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Chromophoric dissolved organic matter (CDOM) absorption characteristics in relation to fluorescence in Lake Taihu, China, a large shallow subtropical lake

Yunlin Zhang · Boqiang Qin · Guangwei Zhu · Lu Zhang · Longyuan Yang

Abstract Absorption measurements from chromophoric dissolved organic matter (CDOM) and their relationships with dissolved organic carbon (DOC) and fluorescence were studied in Lake Taihu, a large, shallow, subtropical lake in China. Absorption spectra of lake water samples were measured from 240 nm to 800 nm. Highest values of $a(\lambda)$, DOC and $F_n(355)$ occurred near the river inflow to Meiliang Bay and decreased towards the central lake basin. A significant spatial difference was found between Meiliang Bay and the central lake basin in absorption coefficient, DOC-specific absorption coefficient, exponential slope coefficient, DOC concentration and fluorescence value. The spatial distribution of CDOM suggested that a major part of CDOM in the lake was from river input. CDOM absorption coefficients were correlated with DOC over the wavelength range 280–500 nm, and $a(355)$ was also correlated with $F_n(355)$, which showed that CDOM absorption could be inferred from DOC and fluorescence measurement. The coefficient of variation between $a(\lambda)$ and DOC concentration decreased with increase in wavelength from 240 nm to 800 nm. Furthermore, a significant negative linear relationship was recorded between S value and CDOM absorption coefficient, as well as DOC-specific absorption coefficient. S value and DOC-specific absorption coefficient were used as a proxy for CDOM composition and source. Accurate CDOM absorption measurements are very useful in explaining UV attenuation and in developing, validating remote sensing model of water quality in Lake Taihu.

Keywords Chromophoric dissolved organic matter (CDOM) · Fluorescence · Dissolved organic carbon (DOC) · DOC-specific absorption coefficient · Spectral slope coefficient · Lake Taihu

Introduction

Chromophoric dissolved organic matter (CDOM) is the light absorbing (ultraviolet and visible) fraction of dissolved organic carbon (DOC) (Kirk, 1994). Spatial and temporal distribution

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Eutrophication of shallow lakes with special reference to Lake Taihu, China

Y. Zhang · B. Qin (✉) · G. Zhu · L. Zhang ·
L. Yang
Nanjing Institute of Geography & Limnology,
Chinese Academy of Sciences, Nanjing 210008,
P. R. China
e-mail: qinbq@niglas.ac.cn

Y. Zhang · L. Zhang
Graduate school of Chinese Academy of Sciences,
Beijing 100039, P. R. China

of CDOM in aquatic ecosystems can affect ecosystem productivity; negatively impacting primary productivity since CDOM absorbs light, while positively impacting secondary productivity by providing a substitute for microbial respiration via photo-degraded CDOM (Benner & Biddanda, 1998; Reche et al., 1998; Mazzuoli et al., 2003). Overlaps of pigment absorption spectra with CDOM absorption at blue wavelengths complicate the use of chlorophyll *a* retrieval algorithms based on remotely sensed ocean color (Carder et al., 1991) and lead to chlorophyll *a* concentration overestimation (Nelson et al., 1998; Rochelle-Newall & Fisher, 2002a). In addition, high CDOM concentrations can act as a photoprotectant against UV damage to aquatic organisms (Williamson et al., 1996). Several studies have identified CDOM as the most important factor controlling UV attenuation in natural freshwaters (Morris et al., 1995; Laurion et al., 2000). However, protection of aquatic biota against UV radiation provided by dissolved humic material may be diminished with CDOM photodegradation by UV radiation (Morris & Hargreaves, 1997) and acidification (Schindler et al., 1996; Yan et al., 1996).

