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RESEARCH ARTICLE

Land use regulates the spectroscopic properties and sources of dissolved organic matter in the inflowing rivers of a large plateau lake in southwestern China: implication for organic pollution control

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Abstract

Dissolved organic matter (DOM) transported by inflowing rivers can considerably contribute to the organic loadings of lakes. The current study characterized the DOM properties and source apportionment in the inflowing rivers of Dianchi Lake, the sixth largest freshwater lake in China suffering from organic pollution, during the rainy season by using spectroscopic and carbon stable isotope techniques, and the regulation role of land use was assessed. The results showed that land use (urbanized, agricultural, or mixed) largely affected DOM properties. Greater concentrations and fluorescence intensities of DOM with low aromaticity and dominant autochthonous sources were observed in the urban rivers than in the agricultural rivers. The proportion of humic-like substances increased, while that of tryptophan-like matter decreased from upstream to downstream of two main urban rivers. DOM in the agricultural rivers was characterized by more amounts of aromatic humic-like substances with dominant allochthonous sources compared to that in the urban rivers. Stable isotope analysis showed that the decomposition of macrophytes and input of terrestrial sources from C3 plant-dominated soil and sewage were the major DOM origins in the rivers. The positive linear relationship between the chemical oxygen demand (COD) concentration and fluorescence intensities of terrigenous DOM components implied the necessity of controlling exogenous inputs to alleviate organic pollution in the Dianchi Lake.

Keywords Dianchi Lake · Inflowing rivers · Dissolved organic matter (DOM) · Land use type · Optical properties · Stable carbon isotopes

Introduction

Dianchi Lake, the sixth largest freshwater lake in China with a water area of more than 300 km^2, is a very important water body located in Kunming city of Yunnan Province and has significant environmental, ecological, and socioeconomic functions (He et al. 2022a). However, this lake has experienced serious pollution and eutrophication during the past two decades, mainly due to the discharge of large quantities of contaminants from the continuously urbanized watershed with an extremely high population density and rapid socioeconomic development (Guo et al. 2017). Moreover, non-point source (NPS) pollution from intensive agriculture in the watershed has considerably and increasingly contributed to the water pollution of this lake (Wang et al. 2020). There are more than 30 rivers directly or indirectly pouring into the lake, carrying many organics and high nutrient loads. Substances leached from soils and litter and NPS waste streams as well as endogenous products from aquatic plant biomass and microbial activities predominantly contribute to pollutant loads in rivers (Zhang et al. 2021). A portion of secondary effluent from wastewater treatment plants (WWTPs) containing refractory organic substances and microbial products is also imported into inflowing rivers (Wang et al. 2019). Intensive human activities can destabilize river hydrologic flows and cause pollution enrichment

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in water ecosystems (Wen et al. 2021a). Although the point source pollution inputs into Dianchi Lake have been effectively controlled in recent years by an assembly of strategies and measures that have spent substantial manpower and material resources, the high concentration level of organic pollution in the lake is still a major challenge for lake environment restoration due to the import of non-point source pollution and high accumulation of endogenous pollutants in the lake sediment (Guo et al. 2017; He et al. 2022a). Therefore, profiling water quality characteristics, including those of DOM, in the inflowing rivers is of significance to provide essential information on pollution control and ecosystem restoration for this lake.

High DOM concentrations can pose a great threat to the health of aquatic ecosystems (Shi et al. 2020). DOM is a chemically complex and heterogeneous organic mixture composed of lipids, proteins, humic acids, and other active organic compounds and can actively be involved into the biogeochemical cycles of C, N, S, and micronutrients (Artifon et al. 2019; Dong et al. 2020). It is well recognized that the quality of DOM plays more significant roles in aquatic ecosystems than the total DOM quantity (Zhang et al. 2021). Some components in DOM have high chemical activity or can act as electron shuttles and thus can influence a series of biological, physical, and chemical processes, such as complexation, biodegradation, and photodegradation, in aquatic environments (Majumdar et al. 2017). DOM quality can determine the distribution, transformation, bioavailability, and toxicity of many exogenous chemicals, such as heavy metals and polyfluoroalkyl substances, via their interactions (Derrien et al. 2019; Zhang et al. 2021). Aquatic trophic status and ecosystem health are also closely associated with DOM features (Wang et al. 2019). Among the DOM components, chromophoric DOM (CDOM) is an optically active component that can strongly adsorb ultraviolet and photosynthetically available radiation (Bai et al. 2017). The penetration of sunlight into the water column, which is crucial for photo-ecological processes such as primary production and the survival of various aquatic organisms, can be restricted by CDOM (Majumdar et al. 2017). Therefore, DOM can be used as a critical tracer of water quality and matter cycling and availability in aquatic environments, and its characterization is thus highly essential for aquatic ecosystem protection and management. An increasing number of studies have focused on the spatiotemporal DOM properties in various aquatic systems and the influencing factors as well as the corresponding driving mechanisms in recent years (He et al. 2021; Zhang et al. 2021). DOM pools in water bodies can originate from allochthonous inputs and autochthonous production. Allochthonous DOM is primarily derived from domestic and industrial wastewater and urban and agricultural runoff containing leachates from wastes, soils, litter, and compost in surrounding terrestrial systems with distinct properties (Nguyen and Hur 2011). Autochthonous DOM mainly originates from the release and microbial decomposition of aquatic organisms (e.g., algae and aquatic plants) and polluted sediments (Qu et al. 2013; Zhou et al. 2016). Therefore, DOM content, properties, and source apportionment in aquatic environments can be regulated by a variety of factors, especially anthropogenic activities, including industrialization, urbanization, and agricultural production (Guo et al. 2017; Cao et al. 2018).

Multiple techniques, such as UV−visible and fluorescence spectroscopic approaches targeting CDOM with advantages of low cost and high sensitivity (DeVilbiss et al. 2016; Bai et al. 2017; Feng et al. 2022) and carbon/nitrogen stable isotope analysis (Phillips and Gregg 2003; Dong et al. 2020; Wen et al. 2020), have been developed to effectively characterize the concentration and chemical composition structure and track DOM sources in various water and sediment samples in past years. The coupling of three-dimensional excitation-emission matrices (EEMs) with parallel factor (PARAFAC) analysis, a spectral-based approach, can reveal DOM chemical compositions at a low cost and rapid rate (Fellman et al. 2010; Derrien et al. 2019). Spectroscopic methods can provide supplementary information on the aromaticity, sources, reactivities, and behaviors of DOM (Zhang et al. 2021). A better understanding of DOM properties can greatly improve our understanding of the underlying mechanisms of changes in water environmental quality and ecological functions (Zhang et al. 2022). Stable carbon isotopes in the environment cannot be transformed or degraded by biological processes; thus, the combination of the $\delta^{13}C$ and C:N ratios is recognized as an effective method to track DOM sources in aquatic systems (Kendall et al. 2001; Dong et al. 2020). Stable isotopes in sediment can provide long-term important environmental information for river ecosystems (Kendall et al. 2001). Isotope mixing models based on mass conservation laws have been developed to calculate the contributions of different DOM sources in samples (Phillips and Gregg 2003; Wen et al. 2020, 2021a).

Although water import through the inflowing rivers into Dianchi Lake is highly critical for the water resource safety and ecological environmental health of the lake, information on the quantity, structure, and sources of DOM in the inflowing rivers of the lake, particularly the influence of land use type (urbanized or agricultural) in the watershed, has been rarely reported. Hence, in the present study, the DOM optical properties in water samples of 35 inflowing rivers and the sole outflowing river of Dianchi Lake were characterized by using spectroscopic approaches of UV absorption and EEMs-PARAFAC. The sources of organic matter (OM) in the river were further explored by a stable C/N isotope method. The main specific purposes of the study were to (1) characterize the quantity and compositions of DOM in the inflowing and outflowing rivers of Dianchi Lake, (2) evaluate the effect of land use types (urbanized, agricultural, and mixed urbanized-agricultural) on the DOM properties in the rivers, and (3) explore the sources of DOM in the rivers by combining spectroscopic properties and C/N isotopes. Overall, this study is aimed at providing basic knowledge on the DOM properties in the inflowing rivers of Dianchi Lake to support the protection and management of the rivers and lake ecosystems.

