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RESEARCH ARTICLE

Unraveling the sources and fluorescence compositions of dissolved and particulate organic matter (DOM and POM) in Lake Taihu, China

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Abstract

Organic matter (OM), a complex entity with diverse functional groups and molecular sizes, has important effects on aquatic systems. We studied the optical compositions and sources of dissolved organic matter (DOM) and particulate organic matter (POM) in Lake Taihu, a large, shallow and eutrophic lake in China. Significant differences in optical compositions and sources occurred between the POM and DOM. The temporal–spatial distribution of the fluorescence indices suggested that the POM in Lake Taihu was mainly from autochthonous sources, but more exogenous characteristics were shown in POM in the river mouths compared with other regions. The chromophoric DOM in Lake Taihu mainly displayed autochthonous characteristics. The POM–DOM PARAFAC model was used to examine OM optical composition and five components were identified, which contained three protein-like components (C1, C2, and C5), a microbial humic-like component (C3), and a terrestrial humic-like component (C4). The POM was dominated by C5 in summer and autumn and C3 in winter and spring, and the DOM was dominated by protein-like components (C1, C2, and C5) through the entire year. The algae-dominated region had a relative higher contribution of tryptophan-like components of POM compared with the macrophyte-dominated region. A conceptual model based on the theory of "four phases of cyanobacteria bloom development" was proposed to fully describe the relationship between POM–DOM exchanges and cyanobacteria bloom development.

Keywords Dissolved organic matter (DOM) · Particulate organic matter (POM) · Eutrophication · Lake Taihu · Fluorescence compositions · POM–DOM PARAFAC model

Introduction

Eutrophication is one of the most important ecological environmental problems with which many lakes in China, especially those in the middle and lower reaches of the Yangtze River, are facing. As an important source of energy and nutrient, organic matter (OM) has great impact on the biological activities of phytoplankton and bacteria in aquatic ecosystems (Wu et al. 2010). First, organic matter, rich in carbon, nitrogen, phosphorus, and other biogenic elements, has numerous geochemical and ecological functions in all kinds of water bodies, which is highly valued in ecological studies of both marine and inland environments (Margot et al. 2018). Second, the formation, transformation, and fate of OM and its accompanying nutrient regeneration process are key links in the biogeochemical cycling of nutrients in aquatic ecosystems. Third, OM, as a provider of nutrients, also plays significant role during the outbreak of algal blooms in inland lakes (Simon et al. 2002; Turner 2002). Lake Taihu, the third largest freshwater lake in China, is not only one of the main sources for nearby residents' drinking water but also one of the most severely polluted freshwater reservoirs in China as the algal blooms have become a critically important issue (Yao et al. 2011).

The OM is categorized and quantified as dissolved organic matter (DOM) and particulate organic matter (POM) based on filtering with a filter pore size of 0.7 μm

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typically used to set the operational boundary between these two components (Verdugo et al. 2004; Roulet and Moore 2006; Azam and Malfatti 2007; He et al. 2016a). The existing form, composition, and chemical characteristics of OM are changed by photochemical degradation, enzymatic hydrolysis, bacterial degradation, and other factors involved in degradation and transformation (McCallister et al. 2006). These processes and factors may also affect not only the transformation of POM into more biologically available DOM and inorganic nutrients but also the role of OM in biogenic circulation in lakes (Mayer et al. 2009). Thus, the compositional differences between POM and DOM should be determined to yield information that can be used to understand the phase transfer between POM and DOM pools, as well as the water quality in lakes. While numerous studies have investigated the optical compositions and sources of DOM, previous studies have not experimentally identified the optical compositions and sources of POM in Lake Taihu, mainly due to a lack of suitable techniques for studying its fluorescence components.

Combined with parallel factor analysis (PARAFAC), the excitation–emission matrix (EEM) fluorescence was used to model base-extracted POM and DOM quality in the Neuse River Estuary, North Carolina, before and after passage of Hurricane Irene in August 2011 and a combined POM–DOM PARAFAC model was developed (Osburn et al. 2012). This model could simultaneously interpret the fluorescence components of POM and DOM, which could qualitatively and quantitatively analyze the content of DOM in the process of POM degradation and transformation, content of residual POM, and composition change of POM (Osburn et al. 2012; Zhang et al. 2013). Yang et al. (2014) revealed the changes in biogenic POM and biogenic DOM derived from wastewater treatment plants during photoexposure. Osburn et al. (2015) observed seasonal variation in the quality of dissolved and particulate organic matter exchanged between a salt marsh and its adjacent estuary. Larsen et al. (2015) tracked the source of organic sediment in restored and unrestored urban streams. However, seldom studies, according to our literature survey, used the combined POM–DOM PARAFAC model to examine POM optical composition in inland lakes of China. To address this lack of information, this model was first applied to evaluate the POM and DOM pools in Lake Taihu from August 2014 to May 2015. The specific objectives of this study were (1) to characterize the similarities and differences of the spatial–temporal variations of the optical components and sources between POM and DOM in different ecological regions of Lake Taihu and (2) develop a conceptual model based on the dynamic exchanges between POM and DOM pools for Lake Taihu.

Material and methods

Study area

Lake Taihu, located in the Yangtze River Delta, has an area of 2338.1 km^2 (Qin et al. 2007), a mean depth of 1.9 m (maximum depth < 3.0 m), and a water retention time of approximately 300 days. The lake has a complicated river and channel network by connected to 219 rivers and channels (Qin et al. 2007). Most of the water enters the lake from west and flows out, primarily through Eastern Taihu Bay, to the east (Zhang et al. 2016).

Due to its location in the most developed region in China, Lake Taihu receives excessive nutrient input from the upstream watershed, leading to algal blooms appearing persistently in the summer for the last three decades (Zhou et al. 2016). The accumulation and subsequent decay of massive algal blooms may result in the outbreak of black water events and frequently high concentrations of POM and DOM (Zhu et al. 2013; Zhou et al. 2015a).

Sample collection

Four extensive sampling campaigns, using the same 32 monitoring sites in Lake Taihu, were conducted in August and November of 2014, and February and May of 2015, which were representative of summer, autumn, winter, and spring, respectively (Fig. 1). The 32 sites, with 18 sites in the Northern Lake, 1 in the transitional zone in the center of the lake, and 13 in the Southern Lake, were evenly distributed in the lake, which were also same with the routine monitoring sites for the Taihu Ecosystem Research Station of the Chinese Academy of Sciences. The division of the Northern and Southern Lakes referred to the division method of CDOM enriched and CDOM depleted region presented by Zhou et al. (2015b).

Water temperature (Temp), salinity (Sal), and turbidity (Turb) were determined in situ at 0.5 m depth using a YSI 6600 Multi-Parameter Water Quality sonde. A total of 128 (32 × 4) surface water samples (0~0.5 m) were collected using 5 L acid-cleaned plastic bottles. The samples were kept on ice in the field and filtered immediately once arrival at the laboratory. All samples were filtered within 2 days, and all measurements were completed within 2 weeks.

