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Spatial variation of dissolved organic matter in the Tuojiang River Basin in Chengdu, China: insights based on EEMs–PARAFAC analysis

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ABSTRACT

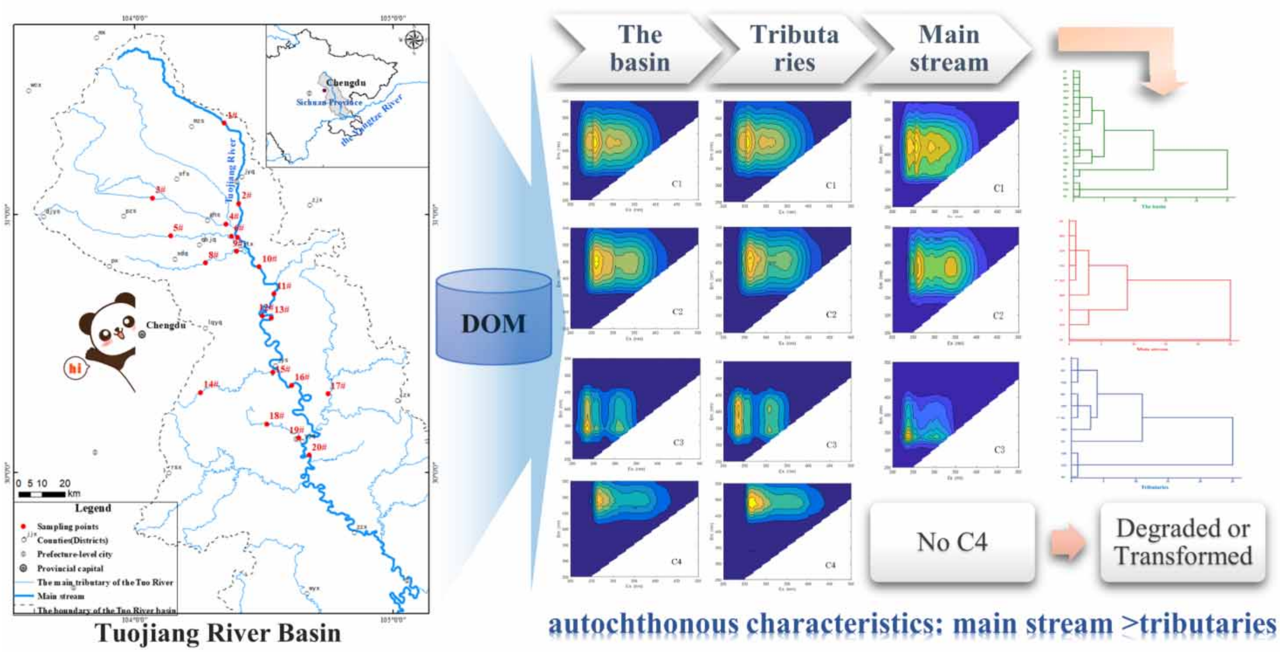
Three-dimensional excitation–emission matrix fluorescence spectroscopy coupled with parallel factor analysis was adopted to investigate the characteristics of dissolved organic matter (DOM) components in water samples collected from the Tuojiang River Basin in Chengdu, including its main stream and tributaries. Four DOM components that matched with three fluorescence peaks were identified in the whole river basin and tributaries; while three components corresponding to four fluorescence peaks were identified in the main stream. In all cases, humic-like components accounted for high proportions of the DOM. Correlation analysis revealed the same sources for four components in the whole river basin and its tributaries, whereas two components had different sources in the main stream. Ultraviolet absorbance parameters (SUVA254, S_R) and fluorescence parameters (BIX, HIX, FI, β:α) indicated the dominant autochthonous sources of DOM in the whole river basin. Higher terrestrial inputs of DOM were observed in the tributaries than in the main stream. In the areas influenced by human activities (6#, 17#, 18#), the sources of DOM showed strong terrestrial characteristics and high degrees of humification and aromatization, as well as serious pollution. The results of this study have potentially far-reaching implications for environmental water management in the area.

Key words: Chengdu section, fluorescent components, fluorescence parameter, main stream–tributary difference, parallel factor analysis, Tuojiang River Basin

HIGHLIGHTS

- DOM of water body can be analyzed by EEMs–PARAFAC analysis.
- The areas with industrial and agricultural emission sources and those affected by human activities have higher degrees of humification and aromatization and are seriously polluted.
- DOM sources in the Tuojiang River Basin in Chengdu were mainly internal, and the terrigenous input of DOM in tributaries was significantly higher than that in the main stream.

