Comparison of spatiotemporal carbon, nitrogen and phosphorus burial in two plateau lacustrine sediments: Implication for N and P control

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Comparison of spatiotemporal carbon, nitrogen and phosphorus burial in two plateau lacustrine sediments: Implication for N and P control

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**Abstract:** The long-term accumulation, burial and release of nutrients, such as carbon (C), nitrogen (N) and phosphorus (P) in lacustrine sediments are responsible for the global lake eutrophication. Interpretation of the spatiotemporal sedimentary record of nutrients (C, N and P) in contrasting trophic level of lakes is helpful for understanding the evolutionary process of water eutrophication. Based on the radiochronology of $^{210}$Pb$_{ex}$ and $^{137}$Cs, a comparative study of spatial and temporal concentrations, burial of total organic carbon (TOC), total nitrogen (TN) and total phosphorus (TP), the sources of organic matter were conducted using sediment cores from two plateau lakes Dianchi (DC) and Fuxian (FX) of SW China. Results showed that concentrations and burial of C, N and P in sediments of DC, a shallow hypertrophic lake with the maximum depth of 5.8 m, were both higher than those in FX, an oligotrophic deep lake with the maximum depth of 155.0 m. For both lakes the molar ratio of TOC/TN increased in the sediments moving from north to south. The values of TOC/TN molar ratios increased over time in DC and was higher than in FX. The extremely high values of TOC/TN appeared in the central and southern parts of FX, indicating the impacts of accumulation effect and sediment focusing in the deeper region and indirect supplement from the Lake Xingyun (XY), an adjoining lake connected with FX via the Gehe River. Time-integrated sources identification in DC indicated the contribution of allochthonsous sources was dominant over the past few decades, which contributed to the increased trophic level of the lake. The comparison of relationships of carbon accumulation rates (CAR), nitrogen accumulation rates (NAR) and phosphorous accumulation rates (PAR), the ratios of N/P and the utilizations of N and P fertilizer between DC and FX implied that both of N and P inputs should be limited for reducing the trophic level, but N control was predominant in comparison with P for both lakes. The results indicated that caution is required in plateau lakes to limit transition from oligotrophic to eutrophic in these lakes.

**Key words:** Accumulation of TOC, TN and TP, Organic matter sources, N and/or P limitation, Lacustrine sediments, Plateau lakes
1. Introduction

To date, eutrophication is still an environmental issue in global lakes (Conley et al., 2009; Schindler et al., 2012; Bajard et al., 2018; Caen et al., 2019). Nutrients, such as total carbon (C), nitrogen (N) and phosphorus (P), are responsible for lake eutrophication through the pathway of accumulation, release and reduction in sediments (Eid et al., 2012; Sanders et al., 2016; Klump et al., 2020). With the changes in climate factors (temperature and precipitation) and human activities (economic development and urbanization) over the past century, C, N and P burial and storage have been found to be increased in lacustrine sediments (Kastowski et al., 2011; Kortelainen et al., 2013; Dietz et al., 2015; Heathcote et al., 2015; Huang et al., 2018). The burial of nutrients in sediments, especially allochthonous organic carbon (OC) sequestration, accelerated the eutrophication in those lakes in North America (Anderson et al., 2014), Europe (Ferland et al., 2012), Australia (Sanders et al., 2016), and China (Zhang et al., 2017a). Therefore, studies on the concentrations and stores of nutrients and sources of OC in lacustrine sediments can help to understand the evolutionary processes and formation mechanisms of lake eutrophication (Wolfe et al., 2013; Hayakawa et al., 2015; Barik et al., 2019; Horppila, 2019).

China has more than 2700 lakes with surface areas greater than 1 km², approximately 70% of which are located in high plateau regions (Wang and Dou, 1998). Plateau lakes can not only provide many functions, such as drinking, irrigation, fishery, and tourism (Lin et al., 2017; Moser et al., 2019), but also act as the important headwaters of the large and long rivers. However, due to population growth coupled with the industrial and agricultural development around these plateau lakes, they have undergone eutrophication similar to lakes elsewhere (Hillman et al., 2014; Dai et al., 2017; Huang et al., 2018). Furthermore, it is difficult to recover a good state in a short period of time because of their relatively vulnerable ecosystems and simple food webs (Wischnewski et al., 2011; Ren et al., 2017). Therefore, the study of plateau lakes has attracted much more scientific attention in recent years (Sitoki et al., 2010; Otu et al., 2011; Jones et al., 2012; Guimarães et al., 2014; Alexandrine et al., 2019).

The Yunnan-Guizhou Plateau (YGP) is located in Southwestern China. With the impact of new tectonic movement, several faulted-type lakes were formed in this region (Wang and Dou, 1998). Of these lakes, Lake Dianchi (DC) and Lake Fuxian (FX) are the two important (largest and deepest) plateau lakes in this region. They are both referred to the “Pearl of the Highland” because of their picturesque scenery. However, different levels of eutrophication in these two lakes also occurred due to the increase of nutrient loadings (Liu et al., 2011; Liu et al., 2014; Tao et al., 2019). In order to investigate the
formation mechanism of water eutrophication, numerous studies were conducted in the YGP of China, including the DC and FX (Liu et al., 2012b). For instance, Zhang et al. (2019a) reported that natural factors, such as precipitation and temperature, played a dominant role in lake environmental changes. Anthropological activity, such as land use change (Zhang et al., 2011; Yang et al., 2016), fish introduction (Liu et al., 2009; Liu et al., 2012a; Ye et al., 2015), watershed domestic and industrial effluents (Wang et al., 2015), and socioeconomic factors (Liu et al., 2011; Huang et al., 2014), have provided higher contributions to the environmental changes in these two lakes. Based on the multi-proxies’ record of C, N, polycyclic aromatic hydrocarbons and lipid biomarkers in lake sediments, the enhanced environmental deterioration occurred in DC and FX after the 1980s, although the trophic level of the latter (oligotrophic) was lower than that of the former (hypereutrophic) (Huang et al., 2018; Tao et al., 2019; Zhang et al., 2019b; He et al., 2020). However, a comparative study of spatiotemporal concentrations, burial and sources of total organic carbon, nitrogen and phosphorus in sediments of the two plateau lakes is scarce. Therefore, two plateau lakes located in the YGP of China with contrasting environmental conditions were selected to investigate the impact of N and P limitation on lake eutrophication.

