



Anthropogenically driven differences in *n*-alkane distributions of surface sediments from 19 lakes along the middle Yangtze River, Eastern China

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Abstract

During the past few decades, the Yangtze River basin has undergone massive anthropogenic change. In order to evaluate the impacts of human interventions on sediment *n*-alkanes of lakes across this region, the aliphatic hydrocarbon fractions of 19 surface sediment samples collected from lakes along the middle reaches of the Yangtze River (MYR) were analyzed using gas chromatography–mass spectrometry. The *n*-alkanes extracted from the sediments contained a homologous series from C15 to C34, with a notable predominance of odd carbon compounds except for sediments from the more intensively industrialized Lake Daye, in which > C21 *n*-alkanes showed no odd/even predominance, and carbon preference index (CPI) approached unity. Abundance values of middle-chain (C21, C23, and C25) and long-chain (C27, C29, C31, and C33) *n*-alkanes in Lake Daye were approximately 4 to 3 times greater than the average for other lakes, reaching 272.4 and 486.3 μg/g TOC, respectively, in the study. Short-chain *n*-alkanes (C15, C17, and C19) in the sediments varied in abundance from 10.0 to 76.2 μg/g TOC across the study and showed a moderate correlation with total phosphorus (TP) concentrations in the overlying water. The results indicated anthropogenic eutrophication enhanced the accumulation of short-chain *n*-alkanes in sediments because the primary producers in which they are synthesized are highly susceptible to nutrient forcing. Middle-chain *n*-alkane abundances were less affected by eutrophication and generally enriched in macrophyte lakes, while long-chain *n*-alkanes tend to be low in sediments from more eutrophic water. In the case of Lake Daye, direct discharges of petroleum products from heavy industry have introduced quantities of petroleum *n*-alkanes (> C21), far exceeding the amounts of biogenic input, and the sediment > C21 *n*-alkanes detected in this study showed typical characteristics of petroleum source. In other lakes, inputs of petroleum products from surface runoff of vehicle/traffic emissions associated with urbanization and economic growth contributed comparatively few *n*-alkanes to sediments, resulting in declines in CPI for > C21 *n*-alkanes, most obviously in Lakes Huanggai, Donghu, and Futou. Calculated CPI values suggest that a major proportion of the *n*-alkanes present in these lakes are derived from biogenic input. The results of this study provided evidences that *n*-alkane profiles of lake sediments respond sensitively to human-induced eutrophication and different sources of petroleum pollution.

Keywords *n*-alkanes · Eutrophication · Sediment · Middle reaches of the Yangtze River · Anthropogenic pollution

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Introduction

The straight-chain alkanes (*n*-alkanes) are among the most widely used classes of molecular geochemical indicators due to their biogenic specificity and relatively resistance to degradation in sediments (Meyers 2003; Jaffé et al. 2006; Wang et al. 2015; Ankit et al. 2017; Derrien et al. 2017; Kim et al. 2017). They show a fairly strong predominance towards odd carbon numbers in living organisms in which the length of the carbon chain in a given *n*-alkane varies depending on the source (Lu and Meyers 2009; Tolosa et al. 2013; Ankit et al. 2017; Li et al. 2018; Wang et al. 2018). Short-chain *n*-alkanes (e.g., *n*-C15, *n*-C17, and *n*-C19) are commonly found in algae and photosynthetic bacteria (Cranwell 1982; Cranwell et al. 1987; Meyers 1997; Lu and Meyers 2009; Ankit et al. 2017), while long-chain *n*-alkanes (e.g., *n*-C27, *n*-C29, *n*-C31, and *n*-C33) are enriched in terrestrial higher plants and emergent macrophytes (Eglinton and Hamilton 1967; Ficken et al. 2000; Ankit et al. 2017). The *n*-alkanes biosynthesized by submerged and floating aquatic macrophytes include a preponderance of middle-length chains (*n*-C21, *n*-C23, and *n*-C25) (Ficken et al. 2000). In addition to biological sources, *n*-alkanes can also be derived from mature organic matter (OM) of geological origin, which is abundant in petroleum (Peters et al. 2005). Petroleum-derived *n*-alkanes generally lack the characteristic odd carbon predominance found in biogenic forms, due to random cleavage of alkyl chains in the kerogen matrix and progressive loss of high odd-over-even components during petroleum formation (Bianchi and Canuel 2011; Kim et al. 2017). The carbon preference index (CPI) is a quantitative proxy developed to indicate odd-over-even predominance among *n*-alkanes, calculated as a ratio of odd carbon forms relative to their even carbon homologs (Meyers 1997). Higher plant-derived *n*-alkanes generally exhibit CPI values > 4, while those of petroleum *n*-alkanes are much lower, tending towards 1.0 (Harji et al. 2008; Hu et al. 2009; Kim et al. 2017). CPI thus offers a reasonable means for evaluating the origin of sediment *n*-alkanes (Meyers 1997, 2003; Hu et al. 2009; Lu and Meyers 2009; Rushdi et al. 2016; Kim et al. 2018).

During the past century, human activities have exerted a significant influence on aquatic environments and consequently on the abundance and composition of *n*-alkanes in sediments (Meyers 1997, 2003; Medeiros et al. 2005; Gao et al. 2007; Zaghden et al. 2007; Harji et al. 2008; Lu and Meyers 2009; Liu et al. 2013; Pisani et al. 2013; Silva et al. 2013; Zhang et al. 2018a, b). For example, nitrogen (N) and phosphorus (P) loading of lakes fuels algal blooms, leading to enhanced input of algal-derived *n*-C17 in sediments (Paerl 1998; Daskalou et al. 2009; Xu et al. 2010; Zhang et al. 2017). The replacement of macrophytes by

algae under eutrophic conditions might drive a decrease in the contribution of middle-chain *n*-alkanes (Scheffer et al. 1993; He et al. 2015) and the supply of terrestrial plant-derived long-chain *n*-alkanes to lakes is greatly impacted by anthropogenic catchment disturbance (Bourbonniere and Meyers 1996; Bragée et al. 2013; Fang et al. 2014; Zhang et al. 2016). In addition to the changes in biogenic input, petroleum pollution can cause drastic increases in *n*-alkane levels in sediments and the odd carbon predominance associated with biogenic *n*-alkanes might be overturned by heavy contamination with petroleum products (Silva et al. 2008; Daskalou et al. 2009; Fang et al. 2014; Zhang et al. 2018b).

