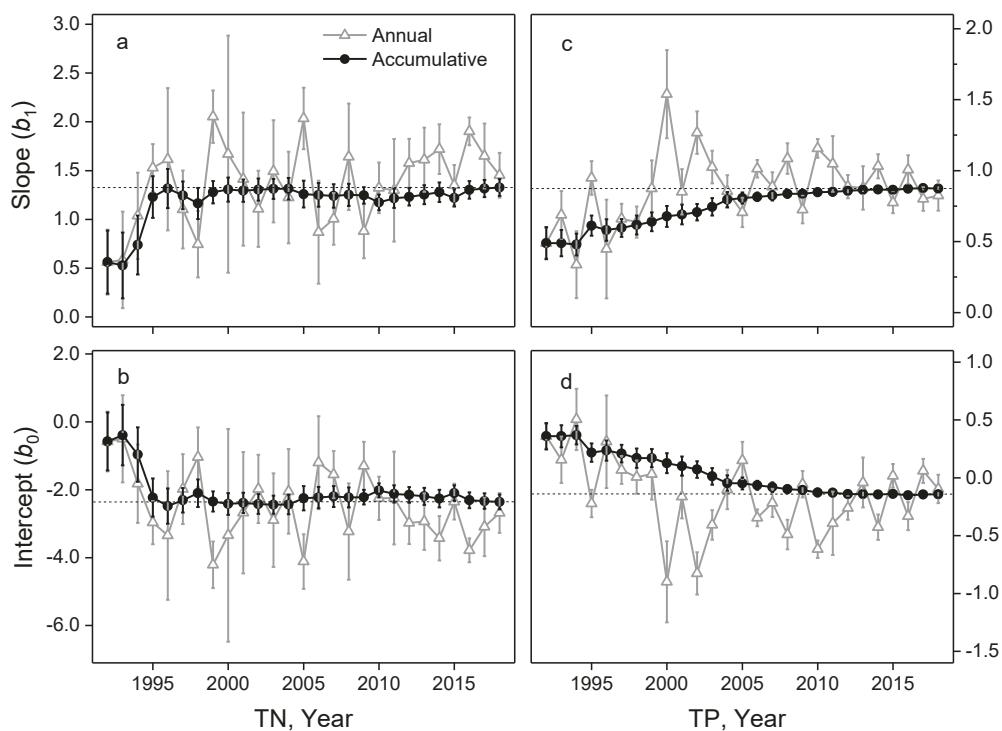


Fig. 3. Estimated coefficients for annual (hollow triangles) and accumulative (solid dots) chlorophyll (Chl) – nutrient relationships in Lake Champlain. The coefficients include slopes (b_1) and intercepts (b_0) for Chl – total nitrogen (Chl-TN, a & b) and Chl – total phosphorus (Chl-TP, c & d) relationship. The horizontal dashed line indicates the relationship for Chl-TN or Chl-TP at the 0.9 regression quantile.

