Estimation of the long-term nutrient budget and thresholds of regime shift for a large shallow lake in China

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\textbf{Abstract}

In this study, we apply an integrated empirical and mechanism approach to estimate a comprehensive long-term (1953–2012) total nitrogen (TN) and total phosphorus (TP) loading budget for the eutrophic Lake Chaohu in China. This budget is subsequently validated, firstly, by comparing with the available measured data in several years, and secondly, by model simulations for long-term nutrient dynamics using both Vollenweider (VW) model and dynamic nonlinear (DyN) model. Results show that the estimated nutrient budget is applicable for further evaluations. Surprisingly, nutrient loading from non-point sources (85% for TN and 77% for TP on average) is higher than expectation, suggesting the importance of nutrient flux from the soil in the basin. In addition, DyN model performs relatively better than VW model, which is attributed to both the additional sediment recycling process and the parameters adjusted by the Bayesian-based Markov Chain Monte Carlo (MCMC) method. DyN model further shows that the TP loading thresholds from the clear to turbid state (631.8 \(\pm\) 290.16 t y\(^{-1}\)) and from the turbid to clear state (546.0 \(\pm\) 319.80 t y\(^{-1}\)) are significantly different (\(p<0.01\)). Nevertheless, the uncertainty ranges of the thresholds are largely overlapped, which is consistent with the results that the eutrophication of Lake Chaohu is more likely to be reversible (74.12%) than hysteretic (25.53%). The ecosystem of Lake Chaohu shifted from the clear to turbid state during late 1970s. For managers, approximately two-thirds of the current TP loading must be reduced for a shift back with substantial improvement in water quality. Because in practice the reduction of loading from a non-point source is very difficult and costly, additional methods beyond nutrient reduction, such as water level regulation, should be considered for the lake restoration.

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1. Introduction

Nutrient enrichment and the subsequent nutrient level elevation are the primary cause for eutrophication in shallow lakes (Rast and Holland, 1988; Cooke et al., 2005). It has led to multiple serious and undesired consequences in lake ecosystems (Smith et al., 1999), thereby declining in the functioning of lake ecological services for human beings in the region of, for instance, the lower Yangtze River basin in China (Dearing et al., 2012). The nutrient level in the lake is an important indicator for the assessment of lake trophic status from the abiotic aspect (Xu et al., 2001a) and is also an essential component for more comprehensive ecosystem-level indicators (Xu et al., 2001b).

Serving as the driver of the variations in nutrient levels and the regime shifts in the lake ecosystem, nutrient loading budget including both point and non-point sources is essential for long-term evaluation of the ecosystem dynamics in a lake (Carpenter, 2005; Smith et al., 2006). However, the acquisition of the budget generally demands a large amount of work, entailing frequently monitoring data for both hydrology and water quality (Dillon, 1975; Bennett et al., 1999; Hargan et al., 2011). Alternatively, multiple models have been developed for basin nutrient loading estimation in case of limited data availability (Zhang and Jørgensen, 2005). Mechanistic models usually have large uncertainty in the results, whereas empirical models depend mainly on data for accuracy (Reckhow and Chapra, 1999; Zhang and Jørgensen, 2005). Nonetheless, to apply a specific mechanistic or empirical model approach for a long-term nutrient loading estimation, certain amount of data is required, which is still very difficult in countries such as China with the restriction of long-term data availability.

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Lake managers are also interested in the thresholds of nutrient loading to the lake in order to maintain in the more favored clear state. Nutrient thresholds of regime shifts also serve as important ecological indicators for evaluation of ecosystem resilience and restoration strategy (Scheffer et al., 2001). Much effort went into nutrient loading reduction and the evaluation of the corresponding effects by field data analysis (Jeppesen et al., 2005) and model studies (Sagehashi et al., 2001; Zhang et al., 2003; Trolle et al., 2008, 2011). Usually a modeling approach is required to build a quantitative understanding of regime shift thresholds (Carpenter and Lathrop, 2008) and implement the loading reduction estimation. However, in China, few studies and projects for lake management have been conducted in the context of regime shift and alternative stable state for shallow lakes, despite the prominence of this approach around the world (Scheffer and Jeppesen, 2007).

Here, we focus on the fifth largest freshwater shallow lake, Lake Chaohu, in China as a case study. This lake provides important ecological services for the 7.6 million people in the basin, but has suffered from serious eutrophication since the 1980s (Xie, 2009). There is an urgent need for scientific research to obtain the long-term annual nutrient loading data and estimate the essential loading reduction for this lake. To deal with the data limitation, we integrate different types of methods (empirical or mechanism) to estimate different components of loadings according to data availability, instead of applying a specific model. We use this method to estimate a long-term (1953–2012) nutrient loading budget for this lake, which is crucial for the evaluation and assessment of the ecosystem dynamics and status. The estimated loading data can be compared with the limited measured loading values in several years. In addition, the estimated loading data were also validated by the long-term water quality data based on the simulation outputs from both the Vollenweider (VW) model (Vollenweider, 1969, 1979) and a modified dynamic nonlinear (DyN) model (Carpenter et al., 1999; Carpenter and Lathrop, 2008). VW model is the first model linking the nutrient loading and mass in lakes, which has been extensively used by water quality managers worldwide due to its simplicity and validity (Mooij et al., 2010). Afterwards, many modified models based on VW model were developed (for a review see Bryhn and Håkanson, 2007), among which the dynamic nonlinear model (DyN model) developed by Carpenter et al. (1999) turns out to be one of the most widely discussed model. By adding the term of sediment recycling process, the DyN model shows a nonlinear feature that exhibits in many shallow lakes (Scheffer and Jeppesen, 2007), thereby being able to describe a variety of changes in lake eutrophication and restoration, and have a better performance than VW model (Carpenter and Lathrop, 2008). Moreover, the DyN model has the ability to quantitatively estimate the regime shift thresholds, which is important theoretically for ecologists and also practically for lake managers to calculate the necessary nutrient reduction. Once the long-term loading budget and the estimated thresholds have been established, it is possible to speculate about the approximate timing of the regime shift of the lake during the eutrophication process, as well as how much the nutrient loading should be reduced for a substantial improvement in water quality.

