Title:

Eutrophication has a greater influence on floodplain lake carbon cycling than dam installation across the middle Yangtze Region

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Highlights:

- C/N ratios and $\delta^{13}C$ suggest regional similarities in floodplain lake carbon cycling one century ago.
- Timings of shifts in sedimentary proxies indicate larger responses to eutrophication than dam installation in the 20^{th} century.
- Eutrophication influences production and burial of autochthonous carbon by stimulating aquatic plant or algal growth.

Abstract:

Carbon cycling in shallow floodplain lakes is complex due to variability in delivery of flood-derived allochthonous organic matter (OM). Human activities have potential to significantly modify the carbon balance of lakes by damming which restricts external OM inputs and via eutrophication which can increase the in-lake production of algae and/or aquatic plants. In order to understand how these human activities influence carbon cycling in shallow floodplain lakes over decadal-centennial timescales, we analysed C/N ratios and $\delta^{13}C$ from terrestrial plants, catchment soils, aquatic plants and dated sediment cores from six heavily modified lakes in the middle Yangtze floodplain. Submerged macrophytes (−21.4 ± 4.6‰) had higher $\delta^{13}C$ than C_3 plants from the catchment (−26.6 ± 0.6‰) and emergent and floating plants (−26.6 ± 4.0‰). Increases in sedimentary chlorophyll *a* (from primary producers) were associated with a decline in sedimentary $\delta^{13}C$ in the severely eutrophic Dongting, Luhu, Wanghu and Poyang Lakes after the 1980s. In contrast, sedimentary $\delta^{13}C$ increased in Honghu and Futou Lakes which have abundant submerged macrophytes. The timing and scale of sedimentary $\delta^{13}C$ changes indicated stronger responses to eutrophication than damming, with eutrophication responses ranging from a macrophyte proliferation to the dominance of phytoplankton.

Key words: carbon cycling; algal production; submerged macrophytes; palaeolimnology; sedimentary $\delta^{13}C$; floodplain lakes

1. Introduction

Floodplain lakes are critical and dynamic hotspots for carbon cycling where particulate organic carbon can be a significant component of the total carbon pool (Cai et al., 2016; Hupp et al., 2019; Sanders et al., 2017). Under natural conditions, floodplain lakes are periodically connected with rivers and this connection is strengthened during seasonal flooding. Floods can be important for the delivery of suspended materials including organic carbon and nutrients (nitrogen, N and phosphorus, P) from rivers to floodplain lakes (Zhang et al., 2022), leading to periods where allochthonous carbon delivery to the water bodies is high (during floods) versus inter-flood periods where relatively more autochthonous carbon sources are generated internally via photosynthesis using (recycled) nutrients derived from the flood pulse (Hupp et al., 2019; Liang et al., 2016). Therefore when floodplain lakes are naturally connected to rivers the relative balance of autochthonous to allochthonous materials in the lake basin should be primarily linked to flood frequency and seasonality, and by extension to the climatic conditions (Aspetsberger et al., 2002; Li et al., 2016; Liang et al., 2016).

Many floodplain lakes across the world are losing connectivity with the main channel associated with human development of river catchments (Yang et al., 2011). Floodplain management to support growing human populations has led to land drainage, damming and realignment of rivers, and shifts in land use and land cover with major consequences for the transport and fluxes of materials between river catchments and lakes (Wu et al., 2007; Yang et al., 2011). Additional to these changes, pollution from nutrients (N, P) has intensified since the acceleration of synthetic fertilizer applications and the centralisation of urban sewage disposal and treatment (Yu et al., 2019). Changes in lake hydrological and nutrient balance can have profound effects on the carbon cycling in lakes (Chen et al., 2017). For example, damming may restrict allochthonous carbon flow to lakes and the restriction of water supply can cause water level lowering and enhance resuspension and decomposition of lake bottom sediments (Catalán et al., 2016; Syvitski et al., 2009). In contrast, increased delivery of N and P is known to stimulate in-lake productivity by enhancing growth of phototrophs (algae, cyanobacteria, macrophytes), stimulating the production of more autochthonous carbon (i.e., lake eutrophication) (Downing et al., 2008; Hupp et al., 2019). Understanding how the combined forces of hydrological change and nutrient pollution interact to affect carbon cycling is key to integrating the function of floodplain basins in the landscape as traps or sources of carbon and informing carbon-sensitive management.

Lake sediment cores can provide multi-decadal scale archives of changes in lacustrine systems, which can be used to learn how past shifts in carbon cycling relate to widespread changes in human activities such as hydrological modification and nutrient pollution (Chen et al., 2017; Tao et al., 2009). Simple and rapid analytical techniques can be used to characterise organic matter (OM) in lacustrine sediments and provide past insights into carbon cycling shifts in lakes. For example, the relative quantities of total organic carbon (TOC) to total nitrogen (TN) (C/N ratios) and sedimentary $\delta^{13}C$ can provide information on sources of OM and how it is processed within lakes, and thus they are widely used in paleoecology to investigate carbon cycling (Meyers and Teranes, 2002). Distinctive ratios of C/N in algae (4 – 10) compared with vascular plants (> 20) mean that this index can help to distinguish whether sedimentary OM derives from predominantly terrestrial or aquatic sources (Meyers and Teranes, 2002). Because algae preferentially assimilate lighter carbon (^{12}C) for photosynthesis sedimentary $\delta^{13}C$ can commonly be driven by algal productivity (Meyers and Teranes, 2002). In hydrologically-stable water bodies, high primary productivity can enrich the remaining inorganic carbon pool available for photosynthesis and thus the newly-generated OM is enriched in ^{13}C (Meyers and

Teranes, 2002; O'Beirne et al., 2017). Interpretation of $\delta^{13}C$ in sedimentary OM requires critical analysis because isotopes integrate OM sources (a) from multiple seasons, (b) across benthic and pelagic lake habitats where carbon availability and isotopic ratios can differ (Brenner et al., 2006; Gu et al., 1996) and (c) from catchment and in-lake sources (Maberly et al., 2013). For instance, a modern survey of over 80 Florida lakes showed that water column chlorophyll *a* (Chl *a*) explained ~65 % of the variance in planktonic $\delta^{13}C$ but only 38% of the variance in sedimentary $\delta^{13}C$, indicating the contribution of OM from non-planktonic sources (Gu et al., 1996). Because floodplain lakes are shallow, have well-developed and sometimes densely vegetated littoral zones, with seasonally- and interannually-variable flooding dynamics affecting the delivery and outwash of catchment-derived OM, interpretation of C/N and $\delta^{13}C$ is not straightforward.