Fluorescence EEM measurement

The POM was collected on the pre-combusted glass fiber filters (Whatman GF/F) with 0.7 μm in porosity and 25 mm in diameter. The filtrate was used for DOM analyze (Osburn et al. 2012). POM fluorescent material was extracted into 10 mL of 0.1 N NaOH for 24 h in the dark at 4 °C. After neutralization with HCl to pH > 6, the resultant solution was

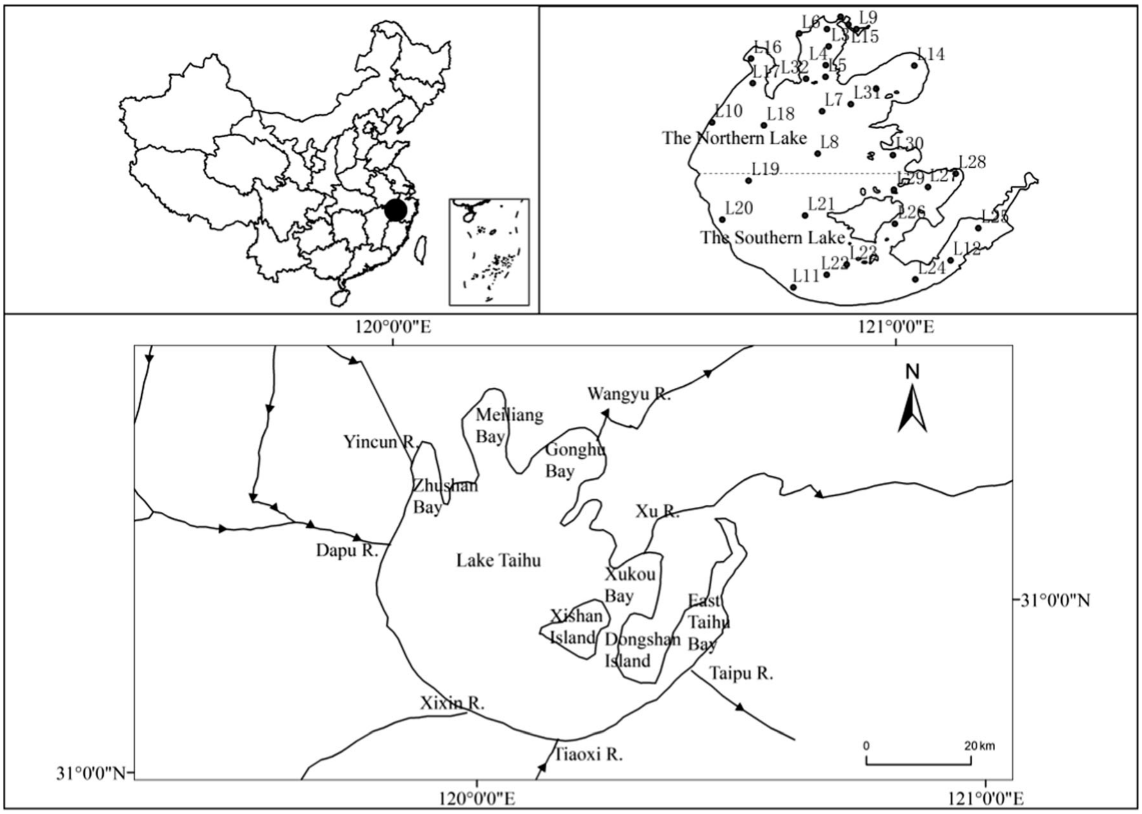

Fig. 1 Map of Lake Taihu showing locations of sampling sites, which were divided into the Northern Lake (sites L 1–10, L 13–14 and L 30–31) and the Southern Lake (sites L 11–12 and L 19–29)

filtered through 0.2 μm polyethersulfone filters (Millipore Sterivex), and then fluorescence was measured.

Fluorescence measurements were carried out using a Hitachi F-7000 fluorescence spectrometer (Hitachi High-Technologies, Tokyo, Japan) with a 700-voltage xenon lamp. Excitation and emission spectra were obtained from 200 to 450 nm (5 nm intervals) and 250–600 nm (1 nm intervals), respectively, and the slit width for both excitation and emission was set to 5 nm. The excitation–emission matrices (EEMs) were Raman calibrated by subtracting daily EEM of Milli-Q water blanks from EEMs of OM solution. All EEMs were modeled with PARAFAC using the DOMFluor toolbox in MATLAB.

Measurements of other parameters

Particulate organic carbon (POC) and particulate nitrogen (PN) were measured using a PerkinElmer 2400 Series II elemental analyzer. Data of seven other parameters, i.e., chlorophyll a (Chl*a*), dissolved organic carbon (DOC), total phosphorus (TP), dissolved organic phosphorus (DOP), total nitrogen (TN), dissolved organic nitrogen (DON), and chemical oxygen demand (COD), were obtained from Taihu Ecosystem Research Station of Chinese Academy of Sciences, with the determination method based on the "Manual for investigating lake eutrophication" (Jin and Tu 1990).

Calculation of fluorescence index

To characterize the sources of the OM and investigate the relative contribution of different sources to the fluorescence intensity of the OM, we used FI (fluorescence index), HIX (humification index), and BIX (biological index) as fluorescence indices. The FI is defined as a ratio of fluorescence intensity at 470 and 520 nm, both excited at 370 nm, and has a value of ~ 1.4 for terrestrially derived OM and a value of ~ 1.9 for microbially derived endogenous fulvic acids (Cory and McKnight 2005). The HIX, a humification index that indicates the degree of OM humification (Zhou et al. 2015b), is defined as the ratio of integral fluorescence intensity at wavelengths 435–480 to 300–345 nm, both excited at 225 nm. Generally, autochthonous and allochthonous sources can be identified if the HIX is below 4 or exceeds 6, respectively (Ohno 2002). The BIX, a biological index that denotes autochthonous biological intensity (Huguet et al. 2009), is defined as the ratio of fluorescence intensity at 380 and 430 nm, both excited at 310 nm. Values of BIX values ≥ 0.4 are related to a predominantly autochthonous origin of OM and to the presence of freshly produced OM, whereas low BIX values (< 0.4) correspond to terrestrial origin (Huguet et al. 2009).

Statistical analyses

Statistical indicators (i.e., the mean value, standard deviation, and *t* test) were analyzed using R-Studio software v.0.97.551. Spatial distribution of sampling sites and OM-related parameters were undertaken using ArcGIS 10.2 and MATLAB R2014a software, respectively. Results of linear regression and *t* test analyses with $p < 0.05$ are reported as significant.

Results

Spatial and temporal variations in water quality parameters

Strong seasonal variation of the TN concentrations was presented in Lake Taihu with the maximum values occurred in winter and spring and the minimum values occurred in summer and autumn, which was consistent with Xu et al. (2010). Spatial variation of TN was also shown in Lake Taihu that with a mean of 2.22 ± 0.12 mg L^{-1}, the TN concentrations ranged from 0.51 mg·L^{-1} in Xukou Bay (the Southern Lake) to 7.63 mg·L^{-1} in Zhushan Bay (the Northern Lake) (Fig. 2a).

The TP concentrations, with a mean of 0.11 ± 0.01 mg L^{-1}, ranged from 0.02 mg L^{-1} in Xukou Bay to 0.32 mg L^{-1} in Zhushan Bay (Fig. 2b). Maximum values were in summer and spring, and minimal values were in winter and autumn. As the asynchronous dynamics of TN and TP, the mass ratio of TN to TP in Lake Taihu presented a seasonal pattern with TN/TP = 27–32 in winter and spring and 14–16 in summer and fall.

The Chl*a* concentrations, ranging from 2.10 μg L^{-1} in Xukou Bay to 298.80 μg L^{-1} in Zhushan Bay, had a mean value of 28.65 ± 3.57 μg L^{-1} (Fig. 2c). Maximum values occurred in summer and spring, and minimum values occurred in winter and autumn. Chl*a* values were significantly higher in Zhushan Bay than in other areas (*t* test, $p < 0.01$).

The COD concentration in Zhushan Bay (i.e., 17.07 mg·L^{-1}) was significantly higher than other areas (*t* test, $p < 0.01$; Fig. 2d). A similar seasonal pattern for the COD concentration was showed with TP, which reached maximum in summer and then decreased in winter. In general, the concentrations and variations of the nutrients mentioned above (i.e., TN, TP, Chl*a*, and COD) were significantly higher in the Northern Lake than in the Southern Lake (*t* test, $p < 0.01$).