The goals of this study are (1) to build the long-term nutrient (TN and TP) budget from 1953 to 2012 for Lake Chaohu in China and validate this budget with the limited measured loading data; (2) to further evaluate the reliability of the estimated loading data by two minimal nutrient mass balances models (VW and DyN model) and compare with long-term water quality observations; (3) to use the DyN model to estimate the probabilistic distribution of the thresholds of lake regime shift from a clear to turbid state and backwards; (4) to estimate the timing of regime shift in the historical development of the lake and the nutrient loading reduction needed for restoration of Lake Chaohu to a good ecological state in the future.

## 2. Materials and methods

### 2.1. Study site

Lake Chaohu (31°34'N, 117°26'E) is the fifth largest lake in China (with a surface area of approximately 780 km²), and it is a typical freshwater shallow lake (mean depth 3.06 m in 1980) (Wang, 1986). The lake was famous for its nice scenery and abundant aquatic products before the 1950s (Xu et al., 1999). However, due to rapid social-economic growth in the lake basin during the last several decades, the lake became seriously eutrophic, with TP loading amounts to 1050 t per year in the late 1980s (Tu et al., 1990) and 1550 t per year during 2002–2010 on average (State Department of People’s Republic of China, 2010). The ecosystem structure was largely altered (Xu et al., 1999), e.g., the macrophyte coverage of the lake significantly decreased (Xie, 2009; Kong et al., 2013) and blue-green algae blooms occurred during the summer in recent years (Kong and Song, 2011). The location of the lake basin is shown in Fig. 1, as well as the land use map in 2011 and all of the towns and cities that are involved in the nutrient loading calculation, including Hefei City (HF), Feidong County (FD), Feixi County (FX), Chaohu City (CH), Hanshan County (HS), Wuwei City (WW), Lujiang County (LJ), Shucheng City (SC) and Lu’an City (LA).

### 2.2. Development of the nutrient loading budget for Lake Chaohu from 1953 to 2012

The framework of the nutrient loading budget is illustrated in Fig. 2. Generally, the total amount of nutrient discharge (both TN and TP) was estimated, and the actual amount of nutrient that entered into the lake water body (loadings) was calculated based on the wastewater treatment ratio (for point source) or lake entering ratio (for non-point source), which are demonstrated in detail in the supplementary materials. The long-term loading budget from 1953 to 2012 determined for each year in the lake will be described below.

#### 2.2.1. Loading from point sources

The total amount of point source loading includes nutrients from municipal, industrial and livestock and poultry breeding wastewater discharge. This part of the nutrient loading was mainly estimated based on empirical relations from the literature and collected social-economic data. For municipal wastewater discharge, the values were the product of the city population in the basin and the nutrient discharge per capita. Similarly, for industrial wastewater discharge, the values were the product of industrial production and the nutrient discharge per 10 thousand RMB of industrial production. Municipal and industrial wastewaters were supposed to be treated in sewage plants before entering into the river and lake, the ratio of which was estimated as 0% from 1953 to 1995, 50% from 1995 to 2005, and gradually increased to 75% by 2012 (State Department of People’s Republic of China, 2000, 2005, 2010). Livestock and poultry breeding wastewater discharge were estimated based on the amount of different types of cultivation (animals) and the corresponding nutrient discharge. Due to the difficulty in determining the wastewater amount and the nutrient concentrations in the wastewater, the values from the “Discharge standard of pollutants for livestock and poultry breeding (GB 18596-2001)” were applied instead. All of the data values and sources are listed in Table S1 in the supplementary materials.

#### 2.2.2. Loading from non-point sources

The total amount of nutrient discharge from non-point sources includes atmospheric dry and wet deposition, fertilizer loss, and nutrient discharge in the dissolved form and also the solid form from soil erosion. Air deposition was estimated according to the
measured wet deposition of TN and TP in Lake Chaohu. Based on the wet deposition of TN (254.5 t) and TP (13.88 t) in 1984 (Wang, 1986) and the ratio of wet deposition and total deposition measured in Lake Taihu (80.3% for TN and 28.2% for TP) (Yang et al., 2007), the air deposition in Lake Chaohu from 1953 to 2012 can be estimated to increase by 5.5% for TN and 2.5% for TP each year. Fertilizer loss was estimated by the product of the collected annual fertilizer usage and the estimated loss rates from runoff. The...
fertilizer usage data were available since 1987. For the previous years (1953–1986), the product of farmland area and the fertilizer usage per unit area (hm²) was applied instead. The fractions of nitrogen (65%) and phosphate (20%) in the fertilizer were based on the average values from Tu et al. (1990) and multiple statistics yearbooks. In addition, according to Wang et al. (2010), the loss percentages of nitrogen and phosphate fertilizer were approximately 6% and 0.45% in the basin farmland, respectively. Dissolved discharge was calculated by the product of the nutrient concentration in the runoff and the runoff volumes from the basin, the latter of which was estimated by the SCS-CN (Soil Conservation Service – Curve Number) method (US Department of Agriculture, 1972). Compared to other models such as Horton infiltration equation (Xie et al., 2003), SCS-CN model is simple with only one parameter required (CN). Thus, this model has been widely used around the world (Ponce and Hawkins, 1996) including Lake Chaohu basin (Wang, 2006). The land use maps for the basin were determined from remote sensing data for the years 1979, 1990, 1995, 2000, 2005 and 2011 (see supplementary materials S2). The CN values were strongly dependent on the different types of land use and also four hydrologic soil groups based on the soil types (with respect to the rate of runoff potential and infiltration rate). Daily precipitation data for the basin from 1953 to 2012 were obtained from the China Meteorological Data Sharing Service System (http://data.cma.cn/2011jg/xw/2011jsgx/index.htm). Solid discharge from soil erosion loss was calculated by the USLE (Universal Soil Loss Equation) model, which was developed by Wischmeier and Smith (1960) and continuously modified by Wischmeier and Smith (1978) and Renard et al. (1997). It is one of the best known and the most widely used method in linking solids transport of field scale to causal and conditional erosion factors (Roose, 1977; Wang, 2006). More advanced models such as G2 model (Panagos et al., 2014) have been developed, but the increased complexity and data demand makes them not applicable here. The efficiency and simplicity make USLE model the most proper method to calculate solid discharge from soil erosion loss. More details of the SCS-CN and USLE model application to the lake basin are described in the supplementary materials (S2).