In shallow, inland lakes and coastal waters, absorption by non-chlorophyllous substances often exceeds that by phytoplankton (Carder et al., 1991; Frenette et al., 2003). CDOM originates from phytoplankton degradation and river inputs and land drainage, especially in inland waters. Although CDOM influences lake optical properties, remotely sensed spectra, and biogeochemical processes, the CDOM dynamics in diverse aquatic environments are not well known. A thorough inventory of regional CDOM distribution and its light absorption in wavebands covering UV and visible radiation spectra have not been conducted in Lake Taihu. This information is needed to interpret satellite and airplane remote sensing imagery to be used to record lake response to changes in environmental conditions, such as water pollution, eutrophication and cyanobacterial blooms. In this paper, we describe the absorption spectrum of CDOM in lake water samples from north Lake Taihu. To examine CDOM, the following aspects are addressed in this paper:

(1) Spatial distribution of CDOM in different lake regions interpreted by absorption spectra;
(2) The relationship between CDOM absorption coefficients and DOC concentrations;
(3) Variations in the absorption spectra with wavelength and fluorescence.

Materials and methods

Study area

Forty sampling sites were chosen in the northern lake. Sampling site 24 was located at the boundary

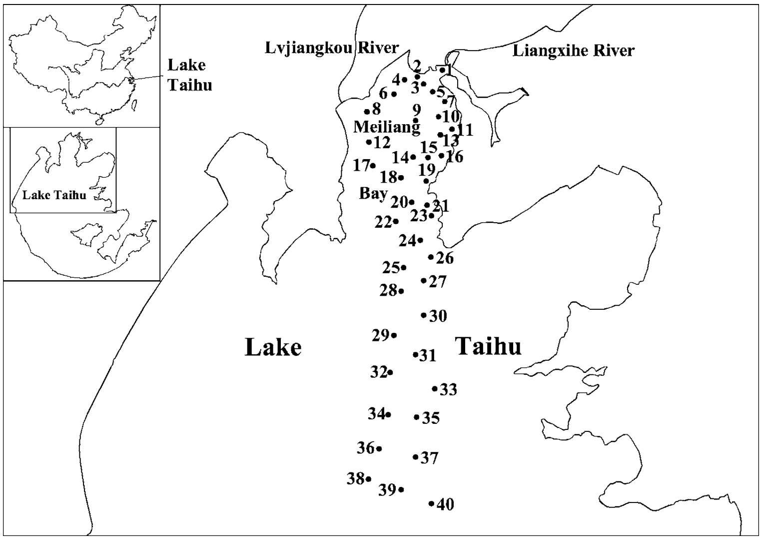

Fig. 1 Position of sampling sites during the cruise

of Meiliang Bay and the central lake. Sites 1–24 were distributed in Meiliang Bay, and other sites were in the central lake (Fig. 1). Meiliang Bay is eutrophic with an area of 132 km^2 and mean depth of 2.0 m. Lujiangkou and Liangxihe Rivers flow into the bay (Fig. 1).

Absorption of CDOM and DOC concentration

Water from 0 m to 0.5 m depth for CDOM absorption and DOC analysis was collected in 250 ml acid-cleaned plastic bottles and held on ice while in the field. In the laboratory, these samples were kept at 4°C until analysis within two days. All samples were filtered at low pressure through pre-combusted Whatman GF/F filters (0.7 μm) into glass bottles pre-combusted at 550°C for 6 h (Del Castillo et al., 1999; Del Castillo & Coble, 2000; Rochelle-Newall & Fisher, 2002b; Yacobi et al., 2003; Frenette et al., 2003; Callahan et al., 2004; Chen et al., 2004). Absorption spectra were obtained between 240 nm and 800 nm at 1-nm intervals using a Shimadzu UV-2401PC UV-Vis recording spectrophotometer with matching 4 cm quartz cells. Milli-Q water was used in the reference cell. Absorption coefficients were obtained by using following expression (Kirk, 1994):

$$a(\lambda') = 2.303D(\lambda)/r \tag{1}$$

where $a(\lambda')$ is uncorrected CDOM absorption coefficient at wavelength λ, $D(\lambda)$ is the optical density at wavelength λ, and r is the cuvette path length in m. Absorption coefficients were corrected for backscattering of small particles and colloids, which pass through filters, using Eq. 2 (Bricaud et al., 1981; Green & Blough, 1994).