At 6300 km long, the vast Yangtze River (only slightly shorter than the Nile and Amazon) plays a hugely significant role in Chinese economics and society (Zhao et al. 2016). Past variations in hydrology and climate have given rise to hundreds of lakes along the river, isolated from the main flow, but servicing millions of people through fishing and aquaculture, tourism and recreation, transportation routes, and drinking water (Wu et al. 2012). The majority of the lakes along the lower Yangtze River (LYR, Hukou to Shanghai City) lie in a highly fertile, populous, and affluent regions of China and have experienced serious eutrophication since the 1980s (Dong et al. 2012; Yan et al. 2018). Along the middle Yangtze River (MYR, Yichang to Hukou City) meanwhile, economic development has been comparatively slow, and the water quality of lakes in this region remains relatively good (Wu et al. 2012). However, pressure for rapid industrialization to improve local economic conditions in recent years has been at the expense of environmental protection and several lakes have been affected by excessive inputs of nutrients, heavy metals, and organic pollutants (Wu et al. 2012; Zhao et al. 2016). Those that lie close to cities are affected by the input of nutrient-enriched domestic sewage and other pollutants, including petroleum products. For example, Lake Donghu, located in the center of Wuhan City, receives 99.6-t phosphorus (P) a year (Gan and Guo 2004). Eutrophication has led to drastic changes in algal productivity and community structure in many lakes, with potential effects on the *n*-alkane profiles of sediments, while the industrialization and urbanization of catchments are likely to have increased direct and indirect inputs of petroleum-derived *n*-alkanes (Zhang et al. 2018b). However, while anthropogenic intervention along the MYR may already have exerted significant changes in lakes across this region, there have as yet been no attempts to map the abundances and composition of *n*-alkanes in sediments. The current study aimed to identify and investigate any correlation between anthropogenic activity and changes of sediment *n*-alkane by conducting a detailed analysis of aliphatic hydrocarbon in the surface sediments of lakes along

MYR. The results will contribute to the development of molecular geochemical proxies for anthropogenic eutrophication and petroleum pollution.

Methods and materials

Study area and sample collection

The MYR spans much of the interior region of China, including the provinces of Anhui, Jiangxi, Hubei, and Hunan (Fig. 1) and includes hundreds of lakes. Given the difficulty in capturing representative samples from very large and spatially heterogeneous water bodies, the two largest lakes in the region, Poyang and Dongting (both exceeding 3000 km² in area), were excluded from this study. Instead, we focused on 19 smaller lakes (Fig. 1) with surface areas from 9.06 to 338.83 km² and mean depths from 1.3 to 5.0 m. In all cases, hydrologic inputs come mainly from small rivers and precipitation (Wang and Dou 1998), and none of the study lakes are connected directly to the Yangtze River itself. Sampling took place in August 2016, when six samples from the top 1-cm layer of sediment were randomly collected within a 200 × 200 m area close to the center of each lake. The sediments from each site were homogenized, packed in prewashed glass bottles, and stored at −20 °C. Pooled water samples were

taken from the surface to 1 m depth of the central zone in each lake on four occasions in 2016 in early February, May, August, and November, representing the four seasons of the MYR region. On all sampling occasions in the course of the year, aquatic macrophytes were absent from the sampling areas of eight lakes, namely Baidang, Longgan, Wuchang, Taibai, Bohu, Saicheng, Daye, and Donghu (Fig. 1).

Sample analyses

Chemical parameters of water samples including total P (TP), total N (TN), and chlorophyll a (Chla) were determined spectrophotometrically, as described in a previous publication (Zhang et al. 2018b). For the sediment samples, total organic carbon (TOC) content was measured by using a CHNS Vario E1 III elemental analyzer after the removal of carbonate by dilute HCl (Hu et al. 2008). Lipids were acquired from sediments by Soxhlet extraction using dichloromethane and methanol as solvents (9:1 v/v) and *n*-tetracosane-D₅₀ as an internal standard for quantitative purposes. The fraction containing *n*-alkanes was eluted by hexane in a silica gel column chromatography and the composition and quantity of *n*-alkanes and other compounds of interest were determined by gas chromatography–mass spectrometry (GC–MS) using the same instrument model, type of chromatograph column,

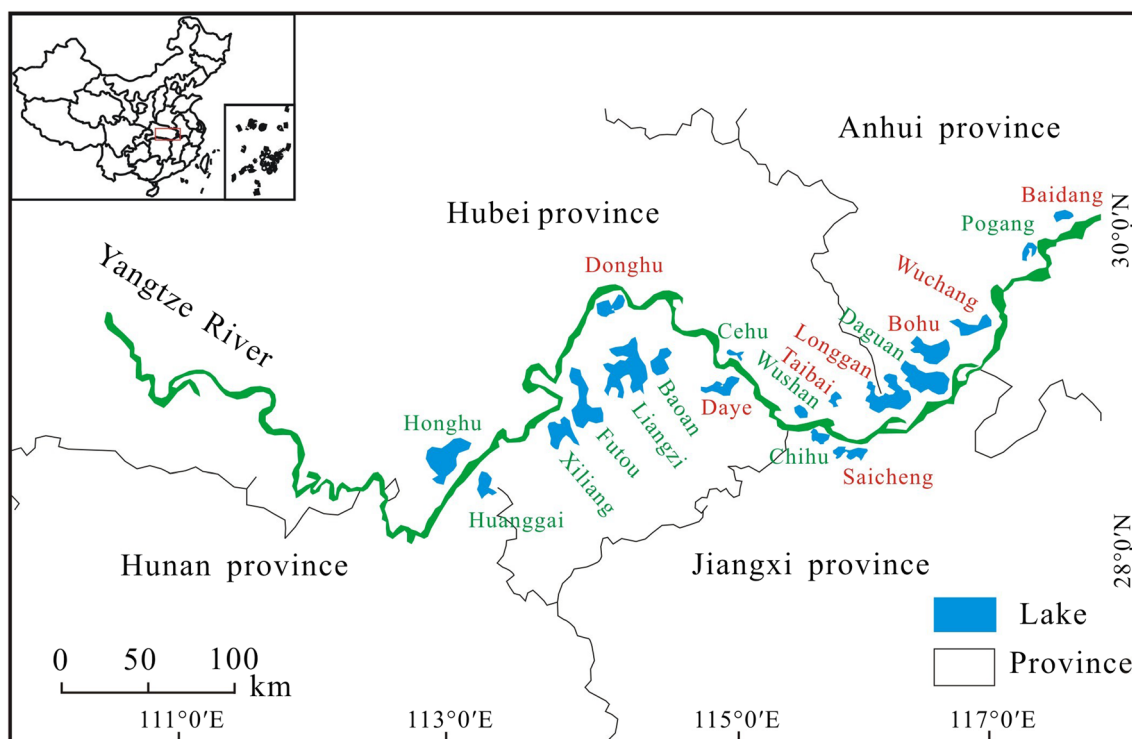


Fig. 1 Location of water and sediment sampling sites in lakes across the MYR region. Lake symbols are printed in red for non-macrophyte lakes and blue for lakes growing macrophytes

and GC temperature program as previously published (Zhang et al. 2018b). Quantities of *n*-alkanes and other compounds were determined by reference to the internal standard and the relative response factors for *n*-alkanes not included in the standard compounds were calculated by linear interpolation. Response factors for hopanes and unresolved complex mixtures (UCM) were calculated by averaging the response factors of nearby *n*-alkanes.

Quality assurance and quality control

One method blank (only solvent), one spiked blank (standard added to solvent), and one matrix blank (standards spiked into a clean matrix) were processed along with each batch of 10 field samples. The analysis of the blanks reveals none of the target compounds were introduced in the experimental procedure. Recovery was determined by adding *n*-tetracosane-D₅₀ to a clean matrix, with the value from 71 to 106%. The detection limit of the current study was 2.3 ng/g sediment. In addition, four samples were selected randomly for further analysis. *n*-Alkanes in five replicates of each sample were extracted and quantified, and the results showed a standard deviation of less than 10% for *n*-alkanes.