Considering that only part of the nutrient discharge from non-point resources will finally enter into the lake and thereby contribute to the loading, a lake-entering-ratio was estimated based on the 10-year loading data (1986–1995) from the literature (Zhang et al., 1997). The data from this period are used for calibration and not further used for validation. The ratio was found to have a positive relationship with the annual precipitation in the lake basin; therefore, the ratio for each year from 1953 to 2012 can be estimated accordingly. By multiplying the lake-entering-ratio, only a fraction of the fertilizer loss and dissolved and solid discharge finally entered the water body and can be considered as part of the loading. In addition, this 10-year dataset was also used to estimate the sedimentation rate for each year (see Section 2.3.2). More details can be found in the supplementary materials (S3).

2.2.3. Uncertainty of the nutrient loading

Due to data limitation, the estimated nutrient loading budget is associated with uncertainty, which is, however, difficult to address. Similar to the method applied in Bennett et al. (1999), we calculate the minimum and maximum values for different components in the nutrient budget to bracket the variations, which is considered as the uncertainty. For point sources, the parameter ranges of the TN and TP discharge per capita were ascertained from collected values (see Table S1 in the supplementary materials); hence, the variation in the municipal wastewater discharge can be obtained. For industrial and livestock and poultry breeding wastewater, however, the uncertainty was difficult to ascertain, and uniform distributions with fixed and relatively large coefficients of deviation (20%) were assumed for both components. For non-point sources, a 20% variation was also assigned for the variations of air dry and wet deposition, fertilizer loss and dissolved discharge in runoff. For solid discharge from soil erosion, however, the uncertainty mainly originated from the factors K and LS (see supplementary materials, S2), as well as the particle fraction of TN and TP in the soil in the lake basin (Table S1 in the supplementary materials).

2.3. Validation by two minimal models

Based on the estimated nutrient budget, two minimal models for nutrient mass balance were applied, and the model outputs were compared with the observations of nutrient concentrations in samples collected from Lake Chaohu in multiple studies (1974–2012; see Table S1 in supplementary materials). The details of the two models are described below.

2.3.1. Model equations

The VW model is a nutrient budget model based on mass balance in lakes (Vollenweider, 1969, 1975; Vollenweider and Dillon, 1974). The model assumes that the changes in the nutrient mass in lakes equal the increase by loading minus the loss from outflows and sedimentation. The model equation is as follows:

\[
\frac{dM_w}{dt} = L - seds \cdot M_w - outh \cdot M_w
\]  

(1)

where \(M_w\) is the mass of either nitrogen (N) or phosphorus (P) (g m⁻²). \(L\) (g m⁻² y⁻¹) is the loading per year, which represents the nutrient discharges that finally enter into the water body. \(seds\) (1 y⁻¹) is the sedimentation rate coefficient, which denotes the fraction of the nutrient that settled in the sediment. \(outh\) (1 y⁻¹) is the outflow rate, which denotes the fraction of the nutrient that moves away from the lake by outflows. The nutrient mass (g m⁻²) can be calculated by simply dividing the concentration (g m⁻³) by the water depth (D; m).

Carpenter et al. (1999) proposed a modified nutrient balance model by adding the sediment recycling process in the Vollenweider model. This model was applied for phosphorus, and the equation is as follows:

\[
\frac{dP}{dt} = L - (seds + outh) \cdot P + f(P)
\]  

(2)

where \(P\) is the phosphorus mass in the lake (g m⁻²), and \(f(P)\) is as follows:

\[
f(P) = \frac{\text{recyc} \cdot P}{P_0 + \text{half}}
\]  

(3)

where \(\text{recyc}\) (1 y⁻¹) is the recycling rate of phosphorus from the sediment, and \(\text{half}\) (g m⁻²) is the \(P\) value where recycling is half of \(\text{recyc}\). \(q(\cdot)\) is the exponent that controls the slope of \(f(P)\) when \(P\) is close to \(\text{half}\). This term suggests that the recycling is low when \(P\) is low, and after \(P\) gets higher than the certain value (\(\text{half}\)), the recycling shows a rapid elevation and approaches the maximum value of \(\text{recyc}\).

By adding the recycling term, this model shows a nonlinear feature and turns into the DyN model. This simple model is able to address many aspects during the eutrophication and restoration of lakes (Carpenter and Lathrop, 2008), and it has been used or modified as a model example to study features of regime shifts and also loading thresholds of eutrophication in lakes (Ludwig et al., 2003; Martin, 2004; Carpenter, 2005; Carpenter and Brock, 2006; Rougé et al., 2013). Because phosphorus was the limiting factor of Lake Chaohu (Xu et al., 1999), the DyN model was further applied for TP simulation, and the results were compared with those from the VW model.
2.3.2. Parameter estimation

In this study, a useful dataset from Zhang et al. (1997) was employed with 10 years (1986–1995) of observations of nutrient loading, concentration and water outflow for Lake Chaohu. The sedimentation rates (seds) for TN and TP can be calculated accordingly, both of which were found to have an exponential relation with annual precipitation. Therefore, the sedimentation rates can be obtained for each year from the precipitation (see supplementary materials, S3); outflow is estimated based on the water balance. Monthly water level data were provided by Anhui Survey and Design Institute of Water Conservancy and Hydropower (1953–2007) and Anhui Hydrological Telemetering Information system (http://yc.wswj.net/ahyc.xjb; 2007–2013). Water depth data were calculated by quotient of the lake volume and surface area, both of which can be estimated by the relation provided in Tu et al. (1990). Water inflow includes both precipitation and river inflows. Monthly inflow data were estimated by the linear relation with precipitation provided in Tu et al. (1990) from May 1987 to April 1988. Then, the river outflow can be calculated according to the total inflow, changes in water depths and estimated evaporation from Tu et al. (1990). Subsequently, with the estimated annual loading data (L), the sedimentation rates (seds) and outflow volume obtained from the water balance (outh) (Kong et al., 2014), it is feasible to let the VW model to calculate the nutrient mass (Mw) from 1953 to 2012.