The DOC concentration, ranging from 2.5 mg L^{-1} in Gonghu Bay to 6.3 mg L^{-1} in Zhushan Bay, had a mean value of 3.92 ± 0.06 mg·L^{-1} over the entire lake (Fig. 2e). Maximum values were in summer and winter, and minimum values in spring and autumn.

The POC concentration averaged over the entire lake was 4.10 ± 0.43 mg L^{-1} (Fig. 2f), which is consistent with Ye et al. (2017) that a mean value of 4.66 ± 5.73 mg L^{-1} was presented in Lake Taihu. The POC/(POC + DOC) ranged from 14.47% in Zhushan Bay to 87.22% in Gonghu Bay, with a mean of $40.37 \pm 0.01\%$. The POC/(POC + DOC) exhibited a similar seasonal pattern to POC, with peak occurring in summer and spring. The DOC and POC concentrations and seasonal variations in the Northern Lake were significantly higher than that in the Southern Lake (*t* test, $p < 0.01$).

To determine whether the POC of Lake Taihu was primarily derived from allochthonous or autochthonous sources, the ratio POC/Chl*a* was calculated for all seasons. The POC/Chl*a* ratios ranged from 51:1 in autumn to 485:1 in winter, with a mean of 200:1 (Fig. 2g). For both northern and southern lakes, POC/Chl*a* increased from summer to winter and then decreased in spring, which was similar to that observed in Jiaozhou Bay in China (Lü et al. 2009). During late autumn and winter, POC/Chl*a* was greater than 200, indicating that higher loading of detrital POC coming into the lake was from external sources (Parsons et al. 1977). However, during summer and spring, the ratio less than 200 suggested most of POC from algal sources within the lake (Cifuentes et al. 1988).

Seasonal variation was presented for the ratio C/N (POC/PN) with the values in autumn and spring significantly lower than summer and winter (*t* test, $p < 0.01$). The C/N in river mouths were significantly higher than other regions (*t* test, $p < 0.01$), and the C/N of POM ranged from 1.78 in Zhushan Bay to 28.43 in the river mouth of Meiliang Bay, with a mean of 5.81 ± 3.59 (Fig. 2h). The relatively lower C/N of POM in Lake Taihu, compared with that of terrestrial higher plants, could be explained by the biodecomposition of both autochthonous POM and allochthonous POM (Ye et al. 2017).

Differences in EEMs between POM and DOM

Previous identified peak regions of EEMs fluorescence are indicated in Fig. 3 which includes their probable origins. Six types of fluorescence peaks containing three humic-like fluorescence peaks (peaks A, C, and M) and three protein-like fluorescence peaks (peaks B, T, and D) were found in OM EEMs in Lake Taihu (Fig. 3). Peaks A and C have traditionally been considered as originating from allochthonous and terrestrial sources, relating to humic acid and fulvic acid (Murphy et al. 2008; Yamashita et al. 2008; Wu et al. 2010; Yao et al. 2011). Peak M is generally assigned to microbial humic-like fluorescence, probably derived from microbial activity, degradation of algae, or human activities (Stedmon and Markager 2005a; Williams et al. 2010). Peaks B and T are considered as tryptophan fluorescence, mainly produced by microbial and phytoplankton, or influenced by microorganisms which were carried by exogenous inputs (Stedmon and Markager 2005b; Kowalczuk et al. 2010; Zhou et al. 2016). Peak D is generally assigned to tyrosine fluorescence and is indicative of recent biological activity (Murphy et al. 2008; Williams et al. 2010).

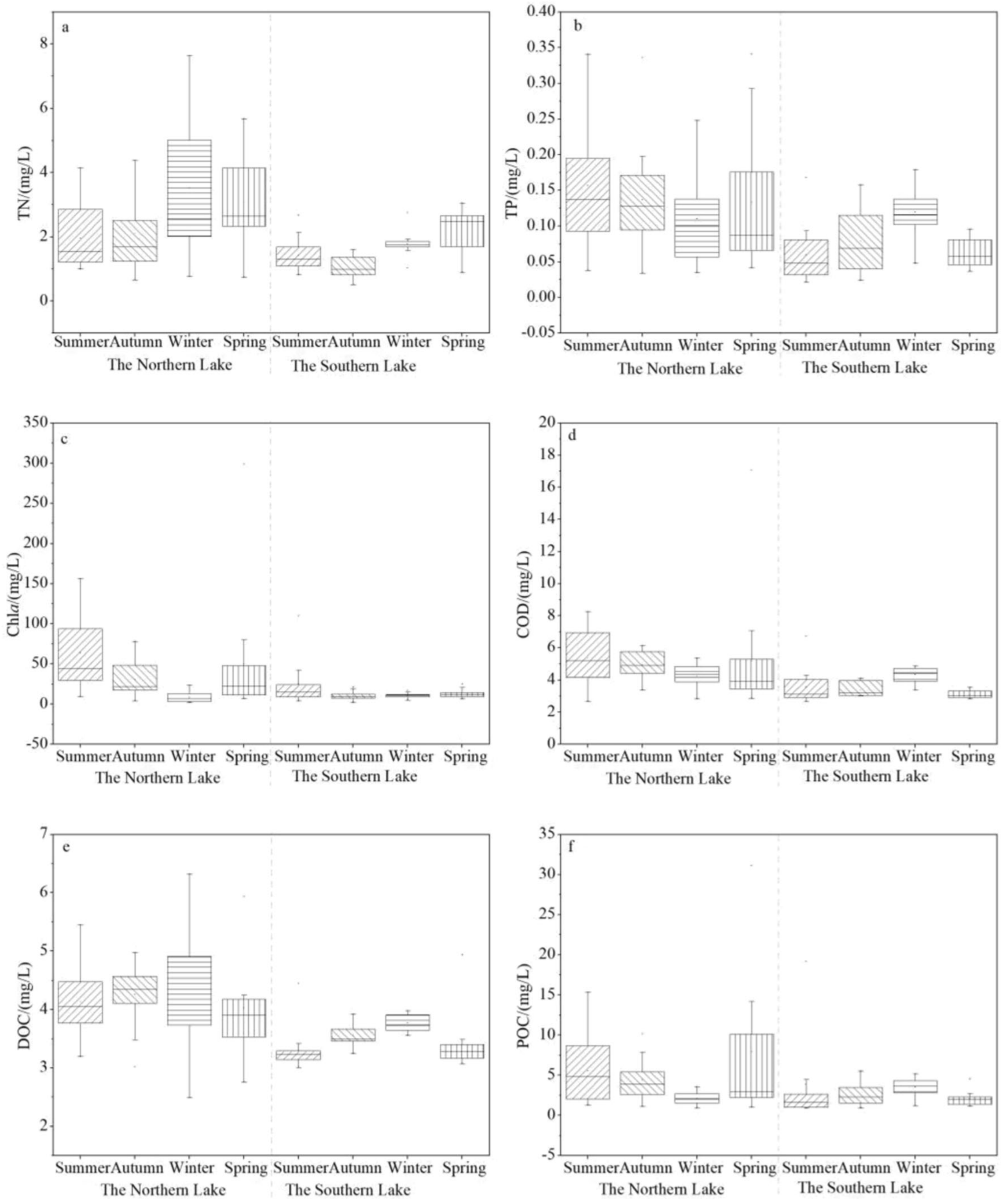

Fig. 2 Box plots of temporal and spatial variation of six water quality parameters in Lake Taihu. **a** TN. **b** TP. **c** Chl*a*. **d** COD. **e** DOC. **f** POC. **g** POC/Chl*a* ratios. **h** C/N ratios

Differences in the number of fluorescence peaks, fluorescence position, and fluorescence intensity existed between POM and DOM, which indicated that POM had more complex components than did DOM. For example, there were six types of fluorescence peaks of POM, but only four types of fluorescence peaks of DOM (Fig. 3). The emission wavelength of peak D of POM in winter was 400 nm (Fig. 3e, f). The position of peak D of POM in winter had an obvious red shift, which may be related to the degradation of POM, whereas the positions of other fluorescence peaks were stable and no obvious shift occurred over the four seasons. The POM was dominated by peak D and M in summer and winter, respectively, and the fluorescence intensity of POM in summer was higher than that in winter (Fig. 3). However, the DOM was dominated by peaks B and D during all seasons, and the fluorescence intensity of DOM in winter was significantly higher than that in summer.