The main objectives of this study were to: 1) compare the spatial and temporal variability of TOC, TN, and TP and carbon accumulation rates (CAR), nitrogen accumulation rates (NAR) and phosphorous accumulation rates (PAR) in sediments, 2) identify the impacts of natural factors (temperature and precipitation) and human activity (population, N and P fertilizer utilization) on the spatiotemporal variations in autochthonous and allochthonous contributors to increased CAR, NAR and PAR, and 3) investigate the priority of N and/or P controlling in the two different trophic level of plateau lakes.

2. Materials and methods

2.1 Site description

The study was carried out in DC and FX, situated in the YGP of SW China (Fig. 1a and b). Lake DC (24°40'-25°02'N, 102°36'-102°57'E), a hyper-eutrophic lake, is 41.2 km long and 13.0 km wide, with a surface area of 297.9 km², a mean depth of 2.9 m, and a water residence time of 3.9 years (Fig. 1c) (Wang and Dou, 1998). Under a typical southwestern monsoon climate, it has a mean temperature of 14.4 °C and a mean annual rainfall of 1036 mm, mainly from May to October. It is a semi-closed lake and nearly 20 rivers flow into the lake, with only the south-west Haikouzi River as its outlet (Fig.1b). Kunming City, the capital of Yunnan Province, is located in the northern shore of the lake (Fig.1b). With
increasing inputs of municipal wastewater over the past few decades, eutrophication is the major environmental problem now (Huang et al., 2018). The levels of N and P concentrations in water increased from 1.15 and 0.132 mg/L to 2.53 and 0.173 mg/L, respectively, from 1980 to 2010 (Zhou et al., 2016).

Lake FX (24°21′-24°38′N, 102°49′-57′102°E), an oligotrophic lake, has a surface area of 212.0 km², a mean depth of 89.6 m, and a water residence time of 167 years (Fig. 1d) (Wang et al., 2018). The lake has the same climate type as DC as mentioned above, the average annual temperature of FX is 15.6 °C, and annual average precipitation is 879 mm. More than twenty short rivers flow into the lake, and the only outflow to the lower catchment is the Haikou River which is located in the mid-eastern shore of the lake (Fig 1b). The Gehe River on the southwestern shore of the lake connects the FX with the adjoining Lake Xingyun (XY), a relatively small, shallow eutrophic lake (Fig. 1d). Chengjiang County, is located in the northern part of the lake (Fig. 1b). The concentration of TN in the surface water of FX increased from 0.1 to 0.223 mg/L, and TP increased from 0.005 to 0.013 mg/L between 1981 and 2005, respectively (Pan et al., 2008). In 2012, the concentrations of TN and TP in the water column (0-50 m) were 0.16 and 0.009 mg/L, respectively (Zhang et al., 2019b). Although it is in an oligotrophic state now, an upward trend of trophic states also occurred in FX (Zhang et al., 2015; He et al., 2020).

2.2 Sediments sampling

Using a 6 cm inner-diameter gravity corer, one sediment core was taken from each of seven sites on each of the lakes (the length for most of cores ranged from 20-30 cm, with an exception of 13 cm in FX6) in each lake were collected in early-October of 2007 at different water depths for DC and FX (named DC1-7 and FX1-7, respectively) (Fig. 1c and d). The moist sediment cores were carefully extruded from the core barrel in the laboratory and sectioned at 0.5 cm equal intervals from top to bottom using a self-designed cutting machine. The slices were oven-dried at 60 °C for approximately 72 h, weighed to obtain the dry bulk density, and then pulverized with a manual agate mortar and pestle to pass through a 2-mm plastic sieve for radionuclide dating and nutrients (TOC, TN and TP) analysis.

2.3 Laboratory analysis

Each dried sediment section was packed into an airtight plastic container, which was sealed and incubated for about 20 days to allow the $^{238}$U decay chain to reach radioactive equilibrium. The $^{210}$Pb, $^{226}$Ra ($^{214}$Pb), and $^{137}$Cs activities were then determined simultaneously using a high-resolution HPGe γ-spectrometry system (GWL-120-15, ORTEC, USA), which had a 62% relative detection efficiency, using a counting time of 40,000 s. The resolution of the spectrometer was 2.25 keV for 1.33 MeV γ-rays from
The γ-ray emissions at 661.6 keV were used to obtain the activity of $^{137}$Cs, whereas $^{210}$Pb was determined from its γ-ray emissions at 46.5 keV, and $^{226}$Ra by measuring the γ-ray emissions from the short-lived daughter $^{214}$Pb at 351.9 keV. The unsupported $^{210}$Pb ($^{210}$Pb$_{ex}$) was calculated by subtracting the $^{226}$Ra-supported $^{210}$Pb activities ($^{214}$Pb) from the total $^{210}$Pb activities. The detector efficiency was derived by analysis of standard samples of soil of known activity and calibrated by the International Atomic Energy Agency (IAEA) and the Institute of Atomic Energy, Chinese Academy of Science. The relative error for this method was less than 5%.