GRAPHICAL ABSTRACT



1. INTRODUCTION

A sound ecological environment is one of the most important contributors to a population's wellbeing. The Tuojiang River Basin – a major first-level tributary that flows into the upper reaches of the Yangtze River – is an essential support point for the Yangtze River protection strategy in China. Its water environmental conditions have a direct influence on the incoming water quality in the middle and lower reaches of the Yangtze River. In recent decades, the dual impacts of sociodemographic and economic development have exerted tremendous pressure on the water ecological environment of the Tuojiang River, making it the most seriously polluted river basin in Sichuan Province.

Despite extensive efforts toward water quality improvement in recent years, parts of the Tuojiang River are still heavily polluted (Fan *et al.* 2022). Persistent problems in this area are exemplified by seasonal overloading of water environmental capacity and failure of water ecological function, which raise serious environmental issues. In particular, discharge of urban domestic sewage as well as industrial and agricultural wastewater has a strong impact on water quality in the Tuojiang River Basin in Chengdu, leading to considerable water quality differences between the main stream and tributaries (Qin *et al.* 2020). The overall water quality of the main stream is satisfactory, in contrast to the much poorer water quality of its tributaries. Black and odorous water was generally eliminated, despite potential risk of recurrence in some small tributaries. Given the direct influence of water quality on the life and productivity of urban residents, addressing the problems of water environmental pollution in the Tuojiang River Basin in Chengdu is imperative.

Dissolved organic matter (DOM) is the largest organic carbon pool in aquatic ecological environments. It plays a significant role in maintaining the carbon cycle in aquatic ecosystems, as well as the degradation and adsorption of heavy metals and organic pollutants (Wu *et al.* 2010; Liu *et al.* 2019). DOM is widely present in surface water and groundwater as a consequence of different hydrological, biological, and geological interactions (Leenheer & Croue 2003; Mayayorga *et al.* 2005). It supports heterotrophic microbial metabolism (Stedmon *et al.* 2011) and, to some extent, affects the integrity and function of riverine ecosystems. The composition of DOM is determined by its source. Riverine DOM can be derived from autochthonous inputs (e.g., phytoplankton, aquatic plants, and heterotrophs), allochthonous inputs (e.g., soil leaching, rock weathering, and atmospheric deposition), and anthropogenic inputs (e.g., industry, wastewater, intensive agriculture, and farms) (Fisher *et al.* 2004; Elliott *et al.* 2006; Giorgio & Pace 2008; Griffith & Raymond 2010; Fashing *et al.* 2014).

Three-dimensional excitation–emission matrix fluorescence spectroscopy coupled with parallel factor analysis (EEMs–PARAFAC) is widely used for DOM source analysis. In this approach, the specific identification of organic compounds is performed using excitation and emission matrices (Zheng *et al.* 2016). It has advantages in terms of high sensitivity and fast detection time in tracking the quality and quantity of natural DOM (Stedmon *et al.* 2006; Mohammad *et al.* 2009). PARAFAC resolves the fluorescence spectra of DOM to obtain the source and characteristics of different components (Zhang 2019). Using EEMs–PARAFAC, Zhang *et al.* (2022) traced the source and composition of DOM in black and odorous rural river water, where the DOM contained six fluorescent components under the influence of both autochthonous and allochthonous sources. Additionally, Chen *et al.* (2022) explored the influence of different flooding conditions on the source of DOM in lake sediments based on EEMs–PARAFAC. They reported that protein-like component accounted for a higher proportion of DOM under seasonal flooding conditions, whereas humus-like components exhibited the opposite pattern. Furthermore, Xu *et al.* (2022) analyzed the spatial variation of DOM in river water by EEMs–PARAFAC and observed remarkable spatial variation of water quality parameters. As an important carrier of pollutants, DOM is closely related to the migration and transformation of pollutants, bioavailability, and nutrient retention and release (Smith *et al.* 2021; Wen *et al.* 2021; Li *et al.* 2022). Therefore, the source, characteristics and other information of DOM can be judged according to different components identified by the water body DOM. Accordingly, these previous studies have demonstrated the effectiveness of EEMs–PARAFAC in the analysis and identification of DOM sources and its compositional characteristics.

Herein, EEMs–PARAFAC was adopted to characterize the variation of DOM characteristics along the Tuojiang River Basin in Chengdu as well as its main stream and tributaries. The aim of this study was to determine the sources of DOM and its material composition in the river water. The results provide useful information to guide source control, sewage interception, and water environmental quality improvement in the Tuojiang River Basin.