Material and methods

Study area and sample collection

Dianchi Lake (24° 27′ – 25° 27′ N, 102° 29′ – 103° 01′ E, altitude: 1886 m) is a typical shallow lake located in Kunming city, the capital of Yunnan Province in southwestern China, with a subtropical plateau mountain monsoon climate. It is the largest lake in the Yunnan-Kweichow Plateau, with a lake area of more than 300 km^2 and a watershed area of more than 2900 km^2. Dianchi Lake plays critical roles in local economic and social development, microclimate formation, ecological service, and tourism value provision and flood prevention. This lake is nearly surrounded by anthropogenic land use such as urban construction area or farmland with intensive human activities (Fig. 1). Surface runoff carrying pollutants in the upstream watershed can be imported into Dianchi Lake directly or indirectly through 35 inflowing rivers. Among them, rivers R1–R23 are located on the north side of the lake and flow through heavily urbanized and densely populated areas with almost all the land use being built-up land except for R1 and R2 (Fig. 1). Rivers R24–R27 on the east side of the lake and R34 and R35 on the west side flow through a mixed urbanized-agricultural land use area, with aquatic macrophytes restored in some sections for water ecosystem amelioration. Rivers R28–R33, located on the eastern and southern sides of the lake, predominantly flow through agricultural land use areas. R36 on the west side is the sole outlet river of Dianchi Lake. The annual precipitation in the lake watershed is approximately

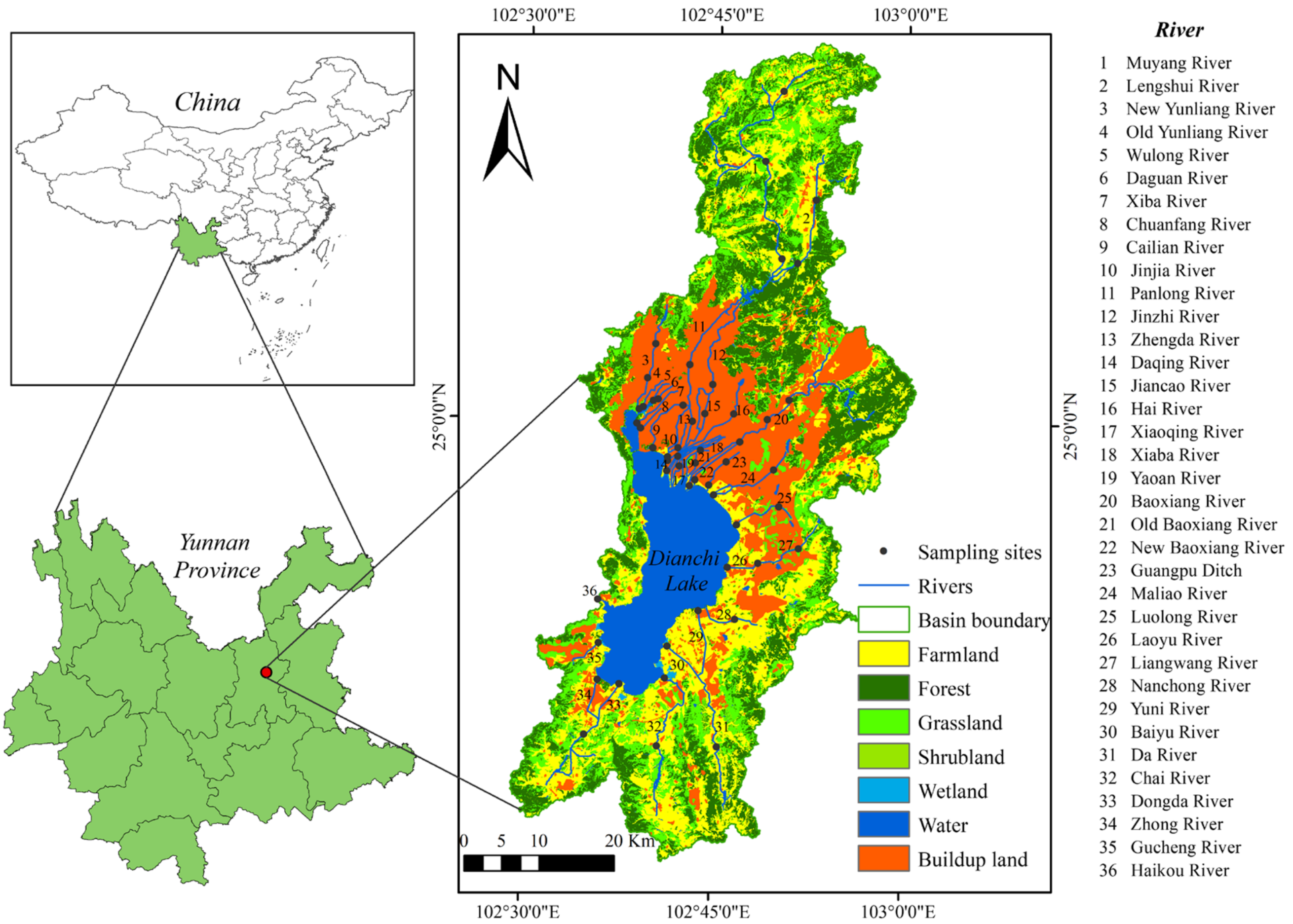

Fig. 1 Map showing the study area of the Dianchi Lake watershed with different land use types and sampling sites in the inflowing and outflowing rivers of Dianchi Lake in Yunnan Province, China

1000 mm, over 80% of which occurs in the rainy season from May to October. Thus, characterizing river DOM in the rainy season is more significant. We collected water samples (0.3–0.5 m in depth) at the estuary of the inflowing rivers (Fig. 1) in July 2021 (during the mid-rainy season). Moreover, water samples in the upstream and midstream of four main inflowing rivers flowing through different land use areas were collected to investigate the spatial variations in DOM properties along the rivers. Geographical information of the sampling points is listed in Table S1. Furthermore, surface sediment samples (0–10 cm if available) in the rivers were collected using a grab sampler, and branches and bulk debris were manually removed.

Water chemical parameter measurement

The collected water samples were stored in clean amber bottles in a portable refrigerator and taken back into the laboratory at Yunnan University for immediate processing. The samples were filtered using a 0.45-μm mixed fiber Millipore filter to remove suspended matter and acidified using HCl to $pH < 2$. Then, the DOC concentration in the filtrate was measured by a total organic carbon analyzer (TOC-L CPN, Shimadzu, Japan). Other water chemical parameters, including COD, ammonia nitrogen (NH_4^+–N), nitrate nitrogen (NO_3^-–N), total nitrogen (TN), and total phosphorus (TP), were measured according to standard methods (SEPA 2002).

DOM absorption and fluorescence analysis and PARAFAC modeling

The UV−visible absorption spectra of the filtered water samples were recorded using a UV spectrophotometer (Shimadzu, UV-2600, Japan) over a wavelength range of 200–800 nm with 1-nm increments. Milli-Q water was employed as a blank control in the reference cell, and the instrument baseline was corrected by subtracting the absorbance at 700 nm before sample analysis. The fluorescence EEM spectra of DOM were measured using a fluorescence spectrophotometer (Shimadzu, RF-6000, Japan) in a 1-cm quartz cuvette. The excitation wavelengths (Ex) ranged from 200 to 450 nm with a scanning interval of 5.0 nm, and the emission wavelengths (Em) ranged from 250 to 600 nm with a scanning interval of 1.0 nm. The scanning speed was 2000 nm/min, and the excitation and emission bandwidths were both set as 5 nm. Milli-Q water was employed as a blank to record EEMs by the same method, and the blank EEMs were subtracted from the sample EEMs to eliminate water Raman scattering. The inner-filter effects were corrected using the CDOM absorbance measured at corresponding Ex and Em. The interpolation method was used to eliminate the effect of Rayleigh scattering. Simultaneously, Raman spectra were recorded at an excitation wavelength of 350 nm, and the Raman peak area was used to normalize the EEM data for regulating Raman scattering to produce corrected fluorescence intensity expressed in Raman units (R.U.).

PARAFAC analysis of the EEM data of the samples was carried out to identify and characterize DOM fluorescent components using the DOMFluor v.1.7 Toolbox in MATLAB R2015b (Natick, MA, USA). After removing outliers and eliminating scattering from the PARAFAC model, four components were finally determined to provide a robust description of DOM fluorescence in the dataset through model validation by split half test and random initialization analysis together with the residuals analysis (Dong et al. 2020). The spectral shapes and locations of the PARAFAC components were compared to previously identified components in aquatic ecosystems using an online spectral library (OpenFluor) recording fluorescent DOM. The maximum fluorescence intensity (Fmax) of each component obtained by modeling was used to quantify its fluorescence intensity (Wen et al. 2020). The proportion of each component was calculated by dividing its Fmax value by the total fluorescence intensities of all components.

The absorbance and fluorescence properties of DOM are defined as "optical indexes" and can be used to profile potential sources and the biochemical structure of DOM (Derrien et al. 2019). Three important indexes are calculated based on the absorbance properties. Specific ultraviolet absorbance ($SUVA_{254}$, L m^{-1} mg^{-1}) was calculated by dividing the UV absorbance at 254 nm (a_{254}) by the DOC concentration, which can represent the humus content and indirectly reflect the aromatic and hydrophobic/hydrophilic properties of DOM (Weishaar et al. 2003). The absorption coefficient a_{254} (m^{-1}) is calculated as $a_{254} = 2.303 \times A_{254}/L$, where L is the path length of the quartz cuvette (1 cm). This parameter represents the CDOM absorption at a wavelength of 254 nm and can be used as a surrogate for the CDOM concentration (Derrien et al. 2019). $S_{275–295}$ is calculated by linear fitting of the logarithm of absorption coefficients over the wavelength interval of 275–295 nm to provide information on DOM molecular weight ($S_{275–295}$ is negatively correlated with the DOM molecular weight) (DeVilbiss et al. 2016).

