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RESEARCH ARTICLE

Dynamics of chromophoric dissolved organic matter influenced by hydrological conditions in a large, shallow, and eutrophic lake in China

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Abstract High concentrations of chromophoric dissolved organic matter (CDOM) are terrestrially derived from upstream tributaries to Lake Taihu, China, and are influenced by hydrological conditions of the upstream watershed. To investigate how the dynamics of CDOM in Lake Taihu are influenced by upstream inflow runoff, four sampling cruises, differing in hydrological conditions, were undertaken in the lake and its three major tributaries, rivers Yincun, Dapu, and Changdou. CDOM absorption, fluorescence spectroscopy, chemical oxygen demand (COD), and stable isotope $\delta^{13}C$ and $\delta^{15}N$ measurements were conducted to characterize the dynamics of CDOM. The mean absorption coefficient *a*(350) collected from the three river profiles ($5.15\pm1.92\ m^{-1}$) was significantly higher than that of the lake ($2.95\pm1.88\ m^{-1}$), indicating that the upstream rivers carried a substantial load of CDOM to the lake. This finding was substantiated by the exclusively terrestrial signal exhibited by the level of $\delta^{13}C$ (−26.23±0.49‰) of CDOM samples collected from the rivers. Mean *a*(350) and COD in Lake Taihu were significantly higher in the wet season than in the dry season (*t* test, $p<0.0001$), suggesting that the abundance of CDOM in the lake is strongly influenced by hydrological conditions of the watershed. Four components were identified by parallel factor analysis, including two protein-like components (C1 and C2), a terrestrial humic-like component (C3), and a microbial humic-like (C4) component. The contribution percentage of the two humic-like components relative to the summed fluorescence intensity of the four components (C_{humic}) increased significantly from the dry to the wet season. This seasonal difference in contribution further substantiated that an enhanced rainfall followed by an elevated inflow runoff in the lake watershed in the wet season may result in an increase in humic-like substances being discharged into the lake compared to that in the dry season. This finding was further supported by an elevated *a*(250)/*a*(365) of CDOM samples collected in the lake in the wet season than in the dry season. Significantly higher mean levels of C3 and *a*(350) were recorded for CDOM samples collected from River Yincun than those from rivers Dapu and Changdou, differing in seasons, suggesting the significance of terrestrial CDOM input from River Yincun.

Keywords Chromophoric dissolved organic matter (CDOM) · Lake Taihu · Parallel factor (PARAFAC) analysis · Hydrological conditions

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Introduction

Freshwater lakes serve as important drinking water sources in many places around the world (Williamson et al. 2008; Williamson et al. 2009); however, high concentration of chromophoric dissolved organic matter (CDOM) present in these water bodies is responsible for the formation of carcinogenic byproducts that can compromise the safety of the drinking water source (Zhang et al. 2011a; Duan et al. 2014). CDOM is also responsible for unpleasant odor and taste during drinking water treatment processes (Baghoth et al. 2011; Meng et al. 2012). The translocation and transformation of CDOM in lakes and coastal waters are mediated by its structure and relative abundance; thus, it can be beneficial to know the temporal–spatial dynamics of CDOM in these waters (Zhang et al. 2011a). CDOM displays conservative mixing behavior in lakes and coastal waters, and its geochemical cycling processes are often coupled with river-borne pollutants (Yamashita et al. 2008; Zhang et al. 2011a; Hosen et al. 2014). Furthermore, CDOM can strongly absorb light in the blue part of visible region and can interfere with the remote sensing estimation of phytoplankton and nonalgal particle matter (Lee et al. 2011; Hu et al. 2012; Song et al. 2013; Huang et al. 2014). Thus, numerous studies have been dedicated to investigating factors that control the source, cycling, and fate of CDOM in lakes and coastal waters (Stedmon and Markager 2005; Ishii and Boyer 2012; Asmala et al. 2013; Bianchi et al. 2013; Guo et al. 2014).

In large, shallow, and eutrophic lakes, however, in addition to terrestrial origin, CDOM may also be produced and transformed through biological metabolism or released from lakebed sediment by wind-induced sediment resuspension (Qin et al. 2007). In inland and coastal waters, upstream tributaries carry substantial CDOM and discharge to downstream-linked water bodies (Stedmon and Markager 2005; Zhang et al. 2011a; Yang et al. 2013; Guo et al. 2014). The presence of high CDOM concentration in coastal waters and bays derived from upstream rivers can result in eutrophication and algal blooms (Pearl 1988). Precipitation may result in an elevated export of terrestrial CDOM fluorescence from upstream tributaries to downstream-linked waters (Stedmon and Markager 2005; Zhang et al. 2011a). How hydrological conditions may mediate CDOM dynamics in large, shallow, and highly eutrophic lakes remains to be elucidated.

Lake Taihu, located in the Yangtze River Delta, has a water area of 2338 km^2 and a mean depth of 1.9 m, ranking it the third largest freshwater lake in China. Rapid economic development over the last three decades has resulted in high spatial heterogeneity in the lake, with its northern and northwestern regions dominated by algae and its southeastern region dominated by macrophytes (Zhu et al. 2008). The lake has an average water level of ~3.3 m above sea level, the lake basin has an area of 36,900 km^2, and more than 40 million people live within the watershed. In the Lake Taihu basin, 172 rivers or channels connect to the lake (Qin et al. 2007); most inflow tributaries are in the western and northwestern regions, and most outflow rivers are in the southern and southeastern subbasins. Economic booms in nearby cities and towns since the 1980s have increased the amount of sewage being discharged into Lake Taihu (Qin et al. 2010). In the large, shallow, and eutrophic lake, algal blooms caused by *Microcystis* spp. have occurred throughout summer since the last decade (Qin et al. 2007).