In addition to L, outh and seds, three more parameters, the recyc, halm and q, need to be determined before the DyN model can be applied. However, few studies have been conducted that provide the information to estimate the values or ranges of recyc, halm and q for Lake Chaohu. Bayesian analysis and Markov Chain Monte Carlo (MCMC) sampling were implemented, which have been used in many model studies for parameter determination and uncertainty analysis (Carpenter and Lathrop, 2008; Saloranta et al., 2008; Kong et al., 2014). This method has several advantages over other methods for uncertainty analysis, e.g. basic Monte Carlo simulation. For example, Bayesian analysis and Markov Chain Monte Carlo sampling can disregard improbable parameter combinations by comparing model outputs and observations, thereby largely reduce the ascertained parameter variations and prevent the overestimation of parameter and model uncertainty (Saloranta et al., 2008; Kong et al., 2014). In addition, the model parameters are updated during the process so that the model is calibrated, which is not possible by basic Monte Carlo simulation. As the first step, the prior distributions for all five parameters were assigned as inputs for the MCMC procedure, and the model subsequently provided the posterior distributions. The probability distribution of outh was determined according to the data from 1953 to 2012. A typical normal distribution was observed (validated by the Kolmogorov–Smirnov (KS) test), with a range from 0.048 to 4.056 and mean and standard deviation values of 1.637 and 0.7343, respectively. Similarly, the probability distribution of seds was also ascertained from the estimated values from 1953 to 2012. The data show a log-normal distribution, and the log- transferred data have been proved to follow a normal distribution (validated by KS test). halm was difficult to ascertain because there is no study concerning the estimation of this parameter. Therefore, we used a uniform distribution ranging from 0.1 to 1.2, which covers the observed P mass in Lake Chaohu during 1975 to 2012 (0.258–1.043; g m$^{-2}$). q was set to obey the normal distribution with the same average value of 4, as in Carpenter and Lathrop (2008), but with a higher dispersion range of 4. recyc was estimated according to Wang et al. (2002) in which the phosphorus release was studied in Lake Chaohu sediment. The results showed that the maximum phosphorus recycling rates were 1.263 mg P m$^{-2}$ (278 K) and 5.639 mg P m$^{-2}$ (297 K) (0.461 g m$^{-2}$ y$^{-1}$ and 2.058 g m$^{-2}$ y$^{-1}$, respectively). Because the annual average temperature for the lake water is approximately 289 K, we assumed that the recycling rate coefficient should be within the range above. A normal distribution was set to this parameter with a large deviation (50% of the coefficient of variation). The other parameters, such as model error for the estimated P mass, were set the same as those in Carpenter and Lathrop (2008).

The posterior distributions of the parameters were estimated using the MCMC method with the software WinBUGS, and the differential equation was solved by the WBBdiff package for WinBUGS with the fourth-order Runge–Kutta method with one-step prediction. Fourth-order Runge–Kutta method is the most classic and most commonly applied one among the family list of Runge–Kutta method for solutions of ordinary differential equations (Hainer et al., 2008). One-step prediction means that the observation at time $t$ was used to estimate the value at time $t + 1$. Similarly to Carpenter and Lathrop (2008), $(10^8)$ samples were generated, and the first $9 \times 10^4$ values were set as the burn-in period and discarded. The last $10^4$ samples were used to determine the posterior distribution of the parameters, the uncertainty of the P mass estimation, and the probability distribution of the thresholds for regime shift.

2.3.3. Model validation

The model outputs were validated by comparing with field observations of nutrient mass in multiple years, which is listed in Table S1 in supplementary materials. Three performance criteria were adopted for the assessment of model performance, i.e. root mean squared error (RMSE; root of $\Sigma[(\text{observed} - \text{simulated})^2/\text{observed}^2]$, relative error (RE; $\Sigma(\text{observed} - \text{simulated})/\Sigma \text{observed}$) and coefficient of determination ($r^2$). These three criteria are mostly utilized for performance evaluation of ecological models (Kong et al., 2013; for a review see Arhonditsis and Brett, 2004).

2.4. Model uncertainty and estimation of the regime shift thresholds for Lake Chaohu

The uncertainty of the model output for the annual P mass can be obtained from the $10^4$ values for each year during the MCMC simulation. The interval of 2.5–97.5% of the posterior samples was considered as the range of the estimation uncertainty. The median values, as well as the uncertainty range, were compared to the observations then.

The last $10^4$ samples from the MCMC results indicated $10^4$ combinations of the parameters, which might correspond to reversible, irreversible and hysteresis types for Lake Chaohu (Carpenter and Lathrop, 2008). In addition, the thresholds can be derived from the steady states of the model equation under the condition of irreversible and hysteresis types, which are the intersections of the straight line (seds + outh)$P = L$ and the sigmoid curve $f(P)$ (Carpenter and Lathrop, 2008). The critical loadings for eutrophy (irreversible and hysterisis) and oligotrophy (only hysteresis) thresholds can be obtained by solving the following equation:

$$\text{seds + outh} = f(P)$$ (4)

This calculation was conducted in MATLAB (The Mathworks Inc., 2006). All of the threshold results should be considered as samples from the distribution of the eutrophication thresholds for Lake Chaohu. Therefore, the statistical characteristics can be addressed for both eutrophic and oligotrophic loading thresholds.
3. Results

3.1. Nutrient loading budget for the Lake Chaohu basin from 1953 to 2012

The nutrient loading budget of TN and TP for the Lake Chaohu basin from 1953 to 2012 is illustrated in Fig. 3. The total loading increased from 1953 to 2000 (Fig. 3A and B) and remained relatively stable during the last decade. The elevation of loadings from point sources was faster than from non-point sources (Fig. 3C–F). Most of the loadings originated from non-point sources, but the fractions of point sources for both TN and TP were also increasing (Fig. 3A and B). Compared to TP loading, non-point sources played a more vital role in TN loading, accounting for a higher percentage of the total loading (85.7% for TN and 77.0% for TP on average). In addition, the loading from non-point sources contributed to most of the fluctuations, especially the extreme events in 1954 and 1991 associated with high precipitation and recorded flooding in the lake basin (Fig. 3E and F).