Four fluorescent proxies, including the humification index (HIX), fluorescence index (FI), biological index (BIX), and freshness index (β/α), were calculated by the ratio of fluorescence intensities. Specifically, the HIX was calculated as the ratio between the sum of fluorescence intensity at the Em of 435–480 nm to the sum of Em intensity between 300–435 and 435–480 nm under an Ex at 254 nm, which can reflect the degree of DOM decomposition and humification (DeVilbiss et al. 2016). The FI was calculated from the ratio of fluorescence intensity at an Em of 450 nm to that at 500 nm under an Ex at 370 nm to discriminate terrestrial or microbial sources of DOM (Dong et al. 2020). $FI > 1.7$ usually indicates dominantly microbial sources, while $FI < 1.2$ indicates terrestrial input (Fellman et al. 2010; Xie et al. 2018).

The BIX is also an indicator of the DOM source that can reflect autochthonous biological productivity in aquatic environments and can be obtained from the ratio of Em intensity at 380 nm to that at 430 nm with an Ex of 310 nm (Bai et al. 2017). Samples with a high BIX value > 1 usually correspond to a predominant freshly self-initiating DOM source in water, while those with a low value < 0.7 are deemed to have a low autochthonous origin (Chen et al. 2018). The freshness index β/α can reflect the contribution of recently produced DOM and is calculated by the ratio of the Em intensity at 380 nm to the maximum Em between 420 and 435 nm when the Ex intensity is 310 nm (Sepp et al. 2019).

C/N stable isotope analysis

The $\delta^{13}C$ and $\delta^{15}N$ in the river sediment samples were analyzed by using an isotope-ratio mass spectrometer coupled with an elemental analyzer (Isoprime 100, trace gas, vario micro cube) with an analytical precision (SD) of 0.999. Before analysis, inorganic carbon was removed from the samples by adding 1 mol/L HCl. The carbon and nitrogen isotope values δ (in ‰) were expressed as the relative difference in isotope ratios between the samples and the conventional standards, where carbon and nitrogen standard reference materials were Vienna Pee Dee Belemnite (VPDB) and atmospheric nitrogen, respectively. The proportional contribution of potential carbon sources to total OM in the rivers was calculated by establishing isotopic multivariate mixture models using IsoSource software (Phillips and Gregg 2003; Phillips et al. 2005). The source increment and mass balance tolerance were set to 2% and 0.05‰, respectively.

Statistical analysis

Based on the digital elevation model (DEM) data (http://www.gscloud.cn/sources) of the Dianchi Lake watershed, the ArcGIS10.2 software package (Redlands, CA, USA) was used to delineate the Dianchi Lake basin and the related rivers and then display the spatial distribution of the sampling sites and DOM properties. The land use data were obtained from GLOBELAND 30. Data plots were produced using Origin Pro 2018 software (Origin Lab, Northampton, MA, USA). The differences in indexes of DOM properties and general hydrochemical parameters were determined by one-way ANOVA followed by Tukey post hoc tests using SPSS, with $p < 0.05$ regarded as significantly different. Pearson's correlation analysis was carried out to explore the correlations between the DOM properties and general hydrochemical parameters. Canoco 5 software was applied for principal component analysis (PCA) of the integrated differences of the river water samples collected from different land use areas based on the DOM properties and general hydrochemical parameters.

Results and discussion

General hydrochemical properties

The concentrations of NH_4^+-N, NO_3^--N, TN, TP, and COD in the inflowing rivers flowing through different land use areas are listed in Table S2. The water quality in the rivers varied in a large range from 0.20 to 5.03 mg/L NH_4^+-N, 0.05 to 18.26 mg/L NO_3^--N, 1.53 to 23.77 mg/L TN, 0.09 to 0.81 mg/L TP, and 0.56 to 24.80 mg/L COD. The high nutrient concentrations in some rivers, such as the Cailian River (R9) and Xiaoqing River (R17), implied the necessity of mitigating nutritional pollution in the rivers to protect Dianchi lake from eutrophication. Land use greatly influenced the water quality characteristics in the rivers. Specifically, the average NH_4^+-N (1.50 ± 1.36 mg/L) and COD (10.40 ± 7.25 mg/L) concentrations in the urban rivers were higher than those in other rivers. Surface runoff in urbanized catchments usually contains high contents of reducing substances, such as organic nitrogen and NH_4^+-N, which can cause the formation of black-odor water bodies (Guo et al. 2017). In the agricultural rivers, higher mean NO_3^--N and TN concentrations of 7.40 ± 6.19 and 11.72 ± 6.63 mg/L were measured compared to those in other rivers. The application of nitrogen fertilizers for agricultural production might be primarily responsible for the high concentration of nitrogen in the dominant form of NO_3^--N in rivers (Feng et al. 2022). Land use displayed only a slight effect on the TP concentrations in the rivers, with the highest average value of 0.32 ± 0.25 mg/L observed in the mixed land use area. Phosphorus is a limiting factor for algal blooms in water bodies, so reducing the high concentration level of P in some rivers is highly necessary.

DOC import into lake by the inflowing rivers

DOC is a good index of organic pollution and is usually used to quantify the DOM concentration in aquatic environments (Tran et al. 2015). The DOC concentrations in the inflowing rivers and DOC mass imported into Dianchi Lake by the rivers are depicted in Fig. 2. The results showed that the DOC concentrations in the rivers were spatially different. Among these rivers, R6 (Daguan River) flowing through a highly urbanized area with a low flow rate presented the highest DOC concentration of 26.87 mg/L, which was notably higher than those in other rivers. R32 (Chai River) in the agricultural area had the lowest DOC concentration of 2.75 mg/L. The DOC import mass of the rivers was calculated by multiplying the DOC concentration by the flow rate (Table S3). R11 (Panlong River) conveyed the highest quantity of DOM into the lake (41.40 g/s) despite its relatively low DOC concentration (5.45 mg/L) because of its high

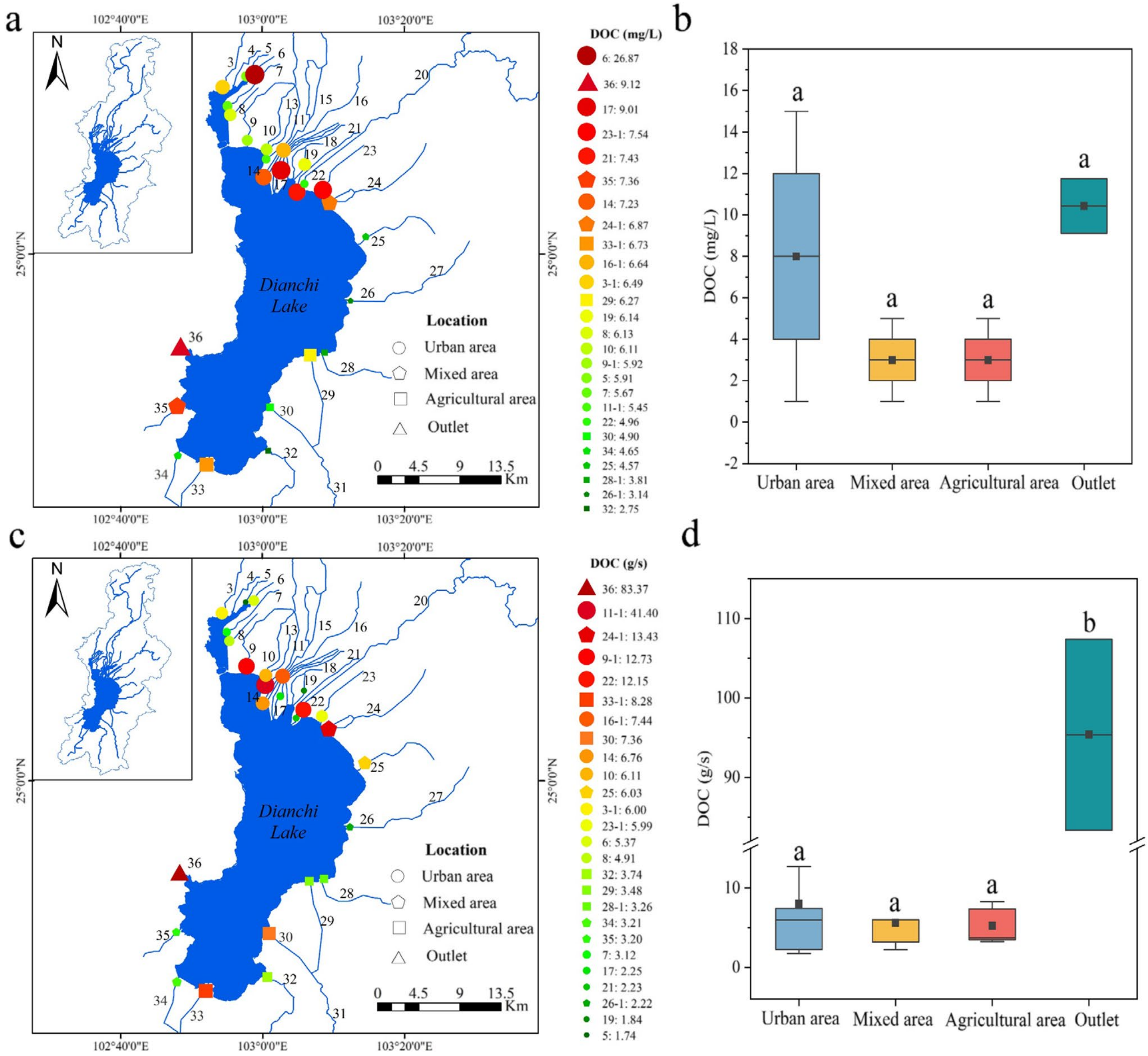

Fig. 2 DOC concentration in the inflowing rivers (**a** and **b**) and the corresponding DOC quantity imported into Dianchi Lake (**c** and **d**)

flow rate. The Panlong River flows through urbanized areas and acts as a channel for the diversion of water resources from the Niulan River outside the watershed to improve the water resource safety and quality of Dianchi Lake (He et al. 2022b). Therefore, maintaining a low DOC concentration in this river by controlling organic pollutant inputs is essential for protecting Dianchi Lake from organic pollution.