In the lake, in addition to in situ production of CDOM from the degradation of phytoplankton and aquatic macrophytes, CDOM export from upstream tributaries is the most important source to the whole CDOM pool of the lake (Zhang et al. 2011b). Previous results indicated that CDOM in lakes and coastal waters may be strongly influenced by hydrological conditions, especially inflow runoff, of upstream watersheds (Stedmon and Markager 2005; Coble 2007; Zhang et al. 2011a; Guo et al. 2014). For example, a substantial increase in inflow runoff in the wet season may result in an elevated CDOM concentration of a downstream-linked lake as a result of high concentration CDOM input. Similarly, CDOM concentration of a downstream-linked lake may drop as a result of a dilution effect by flushing low concentration CDOM waters from upstream tributaries.

A recent study conducted in Lake Taihu found an elevated CDOM absorption coefficient in the wet season compared to the dry season (Zhang et al. 2011b); however, the behaviors of different components of the overall CDOM dynamics remain unknown. Another study performed in the same region provided a snapshot of the relative abundance of parallel factor (PARAFAC) analysis components (Yao et al. 2011), but seasonal dynamics of CDOM mediated by hydrological conditions were lacking. The details of how hydrological conditions, including rainfall and inflow runoff, may alter the spatial and temporal dynamics of CDOM in the lake remain unknown.

To address this knowledge gap, our objective was to characterize how the dynamics of CDOM in Lake Taihu may respond to different hydrological conditions, including rainfall and inflow runoff of the upstream watershed. We used spectral absorption, chemical oxygen demand (COD), fluorescence spectroscopy, and stable isotope $\delta^{13}C$ and $\delta^{15}N$ to characterize the dynamics of CDOM mediated by hydrological conditions in the lake. We assumed that the maximum fluorescence intensities (F_{max}) of individual PARAFAC components are proportional to the concentrations of corresponding fluorescent fractions (Borisover et al. 2009).

Materials and methods

Sample collection

Four sampling cruises were performed in Lake Taihu (Fig. 1), one each in winter (February), spring (May), summer (August), and autumn (November) 2011. Surface water samples (0–0.5 m) were taken from the lake and from profiles of its three major tributaries, rivers Changdou, Dapu, and Yincun (Fig. 1). Thirty-two total sites were distributed evenly in Lake Taihu, composed of 11 sites along each of the three river profiles: sites 1–5 in the lake, site 6 at the three river mouths, and sites 7–11 upstream in the river (Fig. 1). Note that the

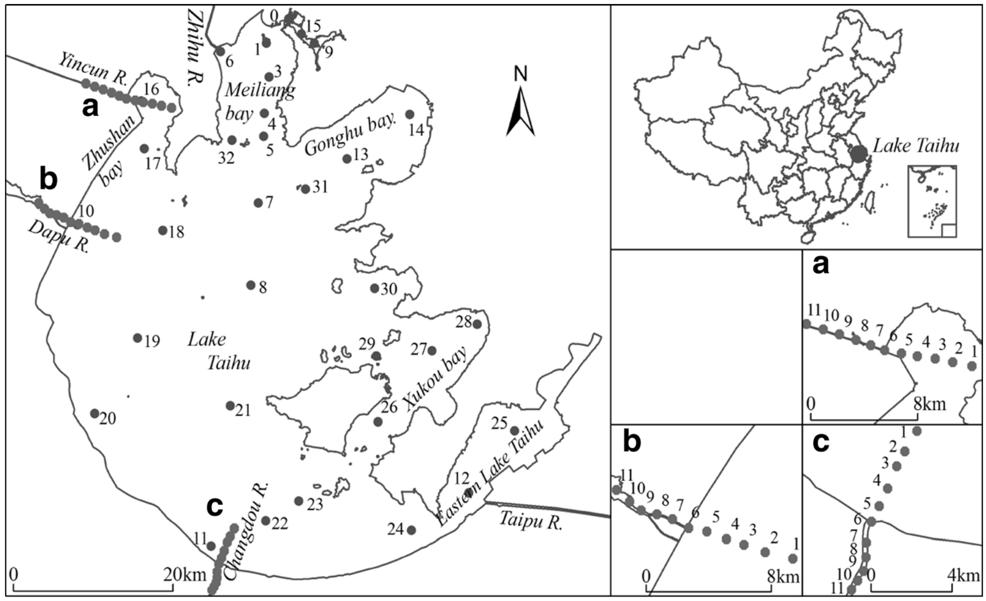

Fig. 1 Locations of sampling sites in Lake Taihu and profiles of its three major tributaries: rivers Changdou, Dapu, and Yincun

locations of site Dapu 5 and Yincun 4 were the same as Taihu 10 and Taihu 16, respectively (Fig. 1).

Thus, 252 total surface water samples were collected: Lake Taihu 128 (32 sites×4 seasons)+Changdou 44 (11 sites×4 seasons)+Dapu 40 (10 sites×4 seasons)+Yincun 40 (10 sites×4 seasons). These samples were grouped into two spatial categories, the lake group (samples collected from Lake Taihu) and the river group (samples collected from the three river profiles; Fig. 1). The four cruises were divided into three seasonal groups according to the rainfall of the watershed and the water level of the lake (Fig. 2): dry season (low water season), February+May (winter+spring); wet season (high water season), August (summer); and wet-to-dry transition season, November (autumn). Note that inflow runoff to the lake was relatively low in November compared to the rest of the year (Fig. 2).

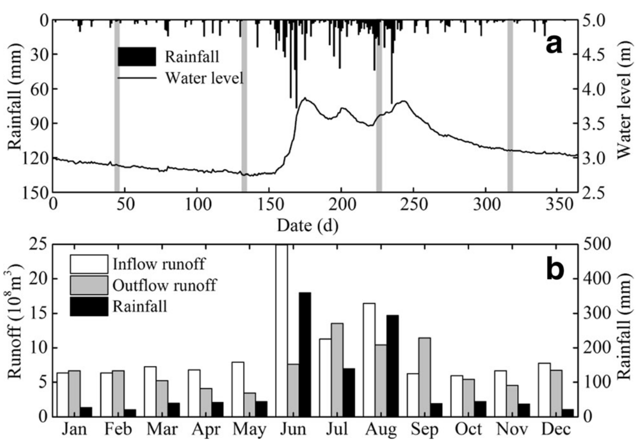

Fig. 2 **a** Variations of daily rainfall (in mm) of Lake Taihu Basin and water level (in m above sea level) of the lake in 2011 *gray bars* denote sampling collection date. **b** Monthly variations of mean rainfall in the lake basin, and inflow runoff to the lake and corresponding outflow runoff

Hydrological data

Hydrological data, including the monthly rainfall of Lake Taihu Basin, monthly inflow runoff to the lake, and the corresponding outflow runoff, were provided by the Taihu Basin hydrological information service system (http://www.tbhi.gov.cn//tba/content/SWJ/sqyb/index.html).