Results

Variation of water chemical parameters and sediment TOC

The monitoring data for the year 2016 reveals marked variations in the average TP concentrations between lakes across the study area. Average TP in Lakes Cehu and Wushan was 0.21 and 0.23 mg/L, respectively, much higher than in other lakes. Moderate concentrations between 0.10 and 0.13 mg/L were recorded in Lakes Taibai and Pogang, while the remaining lakes all exhibited values lower than 0.10 mg/L (Fig. 2a). The pattern of TN concentrations differed from that of TP, with the highest value recorded in Lake Huanggai, at 2.18 mg/L. Lakes Cehu and Wushan exhibited relatively lower TN levels, at 2.11 and 1.94 mg/L, respectively, and TN was lower still in Lakes Taibai, Daye, and Futou, ranging from 1.57 to 1.95 mg/L (Fig. 2b). As for TP, Chla concentrations were highest in Lakes Cehu and Wushan, reaching 49.7 and 51.1 μg/L, respectively. Chla was comparatively less abundant in Lakes Taibai and Pogang, at 42.0 and 36.2 μg/L, respectively (Fig. 2c). TOC in the sediments ranged from 1.14 to 4.77%, with high values (>4%) recorded in Lakes Xiliang, Honghu, and Donghu (Fig. 2d).

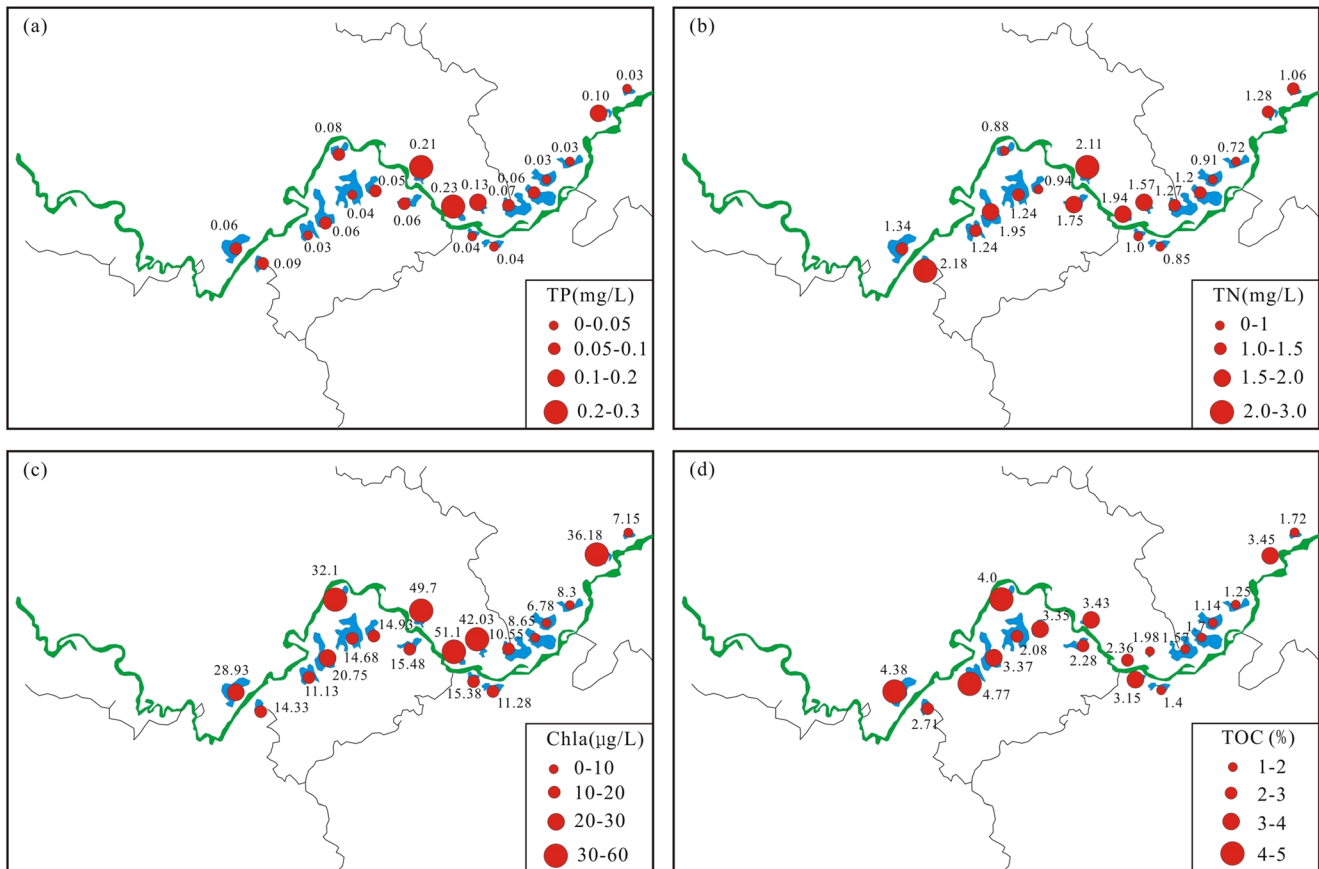


Fig. 2 Variations in water chemical parameters (a–c) and sediment TOC (d) in the studied lakes

Distribution of *n*-alkanes in the sediments

The *n*-alkanes extracted from MYR lake sediments included a homologous series from C15 to C34, with a prominent bimodal distribution (Fig. 3). For the < C20 *n*-alkanes, the dominant peak (C_{max} , *n*-alkane with the highest concentration) occurred at *n*-C17 in all tested samples. The percentages of *n*-C17 relative to total *n*-alkanes ranged from a minimum of 2.41% in Lake Daye to a maximum of 23.45% in Lake Taibai and averaged 9.85% (Fig. 3). Moderate values, exceeding 10% were recorded in Lakes Baidang, Pogang, Cehu, Wushan, Baoan, and Donghu (Fig. 3). Meanwhile, *n*-C15 represented less than 1% of the total *n*-alkanes in all samples, while the values for *n*-C19 varied from 0.78 to 6.39% and averaged 2.22%, being highest in Lake Pogang. This same lake also exhibited the highest percentages of *n*-C18, at 7.18%. The percentages of *n*-C16 were lower than 1% in all except Lake Donghu, where the value was 1.06% (Fig. 3). Short-chain *n*-alkanes (< C20) displayed a predominance towards odd carbon compounds and the CPI calculated for C15–C20 *n*-alkanes ($CPI_{15-20} = (n-C15 + n-C17 + n-C19)/(n-C16 + n-C18 + n-C20)$) ranged from 1.97 to 7.84 and averaged 4.09. CPI_{15-20} values in Lakes Pogang, Daguan, Longgan, Wuchang, and Daye were all < 3.0 (Fig. 4a). For the middle- and long-chain fraction (> C21), C_{max} occurred at *n*-C25 in Lake Daguan; at *n*-C27 in Lakes Pogang, Daye, and Donghu; and at *n*-C29 elsewhere (Fig. 3). The percentages of *n*-C29 relative to total *n*-alkane contents ranged from