Both the average DOC concentration and the DOC mass carried by the urban rivers were higher than those in the agricultural and mixed land use areas. A large amount of anthropogenic carbonaceous substances can be transported into rivers from urbanized landscapes with predominant artificial surfaces and intensive human activities, resulting in a high DOC content in urban rivers (Wen et al. 2021b). Although point source pollution in urban construction areas has been well intercepted and treated by municipal sewage treatment systems in recent years, the NPS pollution contributing considerably to the DOM in the rivers requires more attention. The lowest average DOC concentration and transport mass were observed in the agricultural rivers. Agricultural drainage is usually characterized by a low content of organic carbon and a high concentration of nitrogen mainly in the form of NO_3^--N (Liu et al. 2022), indicating that it is necessary to mitigate the NO_3^--N pollution in these rivers. In addition, the DOC concentration in the sole outflowing river R36 (Haikou River) on the west side of the lake was relatively high (9.12 mg/L), and the DOC export mass (83.37 g/s) was notably higher than the average import mass of the inflowing rivers because of its high flow rate.

Spectral characteristics of DOM in the rivers

Identification and compositions of fluorescence components

Based on the PARAFAC modeling of the EEM data, four DOM components were identified in the rivers, and their corresponding Ex/Em loadings are displayed in Fig. 3. Samples from R19 and R35 were excluded as outliers during the modeling. The four fluorescent components were roughly divided into two categories: humic-like and protein-like substances. Among them, component 1 (C1) displayed a single Ex/Em peak of 300/370 nm and resembled marine-like humic matter, which is commonly present in marine environments and exhibits a low molecular weight and can also be found in wastewaters, wetlands, and agricultural environments (Stedmon et al. 2003; Fellman et al. 2010). Component 2 (C2) was congruent with terrestrial humic acids with a maximum Ex/Em of 280/476 nm and a fulvic acid fluorophore group with strong aromaticity from terrestrial plants or soil and was generally produced by anthropogenic or microbial activities (Hiriart-Baer et al. 2013). This component was found to be widely present in freshwater ecosystems (Bai et al. 2017; Singh et al. 2017). Component 3 (C3) exhibited an excitation maximum at 340 nm and an intense emission peak at 409 nm and could also be categorized as terrestrial humus, which is associated with microbial degradation of fulvic acids (Stedmon and Markager 2005; Dong et al. 2020). Furthermore, component 4 (C4) was congruent with autochthonous protein-like substances with a tryptophan-like fluorophore with a maximum Ex/Em of 280/330 nm, which generally has poor resistance to biodegradation compared to humic-like substances (tedmon and Markager 2005; Bai et al. 2017). Tryptophan can be generated from the biodegradation of polycyclic aromatic hydrocarbons (PAHs) mainly present in wastewater, urban runoff, and industrial and livestock wastes in addition to autochthonous microbial activities; thus, high contents of tryptophan-like and protein-like substances are usually associated with heavy pollution in water (Singh et al. 2017; Dong et al. 2020).

Comparative analysis of the fluorescence intensity of each component in the samples was conducted to determine the relative concentration of DOM components (Zeng et al. 2017). The fluorescence intensities of the four identified DOM components in the inflowing rivers are shown in Fig. 4. R17 (Xiaoqing River) in the urbanized area presented the highest DOM fluorescence intensity signals (1.77 R.U.), with C1 dominating, indicating that humic-like substances substantially contributed to the DOM. R32 (Chai River), located in an agricultural area, exhibited the lowest fluorescence intensity (0.33 R.U.), in which the four components occupied similar proportions, implying a uniform DOM composition distribution in this river. Overall, the total fluorescence intensity of CDOM in most of the urban rivers was higher than that in the rivers in agricultural and mixed land use areas (Fig. 4a), confirming heavier organic pollution in the urban rivers, which could result from more DOM inputs from urbanized areas with intensive human activities and higher microbial activity (Liu et al. 2020). Although it has been reported that intensive anthropogenic land use (both urban construction area and farmland) could

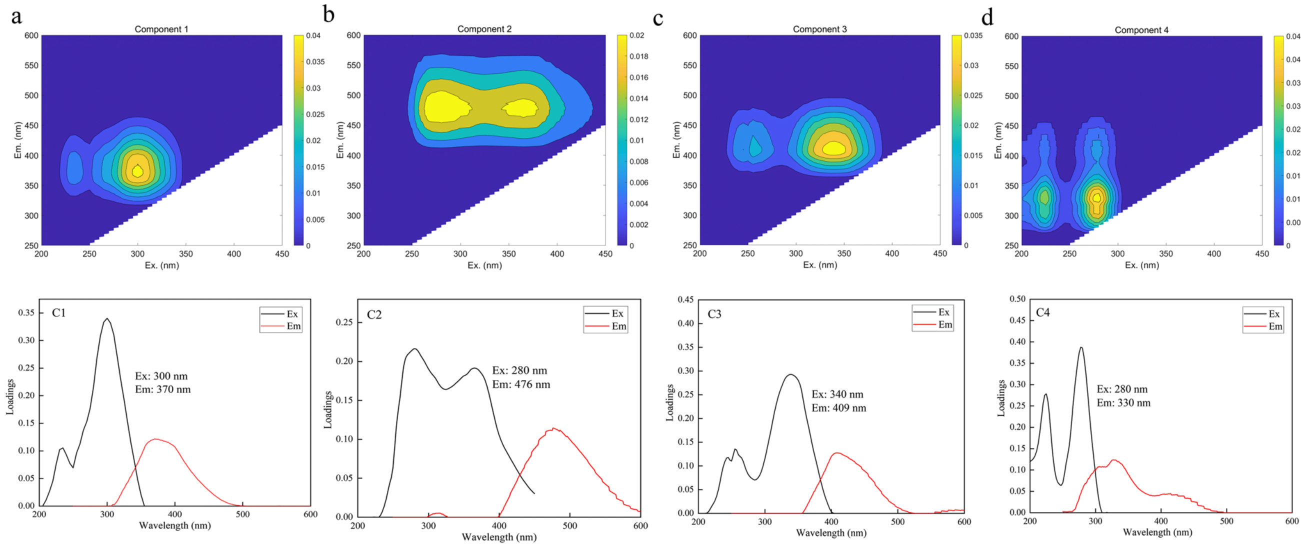

Fig. 3 Fluorescence spectra and excitation/emission loads of fluorescence components of DOM in the inflowing rivers of Dianchi Lake

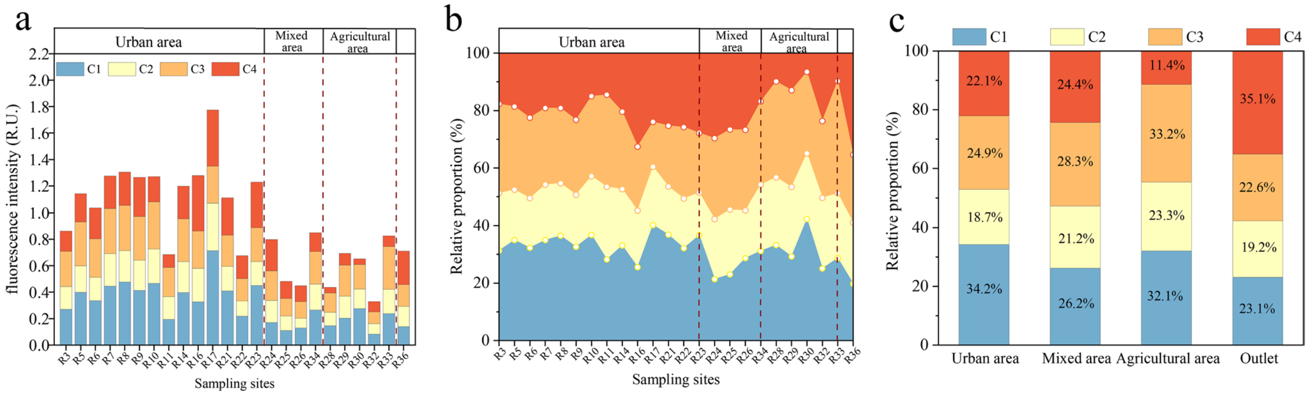

Fig. 4 Fluorescence intensities **(a)** and composition distributions **(b** and **c)** of DOM components in the inflowing and outflowing rivers of Dianchi Lake identified by the EEM-PARAFAC model

result in the strong fluorescence intensity of DOM in recipient aquatic systems (Shi et al. 2020), the results indicated that urbanization could lead to more severe DOM pollution than agricultural activities. The reason might be because the runoff retention and infiltration capabilities of artificial surfaces constructed with cement and asphalt for organic compounds were low.