Measurements of CDOM absorption and COD

Water samples were held on ice while in the field and transported to the laboratory where they were filtered immediately. A small portion of water samples that cannot be filtered immediately in summer was stored at −20 °C to prevent microbial degradation due to high air temperatures and then filtered within 2 days. Our previous study showed that freezing the original water samples for 2 days did not produce a marked change in the CDOM absorption (Zhang et al. 2011a). The filtered samples were stored in the dark at 4 °C for CDOM optical and fluorescence measurements. CDOM optical and fluorescence measurements were finished within 3 days, and all laboratory measurements were completed within 1 week after field sampling.

Samples were first filtered through precombusted 47-mm diameter Whatman GF/F filters (0.7 μm porosity) and then through prerinsed 25-mm diameter Millipore membrane cellulose filters (0.22 μm porosity) to determine CDOM absorption coefficient and fluorescence excitation–emission matrices (EEMs). CDOM absorption spectra were obtained between 200 and 800 nm at 1-nm intervals using a Shimadzu UV-2450PC UV–vis recording spectrophotometer with matching 5-cm quartz cells. Milli-Q water was used as reference. Absorbance measurements at each wavelength (λ) were baseline-corrected by subtracting corresponding absorbance values at 700 nm.

Absorption coefficients were obtained using the following equation:

$$a(\lambda) = 2.303 \times D(\lambda)/r \quad (1)$$

where $a(\lambda)$, $D(\lambda)$, and r are CDOM absorption coefficient at wavelength λ, corrected optical density at wavelength λ, and cuvette path length (in m), respectively. In this study, the abundance of CDOM was expressed using $a(350)$, which has been widely used in CDOM studies (Stedmon et al. 2007; Osburn et al. 2011). Spectral slope of CDOM was calculated by nonlinear fit over the 280–500 nm range, using the following equation:

$$a(\lambda) = a(\lambda_0) \times e^{[S(\lambda_0 - \lambda)]} + k \quad (2)$$

where $a(\lambda)$ and $a(\lambda_0)$ are the corrected CDOM absorption coefficients at wavelength of λ and reference wavelength λ_0=440 nm, respectively. S and k represent CDOM spectral slope and background parameter, respectively. The spectral slope ratio (S_R) was defined as the ratio of spectral slope over wavelength range of 275–295 nm to 350–400 nm (i.e., $S_{275-295}/S_{350-400}$). S_R of <1 and >1 indicated CDOM of terrestrial and in situ autochthonous origins, respectively; solar exposure could result in a notable increase in S_R of CDOM sample (Helms et al. 2008). The ratio of CDOM absorption coefficients at 250 and 365 nm, i.e. $a(250)/a(365)$, was used to semi-quantitatively determine the relative molecular size of CDOM samples; the ratio increases with decreasing of CDOM molecular size (De Haan 1993; Helms et al. 2008; Zhang et al. 2013).

Water samples filtered through 47-mm Whatman GF/F filters were determined for COD concentration using a colorimetric method with potassium dichromate–sulfuric acid as reagent (Yao et al. 2011). COD was only measured for samples collected from the lake (n=128).

CDOM fluorescence measurement

CDOM EEMs were measured using a Hitachi F-7000 fluorescence spectrometer (Hitachi High-Technologies, Tokyo, Japan) with a 700-voltage xenon lamp. The excitation and emission spectra for scanning ranges spanned 200–450 nm (every 5 nm) and 250–600 nm (every 1 nm), respectively, with a 5-nm slit width for both excitation and emission. The EEMs were Raman-calibrated by subtracting Milli-Q water blanks. The spectra were corrected for instrumental response according to the procedure recommended in the Hitachi F-7000 Instruction Manual, which comprised both excitation and emission calibration. Excitation was calibrated using Rhodamine B as standard and a single-sided frosted red filter in excitation scan mode, while emission was calibrated with a diffuser in synchronous scan mode. The subsequent excitation and emission spectra were internally corrected using Fluorescence Solutions 2.1 software (internally installed in the instrument).

Fluorescence intensity was calibrated in quinine sulfate units (QSU), where 1 QSU represents the maximum fluorescence intensity of 0.01 mg/L of quinine in 1 mol/L H_2SO_4 at excitation=350 nm and emission=450 nm (Zhang et al. 2010). Rayleigh scatter effects were removed by replacing the EEMs regions at emission wavelength≤excitation wavelength+15 nm, and at emission wavelength ≥2×excitation wavelength−20 nm, with zeros. The inner-filter effect was calibrated according to the following equation (McKnight et al. 2001):

$$F_{cor} = F_{obs} \times 10^{(A_{Ex.} + A_{Em.})/2} \quad (3)$$

where F_{obs} and F_{cor} are fluorescence intensity before and after calibration, and $A_{Ex.}$ and $A_{Em.}$ are absorbance at corresponding excitation and emission wavelengths.

PARAFAC modeling

A total of 252 EEMs were modeled using PARAFAC in DOMFluor Toolbox (Stedmon and Bro 2008), MATLAB 2012a software. The EEMs after inner-filter effect correction were further processed before PARAFAC modeling by deleting excitation wavelength from 200 to 225 nm and emission wavelength from 250 to 300 nm and 550 to 600 nm to eliminate instrument measurement errors. All 252 EEMs were split into a calibration half and a validation half and were run stepwise from three to seven components on both halves. Outputs of the PARAFAC model, such as sum of squares of residuals and explained variation of the model, indicated that a four-component model was sufficient to explain more than 99 % of total EEM variables and was considered adequate to describe the EEMs dataset.