$$a(\lambda) = a(\lambda') - a(750) \times \lambda/750 \tag{2}$$

where $a(\lambda)$ = absorption coefficient at wavelength (λ) corrected for scattering, and $a(750)$ = measured absorption coefficient at 750 nm corrected for scattering. Because of the chemical complexity of CDOM, concentration is expressed using the absorption coefficient at some reference wavelength, typically 355 nm, 375 nm or 440 nm (Kirk, 1994; Del Castillo & Coble, 2000; Stedmon et al., 2000). Here, absorption at 355 nm was used to analyze the relationship of CDOM and fluorescence. Molecular size of humic molecules was estimated from the ratio of absorption coefficients at 250 and 365 nm [$a(250)/a(365)$] (De Haan, 1993; Peuravuori & Pihlaja, 1997). The apparent DOC-specific absorption coefficient $a^*(\lambda)$ was calculated by the relationship below (Seritti et al., 1998):

$$a * (\lambda) = a(\lambda)/\mathrm{DOC} \tag{3}$$

where DOC is expressed in mg l^{-1}. DOC in the filtrates was measured on a 1020 TOC analyzer.

Exponential slope coefficient calculation

Exponential slope coefficient was calculated from the absorption spectrum between 280 and 500 nm ($S1$), 280 and 360 nm ($S2$), and 360 and 440 nm ($S3$) using the non-linear regression of Equation (4) (Bricaud et al., 1981). The coefficient of determination (r^2) for the fit of S was consistently higher than 0.99.

$$a(\lambda) = a(\lambda_0)\exp[S(\lambda_0 - \lambda)] \tag{4}$$

where $a(\lambda)$, $a(\lambda_0)$ are the absorption coefficients at wavelengths λ and λ_0, respectively, and S is the exponential slope coefficient.

Fluorescence measurements

Fluorescence was measured using a 1-cm quartz cell in a Shimadzu 5301 spectrofluorometer with an excitation wavelength of 355 nm and an emission wavelength of 450 nm. A Milli-Q water blank was used for comparison (Seritti et al., 1998; Rochelle-Newall & Fisher, 2002b).

Data analysis

Statistical analyses were performed with SPSS 11.0 software (Statistical Program for Social Sciences).

Results and discussion

Spectral character and spatial distribution of CDOM

Figure 2a, b shows representative absorption spectra of samples from northern Meiliang Bay (3), the Bay inlet (24), and the central lake basin (31, 40), which represent a range of CDOM concentrations. Spectral curves for CDOM generally showed near zero absorption at the red end of the visible spectrum (700 nm) and exponentially increase through the ultraviolet (UV) wavelength regimes (280–400 nm). Mean corrected CDOM absorption coefficients at 280 nm [$a(280)$] and 355 nm [$a(355)$] were 18.3 ± 5.6 and 4.6 ± 1.8 m^{-1}, respectively, for the 40 sites. CDOM absorption coefficients at 280, 355 and 440 nm for each site are shown in Fig. 3. The highest value of $a(355)$ was 8.6 m^{-1} at site 15 close to shore in Meiliang Bay, and the lowest value was 2.2 m^{-1} at site 40 near the center of the lake (Figs. 1, 3). In general, $a(355)$ decreased away from the river inflow to Meiliang Bay towards the lake center (Figs. 1–3). CDOM absorption in the Meiliang Bay inlet were around twice those in the central basin. All sites were divided into two groups, one representing Meiliang Bay (sites 1–24) and one representing the central basin (sites 25–40). Significant differences were found between Meiliang Bay and the central basin arising from spatial differences in absorption coefficients, DOC-specific absorption coefficients, exponential slope coefficients, DOC concentrations and fluorescence values using ANOVA ($P \leq 0.005$). Values of $a(280)$ and $a(355)$ in Lake Taihu are higher than those in estuarine and coastal waters (Seritti et al., 1998; Chen et al., 2004), but CDOM absorption values are close to those in large, shallow, fluvial Lake Saint-Pierre (Frenette et al., 2003) and others (Kirk, 1994; Morris et al., 1995). The ratio of $a(250)$ to $a(365)$ ranged from 5.28 to 9.24 with a mean value of 7.10 ± 0.94 (standard deviation), which was close to results from other studies (De Haan, 1993; Huovinen et al., 2003). Higher ratios corresponding to smaller molecules were found in the lake center in agreement with findings from Huovinen et al. (2003) and Yacobi et al. (2003).