2. MATERIALS AND METHODS

2.1. Study area

The Tuojiang River is the most important river in Sichuan Province and a major tributary on the left bank of the Yangtze River. The Tuojiang River Basin (103°41′–105°55′ E, 28°50′–31°41′ N) is adjacent to Jiufeng Mountain in the Longmen Mountain range in the northwest and neighbors the Minjiang River in the west. It connects to the Fujinag River in the east and flows into the Yangtze River in the south. The Tuojiang River originates in Dahei Bay under fault rockhead in Mianzhu City, at the southern foot of Jiuding Mountain. It flows through the cities of Deyang, Chengdu, Ziyang, and Neijiang, and then discharges into the Yangtze River through Guanyizui in the urban area of Luzhou City. The basin area (in Sichuan) is 2.56×10^4 km^2, accounting for 5.3% of the total provincial area. The study area is part of the Tuojiang River Basin, comprising the administrative areas of Chengdu, Deyang, Ziyang, Mianzhu, Guanghan, and Jinyang (hereinafter referred to as the Tuojiang River Basin in Chengdu). The Tuojiang River Basin is the area with the most concentrated towns, the most densely populated and the strongest economic strength in Sichuan Province. The economic development in the basin is rapid, with densely covered towns and complete industries of agriculture, forestry, animal husbandry, and fishery. The red layer hilly areas in the middle and lower reaches of the basin (such as Jianyang, Zizhong, Anyue, and other rural areas) are dominated by traditional fragmented farming methods, with high use of pesticides and fertilizers and low utilization rate and serious pollution from agricultural non-point sources. Industry in the basin is developed, such as Deyang heavy machinery manufacturing.

2.2. Sampling point layout

A total of 20 sampling points were selected across the study area, with nine located in the main stream and 11 in the tributaries of Tuojiang River Basin. The number of samples was 20, the sampling volume was 250 mL, and the sampling water depth was less than 0.5 m below the water surface. The basic information of the sampling points is listed in Table 1, and their geographical distribution is shown in Figure 1.

2.3. Sample analysis

2.3.1. Conventional parameter measurements

The main experimental instruments were a HACH portable multi-parameter water quality analyzer (HQ30d, Shenzhen, China), which was used for *in situ* measurements of water temperature, dissolved oxygen (DO), pH, and oxidation–reduction potential [ORP]; a nitrogen gas cylinder; a biochemical incubator; a DR6000 UV–Vis spectrophotometer (Shenzhen, China); a DRB200 digestion system (Shenzhen, China); a total organic carbon analyzer; an ultrapure water system; and pipettes.

Table 1 | Sampling point information

River level	Sampling point	Transect	Water body	Location	Control level
Main stream	1#	Hongyansi	Mianyuan River	Eastern Shidi Town, Mianzhu City	National
	2#	Bajiao	Mianyuan River	Northern Lianshan Town, Guanghan City	National
	7#	201 Hospital	Tuojiang River (north river)	Qingjiang Town, Jintang County, Chengdu City	National
	10#	Sanhuangmiao	Tuojiang River	Sanhuangmiao, Jintang County, Chengdu City	Provincial
	11#	Colmar Town	Tuojiang River	Huaikou Town, Jintang County, Chengdu City	
	12#	Wufengxi Ancient Town	A stream in Wufengxi Ancient Town	Wufeng Town, Jintang County, Chengdu City	
	13#	Hongyuan	Tuojiang River	Hongyuan Town, Jiangyang City, Chengdu City	National
	16#	Xinshi Town	Tuojiang River	Xinshi Town, Jiangyang City	
	20#	Gongchengpu Ferry	Tuojiang River	Gongchengpu, Yanjiang District, Ziyang City	National
Tributary	3#	Lower Luowan Square	Xiaoshi River	Western Majing Town, Shifang City	National
	4#	Sanchuan	Yazi River	Hexing Town, Guanghan City, Deyang City	National
	5#	Sanyi Bridge	Qingbaijiang River	Qingbaijiang District, Chengdu City/Pengzhou City	National
	6#	Qingjiang Bridge	Zhonghe River	Qingjiang Town, Jintang County	National
	8#	Lanheyan	Pihe River	Southerstern Xiangfu Town, Qingbaijiang District	Provincial
	9#	Pihe Bridge 2	Pihe River	Jintang County, Chengdu City	National
	14#	Sancha Reservoir	Sancha Lake	Sancha Town, Jianyang City	Provincial
	15#	Aimin Bridge	Jiangxi River	Jiancheng Street, Jianyang City	Provincial
	17#	Hongruhe Bridge	Yanghua River	Shijia Town, Jianyang City	National
	18#	Jile Village	Jiuqu River/Laoying Reservoir	Jile Village, Linjiang Town, Yandiang District, Ziyang City	Provincial
	19#	Jiuquhe Bridge	Jiuqu River	Yanjiang District, Ziyang City	National

National control level was detected and supervised at the national level; and province control level was detected and supervised at the province level. It is mainly aimed at some construction projects and operating projects with large investment, large emissions of pollutants, and serious harm of pollutants, and construction projects and operating projects that are prone to major environmental hazards.