Stable isotope $\delta^{13}C$ and $\delta^{15}N$ measurements

Three water samples (site 1, 6, and 11) from each of the three river profiles were collected in the dry and wet seasons and stored at −20 °C in the laboratory for stable isotope analysis after filtering through precombusted 47-mm diameter Whatman GF/F filters (0.7 μm porosity). A few drops of HCl solution (1 mol/L) were used to acidify samples to approximately pH=2 to remove the interferences of dissolved inorganic carbon. The samples were dried to constant weight under low temperature (55±5 °C). The solid residue (i.e., DOM) was stored in a desiccator and subsequently combusted using a Flash EA1112 analyzer. CO_2 and N_2 gases were measured using a Thermo FinniganMAT Delta^{plus} dual-inlet continuous flow isotope ratio mass spectrometer, with precision of better than ±0.1‰ for both carbon and nitrogen. Stable isotope values for nitrogen and carbon are expressed as $\delta^{15}N$ and $\delta^{13}C$, respectively. The ratios $^{15}N/^{14}N$ and $^{13}C/^{12}C$ were calibrated against high-purity N_2 gas and the Vienna Pee Dee Belemnite (VPDB) standard, respectively (Pruell et al. 2006; Osburn and St-Jean 2007; Hood et al. 2009). The stable isotope values were calculated using the following equation:

$$\delta A = (R_{sam}/R_{sta} - 1) \times 1000 \quad (4)$$

where δA is either $\delta^{15}N$ or $\delta^{13}C$, R_{sam} represents the isotope ratios of the samples collected (i.e., $^{13}C/^{12}C$ or $^{15}N/^{14}N$), and R_{sta} represents the isotope ratios of the known isotopic standards.

Statistical analyses

Statistical analyses (mean value, standard deviation, *t* test, and linear regression) were performed using R-studio 0.97.551 software. Linear fit, nonlinear fit, and the calculation of spectral slopes (*S*) of CDOM absorption spectra were operated using MATLAB 2012a software. Significance level was reported as $p<0.05$ in *t* test and correlation analysis.

Results and discussion

Variation of CDOM optical properties and COD under different hydrological conditions

The $a(350)$ of all samples ranged from 1.66 to 18.72 m^{-1}, with high levels of $a(350)$ recorded in the Zhushan Bay and Meiliang Bay, especially the river mouths of Yincun and Dapu (Fig. 3). COD concentrations ranged from 2.45 to 14.87 mg/L (mean of 4.54±1.69 mg/L) and shared a similar distribution pattern with $a(350)$, with high levels of COD found in Zhushan Bay and Meiliang Bay. The maximum of both $a(350)$ and COD appeared during the wet season in TH6, located at the river mouth of another tributary, river Zhihu (Fig. 1), a river in the city of Wuxi. This finding implied that CDOM in Lake Taihu, especially in Zhushan Bay and Meiliang Bay, was strongly influenced by upstream terrestrial input. The $a(350)$ of all samples taken from the three river profiles, Changdou, Dapu, and Yincun (5.15±1.92 m^{-1}), were significantly higher than all samples from Lake Taihu (2.95±1.88 m^{-1}) (Table 1), further verifying that a substantial amount of CDOM was carried and discharged into the lake by its tributaries. This was consistent with many studies conducted in other aquatic ecosystems that an increased precipitation and inflow runoff can result in an increased CDOM export to downstream-linked waters (Stedmon and Markager 2005; Fellman et al. 2009; Miller and McKnight 2010; Yang et al. 2013).

Seasonally, mean $a(350)$ values of all the three river profiles were significantly higher than in the lake in the dry season, and only samples collected from River Yincun were significantly higher than in the lake in the wet season (Table 1). Note that the mean $a(350)$ of CDOM samples collected from River Yincun was significantly higher than that of the lake and rivers Changdou and Dapu, differing by seasons (Table 1), indicating that a large quantity of CDOM was accumulated in Zhushan Bay. This was consistent with results that river mouths are considered as the sinks of particulate organic matter and sources of CDOM for inland and coastal waters (Shen et al. 2012).

In Lake Taihu, a significantly higher mean $a(350)$ was found in the wet season than in the dry season ($p<0.0001$; Table 1). Similarly, mean COD values shared a significantly higher level in the wet season (5.29±2.02 mg/L) than in the dry season (4.27±0.95 mg/L, *t* test, $p<0.05$), implying that vast amounts of CDOM were discharged and accumulated in the lake in the wet season compared to the dry season. Spatially, high levels of $a(350)$ expanded from Zhushan Bay in the dry season to the northwestern half of the lake in the wet-to-dry transition season and further to the whole lake in the wet season (Fig. 3), an additional indication that a substantial amount of CDOM was discharged into the northwestern regions of the lake in the wet season compared with that in the dry season. A notable decrease in the mean level of $a(350)$ for samples collected from the three river profiles was recorded in the wet season compared to the dry seasons (Table 1), probably the result of a higher dilution effect caused by a substantial increase in inflow runoff of the three tributaries in the wet season compared with that in the dry season (Zhang et al. 2011a; Guo et al. 2014).

CDOM in Lake Taihu shared a significantly lower $a(250)/a(365)$ in the wet season than in the dry season (Fig. 3; Table 1), probably a result of terrestrial CDOM input. Previous studies indicated that terrestrial CDOM tends to have a higher molecular weight [lower level of $a(250)/a(365)$] compared with CDOM associated with in situ biological metabolisms (Helms et al. 2008; Fichot and Benner 2012). As further support, both CDOM quantity and quality in the lake were strongly influence by runoff-mediated terrestrial CDOM input. Lower $a(250)/a(365)$ levels in CDOM samples collected from the three river profiles compared to the lake further indicated that substantial CDOM was discharged to the lake from upstream watershed (Helms et al. 2008; Zhang et al. 2011a, b).