Relationship between CDOM absorption and DOC

DOC concentrations ranged from a maximum of 10.9 mg l^{-1} at the Liangxihe mouth (site 1) to a minimum of 6.0 mg l^{-1} in the central basin (Site 37), with a mean value of 8.17 ± 1.38 mg l^{-1}. A significant positive correlation was found between DOC and CDOM absorption coefficients in the ultraviolet and blue band (280–500 nm), especially at shorter wavelengths (Fig. 4a), in agreement with other studies where DOC was dominated by CDOM (Seritti et al., 1998; Stedmon et al., 2000; Laurion et al., 2000). Figure 4b shows that the coefficients of determination (r^2) between CDOM absorption and DOC decrease with increasing wavelength from 280 nm to 500 nm. Similar results were found in shallow lakes in the middle and lower reaches of the Changjiang (Yangtze) River (Zhang et al., 2005).

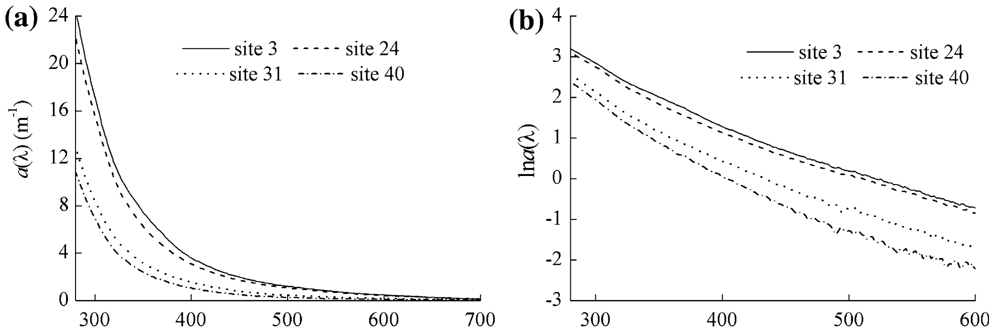

Fig. 2 CDOM absorption spectra of representative sites: (**a**) Wavelength versus $a(\lambda)$ and (**b**) wavelength versus $\ln[a(\lambda)]$