The middle reaches of the Yangtze Basin are within an important social-economic zone in China containing the country's largest and second largest floodplain lakes (Figure 1). Rapid expansion in population and industrial and agricultural activities in China has led to significant nutrient pollution issues (Dong et al., 2012; Yang et al., 2008). Meanwhile, more than 50 thousand dams have been established in this area (e.g., Three Gorges Dam) for hydropower, irrigation, flood control and water supply over the past century (Yang et al., 2011). As a result, the floodplain system has severely deteriorated, especially the floodplain lakes (Chen et al., 2016; Dong et al., 2012, Yang et al., 2008). Lakes are reported to have transitioned from "macrophyte-dominated" to "algal-dominated" states since the "reform and opening-up" period of escalating economic development starting in the late 1970s (Dong et al. 2016; Zhang et al., 2018). Notable increases in carbon burial rates of the middle and lower Yangtze region have occurred since the 1950s associated with eutrophication (Dong et al., 2012). However, in general organic carbon is heavily mineralized in these type of floodplain lakes, and carbon burial is dependent on the size of lakes with larger lakes burying less carbon per unit area (Dong et al., 2012). Better characterization of the OM could help to understand the function of these lakes in carbon processing and to track how these processes have changed through a period of major change spanning the "Anthropocene" (Anderson et al., 2020). Therefore, we aimed to 1) characterise potential sources of OM in the middle Yangtze floodplain (China) by analysing $\delta^{13}C$ and C/N ratios of "end members" (catchment plants, soils, aquatic plants) and lake surface sediments; 2) investigate the relative influences of damming and eutrophication on OM cycling in six shallow mid-Yangtze floodplain lakes by studying C/N ratios and $\delta^{13}C$ and sedimentary Chl *a* spanning the past 200 years. We hypothesise that localised damming of lakes will reduce the relative contribution of allochthonous OM while sedimentary $\delta^{13}C$ and Chl *a* will record eutrophication responses in these lakes which will accelerate during the 1980s when economic development expanded rapidly in this region.

2. Materials and methods

2.1 Study area

The six floodplain lakes in the middle Yangtze floodplain include the two largest in China (Dongting and Poyang, > 2500 km^2) which remain freely connected with the Yangtze River (Figure 1a). The other lakes (Honghu, Futou, Luhu and Wanghu, 40 – 350 km^2) have restricted connectivity with the Yangtze tributaries because of the installation of local dams. There are obvious differences in water quality across the six lakes, with two having abundant macrophyte cover (Futou and Honghu) and the others (Dongting, Poyang, Luhu and Wanghu) experiencing algal blooms with turbid waters which appear to have proliferated in recent years (Figure 1b) (Zong et al., 2019).

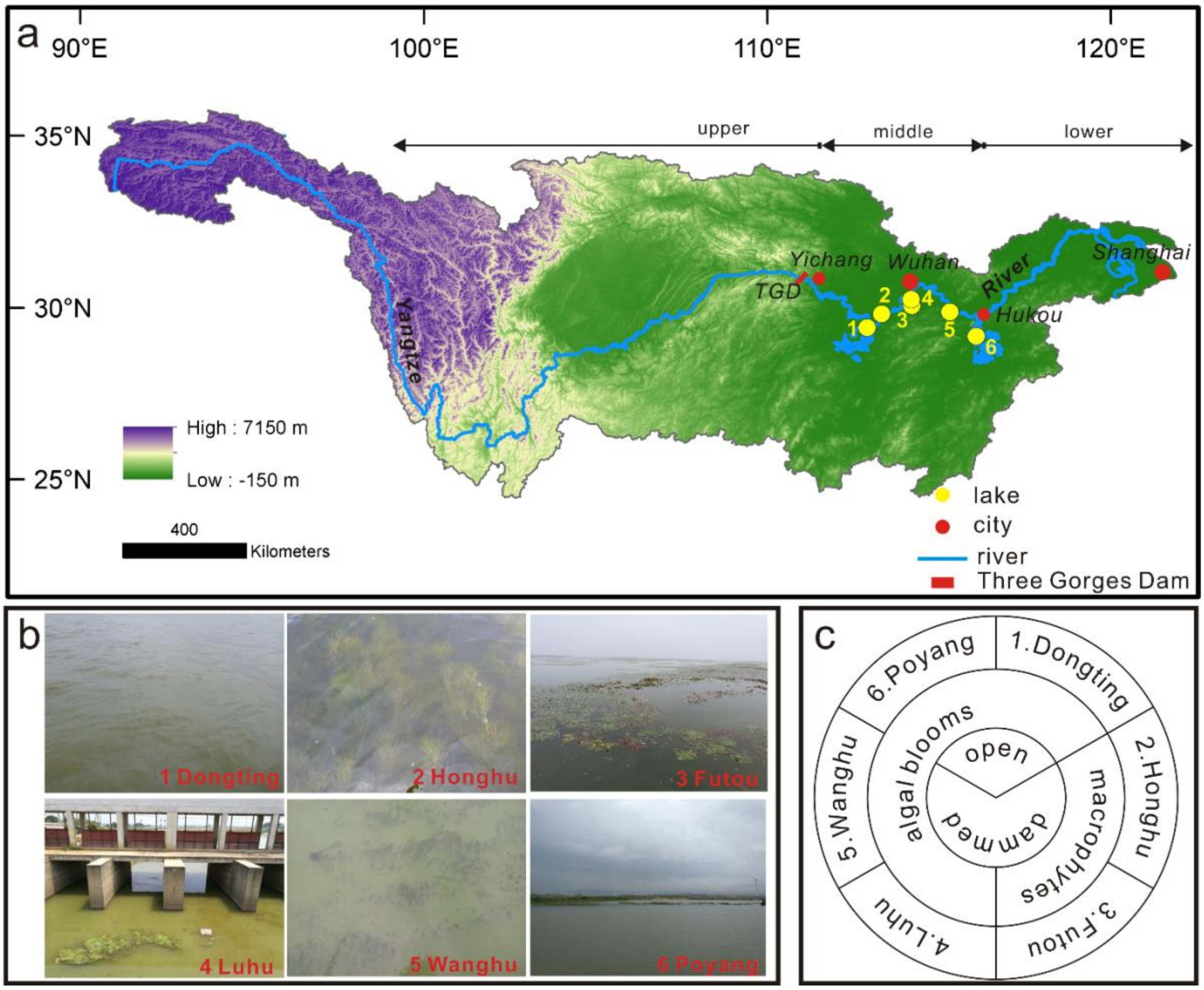

Figure 1 (a) Location of the study lakes (1=Dongting, 2=Honghu, 3=Futou, 4=Luhu, 5=Wanghu, 6=Poyang) in the Yangtze Basin with insets showing (b) the photos and (c) the hydrological and ecological features of the lakes. Based on the geology, climate and the resulting geomorphology, the Yangtze River is divided into the upper (from the headwaters to Yichang), middle (from Yichang to Hukou) and lower (downstream of Hukou) reaches.