After sampling, permanganate index (COD_{Mn}), ammonia-nitrogen (NH_3–N), total nitrogen (TN), and total phosphorus (TP) were determined according to Analytical Methods for Water and Wastewater Monitoring (State Environmental Protection Administration 2002a). Briefly, COD_{Mn} was analyzed using the potassium dichromate method; NH_3–N was quantified using Nessler's reagent spectrophotometry; and TN and TP were determined by alkaline potassium persulfate digestion–UV spectrophotometry and ammonium molybdate spectrophotometry, respectively. Dissolved organic carbon (DOC) was determined using a total organic carbon analyzer (TOC-L CPN, Shimadzu, Japan). Three parallel samples were analyzed for each measurement.

2.3.2. Three-dimensional fluorescence spectral measurement and model analysis

A fluorescence spectrophotometer (RF-6000, Shimadzu, Japan) was used to analyze three-dimensional fluorescence spectral characteristics of the DOM. To eliminate the internal filtering effect, samples containing >8 mg DOC/L were diluted with Milli-Q water to a concentration of 8 mg DOC/L before analysis. Mill-Q ultrapure water was used as a blank. The excitation wavelength (E_x) ranged from 200 to 400 nm in 5-nm intervals and the emission wavelength (E_m) ranged from 250 to 450 nm in 2-nm intervals. The scanning speed was 2,000 nm/min.

2.4. Data analysis

The sampling point distribution map was drawn using ArcGIS 10.2 (Environment System Research Institute Inc., Redlands, CA, USA). Data processing and correlation analysis were performed using Origin 2022 (study version; OriginLab Corp.,

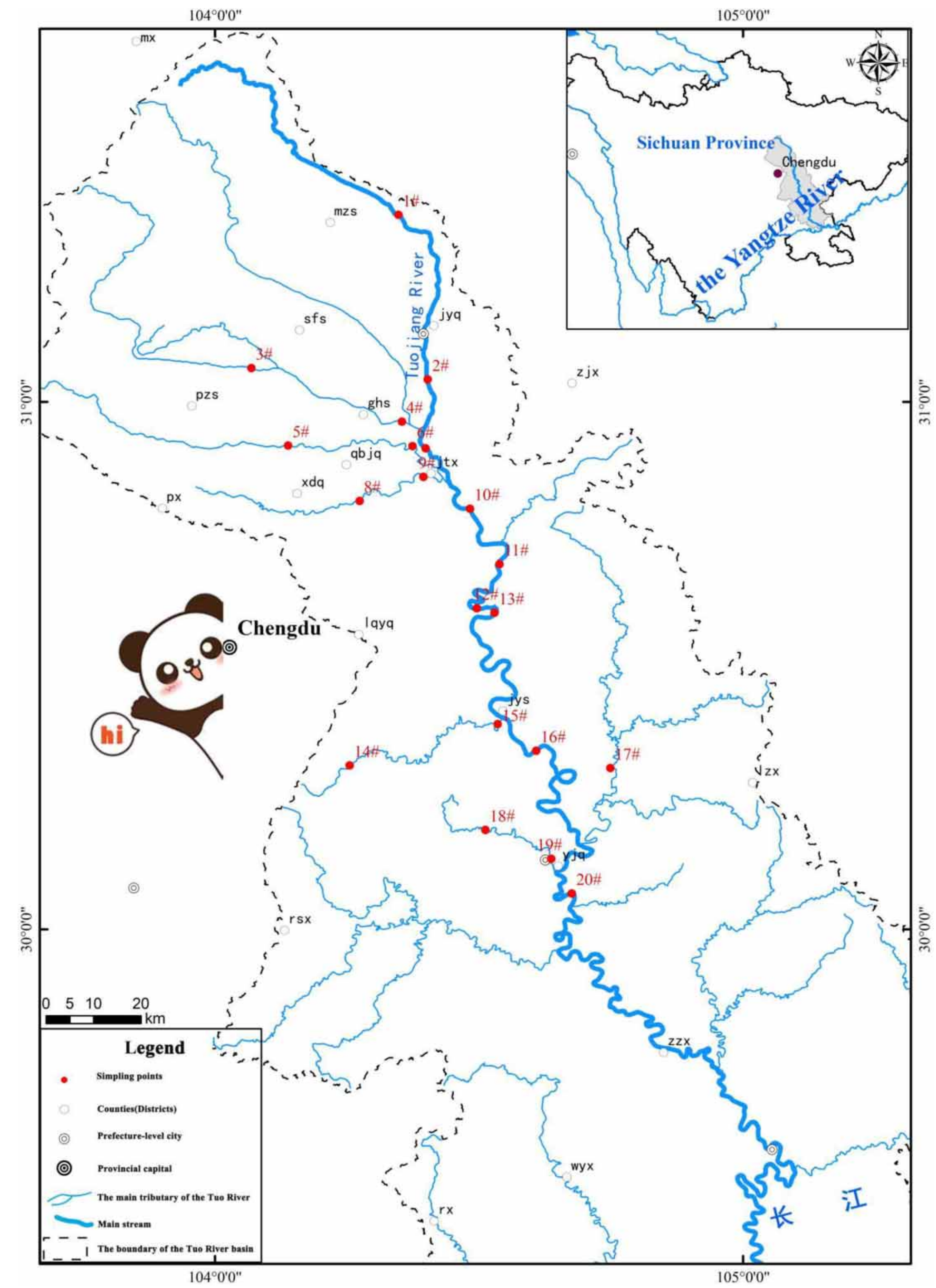


Figure 1 | Sampling points in the Chengdu section of the Tuojiang River Basin are laid out.