TOC, TN, TP concentrations were measured using a concentrated sulfuric acid-potassium dichromate digestion method (Lu, 2000) followed by quantification with a UV-3600 spectrophotometer (Shimadzu Corp., Japan). The analytical data was checked using quality assurance and quality control (QA/QC), including analysis of reagent blanks, duplicate samples and stream sediment standard reference materials (GSD-9 and GSD-4; Chinese Academy of Geological Sciences) for each batch of samples. The differences of the concentrations between the determined and certified values were less than 5%, and the analytical precision for replicate samples was within ± 0.1%.

2.4 Data processing

2.4.1 Sediment chronology and sedimentation rates

The constant rate of supply (CRS) model of $^{210}$Pb$_{ex}$ (Appleby, 2001) was used to calculate the sediment chronology. The detailed formulas can be described as follows:

$$t = \frac{1}{\lambda} \ln \left( \frac{A_h}{A_0} \right)$$

(1)

where $t$ is the age in years, $\lambda$ is the decay constant for $^{210}$Pb (0.03114 a$^{-1}$), $A_0$ (Bq m$^{-2}$) is the inventory of $^{210}$Pb$_{ex}$ in the sediment core, and $A_h$ (Bq m$^{-2}$) is inventory of $^{210}$Pb$_{ex}$ in each depth increment of the sediment. Based on the sedimentary chronology, the sediment accumulation rates (SARs) can be obtained as:

$$SARs = \frac{\partial Md}{\partial t}$$

(2)

where $Md$ is the cumulative mass depth, it is equal to the sediment mass per unit area above a certain depth, and $t$ is the interval time between a certain layer and the initial layer.

2.4.2 C, N and P burial

The accumulation rates of carbon, nitrogen and phosphorus were estimated by the following
equation (Huang et al., 2017):

\[
\text{CAR}(\text{NAR, PAR}) = \text{SARs} \times C(\text{TOC, TN, TP}) \times 10
\]  

(3)

where \(\text{CAR} (\text{NAR, PAR})\) is the carbon, nitrogen and phosphorus accumulation rates (g m\(^{-2}\) a\(^{-1}\)), \(\text{SARs}\) is sediment accumulation rates (g cm\(^{-2}\) a\(^{-1}\)), and \(C (\text{TOC, TN, TP})\) is the concentration of carbon, nitrogen and phosphorus (mg g\(^{-1}\)).

Based on the topographic maps and digital elevation model (DEM, downloaded from http://www.gscloud.cn/) in these two lakes, isobaths were extracted by a geographical information system (GIS) technique (Fig. 1c and d). Combined with the locations of sediment cores, the numbers of bathymetrical regions of the two lakes were divided into 4 for DC and 6 for FX, respectively (Fig. 1c and d). The detailed locations of sediment cores and their area (km\(^2\)) at different water depths (m) of each lake are shown in Table 1. Thus, one or two sediment cores were assigned in each bathymetrical region of the two lakes (Table 1). Here, we assumed that the average \(\text{CAR} (\text{NAR, PAR})\) was similar at the same depth for each lake, and hence, sedimentary fluxes (SFs) at each depth in lake can be obtained on the basis of the average \(\text{CAR} (\text{NAR, PAR})\) (g m\(^{-2}\) a\(^{-1}\)) multiplied by area \(\text{S}_{\text{area}}\) (km\(^2\)) and sediment chronology \(\text{S}_{\text{c}}\) (a). The detailed calculation formula can be expressed as the following:

\[
\text{SF}_{s} = \text{CAR}_{s} (\text{NAR}_{s}, \text{PAR}_{s}) \times \text{S}_{\text{area}} \times \text{S}_{\text{c}}
\]  

(4)

where \(\text{SF}_{s}\) is the total reserves of C, N and P (t), \(\text{CAR}_{s} (\text{NAR}_{s}, \text{PAR}_{s})\) is the average \(\text{CAR} (\text{NAR, PAR})\) at each depth (g m\(^{-2}\) a\(^{-1}\)), \(\text{S}_{\text{area}}\) is the area at each depth interval (km\(^2\)), \(\text{S}_{\text{c}}\) is sediment chronology (yr). If two sediment cores (i.e., DC2 and DC7 in DC, FX1 and FX2 in FX, respectively) were located in the same section, the average a-\(\text{SARs}\) of the two cores were selected to calculate the SFs.

Accordingly, the total sedimentary fluxes in each lake can be estimated by the following equations:

\[
\text{TSF}_{s} = \sum_{i=1}^{i} \text{SF}_{s}
\]  

(5)

where \(i\) is the numbers of isobaths (m), the unit of ton (t) is for \(\text{TSF}_{s}\).

2.4.3 Source identification model

Based on the ratio of C to N in sediments, the binary model was proposed by Qian et al. (1997). This model has been widely used to quantify the contributions of autochthonous and allochthonous sources of lacustrine sediments (Gui et al., 2012; Wu et al., 2016; Zhang et al., 2018). The values of autochthonous organic matter \(C_{\text{auto}}\) and allochthonous organic matter \(C_{\text{allo}}\) can be calculated from the...
following equations:

\[ C_{\text{auto}} = \frac{R_{\text{auto}} \times (TOC - R_{\text{allo}} \times TN)}{R_{\text{auto}} - R_{\text{allo}}} \]  \hspace{1cm} (6)

\[ C_{\text{allo}} = \frac{R_{\text{allo}} \times (TOC - R_{\text{auto}} \times TN)}{R_{\text{allo}} - R_{\text{auto}}} \]  \hspace{1cm} (7)

where \( R_{\text{auto}} \) is the proportion of autochthonous TOC, and \( R_{\text{allo}} \) is the allochthonous TOC, the unit of \( mg \ g^{-1} \) is for \( C_{\text{auto}} \) and \( C_{\text{allo}} \).