For point sources (Fig. 3C and D), the wastewater from municipal and industry were the major components, whereas the wastewater from livestock and poultry breeding was relatively insignificant. The nutrient loading from industry started to grow rapidly from the middle of the 1960s and became dominant in recent years. Meanwhile, the loading from municipal sources was largely reduced from 2000. For non-point sources (Fig. 3E and F), a relatively smooth
loading increment from 1953 to 2012 with much greater fluctuation was observed, which was largely attributed to stochastic hydro-chemical processes (Zhang and Jørgensen, 2005). Dissolved loss from soil runoff dominated for TN, whereas particle loss from soil erosion and fertilizer loss was more important for TP in non-point loadings. Air dry and wet deposition increased rapidly but with less contribution to the lake nutrient budget.

As a validation, the estimated nutrient loading data were compared with measured values (except those from 1986 to 1995). Specifically, the annual TN and TP loadings in 1984, 1987, 1999, 2000 and 2002–2010 (on average) were collected from multiple literatures and compared with the corresponding results in this study (Table 1). The calculated nutrient loading values were consistent with the measured data. Most of the measured values were within the uncertainty band of the estimated loading values, except for TP loading in 1984, and TN loadings in 2002–2010 (on average), where the estimated values tended to overestimate the loading. This could be attributed to overestimation in different components in the loading budget, which is, however, hard to identify. Nonetheless, with respect to the performance of comparison with limited field measurements, the estimated loading budget is generally acceptable. Further validation of this budget will be conducted by model simulation in the following section.

### 3.2. Model simulation and validation

Long-term simulation by the VW model was generally consistent with the observations for both TN and TP (Fig. 4A and B), along with acceptable values of the RMSE, $RD$ and $r^2$ (Table 2). In addition, the DyN model showed a relatively better performance than the VW model for TP (Fig. 4C), with lower values of RMSE, $RE$ and higher values of $r^2$ (Table 2). The established model uncertainty band can explain the deviation of the model outputs from the observations in some years, but it fails in others. In general, for both models, the outputs tend to overestimate the lower level and underestimate the higher level of nutrient concentrations in the lake (Fig. 4D–F).

These results suggest that there is a tendency in the nutrient loading estimation to overestimate in earlier times and underestimate in recent times, which will be further discussed in Section 4.

#### 3.3. Thresholds of regime shift for Lake Chaohu

The prior and posterior frequency histogram and the best fit of the distributions for the five parameters in the DyN model produced from the MCMC method are shown in Fig. 5. The obtained parameter values sampled from their distributions can be used to calculate the distribution of the regime shift thresholds for the lake. From the last $10^4$ combinations of parameter values for the DyN model obtained from the MCMC method, 74.12% were categorized as reversible type of eutrophication, 25.44% were hysteric, and 3.50% were irreversible. The results indicate that in most cases, the eutrophication of Lake Chaohu was reversible and without alternative stable states. Therefore, the lake will most likely continuously respond to nutrient reduction (but not necessarily linearly). In contrast, under the hysteretic cases, the distributions of the forward and backward thresholds (in logarithm units) for Lake Chaohu are depicted in Fig. 6. The phosphorus loading thresholds of the regime shift were $0.81 \pm 0.372 \text{ g m}^{-2} (631.8 \pm 290.16 \text{ t y}^{-1})$ from the clear to turbid state and $0.70 \pm 0.410 \text{ g m}^{-2} (546.0 \pm 319.80 \text{ t y}^{-1})$ from the turbid to clear state with a significant difference ($N=2544$, $p<0.01$). More statistical characteristics of the thresholds are listed in Table 3.

### 4. Discussion

#### 4.1. Pro and con in the methods of nutrient loading budget estimation

In this study, a nutrient loading budget (for TN and TP) from 1953 to 2012 was built for the Lake Chaohu basin. It is generally difficult to determine the long-term nutrient loading of a basin,
primarily due to data limitations. Nonetheless, the approach taken in this study shows a feasible way to estimate the long-term nutrient loading for the Lake Chaohu basin. It resulted in valuable reference material for evaluating the ecosystem dynamics and evolution of the eutrophication in the lake during the last several decades (Xie, 2009). In addition, both the comparison with directly measured loading data in multiple years (Table 1) and validation by model simulations suggest that the nutrient loading estimation method in this study is applicable for this basin and that the results are reliable for further application. Although there are numerous models for evaluating point and non-point nutrient loadings from a basin (Zhang and Jørgensen, 2005), the method applied to a specific location should be very flexible and largely dependent on the data availability.

There are also advantages to the use of the proposed integrated model instead of observed hydrology and water quality data to evaluate nutrient loadings. One of the advantages is the ability to predict the values in the future and to test the effects of various management scenarios (Zhang and Jørgensen, 2005) or climate change (Andersen et al., 2006). For the Lake Chaohu basin, the increasing number of wastewater treatment plants and more advanced water treatment technology will result in a continuous reduction of nutrient loading from municipal and industry in the future. However, the decrease of the farming land area and increasing population will lead to a constrained high usage of fertilizer, which will further enhance the loading from non-point resources. Increasing nutrient loading could also be expected under the context of a warming climate in the future due to higher temperature, higher rainfall and the subsequently higher runoff (Jeppesen et al., 2009) and soil erosion (Lischke et al., 2014). The quantitative evaluation of the future scenarios of nutrient loading is beyond the scope of this study but very much worthwhile to be studied in the future.

The methods for nutrient loading estimation in this study are inevitably associated with disadvantages. Here, we estimate the nutrient loading per year, which is similar to studies in many other areas (Bennett et al., 1999; Hargan et al., 2011; Han et al., 2012).

### Table 2
Evaluations of both Vollenweider model (VW model) and dynamic nonlinear model (DyN model) model in the long-term simulation.