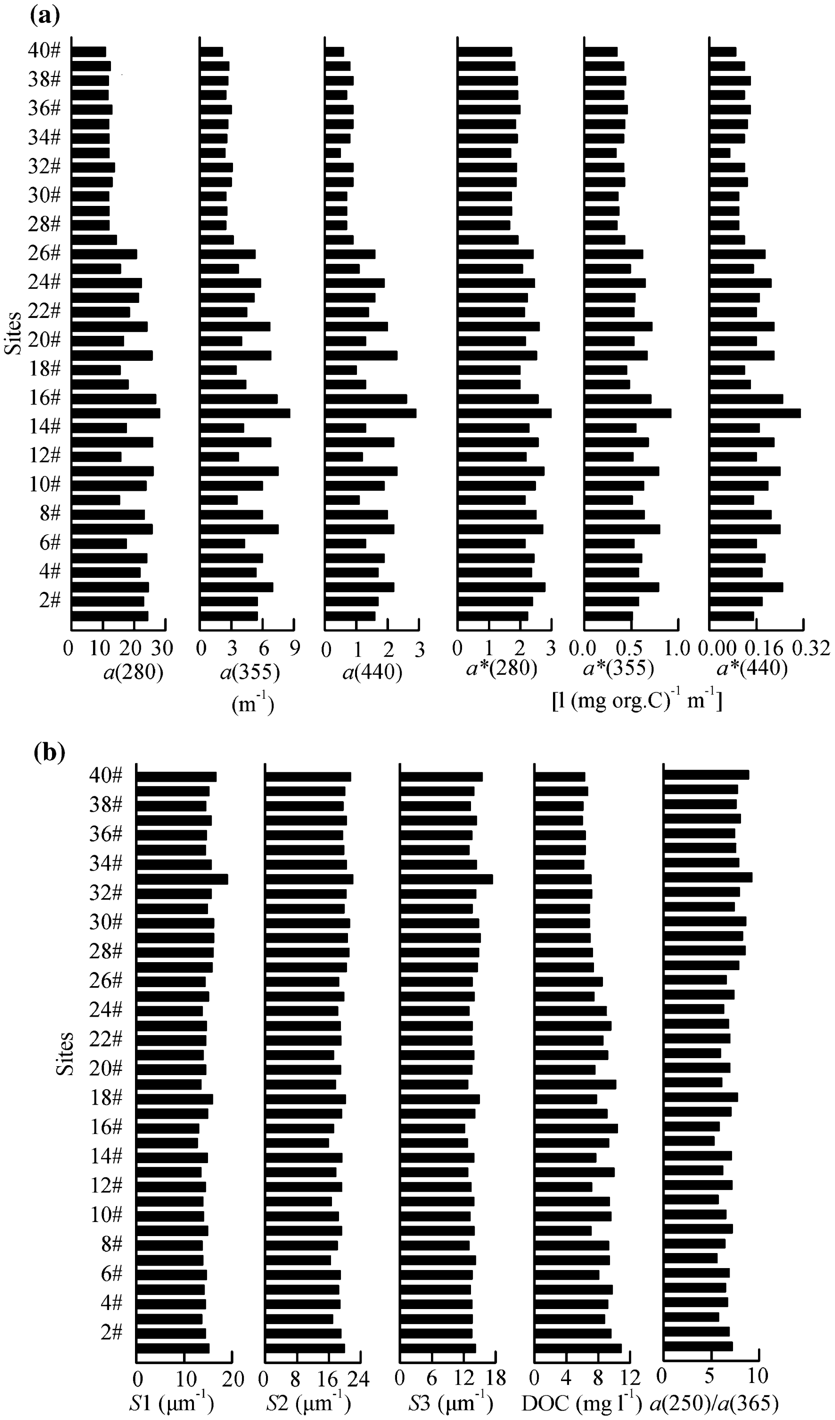

Fig. 3 Absorption coefficients $a(280)$, $a(355)$, $a(440)$, DOC-specific absorption coefficients, $a^*(280)$, $a^*(355)$, $a^*(440)$, spectral slope $S1$ (280–500 nm), $S2$ (280–360 nm), $S3$ (360–440 nm), DOC, fluorescence and ratio of $a(250)$ to $a(365)$

Relationships between DOC and CDOM absorption from this study and other studies are presented in Table 1. Published relationships of DOC and $a(355)$ are valuable tools to model DOC concentration (Seritti et al., 1998; Rochelle-Newall & Fisher, 2002b). However, differences between models suggest regional and lake-to-lake variations in bio-optical characteristics.

Calculated DOC-specific absorption provides an additional means for distinguishing bio-optical

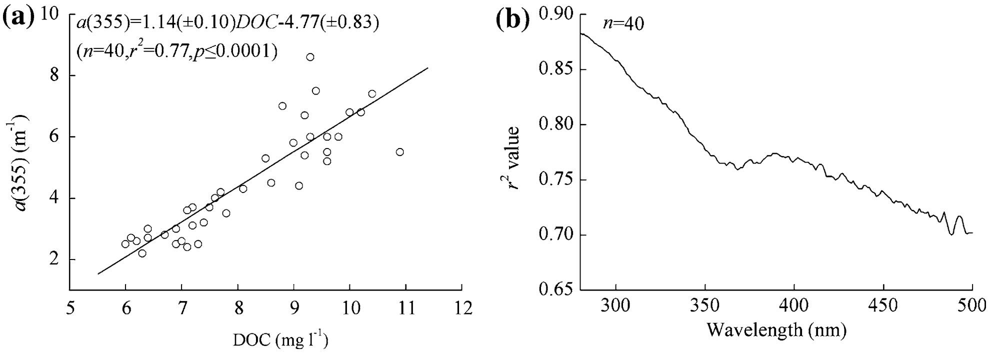

Fig. 4 Plots of CDOM absorption and DOC concentration (**a**) CDOM absorption $a(355)$ versus DOC concentration and (**b**) wavelength versus determination coefficients r^2 value between $a(\lambda)$ and DOC concentration from 280 nm to 500 nm