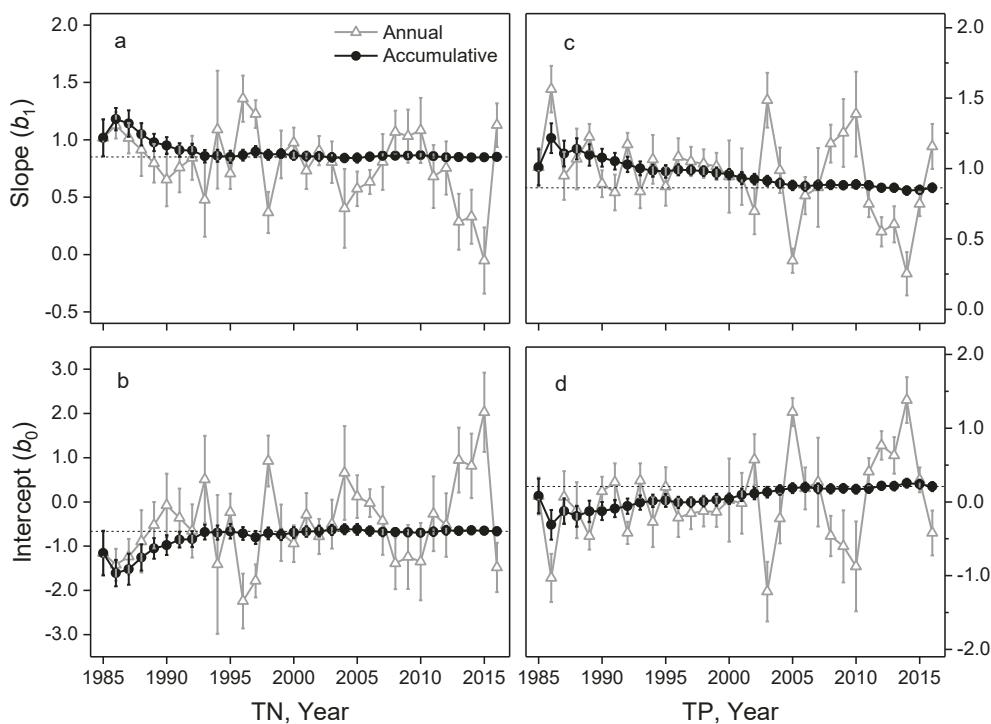


Fig. 4. Estimated coefficients for annual (hollow triangles) and accumulative (solid dots) chlorophyll (Chl) – nutrient relationships in Lake Kasumigaura. The coefficients include slopes (b_1) and intercepts (b_0) for Chl – total nitrogen (Chl-TN, a & b) and Chl – total phosphorus (Chl-TP, c & d). The horizontal dashed line indicates the relationship for Chl-TN or Chl-TP at the 0.9 regression quantile.

suspension of sediments and an increase the non-algal turbidity, thus enhancing light limitation of phytoplankton (Brezonik et al., 2019; Jones and Knowlton, 2005; Zhang et al., 2017). Therefore, it can be reasonably assumed that the hydrodynamic disturbance is the primary factor influencing the nutrient limitation by the result of lower slopes for Chl-nutrients upper boundary equations for LK compared to LC (Fig. 2).

The differences in nutrients limitation of chlorophyll between lakes indicate that lake-specific nutrient criteria are not universally applicable, and the lake-specific upper boundary equation of Chl-nutrient should be calculated using a lake-specific dataset. In this study, the QR-derived Chl-nutrient upper boundaries reflecting peak algal biomass response to nutrients under an ideal condition, fit better than OLSR-derived models which cannot account for unmeasured drivers for Chl (Fig. 2). Therefore, it is reasonable to assume that QR provides more insight into the ecological concept of “law of the minimum”, which could help lake managers to better understand and establish more accurate nutrient criteria. Here, we introduce a Chl standard of $15.0 \text{ mg} \cdot \text{m}^{-3}$ applied by USEPA (2000) as an example to show that the calculation of nutrient thresholds by the upper boundary CNR is different between the two case study lakes. It appears that nutrient criteria derived from the overall Chl-nutrient equations are significantly different between these two case lakes. Given the target Chl concentration of $15.0 \text{ mg} \cdot \text{m}^{-3}$, the thresholds of TN and TP for LC are $458.8 \text{ mg} \cdot \text{m}^{-3}$ ($\log TN = 2.66$), and $32.9 \text{ mg} \cdot \text{m}^{-3}$ ($\log TP = 1.52$), respectively. The thresholds for TN and TP for LK are $146.1 \text{ mg} \cdot \text{m}^{-3}$ ($\log TN = 2.16$), and $13.4 \text{ mg} \cdot \text{m}^{-3}$ ($\log TP = 1.13$), respectively. This suggests that lake-specific nutrient criteria developed using upper boundary QR equations will be useful for informing lake eutrophication management.

4.2. Variation of annual CNR

There was large variations in regressions parameters among annual CNRs in both lakes (Figs. 3 and 4). More importantly, regression parameters in some years differed substantially from regression parameters of the overall relationship. For example, in LC, for the Chl-TN

relationship in 1992, the regression slope was estimated to 0.56, and the difference from the overall value is 1.36 times the slope of 1992. In LK, for the Chl-TP relationship in 2010, the regression intercept was estimated at -0.88 , and the difference from the overall value of 0.21 was 1.24 times the intercept of 2010.