<table>
<thead>
<tr>
<th>Criteria</th>
<th>Unit</th>
<th>VW model for TN</th>
<th>VW model for TP</th>
<th>DyN model for TP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Root mean squared error (RMSE)</td>
<td>g/m²</td>
<td>1.64</td>
<td>0.16</td>
<td>0.13</td>
</tr>
<tr>
<td>Relative error (RE)</td>
<td>%</td>
<td>19.59</td>
<td>27.95</td>
<td>22.25</td>
</tr>
<tr>
<td>$r^2$</td>
<td></td>
<td>−0.59</td>
<td>0.24</td>
<td>0.34</td>
</tr>
<tr>
<td>Log(T Leutro)</td>
<td>ln(g/m²)</td>
<td>−0.31</td>
<td>−0.58</td>
<td></td>
</tr>
<tr>
<td>Log(T Lolig)</td>
<td>ln(g/m²)</td>
<td>−0.65</td>
<td>−1.546</td>
<td></td>
</tr>
<tr>
<td>Minimum</td>
<td></td>
<td>0.13</td>
<td>0.00</td>
<td>0.85</td>
</tr>
<tr>
<td>Maximum</td>
<td></td>
<td>2.34</td>
<td>2.21</td>
<td>0.79</td>
</tr>
<tr>
<td>Percentiles</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>25</td>
<td></td>
<td>0.52</td>
<td>0.39</td>
<td>−0.65</td>
</tr>
<tr>
<td>50</td>
<td></td>
<td>0.72</td>
<td>0.62</td>
<td>−0.32</td>
</tr>
<tr>
<td>75</td>
<td></td>
<td>1.00</td>
<td>0.93</td>
<td>0.01</td>
</tr>
</tbody>
</table>
The results were not considered, and their effects remain unclear. This should lead to larger deviations than the uncertainty band in both the estimation from measured loading data (Table 1) and the model simulation from observations (Fig. 4). For the components from point sources, they were generally the product of the social-economic data and the corresponding emission factors, the latter of which were obtained from very limited literature (Table S1 in the supplementary materials). In addition, for non-point sources, the variations of air dry and wet deposition, fertilizer loss and dissolved discharge in runoff also remain unclear. Therefore, it was difficult to ascertain the variation of these factors and the associated uncertainty in the estimated nutrient budget. Similar to modeling approaches in estimation of pollutant emission inventory.

The temporal resolution may be, however, too coarse for lake managers to determine the strategies for nutrient loading reduction. Seasonal patterns of the nutrient loading are also important if the fluctuations within one year are large. In fact, the non-point sources in our study can be calculated in a smaller step (per month); however, the point sources here are based on annual social-economic data. Therefore, the nutrient budget in this study is given annually and the seasonal variations are not provided. With monthly social-economic data available in the future, it is feasible to calculate the nutrient loading budget with a higher temporal resolution.

Uncertainty in the loading budget estimations can be expected. Despite the uncertainty that has already been determined (Fig. 3A and B), many potential factors that may lead to higher variations in the prior and posterior frequency histogram and the best fit of the distributions for the five parameters in the DyN model produced from the MCMC method. The parameters include (A) halm (phosphorus recycling half-saturation), (B) outh (water outflow rate), (C) q (steepness coefficient), (D) recyc (phosphorus recycling rate from sediment), (E) seds (sedimentation rate coefficient).

Fig. 5. The temporal resolution may be, however, too coarse for lake managers to determine the strategies for nutrient loading reduction. Seasonal patterns of the nutrient loading are also important if the fluctuations within one year are large. In fact, the non-point sources in our study can be calculated in a smaller step (per month); however, the point sources here are based on annual social-economic data. Therefore, the nutrient budget in this study is given annually and the seasonal variations are not provided. With monthly social-economic data available in the future, it is feasible to calculate the nutrient loading budget with a higher temporal resolution.

Uncertainty in the loading budget estimations can be expected. Despite the uncertainty that has already been determined (Fig. 3A and B), many potential factors that may lead to higher variations in
However, the long-term average data estimated from this study well with the results in this study (76.2% for TN and 73.9% for TP), which agreed relatively with the total amount in 1988 (Tu et al., 1990), which can be attributed to both the additional sediment recycling process in extreme loading values (such as 1954 and 1991), but the effect could be weakened by a higher water outflow flushing rate. In addition, fluctuations in the non-point sources from 1953 to 2012 account for most of the fluctuation observed in total loading data (Fig. 3A and B), whereas the loading from point sources was gradually increasing without much fluctuation. Similar to the results in Zhang and Jørgensen (2005), these fluctuations were largely attributed to the stochastic hydro-chemical processes. Precipitation has a large effect on the outputs from the SCS-CN and USLE models because the volume of rain usually determines the volume of runoff and soil erosion. Flooding events usually result in extreme loading values (such as 1954 and 1991), but the effect could be weakened by a higher water outflow flushing rate.

The estimated nutrient loadings from non-point sources (Fig. 3E and F) suggested that for TN, the dissolved phase was relatively more important, while the particle phase was more dominant for TP. According to Wang (2006), the fraction of the TN and TP loss from soil erosion out of the total amount in soil were empirically assigned as 50% and 85%. In reality, the processes to generate nutrient loadings from non-point sources are complicated (Shan et al., 2001; Wang, 2006). Leaching and erosion are the main mechanisms of phosphorus loss from agriculture soil, and multiple studies have suggested that TP loss was mostly in the particle phase (Sharpley et al., 1994; Shan et al., 2001; Zhu, 2004). In contrast, TN loss from non-point sources in two small watersheds in China was mainly in the dissolved phase (68% and 83%) (Zhu, 2004; Wang, 2006), which may be attributed to the chemical properties of nitrate (lower sorption ability to particles and higher dissolvability in water).