2.2 Sampling

In July 2017 samples were taken from soils in the lake catchments (5 paddy soils in total), terrestrial plants (12), and aquatic plants (23, including 6 from emergent plants, 10 from floating-leaved plants and 7 from submerged macrophytes) in the middle Yangtze floodplain (Supporting Information Table S1). In Dongting Lake, surface sediments were collected from

11 points in the lake to represent a spread of depths and distance from the shoreline. In the other lakes, one surface sediment sample was collected from each lake.

Sediment cores (61 – 101 cm) from the central part of each lake were collected using a gravity corer between 2014 and 2017 and sectioned at 1-cm intervals after collection. Subsamples for pigment analysis were stored at −20 °C and subsamples for dating and C/N and $\delta^{13}C$ analysis were stored at 4 °C. Detailed information on the sediment coring were presented in Chen et al. (2019). The sediment cores were dated using ^{210}Pb and ^{137}Cs with a constant rate of supply (CRS) model. ^{210}Pb and ^{137}Cs in the Dongting sediment core were analysed at 2-cm intervals in Nanjing Institute of Geography and Limnology with a gamma spectrometer (Ortec HPGe GWL) (Supporting Information Figure S1). ^{210}Pb activities in the Futou sediment core are from Zeng et al. (2018). ^{210}Pb and ^{137}Cs chronologies in sediment cores from the other lakes are from Chen et al. (2019). Due to the detection limit of ^{210}Pb dating (i.e., older than ca. 150 years), we limited the temporal span of the data to after 1800 CE.

2.3 C/N ratios and $\delta^{13}C$ analysis

C and N mass and $\delta^{13}C$ were analysed at the National Environmental Isotope Facility (NEIF) at the British Geological Survey, after preliminary treatment at the University of Nottingham between October 2017 and May 2018. Freeze-dried samples were soaked in HCl (5%) to ensure that all calcites were removed, and then washed with deionised water three times to eliminate the extra acid before being dried at 40 °C (Brodie et al., 2011). The dry samples were then ground and weighed into tin capsules for analysis. $\delta^{13}C$, TOC and TN content were analysed using Costech Elemental Analyser (EA) and on-line VG TripleTrap and Optima dual-inlet mass spectrometer. TOC and TN content were calibrated using the acetanilide standard. C/N ratios were calculated using corrections for atomic mass. $\delta^{13}C$ was calibrated to the Vienna Pee Dee Belemnite (VPDB) using laboratory standards (BROC 2 and Soil A) which were calibrated against NBS-18, NBS-19 and NBS-22. For each batch of 30 sediment samples, 10 replicates of the internal NEIF standard BROC 2 and 2 replicates of the external standard Soil A were analysed. Analytical precision of $\delta^{13}C$ was to within < ± 0.1 ‰ (1 SD).

2.4 Chlorophyll *a* analysis

Chl *a*, a pigment found in most photoautotrophs, was extracted and analysed in the sediments to provide an estimate of the contribution of autochthonous phototrophic OM to the organic carbon pool (McGowan et al., 2012). High performance liquid chromatography (HPLC) comprising an Agilent 1200 series quaternary pump, autosampler, ODS Hypersil column (250 × 4.6 mm; 5 μm particle size) and photo-diode array (PDA) detector was used to separate and identify Chl *a* extracted from sediments at the University of Nottingham in 2017 (Chen et al., 2001). Firstly, freeze-dried sediments were weighted into vials for extraction. Then 5 ml extraction solvent (acetone: methanol: deionised water 80: 15: 5) was added into the vials to extract the pigments. During extraction, the vials were kept in freezer at −4°C for 12 hours. After that, the samples were filtered through a 0.22 μm PTFE syringe filter, followed by drying down under N_2 gas. Subsequently, the samples were dissolved in injection solvent, a mixture of acetone (70%), ion-pairing reagent (25%) and methanol (5%) and then transferred into HPLC vials and set up for running in the HPLC. Pigment chromatographic peaks were calibrated using commercial standards from DHI (Denmark). Concentrations were expressed as nmole pigment per gram organic carbon (nmol g^{-1} TOC) to allow the relative contribution of autochthonous phototrophic OM to the overall organic carbon pool to be estimated.

3. Results and discussion

3.1 Samples from aquatic plants, catchment plants and soils

To interpret sedimentary carbon profiles, the signatures of potential sedimentary source materials need to be established. Analysis of the modern samples shows that aquatic plants and catchment land plants have similar C/N ratios but distinctive isotope compositions in the middle Yangtze floodplain (Figure 2a and 2b). Similar to values of Meyers and Teranes (2002), *Zea mays*, a common C_4 plant, is characterized by a relatively positive $\delta^{13}C$ value (–12.0 ± 0.6‰, $n = 2$), whereas other catchment plants, such as *Oryza sativa* (–29 ± 0.3‰, $n = 3$) and *Artemisia selengensis*, *Graminae*, and Pinaceae (–29.6 ± 0.7‰, $n = 5$), have a negative $\delta^{13}C$ ($p < 0.05$) (Figure 2a). Terrestrial plants use atmospheric CO_2 (~ –7‰) for photosynthesis (Meyers and Teranes, 2002). C_3 plants (i.e., *Oryza sativa*) use the Calvin photosynthetic pathway (isotopic discrimination ~ –20‰), whereas C_4 plants (*Zea mays*) use the Hatch-Slack pathway (isotopic discrimination –6‰ to –4‰) (Meyers and Lallier-Vergés, 1999). Therefore, OM produced by C_3 plants is characterized by a lower $\delta^{13}C$ value (–27‰) compared to C_4 plants (–13‰ to –11‰) (Meyers and Lallier-Vergés, 1999; Wu et al., 2007).