11.34 to 26.08% and averaged 17.89%, with values > 20% in Lakes Longgan, Wuchang, Bohu, and Saicheng (Fig. 3). The average chain length (ACL) of > C21 *n*-alkanes varied in a narrow range from 26.4 to 28.2. However, the ratios of $n-C31 + n-C33/n-C27 + n-C29$ were relatively high in Lakes Saicheng, Liangzi, Futou, and Huanggai at 0.56, 0.59, 0.53, and 0.73, respectively. Values were more moderate in Lakes Baidang, Baoan, and Xiliang, at 0.48, and lower still elsewhere. As in the short-chain fraction, odd carbon compounds were rather prevalent among the C21–C34 *n*-alkanes, with CPI values ($CPI_{24-34} = 1/2 \times [(n-C25 + n-C27 + n-C29 + n-C31 + n-C33)/(n-C24 + n-C26 + n-C28 + n-C30 + n-C32) + (n-C25 + n-C27 + n-C29 + n-C31 + n-C33)/(n-C26 + n-C28 + n-C30 + n-C32 + n-C34)]$) varying from 1.26 to 8.24 and averaging 4.66. The value was lowest in Lake Daye and also quite low (between 3.0 and 4.0) in Lakes Baidang, Liangzi, Donghu, Futou, Xiliang, and Huanggai (Fig. 4b). The Paq proxy for evaluating aquatic macrophyte input, defined as $(n-C23 + n-C25)/(n-C23 + n-C25 + n-C29 + n-C31)$, ranged from 0.31 to 0.62 and averaged 0.40 across all samples, and values > 0.5 were recorded in Lakes Pogang and Daguan (Fig. 4c). The terrigenous-to-aquatic ratio (TAR) of *n*-alkanes (defined as $(n-C27 + n-C29 + n-C31)/(n-C15 + n-C17 + n-C19)$) ranged from 1.27 to 9.99 and averaged 4.61, exceeding 6.0 in Lakes Daguan, Longgan, Wuchang, Saicheng, Daye, and Xiliang (Fig. 4d). By assuming the C27 *n*-alkane in sediments is of mixed biogenic and petroleum provenance and that

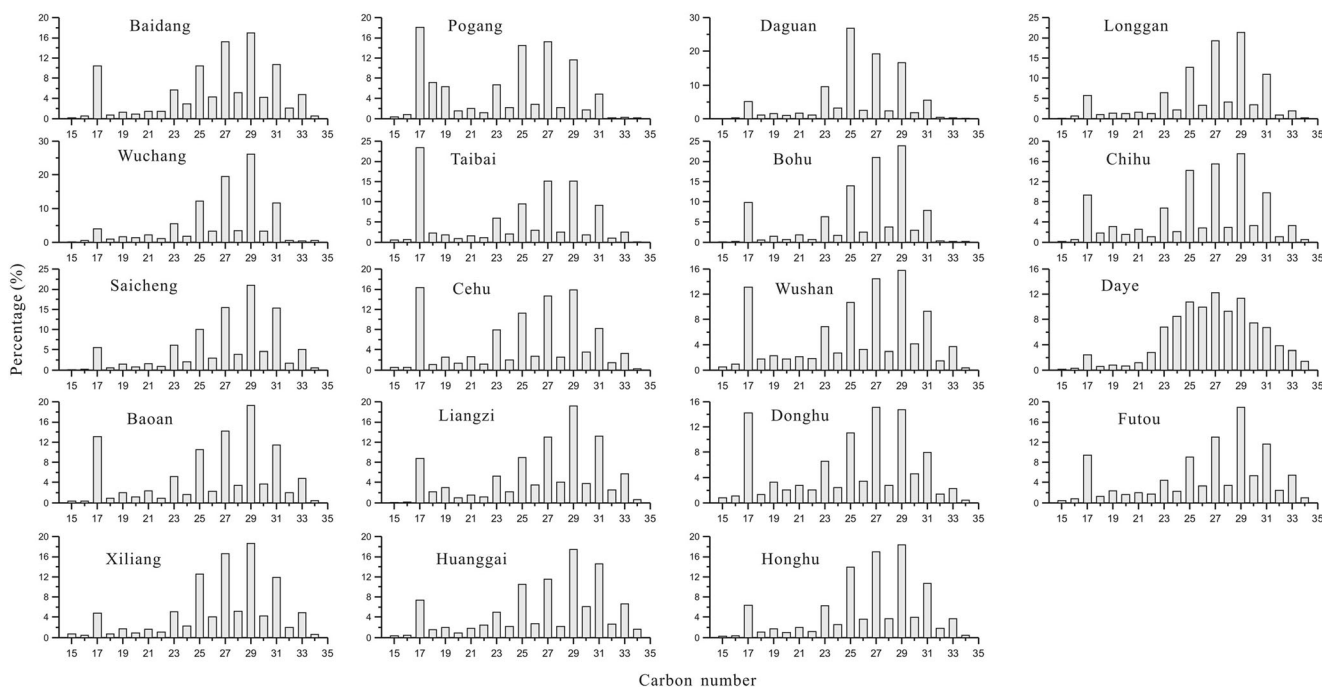


Fig. 3 The percentage of each *n*-alkane relative to total *n*-alkanes in surface sediments of studied lakes

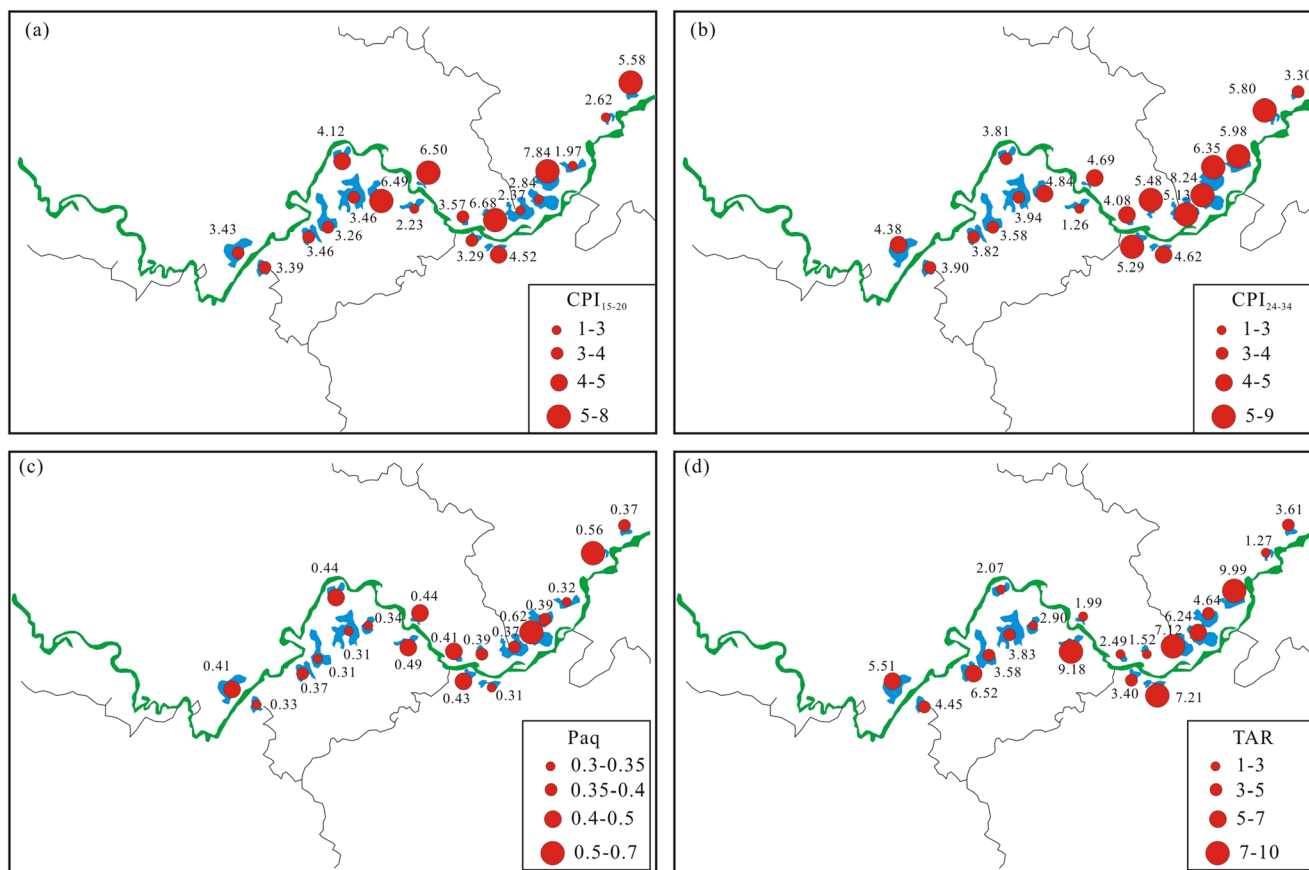


Fig. 4 Variations in CPI_{15–20} (a), CPI_{24–34} (b), Paq (c), and TAR (d) in surface sediments of studied lakes