2.5 Supplementary data

The data of TOC, TN and TP in FX were collected from our previous study (Wang et al., 2018). The meteorological data were obtained from the Kunming and Yuxi Meteorological station, which provides the data from 1974-2014. The data of population were collected from the Yunnan Provincial Bureau of Statistics (http://www.stats.yn.gov.cn). The N and P fertilizer utilization data from 1991-2012 were collected from Huang et al. (2014) and He et al. (2020). The data on vertical profiles of water temperature (WT), pH, and dissolve oxygen (DO) between FX and DC were collected from Wang et al. (2017) and Zhang et al. (2017b). The vertical distribution of TN and TP in water column in Lake Fuxian were collected from Sakamoto et al. (2002). Annual budget of nitrogen, phosphorous and water in Xingyun Lake and Fuxian Lake were collected from Sakamoto et al. (2002).

2.6 Uncertainty analyses

To further investigate the creditability of the measurements in this study, we compared the results with the previous studies of TOC, TN, TP and TOC/TN in sediments from Fuxian Lake (Table 2). Significant heterogeneity existed in these parameters of TOC, TN, TP and TOC/TN in water and sediment from Lake Fuxian due to different environmental conditions. On the whole, most of our data fell within the range of all parameters, which verified that our results were very reliable.

3. Results

3.1 SARs in DC and FX

Based on our previous studies (Wang, 2011; Wang et al., 2018), the vertical profiles of \(^{210}\)Pb\text{ex} and \(^{137}\)Cs, the chronology was corrected using the radiometric dating method and SARs (Eqs. 1 and 2) in each sediment core of DC and FX are shown in Figs. S1-6. Comparisons of the average SARs in multi-sediment cores of DC and FX are shown in Fig. 2a and b. The SARs in DC ranged from 0.007 to 1.162.
Compared with DC, the average SARs in FX was slightly higher, with an average 0.105 ± 0.029 g cm$^{-2}$ a$^{-1}$, but ranged from 0.006 to 0.575 g cm$^{-2}$ a$^{-1}$. For DC, the higher SARs was located in the surrounding region of the lake, whereas the lower value of SARs in DC4 was appeared in the central position of the lake (Fig. 2a). Unlike DC, an increased trend was observed from north to south of FX, with a reception of FX6 (Fig. 2b). The highest value was up to 0.138 ± 0.071 g cm$^{-2}$ a$^{-1}$ in FX7 which was located in the southern region of Fuxian Lake (Fig. 2b).

### 3.2 Spatial and temporal variations of limnological parameters of sediments in DC and FX

#### 3.2.1 Variations of nutrient parameters in DC

On the whole, the concentration of TOC in DC showed an upward trend from the shallow to deep region of the lake, and the higher values of TOC appeared in the sites of DC4 and 5 (Fig. 2c), with an average of 64.34 and 71.92 mg g$^{-1}$, respectively. TN and TP showed upward trends from north to south, with an exception of DC7 in TN (Figs. 2e and g). The average values of TOC/TN ranged from 10.59±4.50 to 21.01±6.59 (Fig. 2i), with a slowly upward trend. For DC, the lowest burial rates of C, N and P (CAR: 28.35 ±14.31 g m$^{-2}$ a$^{-1}$, NAR: 2.56±1.23 and PAR: 0.84±0.39 g m$^{-2}$ a$^{-1}$, respectively) in sediments were located in the central position of the lake (DC4), which is in accordance with the changes in SARs (Fig. 2a). The upward trends from bottom to top over the past ~150 years are found in each nutrients parameters (Figs. 3a-c, 4a and 5a-c), indicating the increasing loads of nutrients in the lake.

#### 3.2.2 Variations of nutrient parameters in FX

To compare the differentiation of nutrients parameters between DC and FX, the previously published data of nutrients parameters (TOC, TN and TP) in FX were used again (Wang et al., 2018). Spatially, the higher concentration of TOC in FX occurred in the central and southern part of the lake, with the sites of FX3 and 7 (Fig. 2d), whereas TN kept a relatively stable tendency in the different sediment cores (Fig. 2f). For TP in FX, a relatively stable tendency was found, with an abnormal value of FX6 (Fig. 2h). The average values of TOC/TN ranged from 12.56±2.12 to 37.33±8.37 with an extreme value of 43.21±6.62 in FX3 (Fig. 2j). C, N and P burial (especially for PAR) in sediments of FX increased gradually from north to south (Fig. 3), which indicated that Lake XY was an important contributor of increased SARs and nutrients burial in FX (Wang et al., 2018). Based on the Eq. 3, the spatial and temporal CAR, NAR and PAR in DC and FX were calculated in the study (Figs. 5 and 6). Compared with the previous results of CAR in Lake Fuxian in 1980 and 2000, the average CAR (73.4 g m$^{-2}$ a$^{-1}$) in this study was higher than 61.7 g m$^{-2}$ a$^{-1}$ (169 mg m$^{-2}$ d$^{-1}$) (Nanjing institute of Geography and Limnology
China, 1990), but lower than 151.5 g m$^{-2}$ a$^{-1}$ (416 mg m$^{-2}$ d$^{-1}$ in F2 and 414 mg m$^{-2}$ d$^{-1}$ in F7) (Hayakawa et al., 2002).