Submerged macrophytes sampled in the middle Yangtze floodplain lakes had higher $\delta^{13}C$ (–21.4 ± 4.6‰, $n = 7$) than emergent and floating plants (–26.6 ± 4.0‰, $n = 16$) ($p < 0.05$) (Figure 2b). This is commonly observed as they use different sources of carbon for photosynthesis (Aichner et al., 2010; Yu et al., 2021). Autotrophs in lakes potentially use atmospheric CO_2, aqueous CO_2 and HCO_3^- for photosynthesis (Meyers and Lallier-Vergés, 1999; Meyers and Teranes, 2002). Aqueous CO_2 has a similar isotopic signature to atmospheric CO_2 (–7‰), whereas HCO_3^- ($\delta^{13}C$ = ~ 1‰) is less depleted in $\delta^{13}C$ due to the fractionation during hydration (Deuser and Degens 1967). Therefore, emergent and floating plants (–26.6 ± 4‰, $n = 16$), living at the surface and margins of lakes, are able to more easily access carbon as CO_2 and have isotopic ratios closer to land C_3 plants (–29.4 ± 0.6‰, $n = 8$) ($p = 0.58$) (Figure 2b). In contrast, submerged macrophytes, inhabiting the bottom of lakes, are more likely to utilize HCO_3^- for photosynthesis when aqueous CO_2 is insufficient in these alkaline lakes.

Therefore, submerged macrophytes have higher $\delta^{13}C$ (Wu et al., 2007; Yu et al., 2021), and are in the range of −21.4 ± 4.6‰ in this study (Figure 2b).

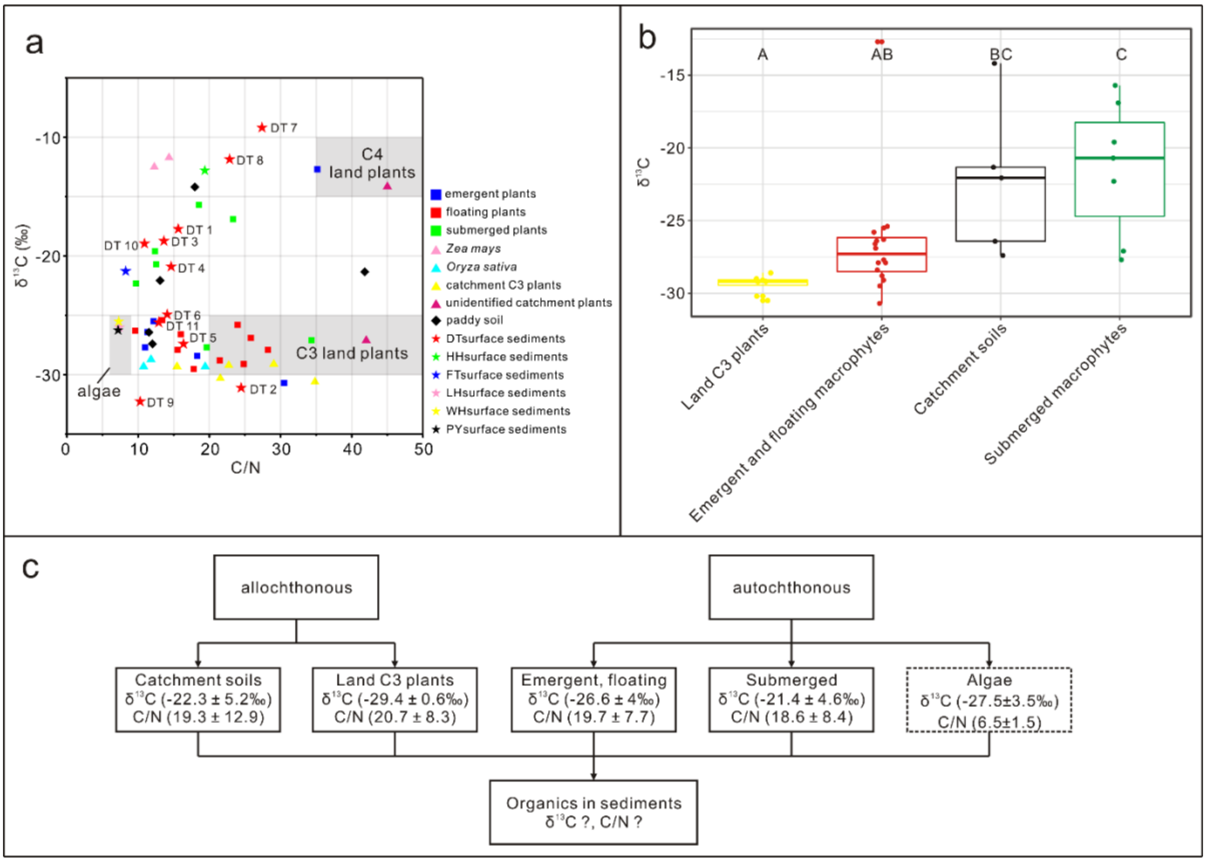

Figure 2 (a) C/N and $\delta^{13}C$ of aquatic and land plants, catchment soils and surface sediments in the six lakes. The grey bars indicate values of algae, C_3 and C_4 plants from Meyers and Teranes, 2002. (b) Boxplot showing $\delta^{13}C$ of different sources. Differences among groups ($p < 0.05$) were assessed using one-way ANOVA. Different letters on the boxes indicate significant differences between groups. (c) Flow diagram showing C/N and $\delta^{13}C$ (mean ± 1sd) of OM from potential sources in the middle Yangtze floodplain. Values of algae (dashed box) are from Meyers and Teranes (2002).

Both $\delta^{13}C$ (−27‰ to −14‰) and C/N (12 to 42) of paddy soils in the middle Yangtze Basin have a wide range of variation (Figure 2a). $\delta^{13}C$ and C/N of catchment soil are determined by the carbon signature of catchment plants and the subsequent processes in soils (Kendall et al., 2001; Wang et al., 2015). In the middle Yangtze floodplain, C_3 plants (mainly *Oryza sativa*) with a lower $\delta^{13}C$ (–29.4 ± 0.6‰, n = 8), are the major agricultural crops (> 90% of the arable land) (National Bureau of Statistics of China, accessed 2020, http://data.stats.gov.cn/). In soils oxidizing reactions and microbial reactions preferentially utilize ^{12}C, which probably causes the observed $\delta^{13}C$ increase in paddy soils (Wang et al., 2015), although the values are highly variable in the middle Yangtze floodplain (–22.3 ± 5.2‰) (Figure 2a).