Table 1 Linear models for the relationship between DOC concentrations and CDOM absorption coefficients

Equations		r^2	n	P	DOC (mg l^{-1})	References
DOC, $a(\lambda)$	$a(355) = 1.465\text{DOC} - 1.315$	0.98	31	≤0.0001	0.89–4.82	Seritti et al. (1998)
	$a(440) = 0.280\text{DOC}$	0.91	6	≤0.0007	4.9–40.7	Yacobi et al. (2003)
	$a(440) = 0.235\text{DOC}$	0.92	6	≤0.0005	7.1–40.5	Yacobi et al. (2003)
	$a(440) = 0.429\text{DOC} - 0.744$	0.73	64	≤0.0001	0.24–23.5	Morris et al. (1995)[a]
	$a(300) = 3.584\text{DOC} - 2.690$	0.71	26	≤0.0001	0.77–3.31	Del Castillo et al. (1999)[a]
	$a(440) = 0.131\text{DOC} + 0.020$	0.58	39	≤0.0001	0.21–3.5	Laurion et al. (2000)[a]
	$a(440) = 0.552\text{DOC} - 1.477$	0.77	12	≤0.0005	4.9–14.9	Huovinen et al. (2003)[a]
	$a(355) = 0.015\text{DOC} - 5.5$	0.90	92	≤0.0001	–	Ferrari et al. (1996)[b]
	$a(355) = 0.00036\text{DOC} + 0.028$	0.66	43	≤0.0001	–	Ferrari (2000)[b]
	$a(355) = 0.744\text{DOC} - 2.176$	0.58	22	≤0.001	2.7–10.1	Zhang et al. (2005)
	$a(355) = 1.14\text{DOC} - 4.77$	0.77	40	≤0.0001	6.0–10.9	Present study
	$a(375) = 0.87\text{DOC} - 3.74$	0.77	40	≤0.0001	6.0–10.9	Present study
	$a(440) = 0.38\text{DOC} - 1.73$	0.73	40	≤0.0001	6.0–10.9	Present study

[a] Fitted with the data in papers ; [b] DOC unit: μM

characteristics of CDOM. Low CDOM absorption was associated with low values of $a^*(\lambda)$. Values of $a^*(355)$ ranged from 0.34 to 0.92 with an average 0.54 ± 0.14 [l (mg org C)$^{-1}$ m^{-1}], which are higher than those observed in estuarine and coastal waters (Seritti et al., 1998). Lowest values of $a^*(320)$ and $a^*(355)$ corresponded with low DOC concentrations and approached those measured in coastal water and mountain lakes (Seritti et al., 1998; Laurion et al., 2000). DOC-specific absorption coefficient represents CDOM composition, which is used to establish relationships with other parameters. Specific absorption was correlated with pH and DOC concentration in three acid lakes (Gallie, 1997). In our study, specific absorption was correlated positively with DOC concentration. The linear regression equation is $a^*(355) = 0.075\text{DOC} - 0.070$ ($r^2 = 0.53$, $n = 40$, $P \le 0.0001$). The linear correlation between DOC concentration and specific absorption is known from other studies (Yacobi et al., 2003). Furthermore, the higher average $a^*(440)$ corresponded to the higher proportion of large and medium fractions in CDOM (Yacobi et al., 2003). In this study, higher $a^*(355)$ and $a^*(440)$ values corresponded to lower $a(250)/a(365)$ values towards increasing molecular size. The regression equation between $a^*(355)$ and $a(250)/a(365)$ values is: $a^*(355) = -0.145a(250)/a(365) + 1.570$ ($r^2 = 0.92$, $n = 40$, $P \le 0.0001$).