The DOM composition results showed that the proportion of C1 was the highest in the urban rivers (34.2%). This component can be found in wastewater, agricultural environments, and wetlands (Fellman et al. 2010). C2 and C3, which were both terrestrial humus, accounted for the highest proportion in the agricultural rivers (23.3% and 33.2%), followed by those in the mixed area (21.25 and 28.3%). It was reported that C3 was associated with agricultural DOM sources, such as field fertilization, and leaching of substantial humus from farmlands could be the primary reason for the high contributions of C2 and C3 to DOM in agricultural rivers (Hiriart-Baer et al. 2013). C4 occupied notably higher percentages in the rivers located in mixed areas (24.4%) and urbanized areas (22.1%) than in the agricultural rivers (11.4%), indicating a higher relative abundance of protein-like DOM in the urban rivers. Sewage that is not fully collected for treatment or dispersedly produced in densely populated urbanization areas could lead to an increase in organic pollutants, including PAHs, leading to a relatively high C4 (tryptophan-like) abundance in water bodies (Dong et al. 2020). In addition, the proportion of C4 in the DOM pools of the outlet river (R36) was markedly higher, while the proportions of C1 and C3 were lower than those in the inflowing rivers, suggesting a dominance of protein-like DOM in the outlet river, which might be primarily derived from extracellular substance excretion and detritus decomposition of algal biomass in the lake (Chen et al. 2018).

Spatial variations in DOM compositions along four main rivers

The variations in DOM chemical compositions along rivers are jointly modified by the integrated effects of external import and internal conversion or metabolization. The spatial variations in DOM compositions along the four main inflowing rivers are presented in Fig. 5. Along R11 (Panlong River), which flows through the urbanized area, the relative abundance of C1, C2, and C3 in the DOM pools increased, while the proportion of C4 largely decreased. Humic-like substances (C1, C2, and C3) with high aromaticity are usually relatively refractory and are liable to accumulate in water environments (He et al. 2022b). In contrast, C4 is a tryptophan-like substance that has high bioavailability (DeVilbiss et al. 2016); thus, its proportion decreased along the river. The result also suggested that sewage containing protein substances might be imported into the upstream of the river, which was degraded with the flow of the river. Similarly, an increase in the C1 proportion and a decrease in the C4 proportion were observed in R20 (Baoxiang River), which flows through urban areas. The results also implied that it is necessary to control sewage discharge in the upstream and midstream sections of urban rivers. A relatively high proportion of C1 and a low proportion of C4 were present in R1 (Muyang River), which flows through a mixed land use of forest, grassland, and farmland, with C1 and C4 accounting for the highest proportion at the midstream, probably because of less anthropogenic influence and/or less import of sewage into the river (Lyu et al. 2021). The C2 proportion decreased slightly along R26 (Laoyu River), while the C4 proportion increased slightly. Only slight variations in DOM compositions occurred in this river because of its short length. In addition to exogenous OM import, DOM in rivers could be

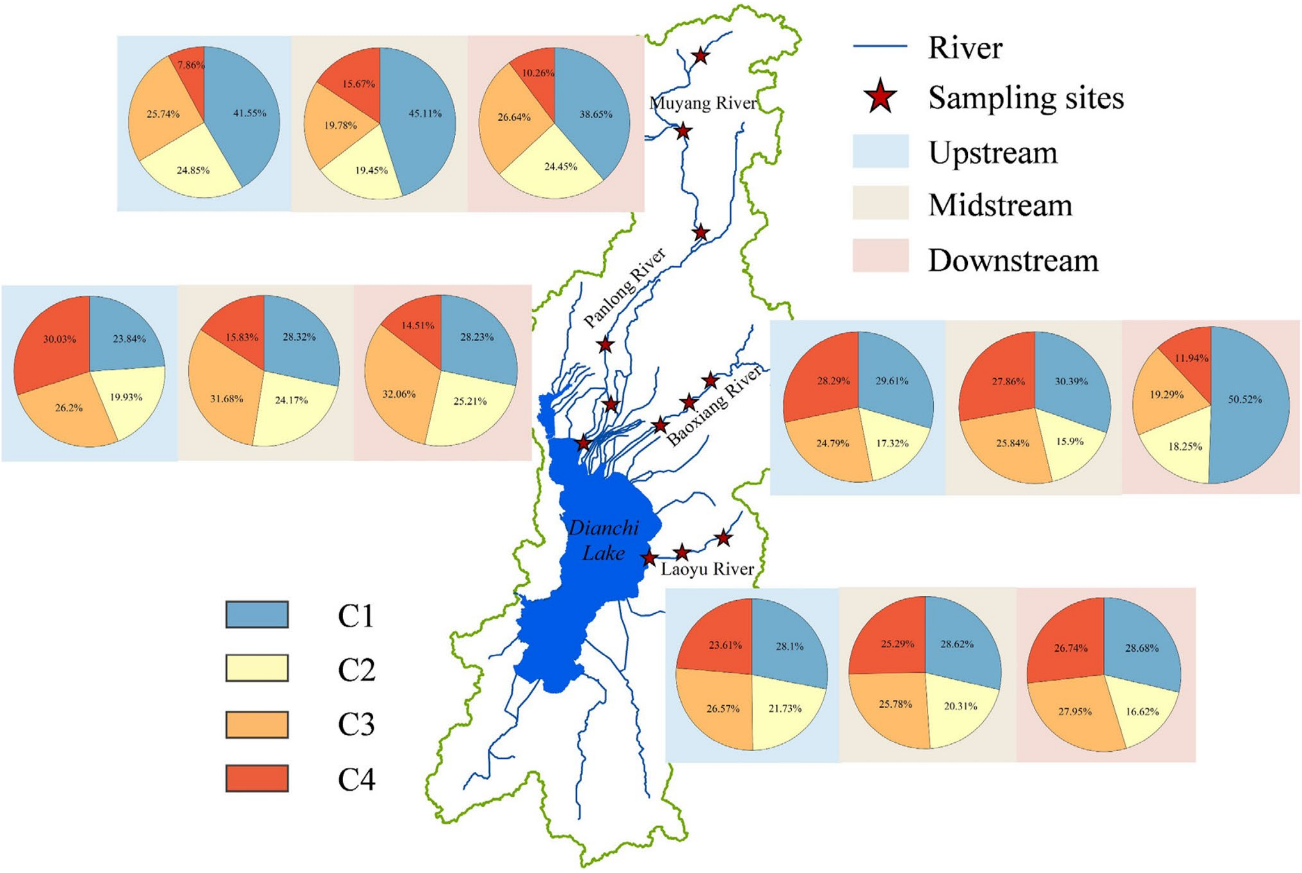

Fig. 5 Spatial variations in DOM compositions along four main inflowing rivers of Dianchi Lake: R1 (Muyang river), R11 (Panlong River), R20 (Baoxiang River), and R26 (Laoyu River)

converted from high molecular substances to low molecular substances by photooxidative degradation and biological processes (Zhang et al. 2021).

Optical indexes

The spectral indexes of DOM in the rivers are presented in Fig. 6. With regard to the absorption characteristics of DOM, the $SUVA_{254}$ values ranged from 0.56 to 14.9 L m^{-1} mg^{-1}, with the highest value observed in R34 (Zhong River), followed by R30 (Baiyu River), and the lowest value was observed in R6 (Daguan River). A high $SUVA_{254}$ value means a large quantity of aromatic substances that are dominantly from allochthonous origination, and the results indicated that aromatic DOM was dominant in R34 and R30, while the DOM in R6 was characterized by low aromaticity. Overall, the $SUVA_{254}$ values in the rivers flowing through the mixed and agricultural land use areas were higher than those in the urban rivers, demonstrating a stronger aromatic structure of DOM dominantly originating from allochthonous inputs in the rivers flowing through agricultural areas. Similar results were recorded by Zhou et al. (2016) and Lyu et al. (2021), and the reasons might be ascribed to more leaching of humic substances from agricultural soil and decomposed vascular plant biomass in the surrounding farmlands and lower microbial decomposition and/or algal growth rates in agricultural rivers than those in urban areas (Artifon et al. 2019). In contrast, the DOM derived from urbanized areas has a relatively low degree of humification but high bioavailability for the autochthonous production of organic compounds (Wen et al. 2021b).