3.2 C/N and $\delta^{13}C$ in lake surface sediments

OM in sediments of Yangtze floodplain lakes have four main potential sources: a) land plants (mainly C_3) and catchment soils; b) emergent and floating plants; c) submerged macrophytes; and d) algae (Figure 2c). Particulate OM (POM) transported in rivers may also derive from these sources, but can undergo significant alteration before it reaches lakes (Kendall et al., 2001; Wang et al., 2015). Lakes which have retained direct connectivity with the Yangtze River (Dongting, Poyang) might therefore be expected to have similar OM signatures that are distinctive from those that have been dammed and derived predominantly from riverine POM (Hupp et al., 2019). However, there was little evidence of differences among the dammed and freely connected lakes. Instead, the $\delta^{13}C$ (ca. –26‰) and C/N (~7) signatures of surface sediments in these Luhu, Wanghu and Poyang Lakes were most similar to algae (Figure 2a), which could reflect the recent increases in algal blooms at these sites (Figure 1b) (Zong et al., 2019). In contrast, Futou and Honghu Lakes which are densely covered with submerged macrophytes (Figure 1b) (Song et al., 2016; Zeng et al., 2018), have surface sediment C/N and $\delta^{13}C$ signatures which are more similar to that source (Figure 2a). This indicates that autochthonous sources of carbon (from either algae or macrophytes) appear to be the most important contributors to the sedimentary OM composition in these lakes.

In Dongting Lake surface sediments, the δ^{13}C and C/N respectively range from ca. −32‰ to −9‰, and ca. 10 to 27 ($n = 11$) (Figure 2a). The δ^{13}C signature in surface sediments of Dongting Lake is positively and significantly correlated with water depth ($p < 0.05$) (Figure 3a), resulting in an increasing δ^{13}C gradient from littoral to the central parts of the lake (Chen et al., 2017). Dongting Lake is the second largest freshwater lake in China, and the annual water surface area fluctuation between the flooding and drought season is >1,200 km^2 (Ke et al., 2017). The advance and retreat of the lake margin during the flooding and non-flooding seasons integrate a large amount of allochthonous OM into the lake (Chen et al., 2017). In addition, the littoral areas are more suitable for emergent and floating plants which have lower δ^{13}C. Therefore, littoral areas in Dongting Lake which receive relatively more allochthonous OM and have abundant emergent and floating-leaved plants have lower sedimentary δ^{13}C. Central parts of the lake are relatively less influenced by terrestrial OM and so have higher sedimentary δ^{13}C (Chen et al., 2017). The relationship between C/N and water depth is not significant (Figure 3b), because the C/N ratios of land plants and soils (20.7 ± 8.3) are similar to emergent and floating plants (19.7 ± 7.7) and there is a high variation of C/N ratios in each group (Figure 2a).

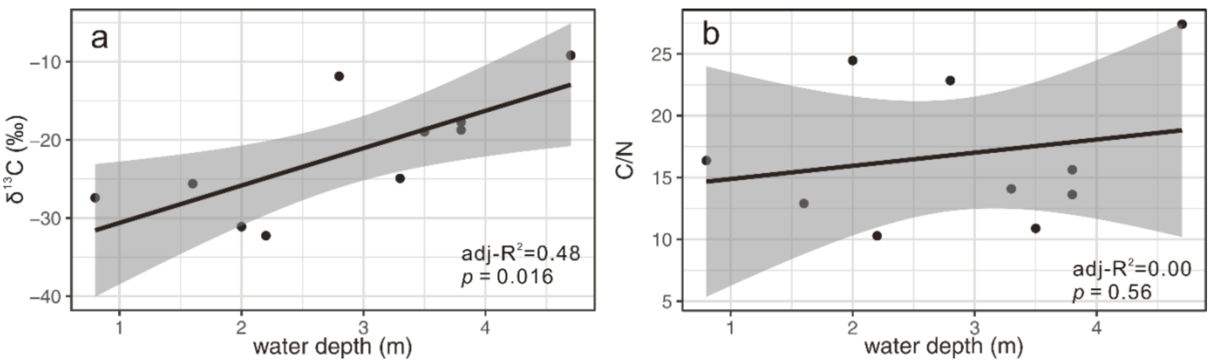

Figure 3 Linear regression between water depth and (a) δ^{13}C and (b) C/N ratios of organic matter in surface sediments in Dongting Lake. The shaded band around the fitted line indicates the 95 % confidence interval.

3.3 Organic matter cycling in the six lakes over the last 200 years

3.3.1 Characteristics of OM under natural conditions

When anthropogenic activities were less intensive in the catchments, the earlier sedimentary $\delta^{13}C$ signature was intermediate (~ –24‰ in Dongting, ~ –23‰ in Honghu, ~ –23.5‰ in Futou, ~ –24.5‰ in Luhu, ~ –24‰ in Wanghu, and ~ –22.5‰ in Poyang), located somewhere between the higher submerged macrophyte values (–21.4 ± 4.6 ‰) and those from other sources (terrestrial plants (–29.4 ± 0.6‰), floating-leaved and emergent plants (–26.6 ± 4‰), algae (–31‰ to –24‰)) (Figure 4 and 5). Although there might be heterogeneity among each individual lake, as indicated by the wide range of $\delta^{13}C$ and C/N ratios in the surface sediments of Dongting Lake, sedimentary records in Dongting Lake sediment core of this study are similar to previously published records by Chen et al. (2017) (Supporting Information Figure S2). This suggests that the single sediment core collected from the central part of Dongting Lake provides a representative record of the history of the lake (Dong et al., 2012).

During the earlier century of lower anthropogenic disturbance, primary producer pigment concentrations were low in the lake sediments (Chl *a*; Figure 4) while water level fluctuations and disturbances from flooding resulted in low to intermediate levels of submerged macrophyte coverage (Zeng et al., 2018). Therefore, the intermediate sedimentary $\delta^{13}C$ values indicate a mixture of OM from different sources. We saw limited evidence to suggest that there were systematic differences in these early OM $\delta^{13}C$ signatures among lakes based on geographic gradients or differences in lakes size (Dong et al., 2012; Wu et al., 2007). This could suggest similarities in the processes of carbon transport and deposition in the wetlands across this region prior to ~ the 1940s (i.e., the time prior to marked shifts in the sediment core profiles).