Estimation of the spectral slope S

It is widely accepted that spectral slope S can be used as a proxy for CDOM composition (Kowalczuk et al., 2003). The values of the exponential slope coefficient (*S*) ranged from 12.7 μm^{-1} to 19.0 μm^{-1} with an average value of 14.74 ± 1.13 μm^{-1}, from 15.9 μm^{-1} to 22.0 μm^{-1} with an average value 19.11 ± 1.42 μm^{-1}, and from 12.2 to 17.4 μm^{-1} with an average value 13.86 ± 0.91 μm^{-1} for the wavelength domain from 280 nm to 500 nm (*S*1), 280–360 nm (*S*2) and 360–440 nm (*S*3), respectively. The coefficient of determination (r^2) for the fit of *S* was higher than 0.99. Although slope values in the present investigation fell within the range of previously reported values (Bricaud et al., 1981; Davies-Colley, 1983; Seritti et al., 1998; Markager & Vincent, 2000), variability of the spectral slope in different wavelength ranges is significant, which illustrates the importance and sensitivity of the wavelength range chosen to calculate the slope coefficient (*S*). *S* generally increases with decrease in the wavelength range and may be influenced by CDOM composition (i.e. humic and fulvic acids affecting spectral properties). Samples collected from the Mississippi River plume in the Gulf of Mexico, where CDOM was comprised predominantly from fulvic acids, had a steeper slope ($S = 0.0194$) than marine samples, where CDOM was mostly humic acids ($S = 0.010$) (Carder et al., 1989). *S* values in Lake Taihu were higher than those for oceanic and coastal waters (Markager & Vincent, 2000).

In our study, there are significant negative correlations between spectral slope *S* and *a*(355) (Fig. 5a) and *a**(355) (Fig. 5b, Table 2). Determination coefficients between *S* and $a^*(\lambda)$ were higher than those between *S* and $a(\lambda)$. It is also interesting that determination coefficients are the highest between *S*2 (280–306 nm) and $a(\lambda)$, $a^*(\lambda)$. Furthermore, significant negative correlations have been obtained between *S* values and *a**(440) (Yacobi et al., 2003), *a*(355) (Rochelle-Newall et al., 2004) and *a*(375) (Del Castillo &

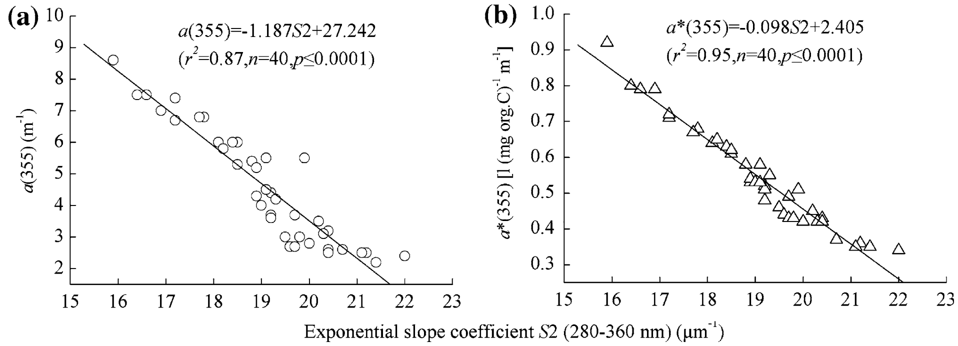

Fig. 5 Plots of exponential slope coefficient versus (**a**) absorption coefficient *a*(355) and (**b**) specific absorption coefficient *a**(355)

Table 2 Linear models for the relationship between *S*1 (280–500 nm), *S*2 (280–360 nm), *S*3 (360–440 nm) and absorption coefficient, DOC-specific absorption coefficient of CDOM ($n = 40$, $P \le 0.0001$)