The values of a254 ranged from 6.11 to 69.34 m^{-1}, with the highest value determined in R34 (Zhong River) which corresponded to the highest CDOM abundance. A similar effect of land use types on the a254 value to that on $SUVA_{254}$ in the rivers was observed, implying that the CDOM abundance in the mixed and agricultural land use areas was higher than that in the urban rivers.

$S_{275-295}$ is one of the markers of DOM molecular weight, and a low $S_{275-295}$ value can characterize a strongly allochthonous source of DOM with a high molecular weight (DeVilbiss et al. 2016; Feng et al. 2022). The largest and smallest molecular weights of DOM were found in R30 (Baiyu River) and R32 (Chai River), respectively. The

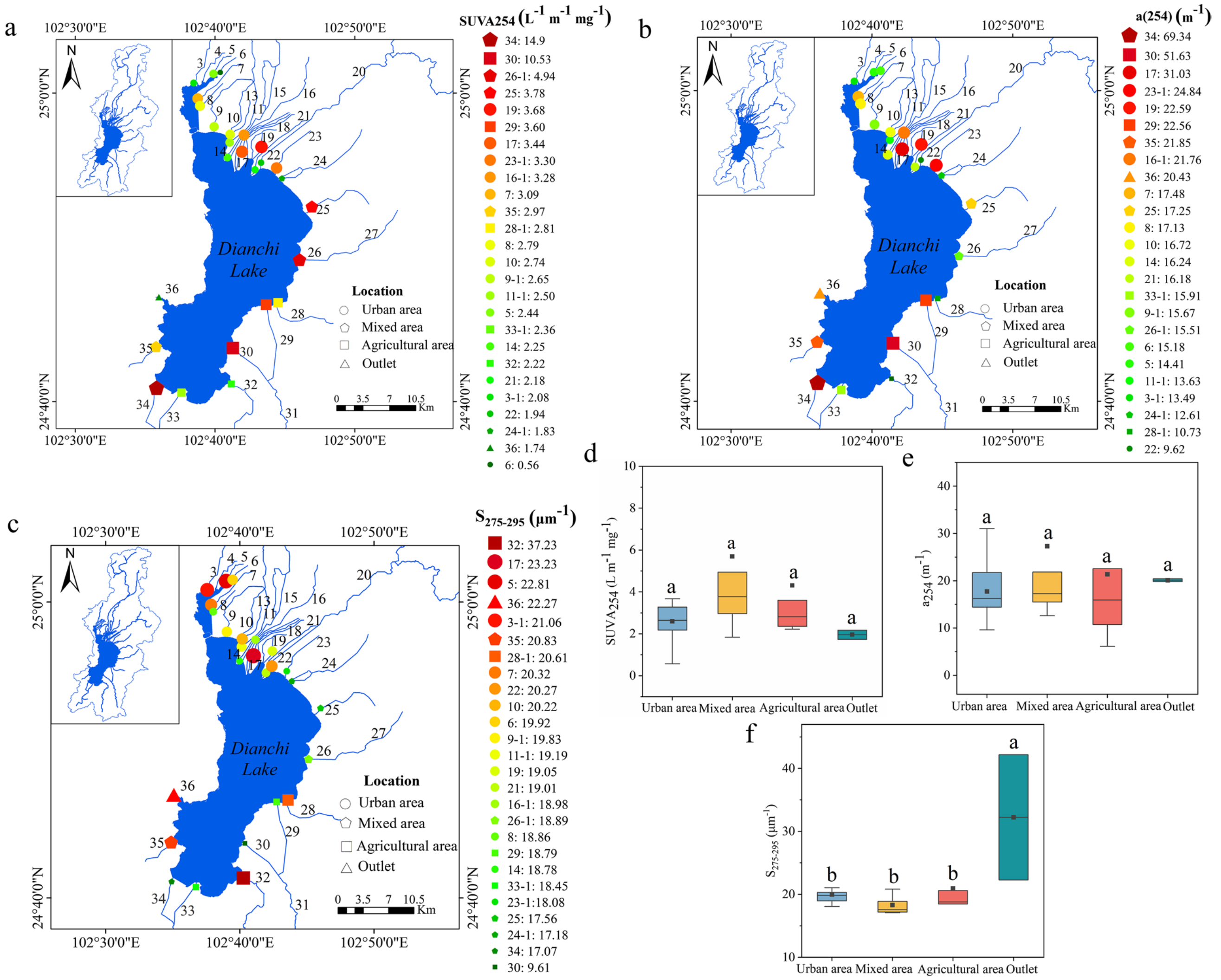

Fig. 6 Spatial variations in DOM optical parameters in the inflowing and outflowing rivers of Dianchi Lake: SUVA254 (**a** and **d**), a254 (**b** and **e**), and $S_{275–295}$ (**c** and **f**)

lower $S_{275–295}$ value in rivers in the mixed areas implied the dominance of high-molecular-weight DOM. In addition, the $S_{275–295}$ value in the outflowing river was significantly lower than that in the inflowing rivers, which signified that low-molecular-weight DOM derived from biological metabolism within the lake was dominant in the outflowing river.

The HIX is an indicator reflecting the humification degree of DOM (DeVilbiss et al. 2016). Its values in the rivers ranged from 0.625 to 0.804, with the highest mean value in the agricultural rivers, followed by those in the mixed land use area (Fig. 6a and e). The results indicated the presence of more humified organic components in the rivers flowing through agricultural areas and were consistent with the $SUVA_{254}$ value, which is indicative of DOM aromaticity. Organic substances with a high humification degree can usually stay for a longer time in natural environments than can those with a low humification degree and dominant autochthonous origins (Zhang et al. 2021; He et al. 2022b). The lowest HIX value in R36 suggested the presence of abundant autochthonous DOM in the outlet river, which mainly originated from algal excretion and microbial decomposition products. The FI values in the rivers were higher than 1.7 except for R17 (Fig. 7b), suggesting a dominance of endogenous sources of DOM in the rivers due to intensive anthropogenic activities in the watershed (Fellman et al. 2010; Xie et al. 2018). A lower FI value in the agricultural rivers than in the other rivers indicated fewer endogenous sources of DOM.

The BIX and β/α can reflect the biological activity and freshness of DOM in water environments (Chen et al. 2018; Sepp et al. 2019). The BIX values of most urban rivers were higher than 1.0 (Fig. 7c), which signified high autotrophic productivity and biological activities producing abundant fresh DOM in these rivers (Chen et al. 2018). The result was consistent with that of the HIX value, probably because the