Because the CNR is determined by the regression slope and intercept together, we further explored the deviation of annual CNRs with the overall relationship (Fig. 5). We found that annual relationships vary substantially among years, and are different from the overall relationships. In fact, the biases between some annual relationships and the overall relationship were very large. For example, for the Chl-TN relationship in LC, the regression equation in 1999 was $\log Chl = 2.05 (\pm 0.27) \times \log TN - 4.21 (\pm 0.69)$, which was different from the overall regression equation $\log Chl = 1.33 (\pm 0.09) \times \log TN - 2.36 (\pm 0.22)$. Assuming that the TN is $1000 \text{ mg} \cdot \text{m}^{-3}$, the Chl derived from the 1999 regression equation is $87.1 \text{ mg} \cdot \text{m}^{-3}$ and that from overall equation is $42.7 \text{ mg} \cdot \text{m}^{-3}$. Additionally, assuming that the TP is $100 \text{ mg} \cdot \text{m}^{-3}$, the Chl calculated from the regression equation in 1986 is $123.0 \text{ mg} \cdot \text{m}^{-3}$, which is 1.45 times that of overall.

Global climate change and regional watershed nutrient dynamics may result in annual variation of Chl-nutrient upper boundary equations. Generally, erratic fluctuation in nutrient limitation on Chl is attributed to annually differences in temperature, which is vital for phytoplankton growth (Huisman et al., 2018; Huo et al., 2019; Paterson et al., 2017). Additionally, differences in annual precipitation is expected to influence concentrations of non-algal suspended solids by watershed inputs, which contributes to variations in the slopes of Chl-nutrient upper boundary equations (Jones and Knowlton, 2005; Yuan and Jones, 2020). As the different forms of nitrogen and phosphorus are transported from the watershed to the lake, variable precipitation intensity alters the input N:P, which further affects the utilization of nutrients by phytoplankton in lakes (Isles et al., 2017).

Considering the large biases between the overall relationship and annual relationships in some years for both lakes and both nutrients, we conclude that the annual CNR do in fact vary. Short-term monitoring

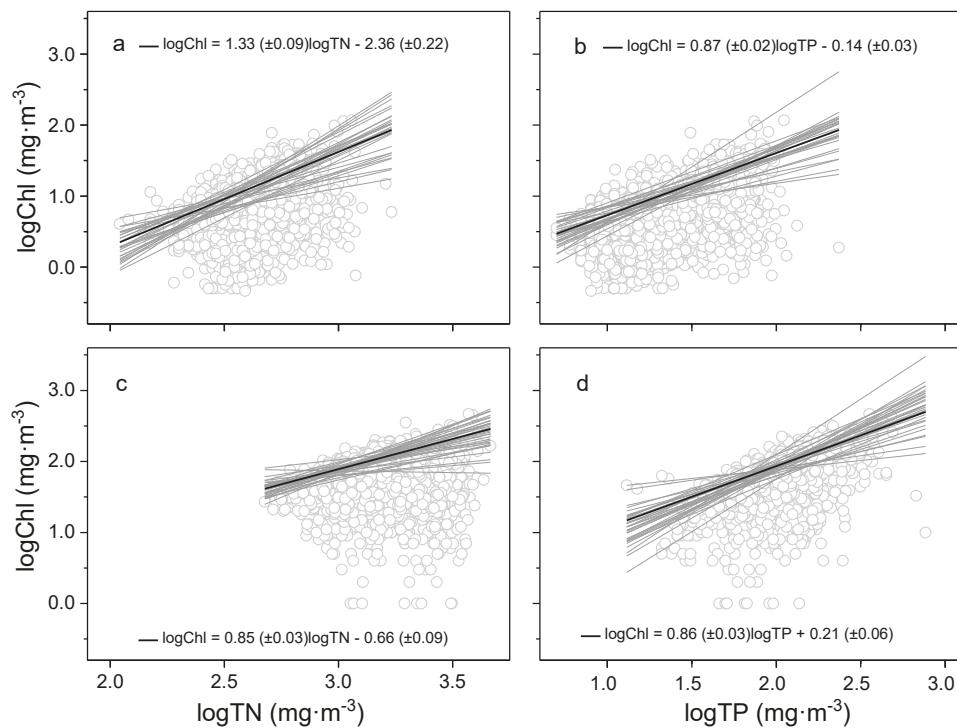


Fig. 5. Scatter plots of \log_{10} chlorophyll (Chl) against \log_{10} total nitrogen (TN, a & c) and \log_{10} total phosphorus (TP, b & d) for Lake Champlain (a & b) and Lake Kasumigaura (c & d). The gray lines represent annual Chl–nutrient relationships, and black lines indicate the overall Chl–nutrient relationships at the 0.9 regression quantile.

might not be able to reflect the overall ecological process influencing nutrient – phytoplankton dynamics over longer temporal scales (Smol, 2019). Despite annual relationships in some years being close to the overall relationship, applying a CNR developed using data from a single

year might mislead lake eutrophication management decisions. To develop a reliable lake-specific CNR, more data collected over more years, rather than more data collected in a single year, are likely required.