petroleum-derived *n*-C27 will be present in similar abundances to *n*-C26 and *n*-C28, we can tentatively calculate a percentage for biogenic *n*-C27 using the formula $(100 \times (n\text{-C27} - 0.5 \times (n\text{-C26} + n\text{-C28}))/n\text{-C27})$. The values for this calculation varied from 22 to 87% and averaged 76%. Similarly, the percentages of biogenic C29 *n*-alkane $(100 \times (n\text{-C29} - 0.5 \times (n\text{-C28} + n\text{-C30}))/n\text{-C29})$ varied from 26 to 88% (average 78%) and C31 *n*-alkane $(100 \times (n\text{-C31} - 0.5 \times (n\text{-C30} + n\text{-C32}))/n\text{-C31})$ from 16 to 84% (average 72%). The biogenic percentages of all three *n*-alkanes were lowest in Lake Daye.

Sediment concentrations of *n*-alkanes and other compounds

Concentrations of *n*-C17 in sediments exhibited significant variation between lakes (Fig. 5a). Abundance was greatest in Lake Taibai at 69.3 μg/g TOC and somewhat lower in Lakes Pogang and Cehu at 54.0 and 53.6 μg/g TOC, respectively. Lakes Baidang, Wushan, Baoan, Donghu, and Futou exhibited values in the range of 40–50 μg/g TOC, and in other lakes, values were lower still, with Lake Wuchang as the lowest of

all at 6.9 μg/g TOC. Short-chain *n*-alkane abundances (sum of *n*-C15, *n*-C17, and *n*-C19) exhibited similar variation, with the exception of Lake Pogang where the abundance was nearly the same as in Lake Taibai (Fig. 5b). Abundances of middle-chain *n*-alkanes (sum of *n*-C21, *n*-C23, and *n*-C25) were lowest in Lake Wuchang at 34.9 μg/g TOC; relatively high (> 80 μg/g TOC) in Lakes Baidang, Daguan, Chihu, Honghu, and Huanggai; and highest of all in Lake Daye at 272.4 μg/g TOC (Fig. 5c). Long-chain *n*-alkanes (sum of *n*-C27, *n*-C29, *n*-C31, and *n*-C33) were also abnormally prevalent in Lake Daye, reaching 486.3 μg/g TOC. Lesser values of 200–300 μg/g TOC were recorded in Lakes Baidang, Liangzi, Futou, Xiliang, and Huanggai, while Lakes Pogang and Daguan exhibited the lowest levels at 95.1 and 96.7 μg/g TOC, respectively (Fig. 5d). In addition, abundances of C27–C33 αβ-hopanes displayed a wide range from 7.0 to 326.5 μg/g TOC, being highest in Lake Huanggai and exceeding 50 μg/g TOC in Lakes Wushan, Daye, Donghu, and Futou (Fig. 5e). Abundances of UCM ranged from 0.1 to 12.1 mg/g TOC, being highest in Lake Huanggai, followed by Lakes Daye, Donghu, and Futou, at 6.8, 4.3 and 4.0 mg/g TOC, respectively (Fig. 5f).

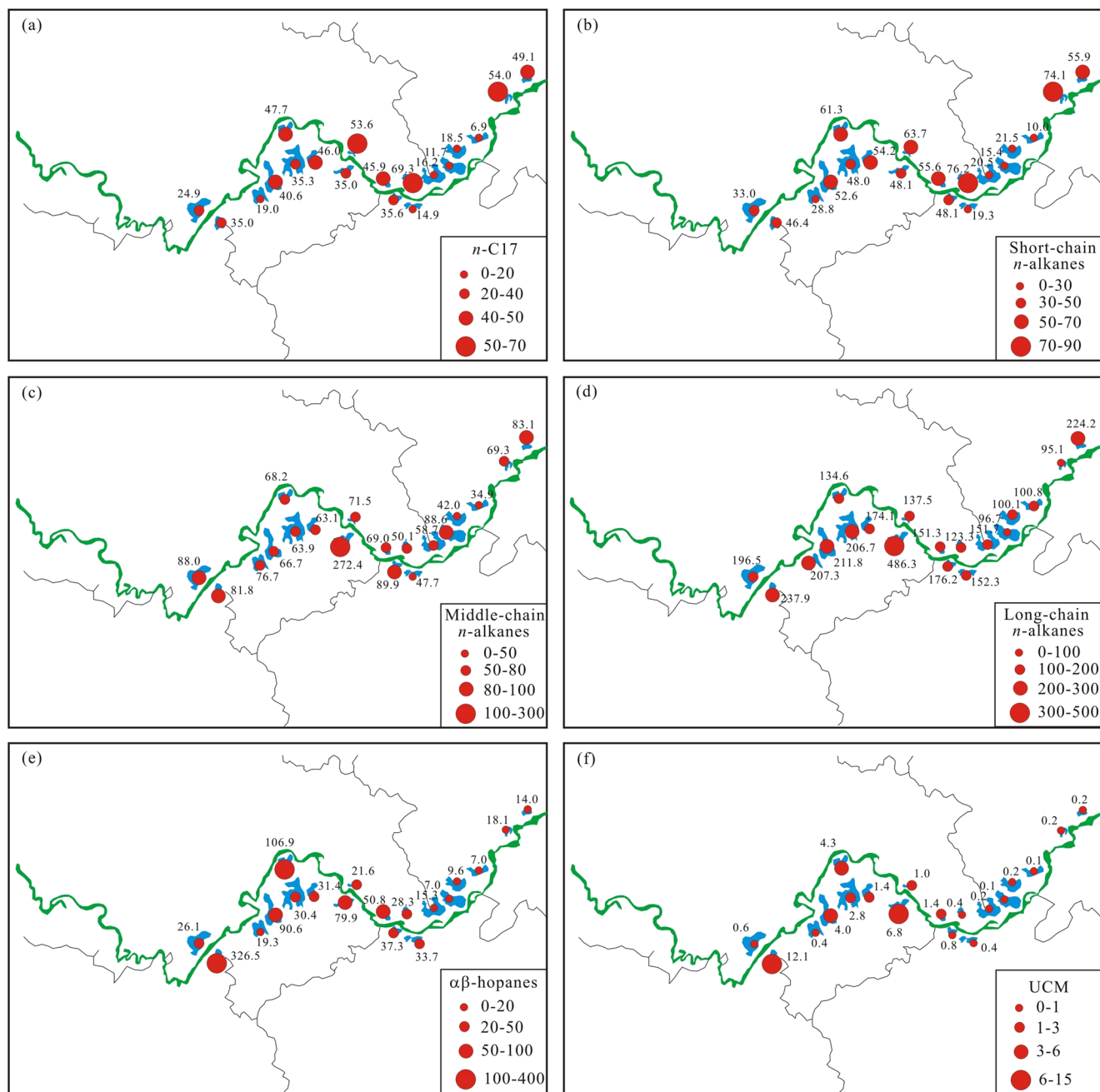


Fig. 5 Concentrations of *n*-C17 (a), short-chain *n*-alkanes (C15, C17, and C19) (b), middle-chain *n*-alkanes (C21, C23, and C25) (c), long-chain *n*-alkanes (C27, C29, C31, and C33) (d), $\alpha\beta$ -hopanes ($\mu\text{g/g}$ TOC) (e), and UCM (mg/g TOC) (f) in surface sediments of studied lakes