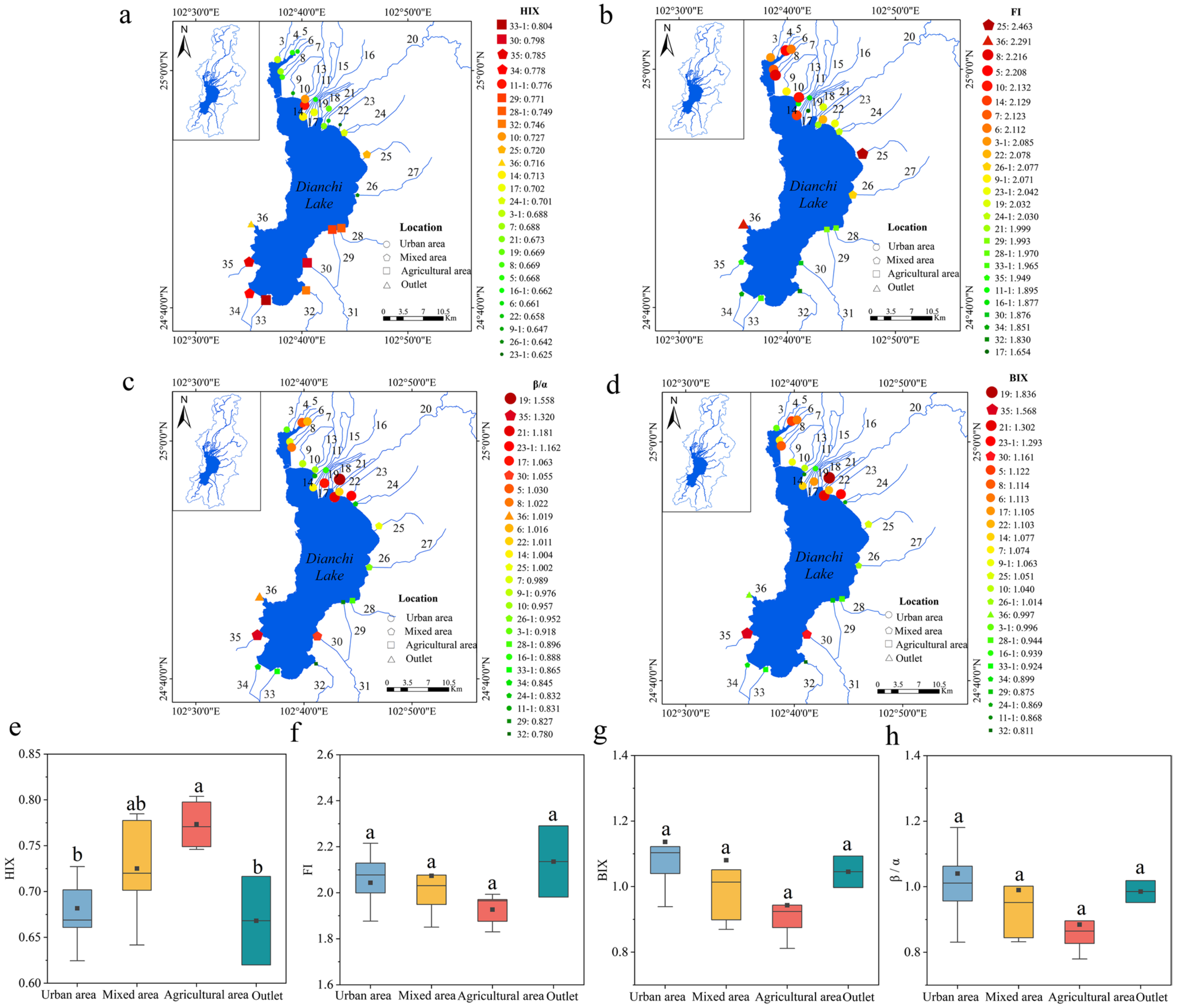

Fig. 7 Spatial variations in the HIX (**a** and **e**), FI (**b** and **f**), BIX (**c** and **g**), and β/α (**d** and **h**) of DOM in inflowing and outflowing rivers of Dianchi Lake

urban rivers received surface runoff rich in multiple nutrients, thus leading to high primary productivity and microbial activities (Derrien et al. 2019). In contrast, the average BIX value in the agricultural rivers was the lowest, indicating relatively low primary productivity and microbial metabolism, which was consistent with the HIX value. The β/α value in the rivers showed a similar spatial variation pattern to that of BIX (Fig. 7d and h), indicating that more recently produced DOM was present in the urban rivers than in the agricultural rivers. Land use plays a significant role in regulating DOM properties, with urbanization increasing DOM content and driving DOM structure to be more protein-like (Cao et al. 2018). In turn, DOM properties can reflect the anthropogenic impacts of land use changes on water ecosystems and provide useful insights into their biogeochemical processes and ecological health (Liu et al. 2020). However, almost no statistically significant difference in DOM parameters was detected in rivers flowing through different land use areas because of the considerable individual variability of the rivers in the same land use area.

Correlations between DOM properties and general hydrochemical parameters

The Pearson correlations between DOM characteristics and general hydrochemical parameters in the rivers were analyzed, and the results are shown in Fig. 8. The HIX and $SUVA_{254}$ values presented a highly significant positive correlation ($P < 0.01$) because both are indicators of DOM aromaticity (Weishaar et al. 2003). A significant negative correlation was observed between $S_{275-295}$ and a254 and between $S_{275-295}$ and humic-like substances (C1 and C2), implying that the humic-like CDOM in the rivers had higher molecular weights (Zhou et al. 2016). Moreover, the significantly positive correlation between the TP and DOC concentrations suggested that a high P concentration could lead to a high content of DOM because P is a basic element for biological productivity. The COD concentration was positively correlated with the terrigenous fluorescence intensities of the C1, C2, and C4 components (C1 and C4 may mainly originate from sewage discharge), suggesting that attention should be given to controlling the exogenous inputs of organic pollutants in the watershed into the rivers and then Dianchi Lake, particularly those from NPS, to prevent the elevation of the COD concentration level in the water bodies. Furthermore, the fluorescence intensities of all DOM components were positively correlated with the NH_4^+-N concentration, and those of C2 and C4 were positively related to TP, implying that higher NH_4^+-N and TP concentrations might result in more fluorescent organic substances in the rivers. He et al. (2022b) found that there was a close relationship between the DOM content and water eutrophication. DOM can be decomposed into simple organic substances and nutrients, and high nutrient contents can also lead to the in situ production of substantial autochthonous DOM. Therefore, DOM characteristics monitored by rapid, accurate, and low-cost spectral methods could reflect nutrient pollution and the trophic status of aquatic ecosystems to some degree (Zhang et al. 2021) and could thus provide important implications for water quality properties and pollution control in the inflowing rivers of Dianchi Lake. In addition, there were highly significant positive correlations among the fluorescence intensities of the four DOM components, especially among C1, C2, and C3, implying partially homologous inputs or production of CDOM components in the rivers and the feasibility of synchronously controlling CDOM fluorescence components.

Figure 9 shows the PCA plot of water samples in the inflowing rivers based on general hydrochemical parameters and DOM properties. The first two PCA axes cumulatively explained 66.62% of the total variance in the dataset, with factor 1 and factor 2 accounting for 40.22% and 26.40%, respectively. The samples from rivers in the urban and agricultural (except for R32 and R33, probably because of relatively strong human activities in the surrounding catchment) areas were separately clustered, demonstrating their distinct water quality characteristics (including DOM) in response to land use type. The samples in the mixed land use area exhibited more similar

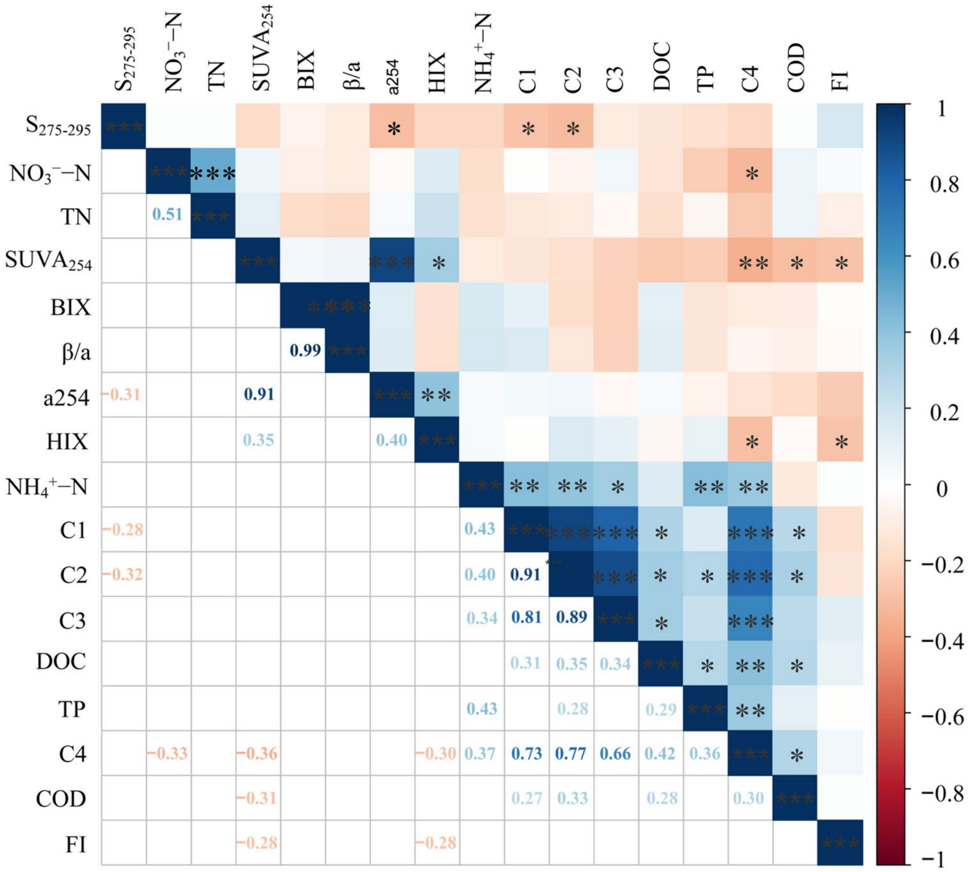

Fig. 8 Correlation matrix heatmap of optical indexes of DOM and general hydrochemical parameters of water samples from the inflowing rivers of Dianchi Lake (blue block indicates a positive correlation, and red block indicates a negative correlation; $^{*}P<0.05$, significant; $^{**}P<0.01$, highly significant; $^{***}P<0.001$, extremely significant)

properties to those in the urban area than to those in the agricultural area. Previous studies have also found a similar regulatory role of land use on DOM properties (Lyu et al. 2021). Most urban rivers were characterized by higher DOC and COD concentrations, BIX, β/α, FI values, and intensities of four fluorescence components of CDOM, implying higher DOM contents with the dominance of freshly generated substances from microbial activities in these rivers. In contrast, the samples in R28, R29, and R30 in the agricultural area showed higher NO_3^--N and TN concentrations and higher values of HIX, $SUVA_{254}$, and a254, which were indicative of strong DOM aromaticity.