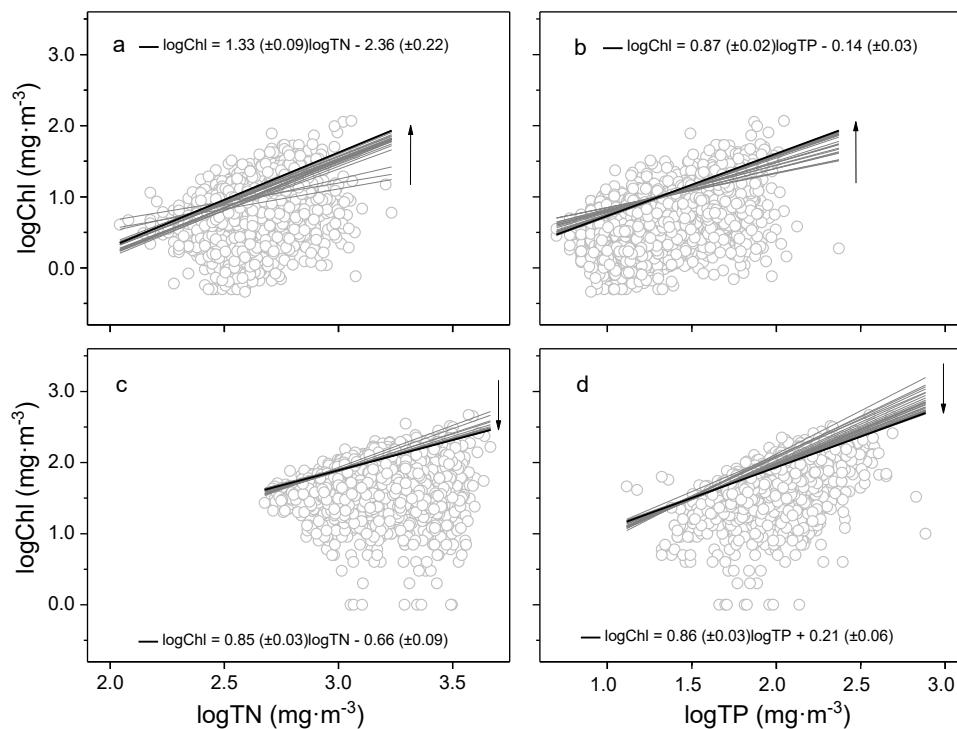


Fig. 6. Scatter plots of \log_{10} chlorophyll (Chl) against \log_{10} total nitrogen (TN, a & c) and \log_{10} total phosphorus (TP, b & d) for Lake Champlain (a & b) and Lake Kasumigaura (c & d). The gray lines represent accumulative Chl–nutrient relationships, and black lines indicate the overall Chl–nutrient relationships at the 0.9 regression quantile. Arrows represent the trends for accumulative Chl–nutrient relationships converging to the overall relationship.

4.3. Stabilization of CNR with data accumulation

After determining that data from a single year might not be adequate to develop a reliable CNR and that additional data collected over more years is desirable, the next question is how to determine whether or not the accumulated data are sufficient to adequately characterize the lake's 'true' CNR. In our framework, we answer this question by comparing the overall relationship with accumulative relationships. We consider the CNR stable when the difference between coefficients for a certain dataset and the overall relationship is 10% or less.

As we mentioned before, parameters of accumulative CNRs generally converge to parameters of the overall relationships. We visually identified the year at which parameters of accumulative relationship and overall relationships become similar (converge). In LC, for the TN-Chl and TP-Chl relationship, the year is approximate 1995 and 2012, respectively. In LK, for the TN-Chl and TP-Chl relationship, the year is approximate 1990 and 2001, respectively.