Northampton, MA, USA) and Excel 2016 (Microsoft Corp., Redmond, WA, USA). PARAFAC analysis of EEMs was carried out using the DOMFluor toolbox in MATLAB 2018b (MATLAB, MathWorks Inc., Natick, MA, USA), and the results were subjected to cluster analysis using SPSS 26.0 (IBM Corp., Armonk, NY, USA).

3. RESULTS AND DISCUSSION

3.1. Comparison of water environmental factors

The values of major water environmental parameters in the Tuojiang River Basin in Chengdu are provided in Table 2. The mean parameter values of DO (8.23 mg/L) and pH (7.61) across the sampling points met the Class I water quality standard

Table 2 | Statistics of environmental parameters in the Chengdu area of the Tuojiang River Basin

Sampling points	DO (mg/L)	pH	Temperature (°C)	ORP (μS/cm)
1#	10.32	7.66	12.60	196.75
2#	11.85	7.68	13.02	181.05
3#	11.79	7.55	13.30	227.00
4#	2.20	7.81	13.30	444.90
5#	2.95	7.53	11.35	214.00
6#	9.74	7.65	10.80	279.00
7#	2.51	7.52	14.05	209.55
8#	10.63	7.73	13.45	310.50
9#	11.41	8.00	12.95	283.00
10#	4.40	7.48	12.45	352.00
11#	0.56	7.78	13.69	212.00
12#	0.64	7.62	12.55	197.60
13#		7.49	13.50	193.35
14#	3.88	7.49	13.40	228.50
15#	0.00	7.75	13.70	202.70
16#	10.87	7.46	12.95	208.55
17#	2.19	7.60	12.97	219.35
18#	4.81	7.46	13.80	230.80
19#	12.16	7.56	13.85	225.30
20#	1.55	7.57	13.10	189.40
Minimum	0.56	7.46	10.80	181.05
Maximum	12.12	8.00	14.05	444.90
Average value ± SD	6.02 ± 4.50	7.61 ± 0.14	13.04 ± 0.79	240.27 ± 63.46
C_v	0.75	0.02	0.06	0.26

according to the Environmental Quality Standard for Surface Water (State Environmental Protection Administration 2002b). The range of water quality category in GB 3838-2002 is: Class I: DO $\geq$ 7.5 mg/L; Class II: DO $\geq$ 6 mg/L; Class III: DO $\geq$ 5 mg/L; Class VI: DO $\geq$ 3 mg/L; Class V: DO $\geq$ 2 mg/L. The DO values at some sampling points, such as 10#, 14#, and 18#, reached Class III water quality standards, but the water quality at tributary sampling points 4# and 5# was still poor after converging into 7#. Additionally, poor water quality of Class V and lower was recorded in the main stream sampling points 11#, 12#, and 20# as well as at the tributary sampling point 17#. The DO values at all other sampling points met Class I water quality standards, and the highest value of 12.12 mg/L was recorded at point 13#.

The DO and ORP values show the coefficients of variation between 10 and 90%, indicating its moderate spatial variability. The maximum ORP of 444.9 μS/cm was observed at point 4#. The pH and temperature values show relatively small coefficients of variation, indicating minor spatial variation in these two parameters across the sampling points, with no significant differences between the main stream and tributaries.

The spatial variations of COD_{Mn} and DOC values across the sampling points in the Tuojiang River Basin in Chengdu are shown in Figures 2, 3 and Table 3, respectively. COD_{Mn} is a comprehensive index that measures the degree of organic pollution in surface water bodies, where a higher COD_{Mn} value indicates more serious pollution of the water body by organic pollutants (Wang 2015). The COD_{Mn} values for the water samples vary spatially from 3 to 13 mg/L. The COD_{Mn} values at sampling points 2#, 4#, 5#, 8#, and 16# are all lower than the standard threshold for Class II surface water (4 mg/L). The COD_{Mn} concentrations at points 3#, 9#, 12#, 14#, and 19# were lower than the standard threshold for Class III surface water (6 mg/L). The COD_{Mn} concentrations at points 6#, 7#, 10#, 11#, and 13# were lower than the standard threshold for Class IV surface water (10 mg/L). Refer to Figure 2 and Table 2 for specific values.