Discussion

Sources of *n*-alkanes in sediments

Short-chain *n*-alkanes

Short-chain *n*-alkanes (< C20) arrive in lake sediments from diverse sources, which can be divided into two major categories: petroleum pollution and biogenic inputs (Meyers 2003). A general lack of predominance of odd carbon number over

even-numbered chains with CPI values close to unity suggests the provenance of the sediment *n*-alkanes is predominantly petroleum-based, for example from fossil fuel emissions (Doskey 2001). The short-chain *n*-alkanes identified in the current study were dominated by *n*-C17, with average CPI_{15-20} values of > 4.0 (Fig. 4a). This suggests the majority are derived from biogenic sources rather than petroleum pollution (Hu et al. 2009; Liu et al. 2013). The lower CPI_{15-20} (< 3.0) calculated for Lakes Pogang, Daguan, Longgan, Wuchang, and Daye (Fig. 4a) was probably indicative of the

presence of petroleum pollution, because the CPI of a biogenically derived hydrocarbon assemblage is reduced by the addition of petroleum residues (Doskey 2001). Interestingly, however, the proportions of *n*-C16, a significant component of petroleum hydrocarbons, were lower than 1% in these samples (Fig. 3). Such low values are generally taken as an indication of unpolluted sources of short-chain *n*-alkanes (Gao and Chen 2008). Thus, the low CPI_{15–20} values in these lakes appear to have resulted from *n*-alkane inputs from diverse biogenic sources. The available data indicate that *n*-C17 is abundant in most types of planktonic algae including diatoms (e.g., *Rhizosolenia setigera*), dinoflagellates (e.g., *Peridinium trochoideum*), euglenophyceae (e.g., *Eutreptiella* sp.), and green algae (e.g., *Derbesia tenuissima*), while *n*-C15 is comparatively lower in these algae, or indeed absent in the case of diatoms (Blumer et al. 1971). The *n*-alkane profiles of benthic algae are dominated by either *n*-C15 or *n*-C17 (Youngblood et al. 1971). In cyanobacteria, *n*-C17 tends to be most abundant, followed by *n*-C18 and lower still in *n*-C19 (Zhang et al. 2017). Compared with algae and cyanobacteria, the *n*-alkanes in bacteria are dominated by even carbon number compounds, peaking at either C18 or C20 (Han and Calvin 1969; Fang et al. 2014). In addition, fungi and yeast species have also been reported to biosynthesize even carbon *n*-alkanes (Elias et al. 2000). Overall, the short-chain *n*-alkanes in the current study were extremely rich in *n*-C17 and relatively deficient in *n*-C15 (Fig. 3). This, together with the high CPI_{15–20} values, e.g., > 6.0 in Lakes Bohu, Cehu, Taibai, and Baoan (Fig. 4a), implies a significant contribution from algal *n*-alkanes, especially diatoms, to the tested sediments. The much lower CPI_{15–20} value of 2.62 calculated for Lake Pogang (Fig. 4a) is a result of the high proportion of *n*-C18, approximately five times the average value for the investigated lakes. Short-chain *n*-alkanes in Lake Pogang showed a gradually declining trend in abundance from C17 to C19 (Fig. 3), supporting the idea that most of the *n*-alkanes present have a cyanobacterial origin. However, the scarcity of strong signals of cyanobacterial input in Lakes Daguan, Longgan, Wuchang, and Daye suggests that the low CPI_{15–20} values seen there reflect mixed inputs of short-chain *n*-alkanes from algae and bacteria.

Middle- and long-chain *n*-alkanes

As with short-chain *n*-alkanes, > C21 *n*-alkanes in lake sediments might originate from petroleum pollution or biogenic input, or both, but the relative significance of the two sources in a given lake can be evaluated by the distributions of various *n*-alkane forms (Meyers 2003). While sediment *n*-alkanes of petroleum source have little odd/even predominance and exhibit CPI_{24–34} values close to 1.0, those derived from biogenic input show a strong predominance of odd carbon compounds (Kennicutt II et al. 1987; Bourbonniere et al. 1997; Gao et al.

2007; Liu et al. 2013; Chevalier et al. 2015). Terrestrial higher plant-derived *n*-alkanes are extremely rich in *n*-C27, *n*-C29, *n*-C31, and *n*-C33, with a CPI_{24–34} of 4–10 (Doskey 2001; Harji et al. 2008; Hu et al. 2009). Sediments with CPI values lower than those of purely terrestrial plant waxes point to inputs of biogenic *n*-alkanes that was affected by additional contributions (Doskey 2001), and large reductions in CPI_{24–34} tend to indicate dominant input of petroleum hydrocarbons (Gao and Chen 2008). CPI_{24–34} levels calculated in the current sediments suggested diverse sources of > C21 *n*-alkanes (Fig. 4b). The minimum value of 1.26 in Lake Daye suggests that *n*-alkanes there were derived mainly from petroleum products. Moderate values from 3.0 to 4.0 in six lakes such as Lake Baidang (Fig. 4b) reflected mixed sources of *n*-alkanes dominated by biogenic inputs, while the higher values > 4.0 in the other 12 lakes (Fig. 4b) suggest their sediment *n*-alkanes are mostly biogenic in origin (Meyers 2003). This interpretation of CPI_{24–34} values is supported by the variation in *n*-alkane abundances observed in sediments. Levels of middle-chain (C21, C23, and C25) and long-chain (C27, C29, C31, and C33) *n*-alkanes in Lake Daye reached 272.4 and 486.3 µg/g TOC, respectively, approximately 4 and 3 times higher respectively than the average abundances recorded elsewhere (Fig. 5c, d), suggesting a distinctly different source of > C21 *n*-alkanes for this particular lake. The calculated 78% of *n*-C27, 74% of *n*-C29, and 84% of *n*-C31 from petroleum input probably overestimated, as partial even carbon *n*-alkanes (> C21) can be derived from eroded soils or higher plant itself (Bourbonniere and Meyers 1996). But the exceptionally high values relative to those elsewhere were much more likely to indicate the predominance of petroleum origin. The organisms contributing to sediment > C21 *n*-alkanes can be further assessed using the proxy Paq. Paq values < 0.1 indicate *n*-alkanes mainly from terrestrial plants, but the proxy rises to 0.1–0.4 when the majority of compounds are derived from emergent macrophytes. Paq values of 0.4–1 indicate material originating in submerged and floating macrophytes (Ficken et al. 2000; Routh et al. 2009). In the lakes studied here, Paq levels exceeded 0.4 in Lakes Pogang, Daguan, Chihu, Cehu, Wushan, and Honghu (Fig. 4c), indicating a mainly submerged and floating macrophyte origin of the > C21 *n*-alkanes, but a small contribution from terrestrial plants and emergent macrophytes cannot be excluded (Xiong et al. 2010). This conclusion was consistent with field observations where submerged and floating macrophytes were observed in the 200 × 200 m sampling areas. In a further 11 lakes in the current study, including six (Baidang, Longgan, Wuchang, Taibai, Bohu, and Saicheng) characterized by an absence of aquatic macrophytes, Paq was lower than 0.4 (Fig. 4c). In the six lakes above, terrestrial higher plants appear to be the principal contributors of > C21 *n*-alkanes (Ficken et al. 2000). In the remaining five lakes (Baoan, Liangzi, Futou, Xiliang, and Huanggai; Fig. 4c), Paq varied from 0.3 to 0.4, slightly lower