Moreover, accumulative relationships tend to converge to the overall CNRs as more years are sampled (Fig. 6). For example, for the Chl-TN relationship in LC, the accumulative relationship for the initial year differs from the overall Chl-TN relationship. As more years are included in the calculation of the relationship, the accumulative relationship becomes closer to the overall relationship. Finally, accumulative relationships for years near the last year are almost coincident with the overall relationship, with variation between the two less than 10%. The Chl-TP relationships in LC and those in LK have similar patterns.

The convergent trend of accumulative CNR to the corresponding overall relationship indicates that the overall relationship is reliable to inform lake eutrophication since it becomes relatively stable after several years from the initial year. In our cases, the number of monitoring years were 27 and 32 for LC and LK, respectively. In fact, considering the earlier convergence of accumulative CNRs, even if we had less data, we could determine the reliability of the overall relationship. For example, for the Chl-TN relationship in LK, if we had approximately 9 years of monitoring data, we could determine that the overall relationship converges and is thus reliable to inform lake eutrophication management.

4.4. Implications and generalizations of the proposed framework

We have shown how our novel statistical framework can reveal the temporal dependence of CNR, including the exploration of variation of annual CNRs and the convergence of accumulative CNR to the overall relationship. Here, we summarize implications of the statistical framework to lake-specific eutrophication management:

Firstly, by comparing the overall with annual CNRs, we can illustrate the variation of annual relationships and how they compare to the overall relationship. In our case, the variation is large, indicating that annual CNRs are likely not adequate to inform lake eutrophication management.

Secondly, via the comparison of the overall and accumulative relationships, we determined whether the current dataset is adequate to develop a reliable CNR, which could be used to inform lake-specific eutrophication management. Since a reliable CNR is essential to lake eutrophication management, our novel statistical framework is of vital importance to ensure the effectiveness of corresponding lake eutrophication control actions.

Given the flexibility of our proposed framework, we suggest that it will be generalizable to many ecological applications through the following considerations. Firstly, with the accumulating of TN, TP, and Chl monitoring data, our framework has great potential for generalization to other lakes. Therefore, the reliability of the framework can be tested in turn. Since phytoplankton biomass may be co-limited by both nitrogen and phosphorus, the N:P ratios (Dolman et al., 2016) or joint nutrients (Liang et al., 2021b; Poikane et al., 2019) can be introduced to generate CNRs to further reveal the temporal dependence. The novel

framework can be further improved by applying the ecologically based quality targets for Water Framework Directive in Europe to assess the temporal dependence of CNRs (Carvalho et al., 2008; Poikane et al., 2010; Poikane et al., 2014). Considering the lack of statistical metrics to determine trend convergence of accumulative CNRs in our framework, a reliable indicator to better assess the trend convergence is an area of future research. The method applied in the framework can also be extended to other methods, such as a Bayesian QR approach (Liang et al., 2021a) and QR neural network (Cannon, 2011).

5. Conclusions

The contribution of this study is the proposed statistical framework which can reveal the temporal dependence of CNR. In the framework, we explored annual variation in CNRs, and the convergence of accumulative CNRs to the overall relationship. The statistical framework is applied in LC and LK. The framework effectively shows the variation of annual CNRs. Our statistical framework can be used to inform lake-specific eutrophication management by revealing the temporal dependence of CNRs, particularly by answering the question about whether or not a developed CNR is reliable. A reliable Chl-nutrient relationship is needed for nutrient criteria establishment which is important for informing eutrophication control decision-making processes. Therefore, our proposed framework can help guide lake-specific eutrophication management.

CRediT authorship contribution statement

Qianlinglin Qiu: Conceptualization, Methodology, Software, Writing – original draft, Writing - review & editing, Visualization. **Zhongyao Liang:** Conceptualization, Methodology, Software, Writing – original draft, Writing - review & editing. **Yaoyang Xu:** Conceptualization, Writing – original draft, Writing - review & editing, Supervision, Project administration, Funding acquisition. **Shin-ichiro S. Matsuzaki:** Data curation, Writing – original draft, Writing - review & editing. **Kazuhiro Komatsu:** Data curation, Writing – original draft, Writing - review & editing. **Tyler Wagner:** Writing – original draft, Writing - review & editing.

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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