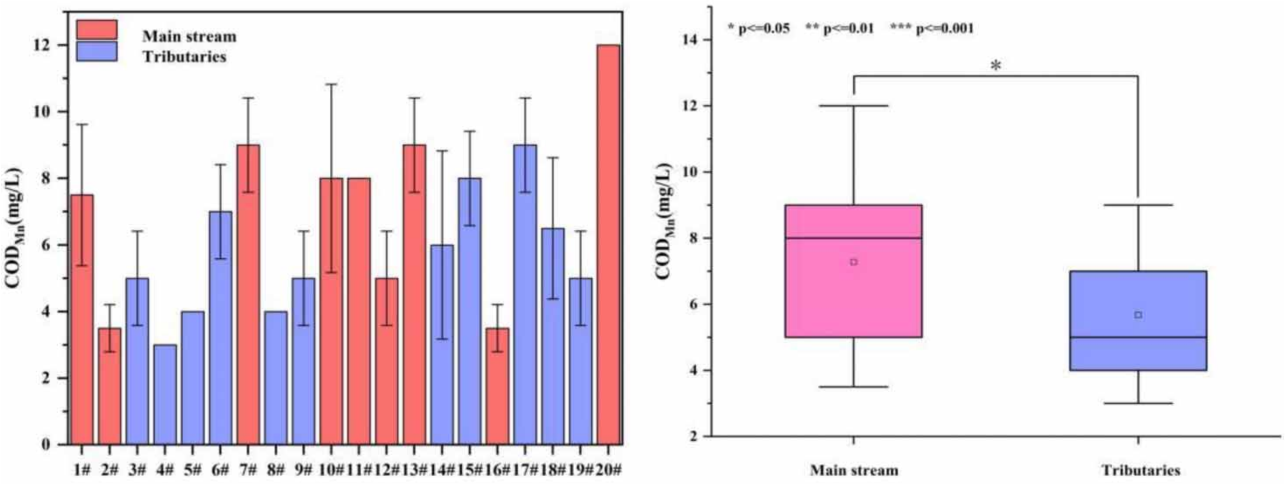


Figure 2 | Spatial variation of COD_{Mn} concentration in Chengdu area of the Tuojiang River Basin.

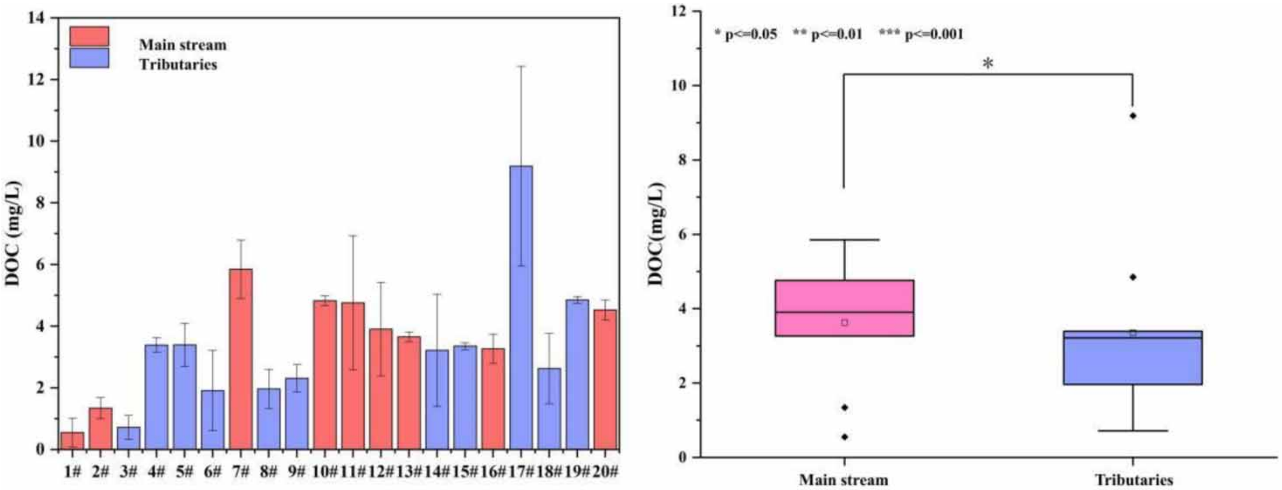


Figure 3 | Spatial variation of DOC concentration in the Chengdu area of the Tuojiang River Basin.

Both COD_{Mn} and DOC values are greater in the main stream than in the tributaries, which mean that the degree of organic pollution is more serious in the main stream than in the tributaries, and there are significant differences between the main stream and tributaries. DOC varies from 0.55 to 9.14 mg/L across the sampling points. It has been reported that higher DOC values occur in water bodies in areas strongly influenced by anthropogenic discharges (Huang *et al.* 2016a, 2016b). In the present study, point 17# is close to an agricultural wastewater discharge channel, leading to it presenting the highest DOC value (9.14 mg/L). In contrast, point 1# is located in a mountainous area with low water flow and sediment deposits, leading to it presenting the lowest DOC value (0.55 mg/L). Owing to the strong influence of human activities at 201 Hospital, point 7# presents a higher DOC value than its adjacent sampling points. After tributary convergence, the DOC values exhibit a decreasing trend along the main stream flow direction. Refer to Figure 3 and Table 2 for specific values.