Stable carbon isotopes in river sediments and OM source tracing

The combination of $\delta^{13}C$ isotopes and the C/N ratio can be used to differentiate the contribution of OM sources based on different OM endmembers (Lu et al. 2013; Wen et al. 2020, 2021a; Zhang et al. 2021). Information on $\delta^{13}C$ isotopes in the inflowing rivers of Dianchi Lake has been rarely reported. The $\delta^{13}C$ and C/N values of six possible OM end-members, including terrestrial C3 and C4 plants, C3/C4 plant-dominated soil, macrophytes, and plankton based on the results of previous studies, are listed in Table S4 (Kendall et al. 2001; Lu et al. 2013, 2016; Luo et al. 2016).

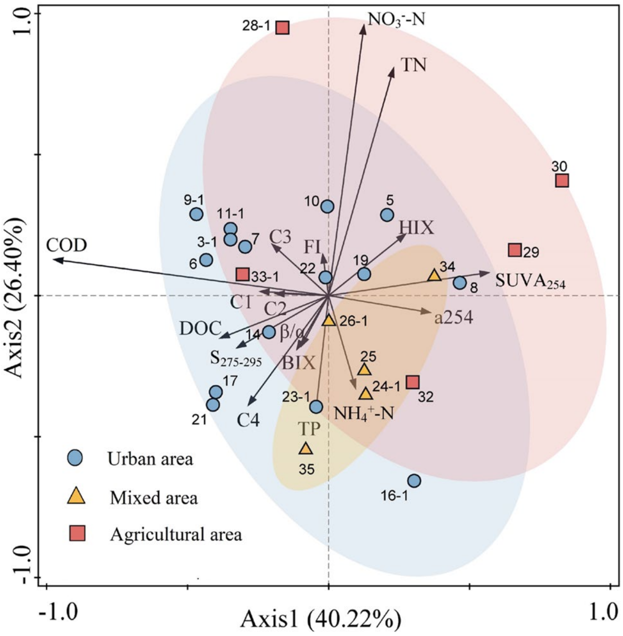

Fig. 9 Principal component analysis (PCA) plot showing dissimilarity of samples in the inflowing rivers of Dianchi Lake flowing through different land use areas based on DOM properties and general hydrochemical parameters

The $\delta^{13}C$ values and C/N ratios of sedimentary OM in the river sediment ranged from −27.86‰ to −3.71‰, with an average of −22.57‰ ± 5.37‰, and from 7.10 to 61.56, with an average of 14.42 ± 11.30, respectively (Fig. 10a; Table S5). The relatively broad $\delta^{13}C$ and C/N ranges reflected complex and varied origins of OM in the river sediments (Lu et al. 2013). Samples with higher C/N ratios usually suffer from stronger anthropogenic disturbances (Lu et al. 2013). The highest C/N ratio was determined in R32 (Chai River) in the agricultural area, probably because of the long-term rural sewage discharge in the river catchment and input of cellulose-rich terrestrial vascular plants. However, the C/N ratios of urban river sediments were almost higher than those in agricultural rivers (except for R32), implying stronger anthropogenic disturbances and more exogenous sewage discharge to urban rivers (Dong et al. 2020). The application of abundant nitrogen fertilizers could lead to a low C/N ratio in agricultural rivers (Kendall et al. 2001).

An isotopic multivariate mixture model was established to quantitatively determine the potential sources of OM in the river sediments. Four major potential sources of OM were present in the river sediments, including plankton, macrophytes, C3 plant-dominated soil and sewage, and their proportional contributions to the sedimentary OM are shown in Fig. 10b and c. The results showed that the main source of organic matter in the river sediments was endogenous aquatic plants (26.5–62.2%), followed by sewage (19.1–33.9%). The contribution of C3 plant-dominated soil was 12.9–28.4%, and plankton contributed the least (6.6–11.3%). Aquatic macrophyte litter can accumulate in river sediment, becoming a primary source of OM. The high contribution of macrophyte sources (62.2%) to OM in the rivers in the mixed land use area might be explained by substantial macrophytes being restored in most of the rivers to improve the river ecosystems. Although macrophyte presence is favorable for water quality amelioration and the normal functioning of river ecosystems (Chang et al. 2021), its biomass is susceptible to decomposition to release OM after withering and decay. Therefore, the proper harvest of aboveground plant biomass in the winter season is needed to prevent organic pollution. In addition, allochthonous inputs had an important contribution to the DOC sink. A large amount of leachable substances could be exported from the lands to rivers by the erosion effect of heavy rainfall in the wet season, resulting in a high contribution of exogenous OM (Galy et al. 2015). C3 plant-dominated soil had a higher contribution (28.4%) to the OM in the river sediments in agricultural areas, probably due to the substantial inputs of soil organic matter carried by surface runoff as a result of erosion from farmlands (Wen et al. 2021a). Sewage also contributed largely (33.9%) to the agricultural rivers, probably because of the discharge of

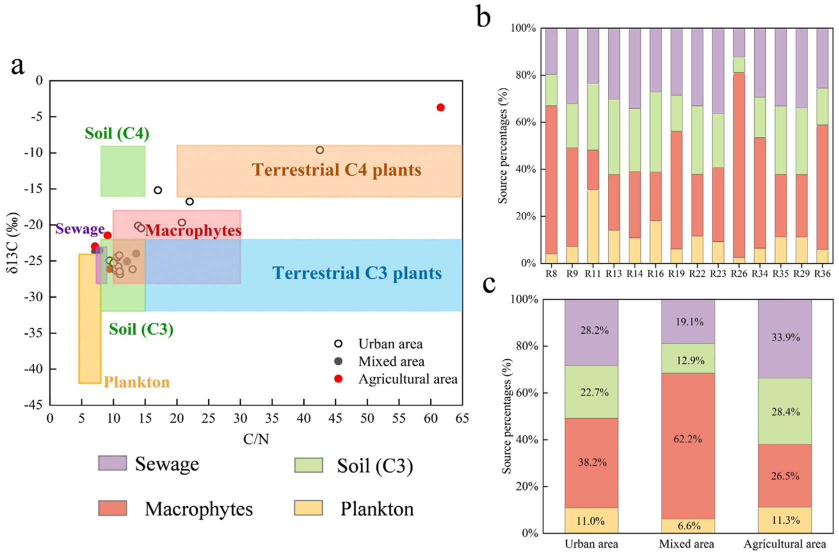

Fig. 10 C/N ratios and δ13C values of sedimentary OM in the river sediment **(a)** and the contribution of different endmember substances to the organic matter based on isotopic multivariate mixture models (**b** and **c**)

dispersed rural wastewater, which is difficult to control. Sewage input presented a moderate contribution of 28.2% to the OM in urban rivers, although centralized municipal wastewater had mostly been treated by WWTPs. Nutrients in sewage can boost phytoplankton reproduction, which displayed contributions of 11.3% and 11.0% to sediment OM in agricultural and urban rivers, respectively. The plankton contribution in the rivers in mixed land use areas (6.6%) was lower than that in other areas, probably because the abundant presence of macrophytes for water ecosystem restoration inhibited algal growth (Chang et al. 2021). Among the rivers, the plankton source was dominant (31.4%) in the DOC pools in R11 (Panlong River), while it accounted for only 2.5% of the DOC in R26 (Laoyu River), in which aquatic vegetation was restored and the macrophyte source contributed greatly (78.6%) to the OM (Fig. 10b). In addition, the flow rate and water quality could influence the contribution of plankton source to the total OM in rivers (Walks 2007).

Conclusion

In the present study, we found that the surrounding land use type largely regulated the properties and source apportionment of DOM in the inflowing rivers of Dianchi Lake in southwestern China. Four fluorescent substances, including marine-like humic matter, terrestrial fulvic acid, terrestrial humus, and tryptophan proteins, were identified by EEM-PARAFAC, and the content and fluorescence intensities of DOM in urban rivers were higher than those in agricultural rivers. The DOM optical properties showed that in the urban rivers, freshly produced DOM with low aromaticity and molecular weight from autochthonous production was dominant. In contrast, the DOM in agricultural rivers was primarily composed of aromatic humus-like substances with high aromaticity from allochthonous inputs, which mainly originated from the leaching of soil humus and decomposition of aquatic plant residues, as revealed by C/N stable isotope analysis. Significant positive correlations between the terrigenous fluorescence intensities of DOM components and COD, NH_4^+-N, and TP concentrations suggested that the DOM optical indexes could well represent the river water quality, and it is necessary to control both the organic and nutrient pollution in the inflowing rivers of Dianchi Lake. More investigation on the temporal variations of the DOM properties in the rivers and the influencing factors is still required in future studies.

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Author contribution Investigation, visualization, validation, and original draft writing were conducted by Rong Wu. Investigation, methodology, and validation were completed by Weijie Guo, Yutong Li, and Shengjiong Deng. Conceptualization, methodology, supervision, original draft writing, review, and editing were conducted by Junjun Chang. All authors read and approved the final manuscript.

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Data availability All data generated or analyzed during this study are included in this article.

Declarations

Ethical approval The manuscript has not been submitted to more than one journal for simultaneous consideration. The submitted work was original and has not been published elsewhere in any form or language. Authors adhered to discipline-specific rules for acquiring, selecting and processing data..

Consent to participate All authors actively participated in this work.

Consent for publication The authors consent to publish this research.

Competing interests The authors declare no competing interests.

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