than expected, on account of the abundant aquatic macrophytes present in the sampling areas. The *n*-alkanes produced by emergent macrophytes typically have Paq values of 0.1–0.4 (Ficken et al. 2000; Routh et al. 2007; Xiong et al. 2010), and so the massive growths of emergent macrophytes such as *Nelumbo nucifera* observed in the sampling areas provide a plausible explanation for the low Paq values recorded. However, an alternative explanation might be an elevated contribution from terrestrial grasses, evidenced by the higher C31 + C33/C27 + C29 *n*-alkane ratio in these lakes (Zech et al. 2013). Overall, the most likely scenario is a mixed source for the > C21 *n*-alkanes. For example, emergent macrophytes and terrestrial plants may have contributed the majority of long-chain (> C27) *n*-alkanes, while the submerged and floating macrophytes may contribute the abundant middle-chain (C21–C25) *n*-alkanes (Zhang et al. 2018b). The higher than expected (> 0.4) Paq value calculated for non-macrophyte Lake Donghu (Fig. 1) was probably caused by the input of OM from more complex sources from the surrounding conurbation of Wuhan City. Unlike other non-macrophyte lakes, the > C21 *n*-alkanes in Lake Donghu exhibited a C_{max} at C27, also implying some differences in OM sources between lakes.

Anthropogenic effects on sediment *n*-alkanes

Eutrophication

It is widely accepted that eutrophication has a profound influence on the productivity of *n*-alkane-producing organisms, including algae, photosynthetic bacteria and aquatic macrophytes, resulting in changes in abundances and composition of the various *n*-alkanes preserved in sediments (Bourbonniere and Meyers 1996; Meyers 1997, 2003; Routh et al. 2007, 2009; Bechtel and Schubert 2009; Choudhary et al. 2009; Lu and Meyers 2009; Xiong et al. 2010; Brag e et al. 2013; He et al. 2015; Zhang et al. 2018a). For example, in Lake Erie, the total *n*-alkanes in sediments increased quickly over the decades from 1850 to 1972 in which excessive nutrients were delivered to the lake then declined with the restoration of the lake to a less eutrophic status (Lu and Meyers 2009). In sediments, sequences from Lake Norrviken in Sweden, both short-chain (C15, C17, C19, and C21) and long-chain (C27, C29, and C31) *n*-alkanes display an elevated trend concomitant with nutrient loading (Routh et al. 2009). In Lake Changdang, however, long-chain *n*-alkane (C27, C29, C31, and C33) abundances appear to be reduced during the period of eutrophication, while the opposite trend is apparent for short-chain (C15, C17, and C19) and middle-chain (C21, C23, and C25) *n*-alkanes (Zhang et al. 2018a). Water column monitoring data from the current study indicates that Chla levels vary in a pattern similar to TP but different from TN (Fig. 2), pointing to P as the key nutrient for aquatic primary producers (Wetzel 2001). Thus, abundances

of TP can be regarded as a proxy for trophic status in these lakes. Our results show a moderate correlation between increasing *n*-C17 in lake sediments and TP in the water column ($R^2 = 0.31$, $n = 19$). Abundances of short-chain *n*-alkanes (*n*-C15, *n*-C17, and *n*-C19) generally show a relatively weak correlation with TP ($R^2 = 0.27$, $n = 19$). The results suggest that eutrophication had a genuine enriching effect on short-chain *n*-alkanes in sediments, although other factors should be also considered in view of the small R^2 . A causal relationship between TP and short-chain *n*-alkanes could be that elevated levels of P fuel phytoplankton growth, resulting in increased production of short-chain *n*-alkanes, especially *n*-C17 (Smith et al. 1999; Meyers 2003). Moreover, cyanobacterial blooms driven by eutrophication might weaken the usual odd carbon predominance of the short-chain *n*-alkanes, because the quantities of *n*-C18 produced by cyanobacteria are almost as great as those of *n*-C17 (Fig. 3). Theoretically, elevated algal productivity forced by nutrient loading is detrimental to the growth of submerged/floating macrophytes (Scheffer et al. 1993) and thereby leads to reduced inputs of the middle-chain *n*-alkanes (*n*-C21, *n*-C23, and *n*-C25) produced by these organisms (He et al. 2015). The results of our field survey, however, suggest a considerable tolerance to P loading by submerged/floating macrophytes, which were still observed in the water column when TP reached 0.23 mg/L (Figs. 1 and 2a). For this reason, TP does not show any correlation with middle-chain *n*-alkane abundance, signifying a lack of impact from eutrophication on these compounds. Instead, with the exception of Lake Daye, the average levels of middle-chain *n*-alkanes in macrophyte lakes are 37% higher than in non-macrophyte lakes, suggesting that inputs of middle-chain *n*-alkanes to sediments are controlled mainly by organism type. Long-chain *n*-alkane abundance values were lower than 160 $\mu\text{g/g}$ TOC in the three most nutrient-enriched lakes, in which TP > 0.1 mg/L. Conversely, the values were generally high in the sediments of lakes with comparatively low levels of nutrients, including Baidang, Liangzi, Futou, Xiliang, and Huanggai (Fig. 5d) where TP varied from 0.03 to 0.09 mg/L and averaged at 0.05 mg/L. These same lakes were also characterized by a rising values of C31 + C33/C27 + C29 *n*-alkane ratio, an indicator of relatively high inputs from terrestrial grasses (Zech et al. 2013), as a consequence of covering the surrounding catchments by dense grasses, and the massive growth of grasses could maintain P in the soil and reduce the input of P to lakes.

Petroleum pollution

During the past century, the *n*-alkane signature of petroleum hydrocarbons from anthropogenic pollution has greatly increased in the sediments of seas and estuaries worldwide (Medeiros et al. 2005; Gao et al. 2007; Zaghden et al. 2007; Gao and Chen 2008; Sojinu et al. 2012; Pisani et al.