3.2. Absorption spectral characteristics of the DOM

The ratio of UV absorbance at 254 nm to DOC concentration ($SUVA_{254}$) characterizes the degree of DOM humification and is proportional to the percentage of aromatics (Spencer *et al.* 2012). Additionally, the spectral slope ratio (S_R) can be used to characterize the source of the DOM. $S_R < 1$ indicates dominant input by terrestrial DOM with high molecular weight and aromaticity derived from vascular plants; $S_R > 1$ indicates dominant input by autochthonous DOM with low molecular weight and aromaticity (Weishaar *et al.* 2003). The $SUVA_{254}$ and S_R variation of DOM across the sampling points is depicted

Table 3 | Water quality and absorption parameters

Sampling points	COD (mg/L)	DOC (mg/L)	S_R	$SUVA_{254}$
1#	7.50	0.55	2.85	2.85
2#	3.50	1.34	5.66	1.44
3#	5.00	0.72	4.85	3.22
4#	3.00	3.39	5.49	0.31
5#	4.00	3.39	4.44	0.30
6#	7.00	1.91	4.33	1.11
7#	9.00	5.85	1.25	0.34
8#	4.00	1.96	4.08	1.11
9#	5.00	2.31	3.79	0.44
10#	8.00	4.83	4.67	0.38
11#	8.00	4.76	5.24	0.39
12#	5.00	3.90	4.79	0.56
13#	9.00	3.66	5.12	0.52
14#	6.00	3.21	6.35	0.73
15#	8.00	3.35	5.17	0.54
16#	3.50	3.27	4.40	0.56
17#	9.00	9.19	5.24	0.97
18#	6.50	2.63	5.81	3.51
19#	5.00	4.85	4.82	0.46
20#	12.00	4.53	5.96	0.50

in Table 2, Figures 4 and 5, respectively. The value range and corresponding meaning of absorption spectral characteristics are shown in Table 4.

The $SUVA_{254}$ and S_R values at various sampling points show significant differences between the main stream and tributaries. The parameter values are higher for the tributaries than for the main stream, indicating a higher degree of DOM humification and aromatization as well as greater autochthonous contribution in the tributaries than that in the main stream. The $SUVA_{254}$ values vary from 0.30 to 3.51. Compared with other sampling points, the $SUVA_{254}$ values at points

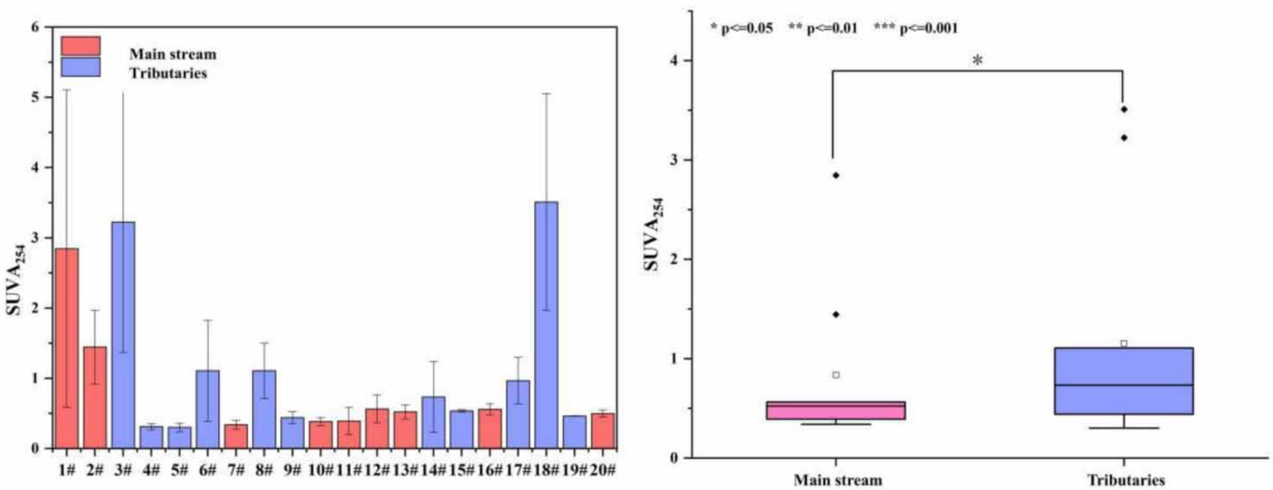


Figure 4 | Spatial variation of the $SUVA_{254}$ of the DOM in the Chengdu section of the Tuojiang River Basin.