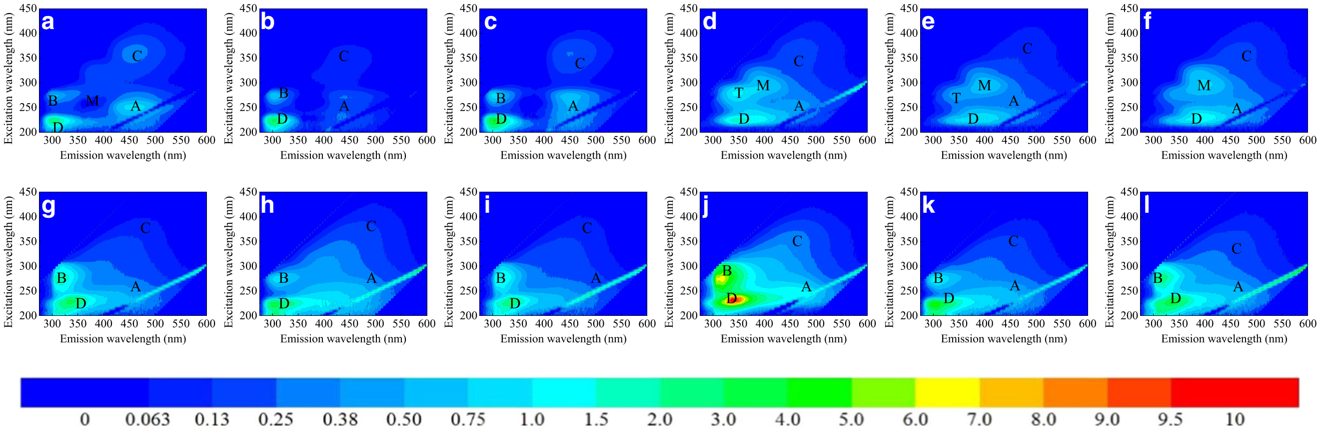

Fig. 3 Examples of EEM fluorescence of POM (**a**–**f**) and DOM (**g**–**l**) from Lake Taihu. The Northern Lake site 6, central lake site 8, and the Southern Lake site 25 samples of POM in summer (**a**–**c**) and winter (**d**–**f**); the Northern Lake site 6, central lake site 8, and the Southern Lake site 25 samples of DOM in summer (**g**–**i**) and winter (**j**–**l**). Major peak regions of EEMs are indicated and identified below the figure

For POM, the types of the fluorescence peaks of the samples collected in the Northern Lake were more than those in the Southern Lake. For example, there were five types of fluorescence peaks at site 6 in the Northern Lake and four types of fluorescence peaks at site 25 in the Southern Lake. In winter, the tryptophan fluorescence peak T was present in the Northern Lake site 6, but not in the Southern Lake site 25. For DOM, the protein-like fluorescence of peaks B and D in the Northern Lake site 6 was stronger than that in the Southern Lake.

Optical compositions analysis of DOM and POM based on DOM–POM PARAFAC model

Five components of the PARAFAC model, i.e., C1–C5 with two humic-like components (C3 and C4) and three protein-like components (C1, C2, and C5), explained over 99.6% of the total EEMs variability, which was well validated using the split-half procedure (Fig. 4). The spectral characteristics of the five components were compared with PARAFAC model analyses conducted for samples from other ecosystems, using the online spectral library "OpenFluor" (Murphy et al. 2014).

The five components (C1–C5) comprised two humic-like components and three protein-like components. Components C1 (Ex/Em = 275 < 225/318 nm) and C2 (Ex/Em = 235/338 (476) nm) were congruent with tryptophan-like fluorophore (peaks T and B) associated with amino acid-like substances (Stedmon and Markager 2005a; Murphy et al. 2008; Yamashita et al. 2008). Components C3 (Ex/Em = < 225(228)/385 nm) and C4 (Ex/Em = 255(365)/455 nm) were categorized as the two traditional types of humic-like fluorescence of marine (microbial) and terrestrial origin respectively (Murphy et al. 2008; Yamashita et al. 2008; Williams et al. 2010; Zhou et al. 2016). The microbial humic-like C3 corresponded to marine fulvic acid peak M, and C4 had a spectral shape similar to the humic-like peaks A and C. The component C5 (Ex/Em = < 225(275)/302 nm) had characteristics of tyrosine-like peak D (Stedmon and Markager 2005b; Murphy et al. 2008; Yamashita et al. 2008).

The amounts of each component, measured as the percentage of the total fluorescence maxima for the five components ($\%F_{\max}$), varied seasonally and spanned from 0 to 96.6% (Table 1). The ranges of $\%F_{\max}$ for C1 (C1p)–C5 (C5p) of POM over the sampling period were 0–42.9%, 0–51.0%, 1.1–66.0%, 1.1–49.4%, and 0.0–96.6%, with means of 7.9 ± 7.5%, 4.0 ± 5.2%, 27.0 ± 21.4%, 14.7 ± 10.9%, and 46.3 ± 31.4%, respectively. The $\%F_{\max}$ of C1p–C5p varied in spatial with the maximum values occurring in the Northern Lake and river mouths and the minimum values confined to the Southern Lake. Seasonal variation in the $\%F_{\max}$ of C1p–C5p occurred in Lake Taihu, and the maximum and minimum values were shown in spring and winter, respectively. The POM pool was dominated by C5p and C3p, with peaks in summer and autumn, and winter and spring, respectively.

The ranges of $\%F_{\max}$ of C1 (C1d)–C5 (C5d) of DOM over the sampling period were 2.1–42.9%, 9.2–51.0%, 4.0–27.3%, 2.1–23.8%, and 0.0–62.0%, with means of 25.8 ± 7.7%, 34.6 ± 7.6%, 11.7 ± 4.3%, 5.8 ± 3.2%, and 22.1 ± 12.3%, respectively. The $\%F_{\max}$ of C1d–C5d of DOM varied seasonally and spatially. Higher values of $\%F_{\max}$ of C1d–C5d were presented in the Northern Lake compared with the Southern Lake. The $\%F_{\max}$ of C1d–C5d revealed an inverse seasonal pattern to POM that the maximum and minimum values occurred in winter and summer, respectively. The DOM pool was dominated by C2d, C1d, and C5d during the whole sampling period.

The contents of the tryptophan-like components (C1p and C2p) in the Northern Lake were significantly higher than in the Southern Lake (*t* test, $p < 0.01$). Moreover, C1p and C2p were significantly higher in winter and spring than in summer and autumn (*t* test, $p < 0.01$).