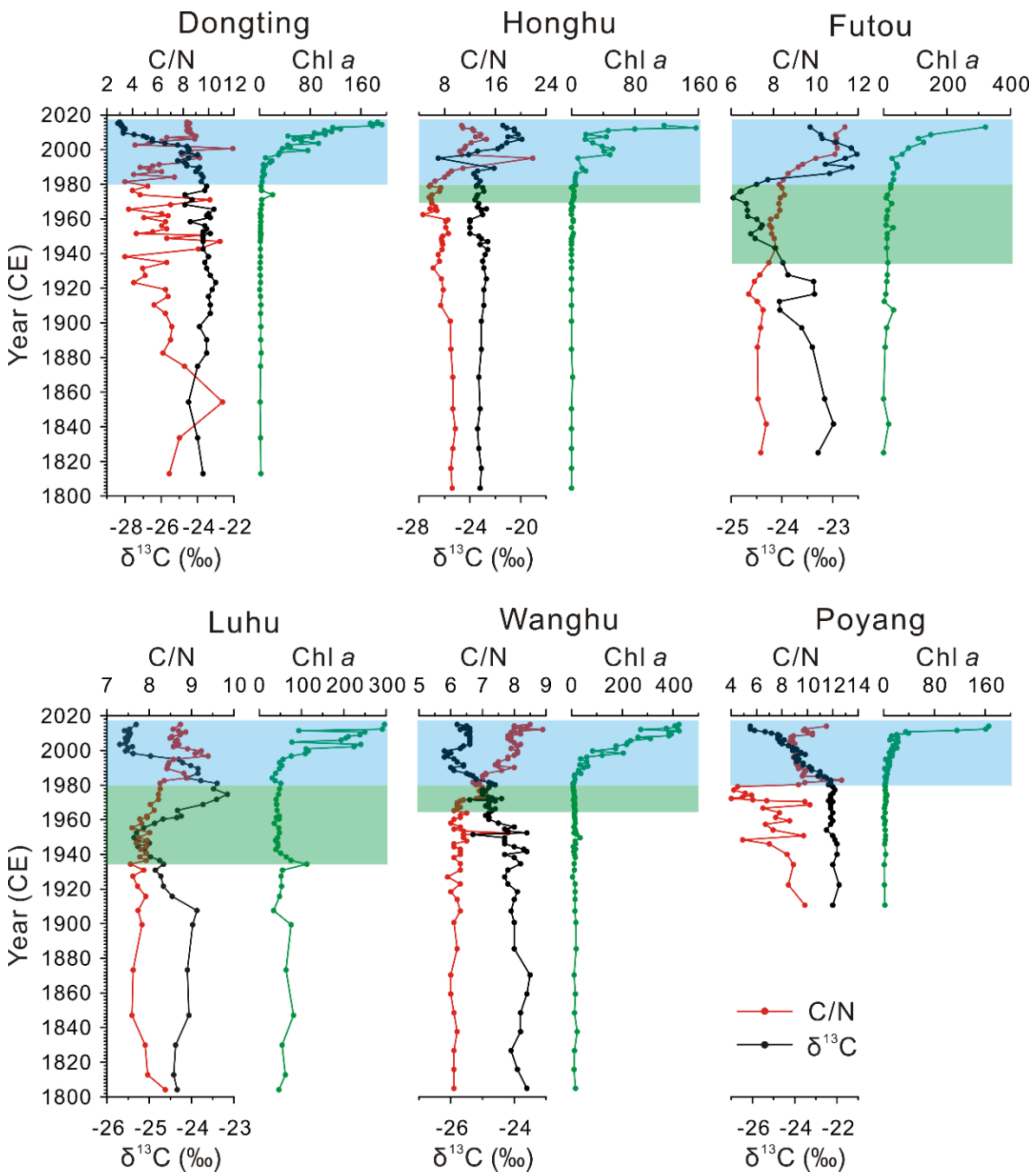

Figure 4 C/N (red), $\delta^{13}C$ (black) and Chl *a* (green, unit: nmol g^{-1} TOC) of the sediment cores from the six lakes since the 19th century. The blue boxes indicate the period after 1980. The green boxes indicate the period after local dam construction.

3.3.2 Responses to hydrological modification

Over the past century, the Yangtze Basin has undergone extensive and intensive hydrological modification due to dam and reservoir constructions (Yang et al., 2011). Local dams have been built in Honghu (~ 1970), Futou (1935, 1973), Luhu (1935, 1967) and Wanghu (1965) Lakes, while the other two lakes maintain free and direct connection with the Yangtze River. Between the 1940s and the 1980s Chl *a* remained at relatively low values in all six lakes (Figure 4), indicating low primary production. C/N ratios were highly variable in the two large, hydrologically open lakes (Dongting and Poyang) (3 – 13) compared with the other four lakes (Luhu, Wanghu, Honghu and Futou) (6 – 8) which have smaller surface areas and restricted connection with the Yangtze main channel due to local dam construction (Figure 4). This might be attributed to the highly variable hydrological conditions in Dongting and Poyang Lakes (Chen et al., 2017; Liang et al., 2016). The difference in water surface area during the flooding and non-flooding season is > 1200 km^2 and > 1000 km^2 in Dongting and Poyang Lakes, respectively (Ke et al., 2017; Shankman and Liang, 2003), which may result in the highly variable C/N ratios through several mechanisms (Harris, 2016; Ke et al., 2017). Firstly, the periodic water level fluctuations may incorporate variable amounts of terrestrial OM (i.e., catchment soils or plants) which have a wide range of C/N ratios (Chen et al., 2017; Woszczyk et al., 2014). Secondly, free and direct connection with the Yangtze River means variable amount of riverine OM can be transported into Dongting and Poyang Lakes during the flooding and non-flooding seasons (Hupp et al., 2019), as well as the amount of Chl *a* being trapped and buried in the lakes (Tockner et al., 1999). Thirdly, water level fluctuations may regulate OM cycling indirectly by changing the redox conditions which regulate the decomposition and degradation of OM, including Chl *a* (Liang et al., 2016). Therefore, the distinct response in C/N ratios between dammed and free lakes implies that damming may be a factor influencing OM cycling in these Yangtze floodplain lakes.