Parameters		Equations	r^2
a(355), *S*	*S*1 (280–500 nm)	$a(355) = -1.254(\pm 0.160)S1 + 23.045(\pm 2.357)$	0.62
	*S*2 (280–360 nm)	$a(355) = -1.187(\pm 0.075)S2 + 27.242(\pm 1.444)$	0.87
	*S*3 (360–440 nm)	$a(355) = -1.158(\pm 0.262)S3 + 20.613(\pm 3.646)$	0.34
a(375), *S*	*S*1 (280–500 nm)	$a(375) = -0.976(\pm 0.119)S1 + 17.772(\pm 1.751)$	0.64
	*S*2 (280–360 nm)	$a(375) = -0.910(\pm 0.057)S2 + 20.779(\pm 1.087)$	0.87
	*S*3 (360–440 nm)	$a(375) = -0.919(\pm 0.197)S3 + 16.133(\pm 2.733)$	0.37
*a**(355), *S*	*S*1 (280–500 nm)	$a^*(355) = -0.103(\pm 0.011)S1 + 2.064(\pm 0.168)$	0.68
	*S*2 (280–360 nm)	$a^*(355) = -0.098(\pm 0.004)S2 + 2.405(\pm 0.067)$	0.95
	*S*3 (360–440 nm)	$a^*(355) = -0.095(\pm 0.020)S3 + 1.858(\pm 0.279)$	0.37
*a**(375), *S*	*S*1 (280–500 nm)	$a^*(375) = -0.083(\pm 0.008)S1 + 1.627(\pm 0.124)$	0.72
	*S*2 (280–360 nm)	$a^*(375) = -0.077(\pm 0.003)S2 + 1.868(\pm 0.049)$	0.96
	*S*3 (360–440 nm)	$a^*(375) = -0.079(\pm 0.015)S3 + 1.495(\pm 0.211)$	0.42

Coble, 2000; Stedmon et al., 2000). Kowalczuk et al. (2003) presented a trend of increased slope coefficient with a decrease in absorption level.

Relationship between CDOM absorption and fluorescence

CDOM absorption may be measured directly using a spectrophotometer or inferred from fluorescence, which can be measured more rapidly and with greater sensitivity than absorption. In previous studies, a strong relationship was established between CDOM absorption and fluorescence intensity (Vodacek et al., 1995; Seritti et al., 1998; Ferrari & Dowell, 1998; Hoge et al., 1993; Kowalczuk et al., 2003) despite numerous sources of CDOM and different chemical characteristics. CDOM fluorescence $F_n(355)$ and absorption coefficients $a(355)$ were correlated positively in Lake Taihu in this study (Fig. 6). The correlation may enable the use of fluorescence as a proxy for CDOM absorption despite different sources from river inputs and autochthonous humic substances and locations within the lake.

Conclusions

(1) Higher CDOM absorption coefficients were found in the Meiliang Bay inlet of Lake Taihu in association with increased DOC concentrations and fluorescence intensity. There were significant differences between Meiliang Bay and the central lake basin in absorption coefficients, DOC-specific absorption coefficient $a^*(355)$, exponential slope coefficient (S), DOC concentrations and fluorescence values.

(2) There is a significant linear relationship between CDOM absorption and DOC concentrations, especially at shorter wavelengths in Lake Taihu samples.

(3) Exponential slope coefficients fall in the range reported in the literatures. There is a significant negative linear relationship between S values and CDOM absorption coefficients, as well as DOC-specific absorption coefficients.

(4) There is a linear relationship ($r = 0.66$, $P \leq 0.0001$) between CDOM absorption and CDOM fluorescence intensity: $a(355) = 2.12(\pm0.33) + 3.72(\pm0.43)F_n(355)$.

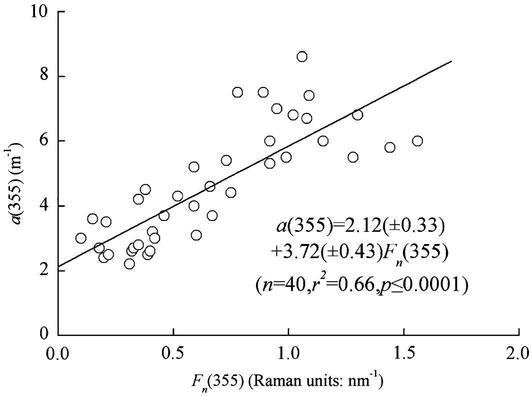

Fig. 6 Plot of $a(355)$ versus $F_n(355)$

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