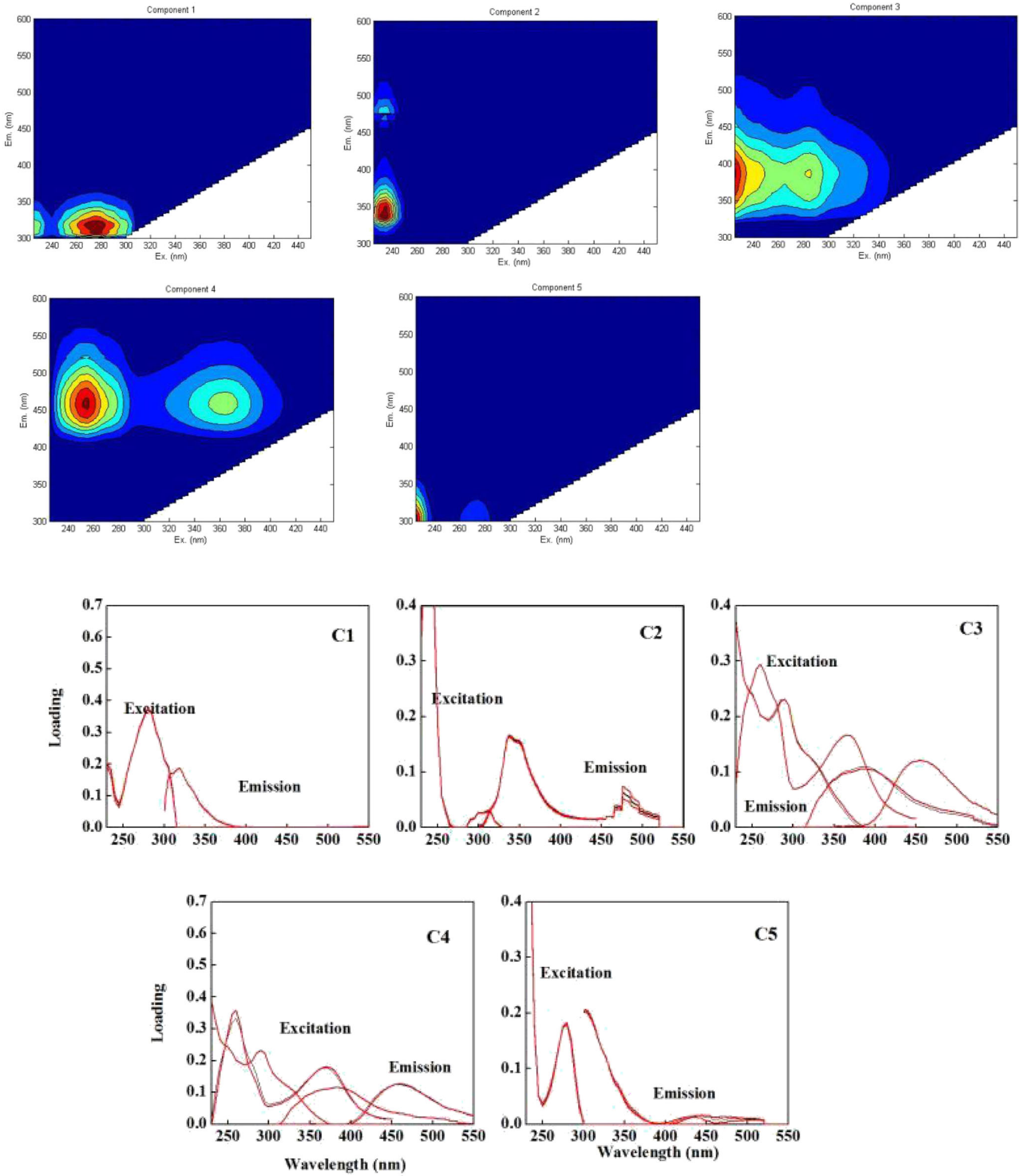

Fig. 4 Five fluorescent components (C1–C5), split-half validated, calculated from the PARAFAC model on POM–DOM EEMs from Lake Taihu. Each EEM was normalized to its maximum fluorescence prior to modeling. The spectral properties of the five components (five upper panels) and the split-half validation results (five lower panels) were given

The protein-like components (C1d, C2d and C5d) dominated the DOM pool during the whole sampling period (Fig. 7e–h). There was no significant difference in the tryptophan-like components (C1d and C2d) between the Northern Lake and Southern Lake. Similar to POM, C1d and C2d in winter and spring were significantly higher than in summer and autumn (*t* test, $p < 0.01$).

Table 1 Relative amount of five components from combined POM–DOM PARAFAC model, for POM and DOM in each of four seasons, and as the average for all four seasons. Data shown are the minimum and maximum for $\%F_{\max}$. C1 and C2: tryptophan-like component; C3 and C4: humic-like component; C5: tyrosine-like component

Fluorescence component		C1	C2	C3	C4	C5
Ex (nm)		275 (< 225)	235	< 225 (285)	255 (365)	< 225 (275)
Em (nm)		318	338(476)	385	455	302
POM	Summer	0–4.8	0–2.2	1.1–18.3	6.3–49.4	32.4–91.3
	Autumn	0–28.1	0–12.4	2.3–18.0	1.1–36.53	33.0–96.6
	Winter	0–21.3	0–15.8	36.8–66.0	1.2–22.5	11.7–26.5
	Spring	0–42.9	0–51.0	1.1–66.0	1.1–49.4	0–96.6
	Average	7.9 ± 7.5	4.0 ± 5.2	27.0 ± 21.4	14.7 ± 10.9	46.3 ± 31.4
DOM	Summer	2.1–31.0	9.6–38.0	7.4–25.8	3.5–23.8	12.0–62.0
	Autumn	5.8–42.9	16.3–51.0	4.0–27.3	2.1–12.3	0–51.0
	Winter	2.6–38.2	9.2–48.6	5.6–19.0	2.5–8.9	0–71.0
	Spring	8.3–35.6	23.6–48.8	5.6–22.4	2.4–18.6	6.0–38.3
	Average	25.8 ± 7.7	34.6 ± 7.6	11.7 ± 4.3	5.8 ± 3.2	22.1 ± 12.3

Correlations between fluorescence components and water quality parameters

PCA was conducted on the fluorescent components and water quality data to seek latent factors and to reveal correlations between fluorescence components and water quality. Figure 5 summarizes the PCA including the loadings and scores plots.

POM and particulate hydration factors such as Chl*a*, POC, COD, BOD, TN, TP, and PN had positive loadings to PC1 (which accounted for 37.38% of the variance) (Fig. 5a). Highly significant, positive correlations were shown between C1p and C2p ($r^2 = 0.98$, $p < 0.01$) and between C3p and C4p ($r^2 = 0.91$, $p < 0.01$), indicating that they had similar sources. The highly significant, positive correlations between C1p ($r^2 = 0.79$, $p < 0.01$), C2p ($r^2 = 0.82$, $p < 0.01$), and C5p suggested that tryptophan-like and tyrosine-like components had similar origins. Significant and positive relationships were also shown between protein-like components (C1p, C2p, and C5p) and humic-like matter (C3p and C4p), indicating a common path of production and depletion.

The Meiliang Bay sites L1, L6, and L32 in summer; L0, L1, L4, L6, and L7 in autumn; and L0 and L18 in spring had positive scores on the PC1, elaborating where relatively higher POM and particulate nutrients occurred (Fig. 5b).

PC2 (28.21% of the variance) showed strong positive loading on the C1d, C2d, C3d, C4d, and dissolved hydration factors such as DOC, TDP, DOP, TDN, and DON, whereas negative loading on the C5d. The C5d showed close negative correlations with C2d, C4d, and the dissolved water factors previously mentioned, suggesting that production and elimination of C5d were different from that of other components. Highly significant, positive correlations were found between C1d and C2d ($r^2 = 0.98$, $p < 0.01$) and between C3d and C4d ($r^2 = 0.88$, $p < 0.01$), respectively. There were positive relationships between protein-like components (C1d and C2d) and humic-like matter (C3p and C4p).