Temporally, the vertical distribution of TOC and TP showed that the nutrient concentrations increased from the northern to the southern except for TP with an extreme value in FX6 (Fig. 3), although a relatively higher value of TOC occurred in FX3 near the maximum depth of the lake (Fig. 3d). Similar to the spatial distribution of TOC/TN in FX, two relatively higher values appeared in the central and southern part of the lake (Fig. 4b). Meanwhile, CAR, NAR and PAR showed an upward trend before 1970s but downward trend after 1970s until the sampling year (Fig. 5).

3.2.3 Comparisons of limnological parameters between DC and FX

As shown in Fig. 2e-h, most of the concentrations of TN and TP in DC were higher than that in FX, with the exception of FX6. For instance, the average contents of TN in Lake DC (5.21 mg g$^{-1}$) were much higher than that in FX (3.39 mg g$^{-1}$) (Figs. 2e and f). Also, the TN ranges of the former (3.35-7.19 mg g$^{-1}$) were larger than that the latter (2.85-4.20 mg g$^{-1}$). Similar to TN, the mean concentrations of TP in DC (2.03 mg g$^{-1}$) were slightly higher than that in FX (1.58 mg g$^{-1}$), with the exception of FX6 (Figs. 2g and h). The TP ranges of the former (1.44-3.30 mg g$^{-1}$) were also higher than that the latter (0.83-2.53 mg g$^{-1}$). Both of the average TOC/TN increased from the northern to the southern part of the two lakes, with the exception of FX3 (Figs. 2i and j). No statistics characteristic can be used to describe the spatial difference in CAR, NAR and PAR between DC and FX (Fig. 6).

Temporally, most of the vertical concentrations of TOC, TN and TP in DC and FX showed the upward trend from bottom to top over the past ~150 years (Fig. 3). Also, the slopes of the increasing trend in DC were relatively larger than those in FX, with a reception of FX6, which may be due to a rapid increasing rate of nutrients in sediments of DC (Wu et al., 2018). With regard to the temporal distributions of TOC/TN in DC and FX during the past ~150 years, obvious upward trends of TOC/TN were found among these sediment cores of DC (Fig. 4a), but no regular trend was observed in the temporal change of TOC/TN in FX (Fig. 4b). According to Meyers (1994), the ratio of C to N was used to distinguish the origin of organic matter in sediments, for example, algae typically have TOC/TN ratios of 4-10 (6 as the origin of algae in this study, dashed line Fig. 4a and b), whereas vascular land plants have TOC/TN ratios of 14-23 (23 as the origin of vascular land plant in this study, solid line in Fig. 4a and b). Here, the TOC/TN ratios of 6 (algae) and 23 (vascular land plants) originated from the binary model of literature as mentioned above (Qian et al., 1997). The increasing trends of TOC/TN over time indicated that the
origin of organic matter in sediments of DC altered from algae to vascular land plants during the past ~
150 years (Fig. 4a). On the contrary, most of the TOC/TN values (>23) in FX are mainly focused on the
central and southern part of the lake (Fig. 4b), which implied that the contributions of vascular land plants
were much higher in those areas. The average values of N/P were 5.82 ± 1.81 (ranging from 1.99 to 11.82)
for DC, and 5.04 ± 1.56 (ranging from 0.70 to 11.65) for FX, respectively. No obvious differences were
found in the vertical distribution of N/P between DC and FX (Fig. 7).

As a whole, the characteristic of CAR, NAR and PAR in DC and FX was intumescent from bottom
to top over time (Fig. 5). Meanwhile, three time periods of CAR, NAR and PAR (pre-1900, 1900-1960
and post-1960 for DC, and pre-1900, 1900-1970 and post-1970 for FX, respectively) were differentiated
in this study. Similar to the vertical distributions of TOC, TN and TP, there was a stable accumulation of
carbon, nitrogen and phosphorus between DC and FX during the period of pre-1900. Afterwards, an
increasing variation tendency was found until 1960 for DC and 1970 for FX, respectively. In contrast,
there was an increasing trend in DC during the period of post-1960 but downward trend in FX after 1970
until the sampling year (Fig. 5).

Although the cores were collected 14 years earlier, the latest researches on both lakes indicated that
TOC, TN and TP and their burial in sediments from DC (Tang et al., 2020) and FX (He et al., 2020) still
have an upward trend. Therefore, the comparative studies of the past environmental changes on these
two adjacent plateau lakes are not out of date in this study.

3.3 Total CAR, NAR and PAR in DC and FX

We estimated the total accumulation rates of carbon (T-CAR), nitrogen (T-NAR) and phosphorous
(T-NAP) in each lake during the past ~150 years (Eqs. 4 and 5). In total, there were 214.0 × 10^4 t of
TOC, 18.6 × 10^4 t of TN and 7.5 × 10^4 t of TP for DC and 211.8 × 10^4 t, 9.3 × 10^4 t and 5.2 × 10^4 t for FX,
respectively. Overall, the stores of N and P in DC during the past ~150 years were higher than those in
FX (Fig. 8). In addition, the estimates of TN and TP burial in DC based on the regional depth-areas in
this study were higher than those of previous studies based on the whole-lake area (Wu et al., 2018) (T-
NAR, 8.1 × 10^4 t and T-PAR, 7.2 × 10^4 t, respectively). In other words, our results of T-CAR, T-NAR and
T-PAR calculated by multiplying the regional depth-areas using lake are more accurate than that in the
single whole-lake area.