4.2. Features of the nutrient loading budget

Loadings from point source accounted for 15.3% (TN) and 23% (TP) on average during 1953–2012, which is relatively lower than expectation. However, the amount of point sources and fraction in the total loading were actually increasing during this period (Fig. 3A and B), due to the rapid increasing of population and economic development. From about 1995, the increasing rates of nutrient loading from the point sources were largely reduced. This is largely attributed to multiple environmental protection projects implemented in Lake Chaohu since then (State Environment Protection Administration of China, 2000). On the other hand, loadings from non-point sources accounted for 74% (TN) and 68% (TP) of the total amount in 1988 (Tu et al., 1990), which agreed relatively well with the results in this study (76.2% for TN and 73.9% for TP). However, the long-term average data estimated from this study suggested that the non-point fractions for TN and TP were significantly higher (85.7% for TN and 77.0% for TP), indicating that nutrient loading from non-point sources was even more important than expectation. Similar to other regions with both agriculture and urban activities (Carpenter et al., 1998), non-point sources in the basin have replaced point sources as the driver of eutrophication in Lake Chaohu. In addition, fluctuations in the non-point sources from 1953 to 2012 account for most of the fluctuation observed in total loading data (Fig. 3A and B), whereas the loading from point sources was gradually increasing without much fluctuation. Similar to the results in Zhang and Jørgensen (2005), these fluctuations were largely attributed to the stochastic hydro-chemical processes. Precipitation has a large effect on the outputs from the SCS-CN and USLE models because the volume of rain usually determines the volume of runoff and soil erosion. Flooding events usually result in extreme loading values (such as 1954 and 1991), but the effect could be weakened by a higher water outflow flushing rate.

4.3. Comparison of the two minimal models

In the VW model, we assumed that the changes in the nutrient mass in lakes equal the increase by loading minus the loss from outflows and sedimentation. Here, the recycling process, or so-called ‘internal nutrient loading’, is not considered. However, this could also lead to immeasurable deviations from reality, and more data are required to evaluate the uncertainty in these factors in the future. In addition, in the long-term estimation, factors including the nutrient discharge per capita for municipal wastewater discharge and the nutrient concentrations in the wastewater for Livestock and poultry breeding wastewater discharge, are supposed to change over time, but they are constant during most of the time. In addition, for TN and TP discharge per 10 thousand RMB of industrial production, only data in 1984 and 1999 were available. Thus, the value of 1984 was applied for the year before 1984, and linearly interpolated between 1984 and 1999. The value of 1999 was applied for the year afterwards (see Table S1 in the supplementary materials). Similar problems also occurred with non-point sources, e.g., the particle nitrogen and phosphorus concentrations in soil were available only recently, and they were fixed at these values throughout the long-term estimation (Table S1 in supplementary materials). These assumptions are simplification for loading estimation, which may lead to overestimation the lower values in earlier times, underestimate the higher values in recent times and increasing uncertainty in the estimation (Fig. 4D–F). However, their temporal variations were considered as not very high so that we ignored in this study. Additionally, in the USLE model for soil erosion estimation, factors such as K and LS were ascertained by the spatial average values from the literature (Wang, 2006). Although spatial variations were considered in the uncertainty analysis, temporary variations of these factors from 1953 to 2012 were considered insignificant and ignored.
calibration period (after 1995). The model outputs from the DyN model were generally more consistent with the observations from the VW model after 1995 (Fig. 4B and C), which is the major reason for the improvement in the model performance (Table 2). Second, the prior distributions of the five parameters are assumed in DyN model according to field observations or empirical value ranges, in which the uncertainty about the parameters is sufficiently considered (e.g. q and recyc). Bayesian–MCMC method will subsequently allow the DyN model to fit the observations with different parameter combinations and find the best one. Among the parameters, two parameters (outh and seds) were shared in both VW and DyN models. However, outh was significantly narrowed, while seds shifted to higher values in DyN model simulation with MCMC procedure (Fig. 5). The recycling from sediment brought more phosphorus to water volume so that higher sedimentation rate coefficient (q) and water outflow rate (outh) remained similar for the three types of eutrophication. However, seds was relatively high compared to the other cases (Carpenter and Lathrop, 2008) due to a sluice for irrigation purposes (built in 1963), which has strongly reduced the hydro-dynamics of the lake and largely enhanced the sedimentation (Zhang and Lu, 1986). On the other hand, the reversible type was associated with the parameter combinations of the lowest recycling rate (recyc) and steepness (q) and also the highest recycling half-saturation coefficient (halm), which led to a better fit to the observations so that they are more favored by the MCMC method and are considered as more consistent with reality. In fact, these results are supported by the field data and observations. The surface layer of the Lake Chaohu sediment is dominated by silt (2–20 μm) or sand (20–2000 μm) (approximately 56.8% and 36.5%, respectively; unpublished data), indicating that sediment is not very prone to resuspension (Janse, 2005). Hence, the phosphorus sediment recycling of the Lake Chaohu ecosystem may not be as strong as expected in literature (Tu et al., 1990). The higher sedimentation than recycling of phosphorus is also consistent with the observations that sediment is a major sink for phosphate in Lake Chaohu (Yang et al., 2013). Moreover, the higher halm and lower q indicated a stronger binding capacity of the lake sediment to the phosphorus, which is supported by the high content of Fe and Al in the sediment (3.2% of Fe and 7.2% of Al; ash free dry weight) (Tu et al., 1990). As a result, the relatively higher values of sedimentation (sed) and water outflow (outh) contributed to a very high slope line of the linear function (left in Eq. (4)), whereas high halm and low recyc and q values led to a less steep sigmoid curve (right term in Eq. (4)). These two reasons may contribute to a higher possibility of the reversible type of eutrophication in which the system responds continuously to the changes in nutrient loading.

Despite the dominant reversible type determined in this study, it is still too early to make a conclusion that the lake does not have hysteresis and the response to the nutrient loading variation in Lake Chaohu is continuously. Van Nes and Scheffer (2005) showed that the chance of large-scale shifts between alternative stable states can be reduced due to spatial heterogeneity, and the large Lake Chaohu was considered as a homogeneous waterbody in this study. In addition, the model applied in this study is so simple that the extensively studied interactions and feedback between phytoplankton and submerged macrophytes (Scheffer et al., 1993; Scheffer and Jeppesen, 2007) are neglected, which should strongly affect the phosphorus cycling in the lake and be significant for the Lake Chaohu ecosystem (Xie, 2009; Kong et al., 2013). This simplification could produce misleading results, which should be considered with caution in further applications.