Spatially, the area with high levels of $a(250)/a(365)$ downsized from the whole lake in the dry season to the southeastern half of the lake in the wet-to-dry transition season and further to Xukou Bay and Eastern Taihu Bay in the wet season (Fig. 3). This systematic decrease exhibited an increased terrestrial CDOM input from northwestern regions of the lake in the wet season compared to the dry season (Zhang et al. 2011b), substantiated by the significantly lower mean

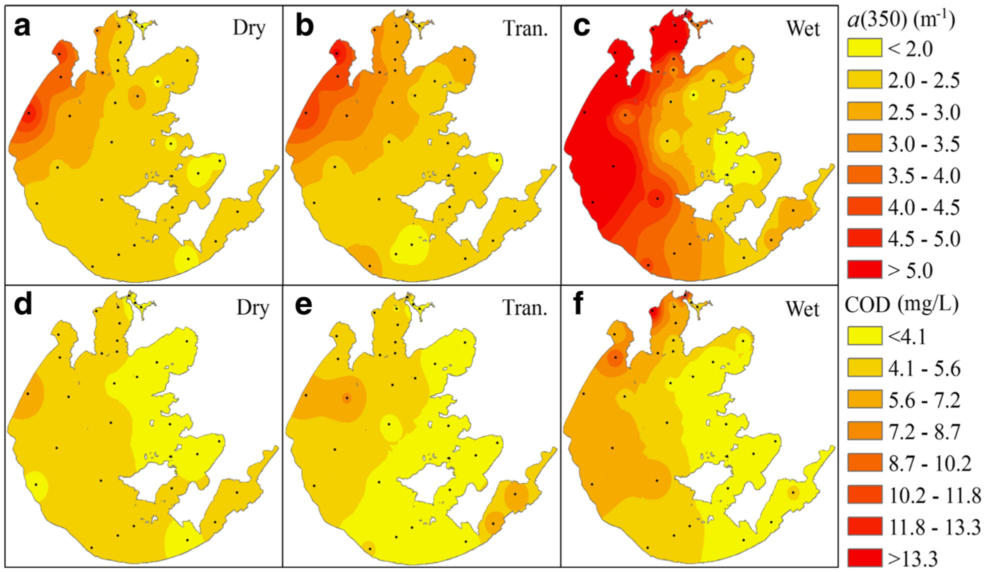

Fig. 3 CDOM absorption coefficient *a*(350) in the **a** dry, **b** wet-to-dry transition, and **c** wet season. Chemical oxygen demand (COD) concentration in the **d** dry, **e** wet-to-dry transition, and **f** wet season

a(250)/*a*(365) and S_R of CDOM samples collected from the river Yincun compared to Lake Taihu in different seasons (Table 1). River Yincun carried high concentration of terrestrial CDOM and discharged into the lake, and this portion of terrestrial CDOM shares lower S_R and *a*(250)/*a*(365) compared to CDOM produced internally from noncolored algal precursors in the lake (Fichot and Benner 2012; Yamashita et al. 2013).

PARAFAC components

The four components of PARAFAC included two autochthonous protein-like components (C1 and C2), one terrestrial humic-like component C3, and one marine humic-like component C4 (Fig. 4). C1 displayed three fluorescent peaks with excitation/emission maxima at ≤230 nm (285, 340 nm)/361 nm, similar to tryptophan-like fluorophore (peak T) (Coble 1996; Coble et al. 1998). C2 exhibited a tyrosine-like fluorophore, which was similar to peak B. These two components, C1 and C2, together demonstrated the autochthonous properties contained in CDOM (Coble 1996; Coble et al. 1998; Stedmon and Markager 2005; Zhang et al. 2010). C3 had similar excitation/emission maxima as peaks A (ultraviolet region) and C (visible region) and had characteristics of terrestrially derived humic-like material (Coble 1996; Coble et al. 1998; Stedmon and Markager 2005; Zhang et al. 2010).

Table 1 Optical properties *a*(350) (m^{-1}), *MS* (arbitrary unit, AU) and S_R (AU) of CDOM and fluorescence intensity of the four components (in QSU) identified by PARAFAC model of water samples taken from Lake Taihu and rivers Changdou (CD), Dapu (DP), and Yincun (YC) in the wet and dry season, and significance levels of difference between parameters using *t* test under these two hydrological conditions

Parameters		CDOM optical properties			PARAFAC components			
Sites	Seasons	*a*(350)	*MS*	S_R	C1	C2	C3	C4
Lake Taihu	Dry	2.46±0.82	9.60±1.12	1.21±0.10	37.95±16.61	18.34±6.30	8.23±5.37	2.41±1.39
	Wet	4.22±3.20	7.46±1.24	1.37±0.17	24.15±13.40	34.16±19.53	13.60±10.16	9.81±2.93
	p value	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
River CD	Dry	4.33±1.69	6.37±2.26	1.34±0.05	29.90±7.10	21.48±2.83	6.57±0.57	6.00±1.52
	Wet	2.44±0.17	8.80±0.36	1.32±0.14	11.62±11.87	13.34±3.28	10.55±0.51	7.88±1.99
	p value	<0.0001	<0.0001	>0.05	<0.0001	<0.0001	<0.0001	<0.0001
River DP	Dry	6.26±1.56	6.01±1.29	1.22±0.08	56.69±18.30	23.15±8.62	17.50±3.47	6.50±1.17
	Wet	4.47±0.33	7.63±0.66	1.35±0.16	23.64±7.90	13.24±5.35	17.07±3.82	9.63±2.48
	p value	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	>0.05	<0.0001
River YC	Dry	7.08±1.50	5.55±1.06	1.13±0.11	80.18±31.82	33.48±13.26	33.75±9.20	5.29±10.46
	Wet	5.87±0.81	6.26±0.73	1.21±0.03	29.18±8.08	9.87±4.17	22.42±1.63	7.96±2.57
	p value	<0.0001	<0.0001	<0.001	<0.0001	<0.0001	<0.0001	<0.001