4. Discussion
4.1 Accumulation effect and external inputs of nutrients in sediments

As shown in Fig. 2i and j, higher values of TOC/TN ratios were focused in the central and southern part of the Fuxian Lake. For FX3, the abnormal higher value of TOC and TOC/TN maybe due to the accumulation effect in the deeper region of the lake. According to Sakamoto et al. (2002), regardless of the dry (November 2000) and rainy (June 2001) seasons, the concentrations of TN and TP in water column exhibited a marked increase with depths, although a remarkable decrease was also found above the depth of 50 m in both seasons (Fig. 9). Meanwhile, the sampling time (October) for this study was just after the wet season, the river waters were markedly turbid and flowed at a considerably higher rate, and thereby resulting in higher nutrient loadings in the deeper regions of the Fuxian Lake (Sakamoto et al., 2002). These facts suggest increased discharge of nutrients by soil-bound via excessive runoff from land into the lakes and finally settled down in deeper region of the lakes.

In the southern region of FX7, the previous research indicated that the external inputs (mainly inputs from the Xingyun Lake) were the main sources of nutrient loading (Wang et al., 2018). According to Hayakawa et al. (2002), approximately 5.5% of the total organic carbon in the eutrophic Lake Xingyun was transported into Lake Fuxian through the Gehe River, thereby causing the higher CAR in FX. Additionally, the concentrations of TN (0.1-0.27 mg L⁻¹) and TP (2.1-12.0 μg L⁻¹) in Fuxian Lake were around one tenth to twentieth of those of TN (0.8-1.88 mg L⁻¹) and TP (70-88 μg L⁻¹) in Xingyun Lake, respectively (Sakamoto et al., 2002). The higher TP and lower TN/TP ratio in the surface water towards the mouth of Gehe River in Lake Xingyun further indicated the waters in the south basin of Lake Fuxian were affected by the outflow waters from Xingyun Lake (Sakamoto et al., 2002).

Additionally, the annual nitrogen and phosphorus budget was estimated by Sakamoto et al. (2002), who reported that approximately 91% of the total inputs for TN and 80% for TP flowed out of the Xingyun Lake and only 9% of TN and 20% of TP remained in the Lake (Table 3). In Fuxian Lake, the outflow waters from Xingyun Lake accounts for 53% of the river input of TN and for 57% of that of TP (Table 3). Of the total inputs into Fuxian Lake, only 8.7% for TN and 2.5% for TP flowed out, and more than 90% were remained inside of the lake (Table 3). This finding further verified that Xingyun Lake is a flowing system with a shorter residence of the external pollutants and works as an important pollutant source for Fuxian Lake, and Fuxian Lake behaves as a sink for imported nitrogen and phosphorus.

4.2 Impact on variation of CAR (NAR and PAR) in DC and FX

4.2.1 Spatial distributions and regional variabilities
The spatial distributions of carbon, nitrogen and phosphorus burial in lacustrine sediments are complex (Łukawska-Matuszewska et al., 2014). In this study, the CAR, NAR and PAR had high spatial variability between DC and FX (Figs. 5 and 6). For the shallow eutrophic lake of DC (average water depth of 2.9 m), the spatial patterns of CAR, NAR and PAR were similar to that of the changes in SARs (Fig. 2a and Fig. 6a, c and e), which had relatively higher values in surrounding regions and lower ones in central position of the lake. Increased catchment erosion and nutrient inputs accelerated inevitably in SARs and nutrient burial on the nearshore of lake, whereas relatively smaller hydrodynamic condition in center/offshore of lake caused the lower accumulation rates of carbon, nitrogen and phosphorus, respectively (Koziorowska et al., 2018; Worthington et al., 2018).

The average CAR, NAR and PAR were ~51, 5, 2 g m\(^{-2}\) a\(^{-1}\) in DC, and ~73, 4, 2 g m\(^{-2}\) a\(^{-1}\) in FX, respectively. By comparisons, CAR and NAR are lower than the global mean burial rates of 163 and 9 g m\(^{-2}\) a\(^{-1}\), but PAR are slightly higher than that the global mean burial rate of 1 g m\(^{-2}\) a\(^{-1}\) (Breithaupt et al., 2012; 2014). For CAR, both of these two lakes are higher than those in the adjacent Lugu Lake located in Yunnan-Guizhou Plateau (~27 g C m\(^{-2}\) a\(^{-1}\)) (Lin et al., 2021), the eastern humid regions (~15 g C m\(^{-2}\) a\(^{-1}\)) (Dong et al., 2012), and the northwestern arid regions of China (~50 g C m\(^{-2}\) a\(^{-1}\)) (Lan et al., 2015), respectively. For NAR and PAR, both of them are comparable with 2.6 g N m\(^{-2}\) a\(^{-1}\) (3.4 g P m\(^{-2}\) a\(^{-1}\)) and 2.1 g N m\(^{-2}\) a\(^{-1}\) (2.2 g P m\(^{-2}\) a\(^{-1}\)) in DC and FX from the latest studies (Wu et al., 2018; Tang et al., 2020).

The generally lower CAR in these lakes of China than the global estimated averages could be related to the differences in limnological parameters in lakes and their catchments, such as temperature, evaporation, precipitation, location (latitude and longitude), lake size/area, water depth (Zhang et al., 2017a).

For the deep lake of FX, the increasing amounts of SARs and CAR, NAR and PAR from north to south were mainly attributed to the contributions of XY (Liu et al., 2014; Wang et al., 2018). The Gehe River was dredged in 1923 (Zhang et al., 2010) and closed until 2000 or so (personal communication with Mr Luo, from the research station of plateau-deep Lake Fuxian, CAS). The opening of the Gehe River provided the perfect pathway for the prevailing southwest monsoon, allowing the water of Xingyun Lake to flow into Fuxian Lake with an average flowing rate of 5 m\(^{3}\) s\(^{-1}\) (Hayakawa et al., 2002; Hillman et al., 2018), which led to the elevated SARs, CAR, NAR and PAR in FX, especially in central and southern part of the lake (Fig. 6b, d and f). For example, the average values of stable carbon and nitrogen isotopes (δ\(^{13}\)C and δ\(^{15}\)N) in XY were respectively 1.8% and 1.2% higher than those in FX (Xu et al.,...
Previous research found that, similar to TOC, TN and TOC/TN, the variations of δ13C and δ15N in lacustrine sediments can provide the powerful evidence for interpreting the recent eutrophication and source identification (Wu et al., 2007; Yuan et al., 2020). Once again, the recent research of Hillman et al. (2018) indicated anthropogenic activity, at a millennial scale, also had an important impact on the carbon cycle of Xingyun Lake. Consequently, CAR, NAR and PAR in FX are influenced by anthropogenic forcing due to their connectivity between FX and XY.