2013; Silva et al. 2013; Wang et al. 2018). In the current study, however, petroleum-derived *n*-alkanes were only found to reach a level sufficient to mask biogenic input in Lake Daye (Gao et al. 2007; Zhang et al. 2018b). This unique situation might be related to direct petroleum pollution from mining and smelting industries around the lake. The catchment contains 12 metal mines for extraction of copper and iron (Zhang et al. 2014), and direct discharges of petroleum products used in mining and smelting into the lake have been a routine (Wang et al. 2018). The speed and directness of these inputs allow little opportunity for *n*-alkanes derived from petroleum to degrade before reaching the sediments, though more rapidly degraded short-chain *n*-alkanes (< C20) are more likely to be lost than their long-chain homologs (Cranwell 1984; Routh et al. 2007). Compared with the situation in Lake Daye, petroleum hydrocarbon pollution in other lakes of the current study is more indirect, coming mainly from surface runoff of vehicle emissions associated with urbanization (Hostettler et al. 1999; Rushdi et al. 2016). These compounds are mainly composed of geologically configured $\alpha\beta$ -hopanes and UCM (Zhang et al. 2018b), with *n*-alkanes making up a smaller proportion of the total hydrocarbons because *n*-alkanes are much more effectively degraded by long-term transport than $\alpha\beta$ -hopanes and UCM, which are comparatively resistant to microbial degradation and persist much longer in the environment (Lytle et al. 1979; Fang et al. 2014). For example, in Lakes Huanggai, Donghu, and Futou, abundances of $\alpha\beta$ -hopanes and UCM exceeded 90 $\mu\text{g/g}$ TOC and 4 mg/g TOC (Fig. 5e, f), but their

CPI_{24–34} values were just a little lower than those in higher plants (4–10) (Hu et al. 2009). Thus, the amounts of *n*-alkanes associated with petroleum input in these lakes are not large enough to overwrite the biogenic characteristics of sediment *n*-alkanes, despite massive discharges of petroleum products into the lakes. Instead, the input is reflected in a reduced odd carbon predominance in sediment *n*-alkanes, creating a relatively small CPI, similar to those observed in other Chinese lakes (Xiong et al. 2010; Fang et al. 2017; Zhang et al. 2017, 2018b). For example, the CPI of long-chain *n*-alkanes (> C24) in Lake Kuncheng is 2.06 as a result of petroleum pollution from shipping (Zhang et al. 2018b). The values in Lakes Taihu (northern area), Chaohu, and Dianchi, all infamous for nutrient pollution and cyanobacterial blooms, were 2.25, 3.02, and 2.40, respectively, due to introduction of petroleum *n*-alkanes by sewage discharge (Xiong et al. 2010; Zhang et al. 2017, 2018b). Lower CPI values are also found in some other eutrophic lakes, for example, 2.8 in Chenghai, 2.9 in Sugan, and 3.5 in Erhai (Fang et al. 2017), but these values reflect the biogenic origin of long-chain *n*-alkanes, because direct inputs of petroleum products, such as from heavy industry, have been much more closely controlled in the associated catchments during the past decade (Wu et al. 2012). The input of *n*-alkanes from petroleum versus biogenic sources in the current sediments was also described in a ternary plot (Fig. 6), constructed by relative variation of total abundances in odd-carbon *n*-alkanes, even-carbon *n*-alkanes and $\alpha\beta$ -hopanes. The distribution of the data points clearly exhibited the *n*-alkanes in Lake Daye (Fig. 6) and reflected

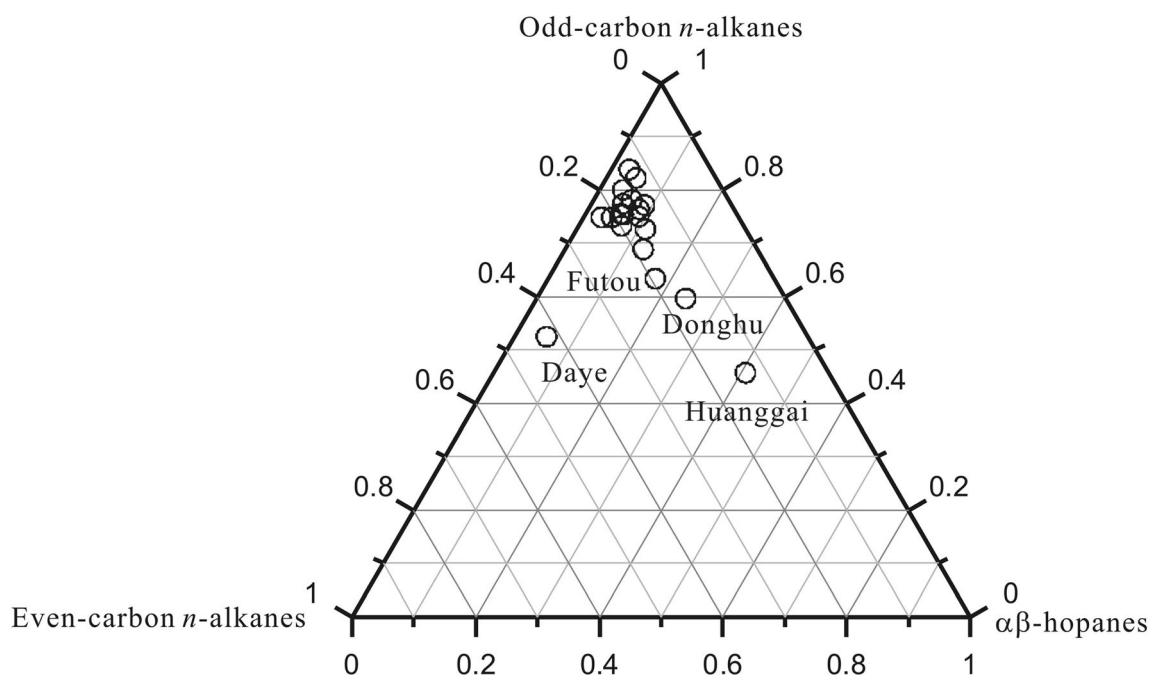


Fig. 6 Ternary plot showing the relative variation of total abundances in odd-carbon *n*-alkanes, even-carbon *n*-alkanes, and $\alpha\beta$ -hopanes

input from fresh petroleum products, while those in other lakes were mainly biogenic in origin. Indirect input of petroleum products, as shown in Lakes Huanggai, Donghu, and Futou (Fig. 6), will not drive a significant variation in *n*-alkanes.

Conclusion

The results of the current study demonstrate the impact of human activities on the abundances and composition of *n*-alkanes in sediments of lakes across the MYR region. The massive growth of algae and photosynthetic bacteria driven by anthropogenic eutrophication has led to enhanced production and accumulation of short-chain (< C20) *n*-alkanes in sediments, but elevated nutrient loading has had little influence on the middle-chain (C21, C23, and C25) *n*-alkanes produced by submerged/floating macrophytes. Anthropogenic petroleum pollution has exerted influence on *n*-alkanes with carbon chains longer than C20. Direct inputs of petroleum fuels have increased the abundances of *n*-alkanes greatly, largely eliminating the odd carbon predominance associated with previously the dominant biogenically derived *n*-alkanes. Conversely, indirect inputs of petroleum hydrocarbons have less of an effect on sediment *n*-alkane profiles, leading to only slight reductions in the odd carbon predominance. The results presented here suggest that sediment *n*-alkane proxies might be used to assess anthropogenic impacts on lake systems, including eutrophication and petroleum pollution. In particular, the CPI values of > C21 *n*-alkanes can be used to evaluate pollution of lakes by fresh petroleum products or aged materials in the catchment. Moreover, this investigation gives a new view of the current impacts of nutrient and petroleum pollution on lakes across the MYR region. The obtained results may inform environmental monitoring and management in the near future.

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