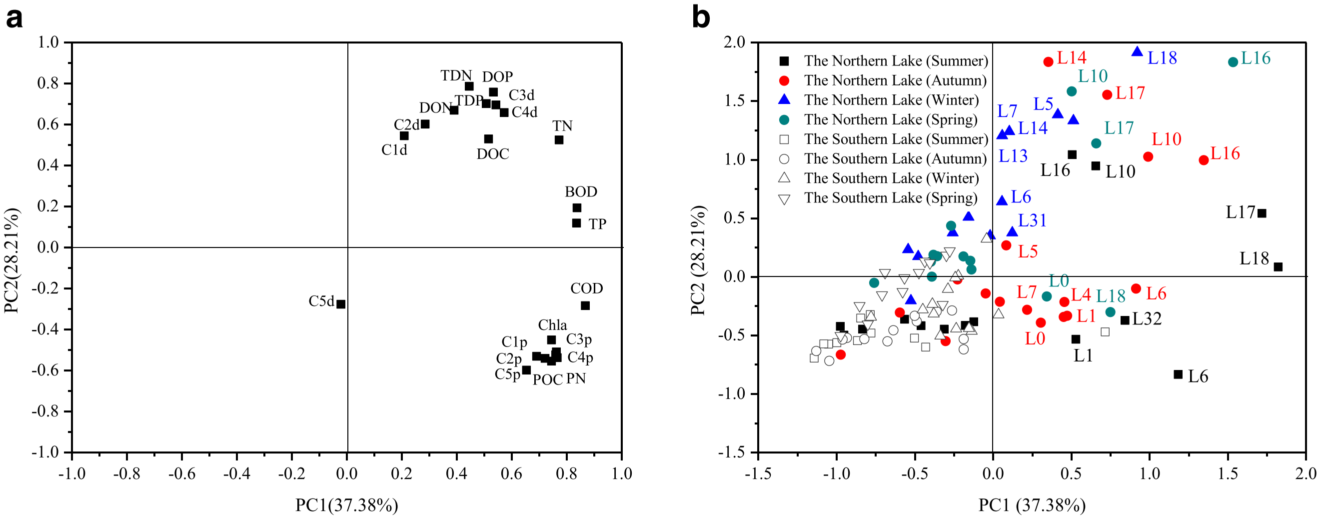

Fig. 5 Loading plot and scores plot for PC1 and PC2 of the DOM–POM and water quality (**a**) and samples (**b**) in Lake Taihu

The river mouth sites L10, L14, L16, and L17 in summer and autumn; L10, L16, and L17 in spring; and the Northern Lake sites L5, L6, L7, L13, L14, L18, and L31 in winter had higher positive scores on the PC2, revealing the region where the relative higher C1d–C4d and dissolved nutrients but lower C5d occurred. The central lake and the southern lake sites during all seasons had lower negative scores on the PC2, indicating where relatively higher abundance of the C5d appeared, while the lower C1d–C4d and dissolved nutrients compared to the other sites.

Fluorescence indices

The fluorescence indices of OM are shown in Fig. 6. The FI, HIX, and BIX of POM ranged 1.57–3.38, 0.27–11.93, and 0.12–1.61, with means of 2.11 ± 0.04, 2.40 ± 0.20, and 0.96 ± 0.04, respectively (Fig. 6a, b). The samples in the circle were mainly concentrated in the river mouths, approaching the characteristics of exogenous material. The samples not circled in Fig. 6a, b were far away from the river mouths, showing strong autochthonous characteristics.

For DOM, the FI, HIX, and BIX ranged between 1.65–2.50, 0.20–2.66, and 0.84–1.48, with means of 1.93 ± 0.01, 0.56 ± 0.03, and 1.12 ± 0.01, respectively. Unlike POM, the DOM we studied was concentrated in the autochthonous region.

Discussion

Differences in optical compositions and sources between POM and DOM

The OM in the aquatic environment has many sources, including (a) allochthonous sources: riverine inputs of terrigenous OM (Spencer et al. 2009; Zhang et al. 2011), rainwater and groundwater (Miller et al. 2009; Birdwell and Engel 2010), and sediment resuspension (Conmy et al. 2015) and (b) autochthonous sources: bacterial release and uptake (Romera-Castillo et al. 2011), biological degradation release from phytoplankton and macrophytes (Lapierre and Frenette 2009; Zhang et al. 2009), and zooplankton grazing (Steinberg et al. 2004). Our present study showed that the POM and DOM in Lake Taihu had different optical components, indicating that they had different sources.

The samples aggregated in the river mouths (marked within circles in Fig. 6a, b) displayed lower FI changing from 1.57 to 1.70 in autumn and winter and higher HIX ranging from 4 to 11.93 in summer and spring compared with the samples (not circled in Fig. 6a, b) far away from river mouths and possessing strong autochthonous characteristics. As Lake Taihu is located in the most developed region in China, excessive nutrient from the upstream watershed flows into the lake, leading to significantly exogenous characteristics in the river mouths (Yao et al. 2011).

The results of C/N of POM were consistent with fluorescence indices mentioned above, indicating that the samples far away from the river mouths had stronger autochthonous characteristics than the samples in the river mouths. Highly significant, positive correlations were shown between POM and Chl*a*, confirming that phytoplankton was the important component of POM, which was also consistent with Zeng et al. (2007) and Pace and Manahan (2007).

Unlike POM, the DOM we studied was concentrated in the autochthonous region, a similar result with Jiang et al. (2017). Previous research has suggested that DOM with low FI, HIX, and BIX indicates DOM with dominant autochthonous characteristics (Huguet et al. 2009; Zhang et al. 2010). Previous study suggested that tryptophan and tyrosine-like components are generally considered to be of autochthonous origin, associated with biological production of DOM (Stedmon and

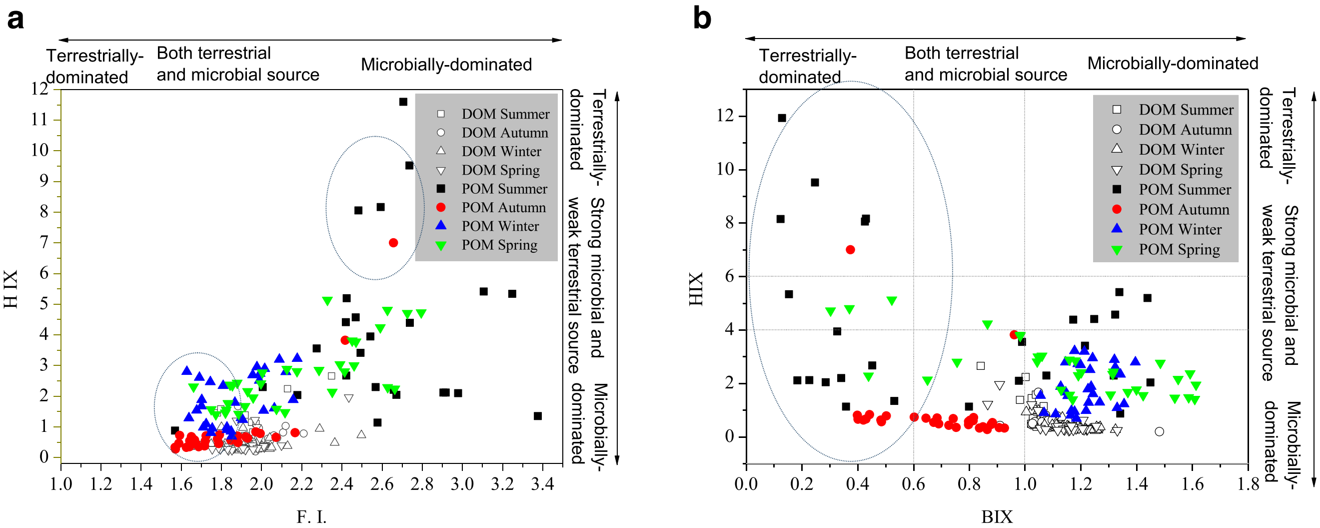

Fig. 6 FI-HIX (**a**) and BIX-HIX (**b**) distributions of POM and DOM in Lake Taihu

Markager 2005a; Murphy et al. 2008). With high concentration of C1d, C2d, and C5d during the whole sampling period, the DOM in Lake Taihu had autochthonous characteristics.