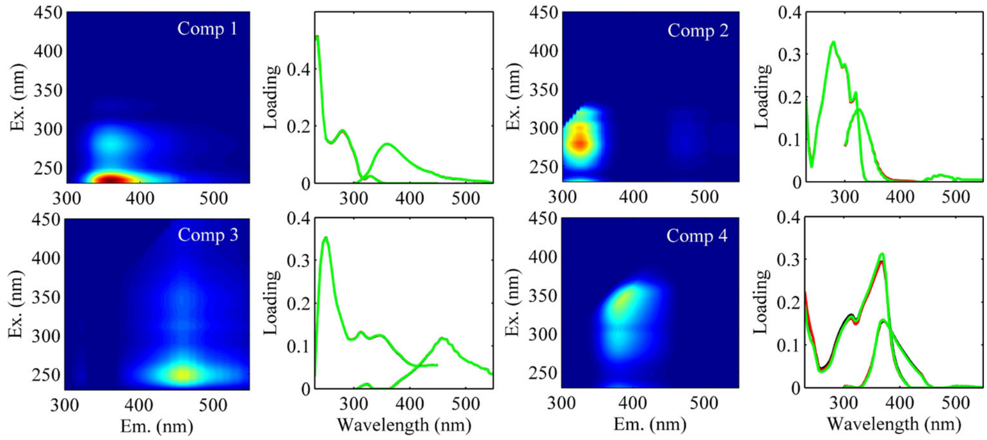

Fig. 4 Four robust components were identified using PARAFAC model. Highly overlapping excitation (*left*) and emission (*right*) spectra were estimated using split-half validation procedure, with *red* and *green lines* representing two independent halves dataset and the *black line* representing the complete dataset

C4 displayed fluorescence spectrum similar to peak M (Coble 1996; Coble et al. 1998) and was considered microbially transformed products from terrestrially derived components (Stedmon and Markager 2005; Zhang et al. 2010).

Variations of CDOM fluorescence intensity under different hydrological conditions

All the four components identified by PARAFAC were well validated by the split-half validation procedure (Fig. 4). Mean fluorescence intensity (F_{max}) and contributions of C1, C2, C3, and C4 for all datasets were 45.05±32.66, 24.03±13.20, 16.52±12.75, and 8.81±10.46 QSU and 47.71, 25.46, 17.50, and 9.33 %, respectively (Table 1). In Lake Taihu, mean F_{max} of C2, C3, and C4 were significantly higher in the wet season than in the dry season; the opposite trend was recorded for C1, with high levels found in the dry season compared with the wet season (Table 1). Our results imply that the dynamics of CDOM composition in Lake Taihu is strongly influenced by hydrological conditions of the watershed, and was consistent with observations in other aquatic ecosystems (Stedmon and Markager 2005; Fellman et al. 2009; Yang et al. 2013). Mean F_{max} of protein-like C1 and C2 in the river profiles was significantly higher in the wet season than in the dry season (Table 1). Note that the mean F_{max} of terrestrial humic-like C3 was significantly higher in river Yincun than in the other two river profiles (Table 1), implying that river Yincun was responsible for the high level of CDOM in Zhushan Bay. Seasonally, mean F_{max} of C3 in river Yincun was significantly higher in the dry season than in the wet season, probably the result of the dilution effects of the inflowing rivers (Table 1). For the microbial humic-like C4, mean F_{max} for samples collected from Lake Taihu and the three river profiles had significantly higher levels in the wet season than in the dry season (Table 1), most likely the result of the close association between the presence and abundance of microbial humic-like component with the allochthonous precursors (Yamashita et al. 2011; Yao et al. 2011). The microbial humic-like C4 is often considered to be associated with microbial transformation from terrestrial humic-like substances (Yamashita et al. 2008; Yamashita and Tanoue 2009; Williams et al. 2010). During the wet season, CDOM microbially transformed from algal substrate in the lake can also result in an elevated level of F_{max} of the microbial humic-like component (Zhang et al. 2009). Note that the mean F_{max} of C3 collected from the river Changdou profile was significantly higher in the wet season than in the dry season (Table 1). River Changdou (also referred to as River Tiaoxi) is the largest tributary of Lake Taihu, and its watershed land use is dominated by mountain forests (Qin et al. 2007). An enhanced rainfall and followed by an elevated inflow runoff in the wet season can enhance flushing of soil organic matter (Asmala et al. 2012; Kothawala et al. 2014; Shafiquzzaman et al. 2014) in the watershed.

Previous results indicated that humic-like components are closely associated with terrestrial CDOM input, and their dynamics are often strongly influenced by upstream hydrological conditions, especially inflow runoff (Stedmon and Markager 2005; Guo et al. 2014). Thus, the contribution percentage of humic-like components (C3 and C4) with regard to the summed F_{max} (C_{humic}, in %) of the four components was investigated to trace the dynamics of CDOM influenced by hydrological conditions in the lake watershed. C_{humic} in Lake Taihu in the wet season (35.76±12.45 %) was significantly higher than in the dry season (16.49±5.86 %, t test, $p<0.0001$) (Table 1). Spatially, the area of high level C_{humic} expanded from the river mouths of tributaries in western sub-basins in the dry season to the whole lake in the wet season (Fig. 5), attributed to a higher inflow runoff in the wet season. An elevated inflow runoff from the upstream watershed in the wet season compared to the dry season resulted in an elevated F_{max} of humic-like materials and an elevated C_{humic} in the downstream-linked Lake Taihu, especially Zhushan Bay, influenced by upstream river Yincun. In the present study, C3 has exclusively terrestrial characteristics (Kowalczuk et al. 2009; Osburn et al. 2012; Andrew et al. 2013), and C4 was