4.4. Lake Chaohu as a shallow lake: reversible or hysteretic?

Among the three types of eutrophication categorized by the DyN model, the probability of irreversible cases was lowest (3.50%), suggesting that eutrophication of Lake Chaohu is not likely an irreversible case. The eutrophication of the lake appears to be a reversible (74.12%) rather than a hysteretic (25.53%) case, which is contrary to the expectation that the hysteretic type was the most likely scenario for any shallow lake (Carpenter and Lathrop, 2008). Parameter value combinations are decisive for determining to which eutrophication type the system belongs (Table 4). The sedimentation rate coefficient (sed) and water outflow rate (outh) were the most likely scenario for any shallow lake (Carpenter and Lathrop, 2008). Parameter value combinations are decisive for determining to which eutrophication type the system belongs (Table 4). The sedimentation rate coefficient (sed) and water outflow rate (outh) were

4.5. Thresholds for Lake Chaohu eutrophication: uncertainty and implication

In spite of the relatively dominant reversible type, the possibilities of the lake ecosystem to exhibit regime shift with thresholds was still large (25.53%). However, these two phosphorus loading thresholds (forward and backward) determined by the model were
similar (from the clear to turbid state (631.8 ± 290.16 t·y⁻¹) and from the turbid to clear state (546.0 ± 319.80 t·y⁻¹)), indicating that even though there is hysteresis in the lake, the range of phosphorus concentration where an alternative stable state exists was narrow. Moreover, the thresholds of the lake regime shift were determined with large variations (Fig. 6), suggesting that the uncertainty range of the two thresholds was greatly overlapped. The large variations of the estimated TP thresholds, particularly from turbid to clear water, are largely attributed to the model uncertainty. Generally, the uncertainty of the model included both inherent variability and true uncertainty of the model estimates (McKone, 1996). The model we applied in this study is a simple nutrient budget model, thus inherent uncertainty is inevitably large. In addition, multiple sources for the variations of all of the parameters in the DyN model due to fluctuations in environment conditions, including stochastic drivers such as climate events (for outh), biogeochemical processes such as nutrient sedimentation and recycling (for recyc and seds) (Carpenter and Lathrop, 2008), and characteristics of the lake sediment (phosphorus binding capacity that determines halm and q), may contribute to inevitable true uncertainty of the model estimates. Scheffer and van Nes (2007) also show that the nutrient loading thresholds largely depend on factors such as lake depth and climate, thereby changing over the time with the large variations in the external environment. Therefore, the high standard deviations of the estimated thresholds in this study are reasonable.

Even though the evidence for regime shift for Lake Chaohu is not as strong as expected, the result of the model study presented here has important implications because (1) phosphorus loading was fluctuating around the threshold from the clear to turbid state (631.8 ± 290.16 t·y⁻¹) until the late 1970s, and the loading was always higher than the upper bound of the threshold (about 920 t·y⁻¹) thereby, suggesting that the lake transitioned to a turbid state during late 1970s; and (2) according to the estimated TP loading in 2013 (1548.8 t), approximately two-thirds of the current TP loading must be reduced for a significant improvement in water quality by crossing the backward threshold (546.0 ± 319.80 t·y⁻¹).

Under any circumstances, reduction of the nutrient loading will always benefit the lake ecosystem (Smith et al., 1999), keeping it away from the threshold and reducing the possibility of deteriorated water quality and ecological risk (Jeppesen et al., 2005; Carpenter and Lathrop, 2008). In addition, even if the lake system is reversible without alternative stable states, the response of TP concentration in water to phosphorus loading in the basin will still be rather discontinuous (sigmoidal curve), indicating a large improvement of water quality in certain critical range of the loading. This is similar to the response of vegetation coverage to the nutrient loading when alternative stable state did not arise, based on the classic vegetation–turbidity model study in Scheffer (2004).

However, according to the nutrient budget presented in Section 3.1, on average, approximately 85% of the TN and 77% of the TP loading was from non-point sources, which are difficult to control in lake management. Even if fertilizer usage in the basin could be completely forbidden (which is not economically profitable), the residues in the farmland would remain for a long time, and the non-point loading will still be substantial. Carpenter (2005) notes that soil phosphorus dynamics can be more important than sediment internal loading in lake eutrophication control. Therefore, soil management could be the core issue in loading reduction for the agricultural area of the Lake Chaohu basin. Other restoration methods, such as water quality rehabilitation with rainwater utilization (Wu and Chau, 2006), are also interesting for lake management. This method can significantly reduce nutrient loading with low investment, while the conflict between drainage and rainwater utilization can be resolved with appropriate drainage plan scheme (Wu and Chau, 2006). In addition, restoration methods beyond nutrient reduction should be considered for the lake, especially the water level regulation (Kong et al., 2013). It is argued that the regulated seasonal water level was a primary factor in macrophyte extinction (Xie, 2009; Zhang et al., 2014). The management of the water level will enhance the restoration of macrophytes, allowing them to store large amounts of nutrients. It is suggested that an integrated environment impact assessment (Zhao et al., 2006) should be implemented for Lake Chaohu water level regulation strategy selection. Moreover, the long-term effect of the water level regulation on the lake ecosystem should be evaluated by more comprehensive ecological models that consider a full aquatic food-web, while also including the effects of macrophytes on the lake, such as the well-evaluated ecosystem model PCLake (Janse, 2005). It is also interesting to compare the outcomes from the complex model PCLake with the results obtained in this study with two minimal models (Mooij et al., 2009). Such comparison will help in developing an integrated view on the behavior of the Lake Chaohu ecosystem.

5. Conclusion

In this study, we use an integrated empirical and mechanistic modeling approach to build a long-term nutrient loading budget from 1953 to 2012 for eutrophic Lake Chaohu in China, which agreed relatively well with limited observations. On average, 85% of the TN and 77% of the TP loading originated from non-point sources, which pointed to the importance of nutrient
Acknowledgments

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at http://dx.doi.org/10.1016/j.ecolind.2014.12.005.

References


Vollenweider, R., Dillon, P., 1974. The Applications of the Phosphorus Loading Concept Eutrophication Research. NRCC.