Overall, the POM in Lake Taihu had dominant autochthonous characteristics, and the POM at the river mouths had more obvious exogenous characteristics. Nevertheless, the chromophoric DOM mainly displayed endogenesis. The extraneous features of POM were notable in summer, and there was a large spatial heterogeneity, which may be related to the outbreak of cyanobacteria bloom in summer.

Differences in optical compositions between the Northern and Southern Lakes

The POM showed strong spatial–temporal variation in Lake Taihu (Fig. 7a–d). In summer and autumn, POMs in both the Northern Lake and Southern Lake were all dominated by C5p, indicating that the tyrosine-like component was the major fraction in the POM when phytoplankton blooms became dominant. In contrast, in winter and spring, POMs in both the Northern Lake and Southern Lake were all dominated by C3p, which could be explained by the by-products of bacterial respiration in the process of in situ bacterial degradation (Karmer and Herndl 2004; Nieto-Cid et al. 2006). The river mouths were dominated by C4p in summer and autumn, indicating that a large portion of humic-like C4p drained into the river mouths during these seasons.

The protein-like components (C1d, C2d, and C5d) dominated the DOM pool during the whole sampling period (Fig. 7e–h), in contrast to that C5p and C3p, respectively, dominated the POM pool in different seasons. In particular, the site 9 and site 15 (both location in Lake Lihu, north of Lake Taihu) were dominated by C5d (Fig. 7e–h). This dominance at these two sites may be related to water quality, which has been greatly improved in recent years due to the comprehensive treatment of the aquatic environment (Wang et al. 2016).

The contents of the tryptophan-like components (C1p and C2p) in the Northern Lake were significantly higher than in the Southern Lake. The different components of POM derived from phytoplankton and macrophytes indicated the different properties of POM in the Northern Lake and Southern Lake (Yao et al. 2014). Moreover, C1p, C2p, C1d, and C2d were significantly higher in winter and spring than in summer and autumn.

The tryptophan-like components (C1p and C2p) of POM were significantly higher in winter and spring than in summer and autumn, which might be related to the changes in river loading in the winter/spring vs. summer/autumn and the degradation of POM. The contents of C1p and C2p decreased from the mouth of the inflow rivers to the mouth of Meiliang Bay and to the center of the lake, and the results of fluorescence indices showed that the samples collected in the mouth of the inflow rivers showed significantly exogenous characteristics. This revealed that the inflow rivers brought in the tryptophan-like material and one of the reasons that the contents of C1p and C2p were higher during the high water season (summer and autumn) than during the low water season (winter and spring) might be that the river diluted the contents of C1p and C2p during the high water season.

Another reason causing the seasonal variation of C1p and C2p might be related to the degradation of POM. The C1p, C2p, C3p, and C4p increased from summer and autumn to winter and spring, with POM and C5p decreasing. Highly significant, positive correlations were found between C5p and C1p, C2p, C3p, C4p, respectively, indicating that the tyrosine-like component C5p was biodegraded and partially transformed into tryptophan-like and humic-like components. This was similar to that observed in two urbanized rivers in China (Yu et al. 2016).

The tryptophan-like components (C1d and C2d) of DOM were significantly higher in winter and spring than in summer and autumn, which might be due to the degradation of phytoplankton. The results of fluorescence indices showed that DOM had strong autochthonous characteristics. Yao et al. (2014) also demonstrated that the tryptophan-like fluorescence was produced in the degradation experiment of phytoplankton.

Conceptual model of the exchange of POM and DOM based on “four phases of cyanobacteria bloom development”

Much attention has been paid to the important role played by POM in the biogeochemical cycle of nutrients in a lake (Shank et al. 2011; He et al. 2016a). However, due to the complicated composition and structure of POM, as well as limited techniques for its study, research on the relationship between the nutrient supply mode of POM and the outbreak of cyanobacteria blooms has been limited. For example, how do the nutrients of autochthonous POM in the lake migrate, transform, and finally enter the process of nutrient cycling in the lake? Therefore, answering these questions is key to deepening our understanding of eutrophication.

The dynamic exchanges between DOM and POM play critical roles in organic carbon cycling (Del Giorgio and Duarte 2002; Verdugo et al. 2004; Hopkinson and Vallino 2005), interactions with aquatic organisms (Simon et al. 2002; Azam and Malfatti 2007; Mayer et al. 2011), bioavailability of pollutants (Eriksson et al. 2004), and aquatic phenomena such as eutrophication (Giani et al. 2005; Mecozzi et al. 2005). The carbon flux from the hydrosphere to the atmosphere is also affected by the exchange efficiency of labile DOM from POM in the aquatic systems (Mayorga et al. 2005).

In our present study, we developed a conceptual model of the exchange of POM and DOM, based on “four phases of cyanobacteria bloom development,” to describe all the exchanges in Lake Taihu (Fig. 8). The theory of four phases of

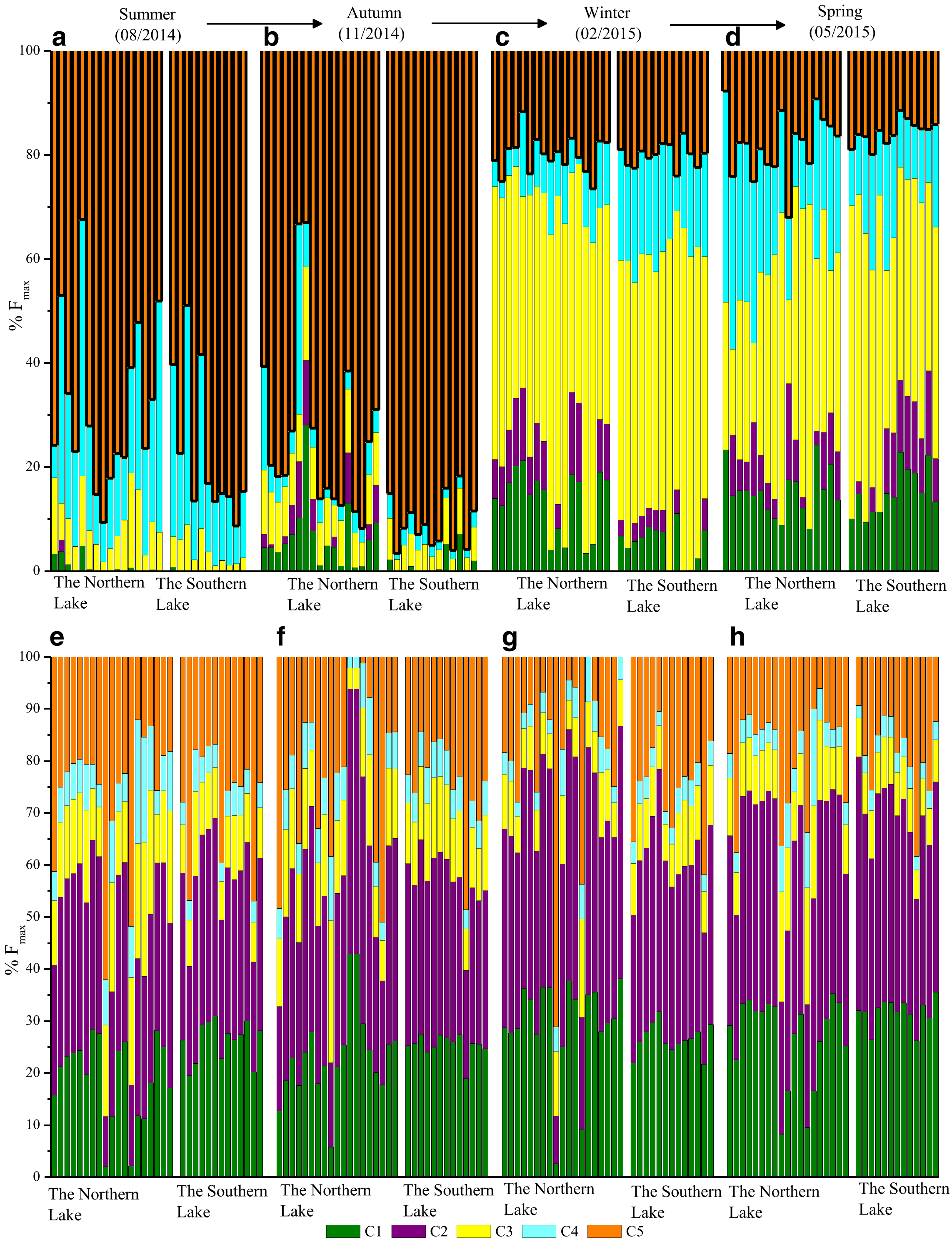

Fig. 7 Relative proportions of five fluorescence components, expressed as $\%F_{max}$, in OM of 32 samples from Lake Taihu, presented according to season, POM (upper panel, **a**–**d**) and DOM (lower panel, **e**–**h**), and region of the lake (the Northern Lake, or the Southern Lake)