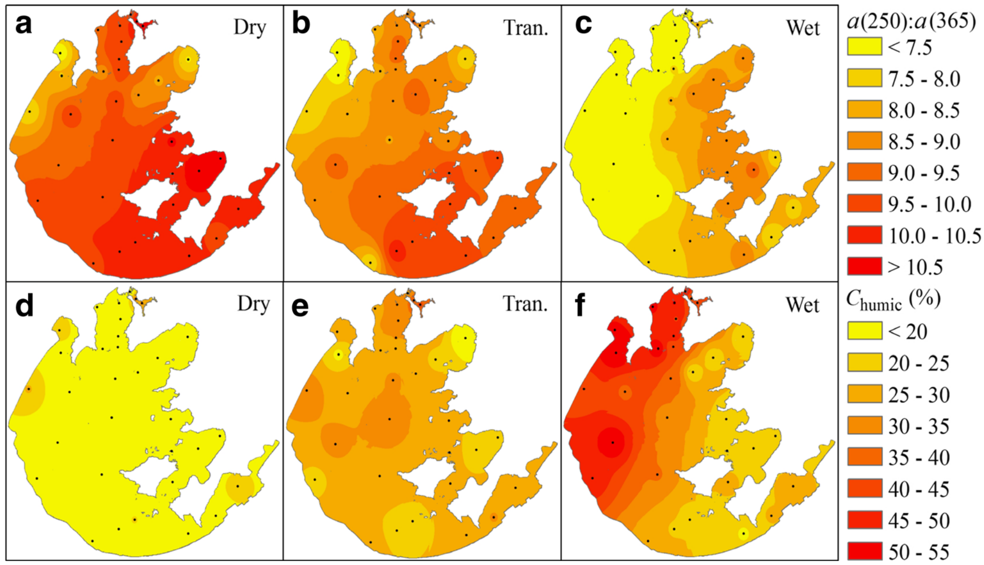

Fig. 5 CDOM absorption ratio *a*(250)/*a*(365) (in arbitrary unit, AU) in the **a** dry, **b** wet-to-dry transition, and **c** wet season. Contribution percentage of humic-like (C3+C4) fluorescence to the summed fluorescence intensity of the four components identified (C_{humic}) in the **d** dry, **e** wet-to-dry transition and **f** wet season

closely associated with the terrestrial humic-like precursor (i.e., C3) in this study (Yamashita et al. 2008; Yamashita et al. 2011; Yao et al. 2011). Thus, the elevated mean C_{humic} in the lake in the wet season compared with that in the dry season substantiated that a large quantity of humified CDOM was discharged into the lake in the wet season.

An elevated lakewater level in the wet season compared to the dry season weakened the process of CDOM released from lakebed sediment by wind-induced waves (Liu et al. 2013). Further, previous study in Lake Taihu showed that the concentration of suspended substance in the lake caused by wind-induced waves was much higher in spring (dry season) than in summer (wet season) (Zhang et al. 2014), indicating a stronger sediment resuspension and CDOM release from sediment in the dry season. Therefore, the elevated mean levels of *a*(350) and C_{humic} observed in the lake in the wet season were most likely the result of allochthonous CDOM input instead of CDOM released from lakebed sediment.

Correlation analysis and PCA of CDOM-related parameters

Significantly positive correlations were recorded between *a*(350) and COD (COD=0.77*a*(350)+2.26, r^2=0.74, n=128, p<0.0001), consistent with previous studies in which close relationships were found in polluted waters (Holbrook et al. 2006; Zhang et al. 2011a). Close relationships were also found between *a*(350) and fluorescent components C1, C2, and C3 (Table 2). CDOM abundance-related parameters including *a*(350) and fluorescent components C1, C2, and C3 were significantly and negatively correlated to *a*(250)/*a*(365) and *S* (Table 2). Similar results were also observed in other aquatic ecosystems (Zhang et al. 2011a; Yang et al. 2013).

Principal component analysis (PCA) was performed to investigate the quantitative and compositional difference of CDOM samples collected from the river profiles and the lake. The first two factors (factors 1 and 2) identified by PCA explain 64.4 and 17.0 % of the total variance, respectively. CDOM abundance-related parameters, including *a*(350) and the four fluorescent components, showed positive factor 1 loadings, whereas *a*(250)/*a*(365) and *S* displayed negative factor 1 loadings (Fig. 6), implying that PCA factor 1 was probably positively associated with CDOM abundance. The *a*(250)/*a*(365) and *S* and the two protein-like components C1 and C2 exhibited positive factor 2 loadings, while *a*(350) showed a negative factor 2 loading (Fig. 6), indicating that PCA factor 2 was possibly positively related to protein-like components. A higher mean factor 1 score was observed for samples collected from the three river profiles than that of the lake (*t* test, p<0.0001, Fig. 6), further substantiating that vast amounts of CDOM were discharged into the lake from upstream tributaries, especially river Yincun. This hypothesis was supported by the notably higher mean levels of PCA factor 1 loading in the northern part of the lake, especially

Table 2 Determination coefficients and significance levels of linear relationships among CDOM-related parameters for all samples (n=252).

Parameters	*a*(350)	*S*	*a*(250)/*a*(365)	C1	C2	C3
S	0.84*					
a(250)/*a*(365)	0.82*	0.92*				
C1	0.34*	0.25*	0.23*			
C2	0.18*	0.17*	0.17*	0.73*		
C3	0.41*	0.28*	0.26*	0.63*	0.34*	
C4	0.00	0.01	0.02	0.01	0.02	0.02

*p<0.0001

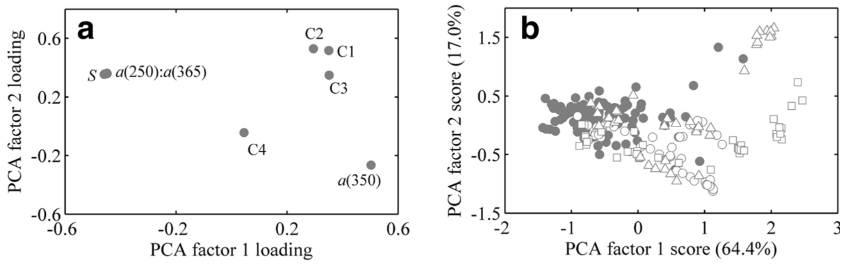

Fig. 6 **a** Property–property plots of PCA factor loadings, **b** property–property plots of PCA factor scores of all samples; *closed circles*, *open circles*, *quadrates*, and *triangles* denote samples taken from Lake Taihu, and river profiles from Changdou, Dapu, and Yincun, respectively

Zhushan Bay compared with the rest lake regions. A higher mean factor 2 score was recorded for samples collected in Lake Taihu than from the three river profiles (t test, $p<0.01$, Fig. 6), indicating that a large amount of protein-like materials were microbially produced from algal remnants in the lake compared with that in the river profiles.