4.2.2 Environmental impacts on nutrients retention

The burial, mineralization and release of nutrients are influenced by many physical parameters in water column, such as WT, pH, and DO (Mosello et al., 2018; Lin et al., 2021). Warmer WT and higher DO in shallow-lake often contribute to increased carbon mineralization and CO2 production (Marotta et al., 2014; Beaulieu et al., 2019). Compared with the vertical profiles of WT, pH, and DO in water column between FX and DC in July (Fig. 10) (Wang et al., 2017; Zhang et al., 2017b), nearly all proxies showed a constant with depth in DC, whereas the downward trends of each parameters from top to bottom were found in FX. In comparison with cooler WT and lower DO in water column of the deep-lake FX, relatively warmer WT and higher DO in shallow-lake DC are prone to contributing to increased nutrients mineralization and release (Gudasz et al., 2010). Similarly, pH is a key factor in influencing the CAR, NAR and PAR (Hutcheon et al., 1993). Due to the strong alkaline condition with high pH, N and P in sediments are easily released to overlying water in DC in comparison with FX (Kwak et al., 2018; Ma et al., 2021), and thereby resulting in the higher trophic levels in DC (Huang et al., 2018). These comparative results indicated that CAR, NAR and PAR in sediments may be an explanation for the differentiation of water trophic levels between the two plateau lakes.

4.3 Time-integrated source identification in DC and FX

The Qian’ model (Eqs. 6 and 7) was used to estimate the time-integrated proportions of autochthonous and allochthonous sources of OM in sediments (Fig. 11). For DC, most of the sediment cores indicated that the CAR, NAR and PAR buried in the sediments originated from the contributions of allochthonous sources after 1960, with the exception of mixed sources for DC5 after 1980 (Fig. 11a). Although many previous studies indicated that water eutrophication in DC began in the 1980s (Huang et al., 2014; Zhang et al., 2014; Gao et al., 2015; Ma et al., 2020), the aquatic plant community evolved from algal-dominant to vascular-dominant in DC after the 1960s. Since then, the enhanced inputs of allochthonous matter inevitably led to the occurrence of algal blooms along with many influencing factors.
factors, such as short residence time (3.9 years), increased temperature, decreased precipitation and increased population, N and P loadings (Fig. 12), and thereby improving the primary productivity and trophic levels (Lewandowska et al., 2012; Evans et al., 2017; Fricke et al., 2018).

Unlike DC, the time-integrated sources in FX are very complex (Fig. 11b). The accumulation of organic carbon in FX was endogenously derived before the 1920s, which further confirmed that Lake Fuxian was in a quasi-natural state with limited human activity occurring within the catchment (Wang et al., 2018). Afterwards, the mixed contribution of autochthonous and allochthonous source occurred in FX until the 1970/80s. Undoubtedly, the dredge of the Gehe channel altered the compositions of organic carbon in the sediment cores of Lake Fuxian (Hayakawa et al., 2002; Zhang et al., 2010; Hillman et al., 2018). Similar to DC, the contribution of allochthonous source was also found in recent years, especially in the northern part of Lake Fuxian (FX1). This might due to the impact of the increase of population, N and P fertilizer use from the Chengjiang County (Fig. 12). Additionally, the latest research of He et al. (2020) also supported the results that human activity was the dominant factor influencing the burial of autochthonous organic carbon in Lake Fuxian after 2000. Combined with the molar ratio of TOC to TN in FX3 and FX6 (Figs. 2j and 4b), the contribution of allochthonous source (terrestrial organic matter) in the central and southern position of the lake was close to 100 %, which may be due to the accumulation effect in the deeper region of the lake and external inputs of the adjacent lake Xingyun (Xu et al., 2005; Barroso et al., 2014). Of course, further work is needed for this. The mix of autochthonous and allochthonous sources were observed in other regions of the lake, which implied that organic carbon accumulation in this region of FX was complex due to the linkage between FX and XY. Similar to other lakes in China (Lan et al., 2015; Zhang et al., 2017a), North America (Ferland et al., 2012; Heathcote et al., 2015) and Europe (Anderson et al., 2014; Lacey et al., 2018), organic carbon burial in sediments of Lake Fuxian were increased with enhanced anthropogenic sources.

4.4 N and P control in DC and FX

N and P concentrations are an important limitation factors in influencing lake eutrophication (Conley et al., 2009; Pennuto et al., 2014; Paerl et al., 2016). To reduce trophic levels of lakes, terrestrial N and P loadings should be predominately limited (Vitousek et al., 2010; Du et al., 2020). The proxy of N/P ratio was used as an indicator of potential N-only, P-only and combined N and P limitation. According to the previous study by Paerl et al. (2016), the limitation of N and/or P were divided into three categories: N only limitation if N/P < 9, N + P colimitation if 9≤N/P < 22.6, and P only limitation.
if N/P\(\geq\)22.6, respectively.