cyanobacteria bloom development was proposed by Kong et al. (2009). The theory divided cyanobacteria bloom formation into four stages: overwintering dormancy, spring recovery, summer growth, and autumn aggregation (Kong et al. 2009).

The conceptual model of the exchange of POM and DOM is shown in Fig. 8. The seasonal variation of the phytoplankton, POM, DOM, and TN:TP ratio could be related to that they interacted with each other through aggregation/dissolution, adsorption/desorption, photo-induced exchange, and organism-involved exchange (He et al. 2016a).

The TN:TP ratio has been widely proposed as a means to identify whether phytoplankton is nitrogen (N) or phosphorus

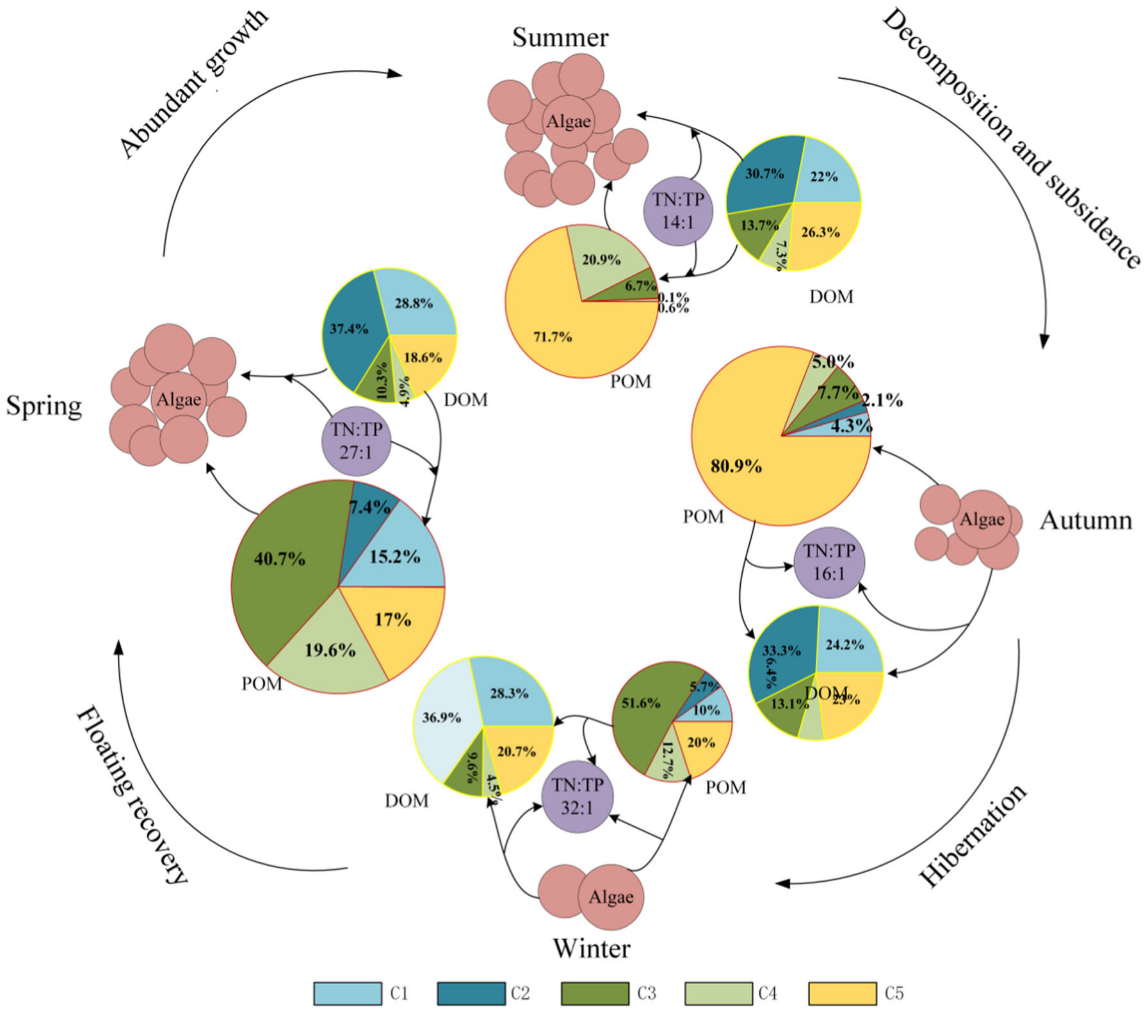

Fig. 8 Conceptual model of the exchange of POM and DOM based on "four phases of cyanobacteria bloom development" (Kong et al. 2009). The model is divided into summer, autumn, winter, and spring and displays: the content of algae, $\%F_{\max}$ of each of the five components C1-C5 of POM and DOM, and TN:TP ratios. The size of the POM and DOM circles represented the total $F_{\max}$ of POM and DOM, respectively. The number of algae (red circles) represented the Chl*a* content

(P) limited (Smith 2006; Verburg et al. 2013). According to Liebig's law of the minimum (Liebig 1842), algae should be limited by N if the water N:P ratio is lower than 16. In contrast, algae should be limited by P if the ratio is above that. During summer and fall, TN:TP ratios ranged from 14:1 to 16:1 in Lake Taihu, but in winter and spring, the ratio increased to 32:1 and 27:1, respectively. Thus, P limitation occurred in winter and spring, and N limitation in summer and fall, which was also consistent with Xu et al. (2010). According to the TN:TP ratio, controlling N when N is limiting and controlling P in case of P deficiency will prevent cyanobacterial blooms effectively in the short term (Ma et al. 2015).

The total $F_{\max}$ of POM increased slightly from summer to fall, decreased significantly from fall to winter, and then increased significantly from winter to spring. The tyrosine-like fraction (C5p) in POM pool increased slightly from summer to autumn and then decreased significantly from fall to winter, displaying only slight variations afterwards. However, the tryptophan-like components (C1p and C2p) increased continuously during the whole period. Unlike C1p and C2p, the humic-like fraction (C3p and C4p) decreased slightly from summer to fall and then increased significantly from fall to winter.

Unlike the POM pool, the total $F_{\max}$ of DOM increased continuously from summer through autumn to winter and then declined significantly in spring. However, the protein-like and humic-like fractions of DOM varied only slightly during the whole period.

We noted that most related studies have focused on POM–DOM exchanges on a global or a regional scale (Engel et al. 2004; Druon et al. 2010; Wang et al. 2013). However, little effort has been made to pay attention on the mesoscale, such as Lake Taihu. The complexity of the POM–DOM exchange at the mesoscale has been often neglected, which is affected by many factors, such as lake area, complex hydrological conditions, and the influence of human activity (He et al. 2016b). In our newly presented conceptual model, special attention has been paid to description of the POM–DOM exchanges during cyanobacteria bloom development.

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