Stable isotope $\delta^{13}C$ and $\delta^{15}N$ characteristics

The $\delta^{13}C$ values of the three river profiles ranged from −26.36 to −25.83‰ with a mean of −26.07±0.25‰ in the dry season. In the wet season, $\delta^{13}C$ ranged from −26.78 to −25.11‰ with a mean of −26.38±0.62‰, and no significant difference was found between mean $\delta^{13}C$ during these two hydrological seasons (Table 3). No significant difference was found among the mean $\delta^{13}C$ of CDOM samples collected from the three river profiles (ANOVA, $p>0.05$) or between samples collected from the river end (site 11, −26.32±0.41‰) and the lake end (site 1, −25.97±0.59‰) sites (pair t test, $p>0.05$). The $\delta^{13}C$ levels of CDOM samples collected from the lake end sites (site 1 of the three profiles) exhibited exclusively terrestrial signals (Vizzini et al. 2005; Zeng et al. 2008). We assumed the $\delta^{13}C$ levels of CDOM samples collected from the three river profiles to be a mixed model of the $\delta^{13}C$ values for both terrestrial plant residues and algal remnants. The depleted $\delta^{13}C$ exhibited at site 1 in the profiles of rivers Yincun, Dapu, and Changdou further supported our finding that substantial amounts of CDOM were discharged into the lake from the three tributaries (Zeng et al. 2008). This hypothesis is consistent with previous studies showing that $\delta^{13}C$ was lighter by about <−25‰ at locations influenced by upstream discharge than those influenced by microbial metabolism (Cole et al. 2002; Vizzini et al. 2005).

Table 3 Comparisons of stable isotope $\delta^{13}C$ and $\delta^{15}N$ (in ‰) of CDOM samples taken from the three river profiles in the dry and wet season, 2011

Sampling sites		Dry season		Wet season	
River	Sites	$\delta^{13}C$	$\delta^{15}N$	$\delta^{13}C$	$\delta^{15}N$
CD	CD1	−26.27	−0.30	−25.52	−3.49
	CD6	−25.94	−0.04	−26.76	1.66
	CD11	−25.63	−0.06	−26.69	3.68
DP	DP1	−25.83	10.56	−25.11	−2.72
	DP6	−26.16	9.54	−26.64	7.58
	DP11	−26.28	9.14	−26.66	2.75
YC	YC1	−26.36	8.02	−26.70	4.02
	YC6	nd	8.06	−26.78	6.99
	YC11	−26.11	8.28	−26.58	6.15

CD river Changdou, *DP* river Dapu, *YC* river Yincun, *nd* no data available

$\delta^{15}N$ ranged from −0.3 to 10.56‰ with a mean of 5.91±4.60‰ for CDOM samples collected from the three river profiles in the dry season. A significantly higher mean $\delta^{15}N$ was recorded in Yincun and Dapu river profiles than in Changdou in the dry season (Table 3), likely explained by the discharge of industrial sewage and domestic wastewater from numerous towns in the river catchment. In the dry season, the water in the Yincun and Dapu rivers was mainly derived from the discharge of industrial sewage and domestic wastewater without precipitation and flooding. Previous studies indicated that CDOM originating from industrial sewage and domestic wastewater shares a higher level of $\delta^{15}N$ compared with CDOM associated with agriculture drainage, precipitation, or microbial metabolism (McClelland et al. 1997; Vizzini et al. 2005). In contrast, in the wet season, $\delta^{15}N$ ranged from −3.49 to 7.58‰ with a mean of 2.96±3.95‰ (Table 3), which was markedly lower than in the dry season, indicating that CDOM was mainly from the agricultural drainage and precipitation. In addition, no significant difference was recorded among mean $\delta^{15}N$ for CDOM samples taken from the three river profiles in the wet season (ANOVA, $p>0.1$), indicating a similar source from the agricultural drainage and precipitation. Spatially, no significant difference was found between the mean $\delta^{15}N$ values of samples collected from the river end (site 11, 4.99±3.51‰) and the lake end (site 1, 2.68±5.81‰) sites (pair t test, $p>0.05$). This finding further verified that vast amounts of CDOM were discharged into the lake by the three tributaries.

Implications for Lake Taihu management

Conservative mixing behaviors observed for both a(350) and the terrestrial humic-like fluorescent component C3, differing in seasons implied that allochthonous CDOM input was probably the primary contributor to the whole CDOM pool in the lake. Although algal blooms occurred in the lake throughout the wet seasons (Qin et al. 2007), CDOM microbially produced from algal remnants probably contributed secondarily to the CDOM pool in the lake compared with terrestrial-derived CDOM subpool (Zhang et al. 2009). This hypothesis was substantiated by the exclusively terrestrial signal exhibited by the level of $\delta^{13}C$ at site 1 of the three river profiles in the wet season. Our results indicated that, in the large, shallow, and highly eutrophic Lake Taihu, the problem from eutrophication and nuisance algal blooms in the lake can only be solved by external loading reduction (Jeppesen et al. 2007). Lake Taihu serves as a drinking water source for at least 10 million people in the most developed region in China, and the Lake Taihu watershed as well as the lake provide a wide range of ecosystem services, ranging from food to recreation and wildlife habitat (Downing et al. 2006). An external CDOM loading reduction would be beneficial to the Lake Taihu ecosystem. The spatial distribution of C_{humic} indicated that river Yincun was probably responsible for the high level of F_{max} of humic-like CDOM components in Zhushan Bay and implies that allochthonous CDOM loading reduction in the Yincun River catchment can provide a safer drinking water source in Lake Taihu.

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