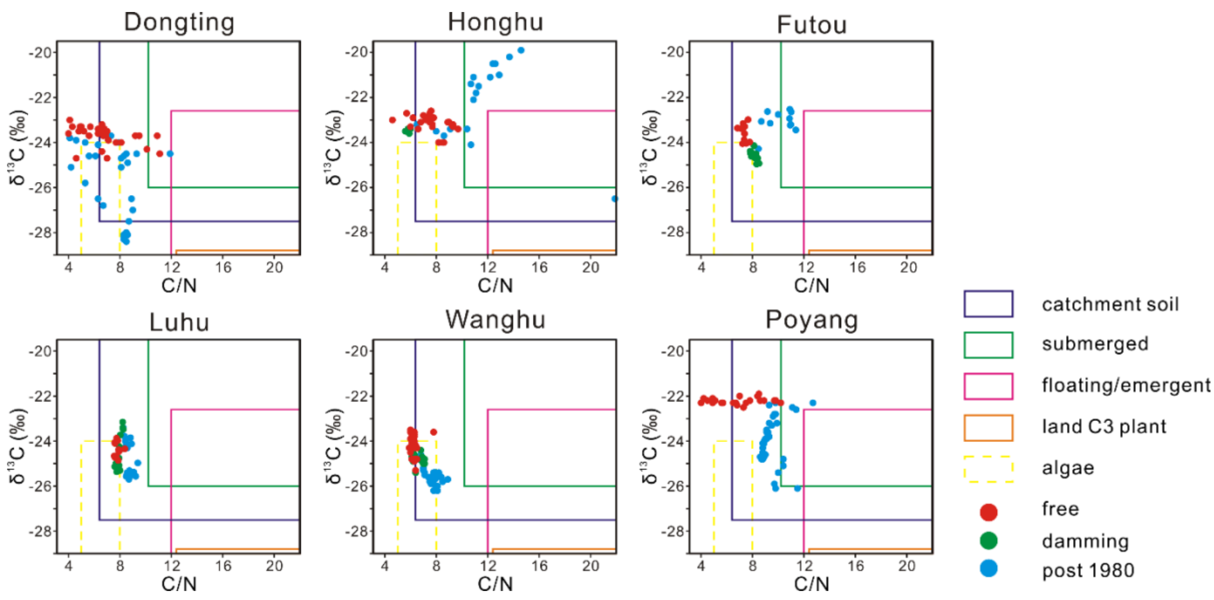

Figure 5 Sedimentary C/N vs. $\delta^{13}C$ in the middle Yangtze floodplain lakes since 1800 CE against values of land plants, submerged, emergent and floating plants in the middle Yangtze floodplain (rectangles in solid line, showing the mean and 1 standard deviation of $\delta^{13}C$ and C/N ratios for each group) and algae from Meyers and Teranes (2002) (rectangle in dashed line).

Hydrological restriction of lakes through damming can promote the development of submerged macrophytes (which have higher $\delta^{13}C$) because they thrive when water levels become more stable and waters become clearer as suspended sediments inputs are reduced (Cao et al., 2014; Zeng et al., 2018). Meanwhile, reductions in riverine POM inputs which have lower $\delta^{13}C$ would be predicted to result in elevated sedimentary $\delta^{13}C$ in lakes with local dams (Hupp et al., 2019). However, there were no obvious and coherent responses in sedimentary $\delta^{13}C$ to dam installation, except in Luhu Lake where sedimentary $\delta^{13}C$ increased (Figure 4). In contrast, sedimentary $\delta^{13}C$ decreased in Futou Lake and there were no obvious changes in Honghu Lake after dam installations. The inconsistent changes in sedimentary $\delta^{13}C$ argue against coherent and overriding impacts of dam installation on sedimentary $\delta^{13}C$. This unexpected pattern may be explained by the synchronous intensification of agricultural activities across the Yangtze floodplain since the late 1940s (Dearing et al., 2012), which could have increased the contribution of terrestrial OM from the catchments, offsetting the decreases in riverine terrestrial OM (Chen et al., 2017). In addition, stabilized water levels after dam construction might also promote the development of emergent macrophytes which are characterized by low $\delta^{13}C$ (Azza et al., 2006). Although speculative, we infer that the concurrent increases in submerged and emergent and floating macrophytes, as well as terrestrial OM, may underlie the observation that dam construction was not marked by a concomitant increases in sedimentary $\delta^{13}C$ in lakes with local dams.

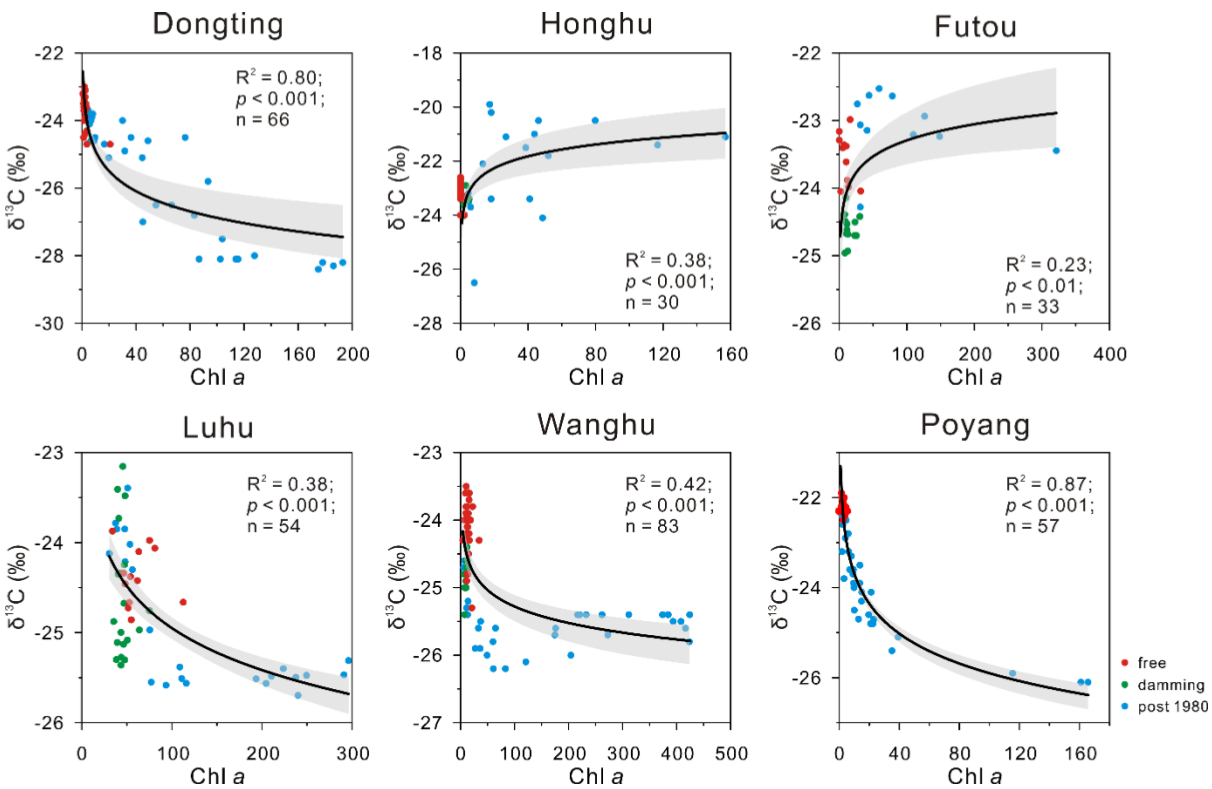

Figure 6 Regression between $\delta^{13}C$ and Chl *a* (unit: nmol g^{-1} TOC) in the sediment cores from the lakes. The colour band around the fitted line indicates the 95 % confidence interval. “free” indicates period when the lakes were freely connected with the Yangtze River.