As mentioned above, concentrations and accumulation rates of C, N and P in sediments of the hyper-eutrophication Lake DC are higher than those in oligotrophic Lake FX, which is also in agreement with the calculated Carlson’s trophic state index (TSI) for DC (71.1) and FX (25.8) (Tables S1 and S2), respectively. Meanwhile, the proportions of allochthonous contributor to increased CAR, NAR and PAR in DC are higher than that in FX after 1960s (Fig. 9). The amounts of N and P fertilizer use in DC are both higher than those in FX (Fig. 10), which further indicated that the lake trophic states were both impacted by the utilizations of N and P fertilizer within the catchments of these two lakes, including their affiliated reservoir and rivers (Wang et al., 2015; He et al., 2020; Zhou et al., 2021). This result also implied that nutrients derived from fertilizer use are responsible for the increased trophic states of these two plateau lakes. On the other hand, the relationships between CAR, NAR and PAR in DC are significantly stronger than those in FX (Fig. 13), which indicated that the impacts of CAR, NAR and PAR in sediments on hyper-eutrophication lake DC are higher than that on oligotrophic lake FX.

Combined with the time-integrated ratios of N to P between DC and FX (Fig. 7), most of the values <9 further indicated that N limitation should be predominant in these two plateau lakes. According to Qin et al. (2020), who reported that water depth played an important role in controlling lake eutrophication based on the statistical results of the global 573 lakes. In general, N was limited in shallow lakes but P in deep lakes. However, our results indicated N limitation should be predominated in FX as discussed above. It is not contradictory because the oligotrophic deep Lake FX was connected to the shallow trophic Lake XY (average depth of 5.3 m) through the Gehe River during the periods of 1920s-2000s (Hillman et al., 2018; Wang et al., 2018). The exchange of water and sediments via the Gehe River altered the concentrations of particulate organic matter between FX and XY (Xu et al., 2005; Wang et al., 2018). Furthermore, serious eutrophication has occurred in XY since the early 1990s due to the increase of anthropogenic loading (Zhang et al., 2010; Hillman et al., 2018). On the other hand, water depth was not the only factor for controlling eutrophication in FX in this study. Integrated with the comprehensive assessment the whole-lake characteristics of FX and XY, N limitation should preferably be taken into consideration in the deep lake of FX in comparison with P.

5. Conclusions

The spatial and temporal concentrations and accumulation rates of carbon, nitrogen, and phosphorus recorded in the sediments of two plateau lakes (Dianchi and Fuxian) of SW China suggested
that, regardless of the lakes, CAR, NAR and PAR had high spatial variability and an increasing trend with time over the past ~150 years. The total storage of CAR, NAR and PAR in DC are both larger than those in FX, which are in accordance with the trophic levels of the two lakes. The TOC/TN indicated the inputs of allochthonous sources after 1960 was the main driving factor for causing the increased trophic states in DC. By contrast, higher TOC/TN in FX in the central-southern part of FX originated from the accumulation effect in the deeper region and was a contributor of the adjoining shallow eutrophic Lake XY thought the connected Gehe River during the periods of 1923-2000s. The mixed autochthonous and allochthonous sources were not distinguished clearly owing to the dual impact of water depths and nutrient inputs from XY.
Declarations:

Ethics approval and consent to participate: Not applicable.

Consent for publication: The Authors confirm: 1) that the work described has not been published before; 2) that it is not under consideration for publication elsewhere; and 3) that its publication has been approved by all-authors.

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The study area and sampling sites. a. Map of China. b. Location of Lake Dianchi and Lake Fuxian. c. The isobaths and sampling sites of Lake Dianchi. d. The isobaths and sampling sites of Lake Fuxian. Note: The designations employed and the presentation of the material on this map do not imply the expression...
of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 2

Comparison of variation of average SARs, TOC, TN, TP and TOC/TN in different sampling sites between DC and FX. From north to south, the orders of sampling sites in each lake were DC2, DC1, DC3, DC4, DC5, DC6, DC7, FX1, FX2, FX3, FX4, FX5, FX6, FX7.
DC6, DC7 and FX1, FX2, FX3, FX4, FX5, FX6, FX7, respectively.

Figure 3

Comparison of temporal distribution of TOC, TN and TP in DC and FX. Dashes lines meant the age in 1960s and 1900s for DC and 1990s and 1900s for FX, respectively.
Figure 4

Comparison of temporal distribution of C/N in DC and FX. Dashes lines meant the source of organic matter from algae and solid lines meant the sources of organic matter from vascular land plants.
Figure 5

Comparison of temporal variation of CAR, NAR and PAR in DC and FX. Dashes lines means the age in 1960s and 1900s for DC and 1970s and 1900s for FX, respectively.
Figure 6

Comparison of spatial variation of CAR, NAR and PAR in DC and FX
Figure 7

Comparison of temporal distribution of N/P in DC and FX
Figure 8

Comparison total sediment storage of CAR, NAR and PAR in DC and FX
Figure 9

Variations of total nitrogen (TN) and phosphorus (TP) in water column with depths in Lake Fuxian (Sakamoto et al., 2002).
Figure 10

Comparison of vertical profiles of WT, pH, DO and CO in water column with depths between FX and DC (Wang et al., 2017; Zhang et al., 2017b).
Figure 11

Comparison of the contributions of the autochthonous and allochthonous sources to organic carbon accumulations for each sediment cores between DC and FX.
Figure 12

Time series of meteorological data (temperature and precipitation) and human activities data (population, N and P fertilizer utilization). The meteorological data were obtained from the Kunming and Yuxi Meteorological station. The data of population, N and P fertilizer utilization were from the Yunnan Provincial Bureau of Statistics (http://www.stats.yn.gov.cn).
Figure 13

Plot of relationships among CAR, NAR and PAR in DC and FX.

Supplementary Files

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