3.3.3 Responses to eutrophication

Lakes have undergone severe eutrophication in the Yangtze floodplain since the 1980s due to intensive and extensive human activities in the catchments (Dong et al., 2016; Zhang et al., 2018). Elevated burial due to increased primary production enhances sedimentary Chl *a* through better preservation (Figure 4) (McGowan et al., 2012). At the same time, the $\delta^{13}C$ and C/N of sediments gradually moved from the previously intermediate values to those similar to algae in Dongting, Poyang, Luhu, and Wanghu Lakes (Figure 5), indicating that algae became the main source of OM. This resulted in a significant negative relationship between $\delta^{13}C$ and Chl *a* in these four lakes (Figure 6).

The association of elevated primary production with the depletion of $\delta^{13}C$ (> 3‰ decrease) in Dongting, Luhu, Wanghu and Poyang Lakes differs from the trends in some deeper lakes (Meyers and Teranes, 2002) but is commonly associated with eutrophication in shallow lakes (Brenner et al., 1999; Bunting et al., 2007; Wu et al., 2007). Lakes in the middle Yangtze floodplain are shallow and hence suitable for submerged macrophytes (Cao et al., 2014; Zeng et al., 2018). Since the ~1980s, the substantial increases in production caused the decreases in submerged macrophytes in these four lakes (Chen et al., 2017; Scheffer et al., 1993; Zong et al., 2019) (Figure 1b). Therefore, the decrease in sedimentary $\delta^{13}C$ in these four lakes after 1980 might reflect changes in the sources of OM from $\delta^{13}C$-enriched submerged macrophytes to $\delta^{13}C$-depleted algae (Wu et al., 2007).

Consistent with this interpretation, C/N and $\delta^{13}C$ were high and gradually moved to values closer to submerged macrophytes in Honghu and Futou Lakes which have abundant submerged macrophytes after the 1980s (Figure 1b), although algal production increased in these two lakes (Figure 5). Honghu and Futou Lakes are in earlier stages of eutrophication and have abundant submerged macrophytes (Cao et al., 2014; Zeng et al., 2018). The coverage of *Myriophyllum* spp. and *Potamogeton* spp. in Honghu Lake increased from 6% and 10% in the 1950s to 65% and 65% in the 1990s, respectively (Song et al., 2016). Therefore, the concurrent increases in

$\delta^{13}C$ and C/N ratios in Honghu and Futou after the 1980s might be attributed to the increasing contribution of submerged macrophytes, enriched in $\delta^{13}C$, to the OM pool in the early stage of eutrophication.

Despite the low values, C/N ratios slightly increased in Dongting, Poyang, Luhu and Wanghu Lakes since the 1980s as revealed by the positive Mann-Kendall coefficients ($p < 0.05$), likely reflecting the influence of allochthonous OM. Increasing C/N ratios normally indicate increasing contribution of cellulose-rich OM to the sediments (Chen et al., 2017; Meyers and Teranes, 2002). As noted above, the intensification of human activities such as land reclamation, deforestation, aquaculture and lake area shrinkage during this time has probably increased the influxes of allochthonous OM (land C_3 plants and catchment soil) (Dong et al., 2012; Chen et al., 2017), which are rich in cellulose and capable of explaining the observed increase in C/N and depletion of sedimentary $\delta^{13}C$ (Figure 4). This is consistent with the previous study on Dongting Lake which illustrated that increased contribution of allochthonous OM resulted in recent increases in the C/N ratios (Chen et al., 2017).

Selective degradation of OM during sedimentation and early diagenesis may modify the C/N ratios and sedimentary $\delta^{13}C$. It has been reported that the preferential mineralization of N relative to C during early diagenesis causes the increases in C/N ratios of lake sediments as diagenesis proceeds (Gälman et al., 2008; Herczeg et al., 2001). In contrast, the diagenetic loss of ^{13}C-enriched components (e.g., carboxyl carbon) and the selective preservation of ^{13}C-depleted refractory components are reported to cause the depletion of sedimentary $\delta^{13}C$ during diagenesis (Lehmann et al., 2002; Spiker and Hatcher, 1984). In this study C/N ratios exhibit an increasing trend ($p < 0.05$) from the bottom to the top of the sediment cores from all the lakes and sedimentary $\delta^{13}C$ showed a decreasing trend from the bottom to the top of the sediment cores from Dongting, Luhu, Wanghu and Poyang Lakes ($p < 0.001$) (Figure 4), which are opposite to that expected if preferential degradation of N than C and the loss of ^{13}C-enriched components had occurred during diagenesis. Therefore, we think the alternations in C/N ratios and sedimentary $\delta^{13}C$ mainly reflect the changes in the source of OM rather than degradation processes in these Yangtze floodplain lakes over the last 200 years.

4. Conclusions

Our study provides clear evidence that eutrophication, rather than damming, was the main factor regulating OM cycling in the Yangtze floodplain lakes over the last 200 years. Since the 1980s the six lakes exhibited contrasting trajectories of OM cycling from similar baseline values, depending on the degree of eutrophication. In less polluted lakes (Honghu and Futou), which are in the early stage of eutrophication, submerged macrophytes were the main source of OM and hence sedimentary $\delta^{13}C$ is higher. In lakes which have undergone ecosystem state transitions and are suffering from algal blooms (Dongting, Poyang, Luhu and Wanghu), algae are the main source of OM and hence the sediments are characterized by depleted $\delta^{13}C$. Our results indicate that eutrophication induced increases in sedimentation of autochthonously-derived POM and this had a more pronounced effect than damming on regulating sedimentary OM.

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Data Availability Statement

The C/N ratios and $\delta^{13}C$ data used in the study have been submitted to the repository PANGAEA Data Archiving & Publication (PDI-29663). The data submission is now being checked and processed. Temporarily a copy of the data is uploaded as Supporting Information for review purposes.

Declaration of competing interest:

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

Author contribution statement

Linghan Zeng: Conceptualization, Methodology, Data curation, Formal analysis, Visualization, Writing – original draft; **Suzanne McGowan**: Conceptualization, Methodology, Funding acquisition, Writing – review & editing, Supervision; **George Swann**: Data curation, Visualization, Writing – review & editing, Supervision; **Melanie Leng**: Methodology, Formal analysis, Data curation, Writing – review & editing; **Xu Chen**: Funding acquisition, Writing – review & editing.

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