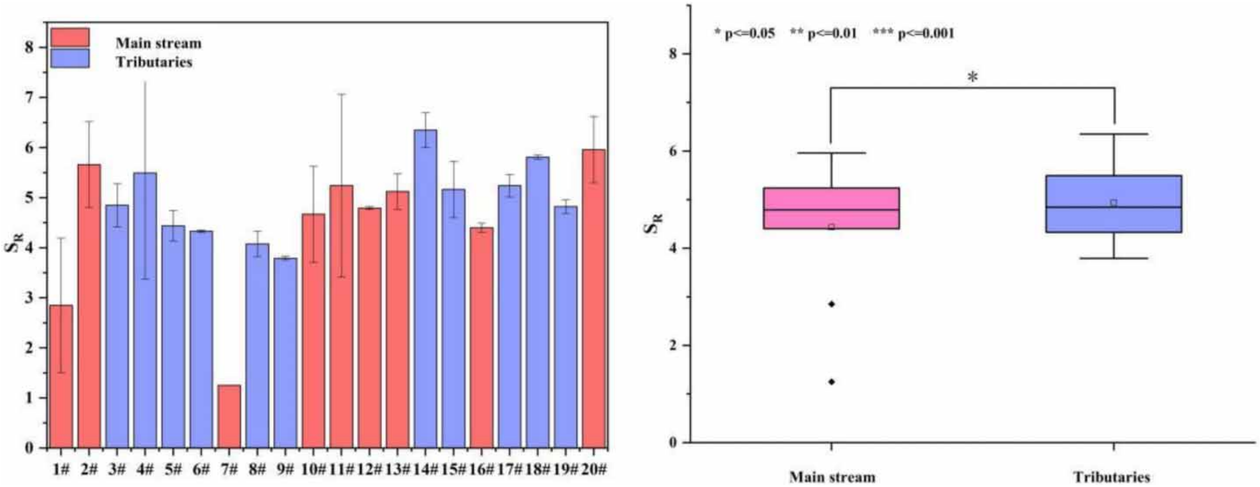


Figure 5 | S_R spatial variation of DOM in the Chengdu section of the Tuojiang River Basin.

Table 4 | The value range and corresponding meaning of absorption spectral characteristics and fluorescence spectral parameters (Li *et al.* 2022)

Parameters	Autochthonous inputs	Terrestrial inputs
FI	FI > 1.9	FI < 1.4
BIX	BIX > 1 indicates large autogenetic contribution, and 0.6 ≤ BIX ≤ 0.7 indicates small autogenetic contribution	
HIX	HIX < 4	HIX > 10
SUVA254	The higher the $SUVA_{254}$ value, the higher the aromatization degree	
S_R	$S_R > 1$	$S_R < 1$
$\beta{:}\alpha$	The larger the $\beta{:}\alpha$ value, the higher the proportion of newly produced DOM	

1#, 2#, 3#, and 18# are remarkably larger, and the higher degrees of aromatization and humification are due to the strong influence of human activities. DOM aromatization in the tributaries decreases along the flow direction. S_R values range from 1.25 to 6.35 (all > 1), indicating that the DOM in this water body is mainly from autochthonous sources. At point 7#, the autochthonous input is relatively low compared with that at other sampling points, which may be due to its proximity to a sewage outlet and low DO concentration.

3.3. Three-dimensional fluorescence characteristics of the DOM

3.3.1. Fluorescence spectral parameters

To identify the source of DOM in the water bodies, the DOM fluorescence parameters of the Tuojiang River Basin in Chengdu, its main stream, and tributary water bodies were plotted (Figures 6, 7 and Table 1 of supplemental materials). Fluorescence index (FI) is used to characterize the source of the DOM (Yang *et al.* 2019). The DOM in the water bodies is mainly derived from terrestrial inputs at FI < 1.2 and autochthonous release at FI > 1.80 (Lavonen *et al.* 2015). In the whole river basin and its main stream, the DOM is basically from autochthonous inputs (FI > 1.8), with a relatively small proportion of autochthonous inputs in the tributaries (Figure 6(a)). The value range and corresponding meaning of fluorescence spectral parameters are shown in Table 4. The FI values show minor variation in the range 1.75–2.04 across the basin, indicating the substantial influence of autochthonous sources at the sampling points. The smallest FI value is presented for point 18# (1.75) and the highest at point (2.04). The FI values increase after the tributaries merge into the main stream, which means a more pronounced autochthonous contribution to DOM in the main stream, consistent with the results shown in Figure 6(a).

The biological index (BIX) measures the autochthonous characteristics of DOM in water bodies, which are proportional to each other (Chen *et al.* 2017). The results indicate both autochthonous and allochthonous sources for DOM in the Tuojiang

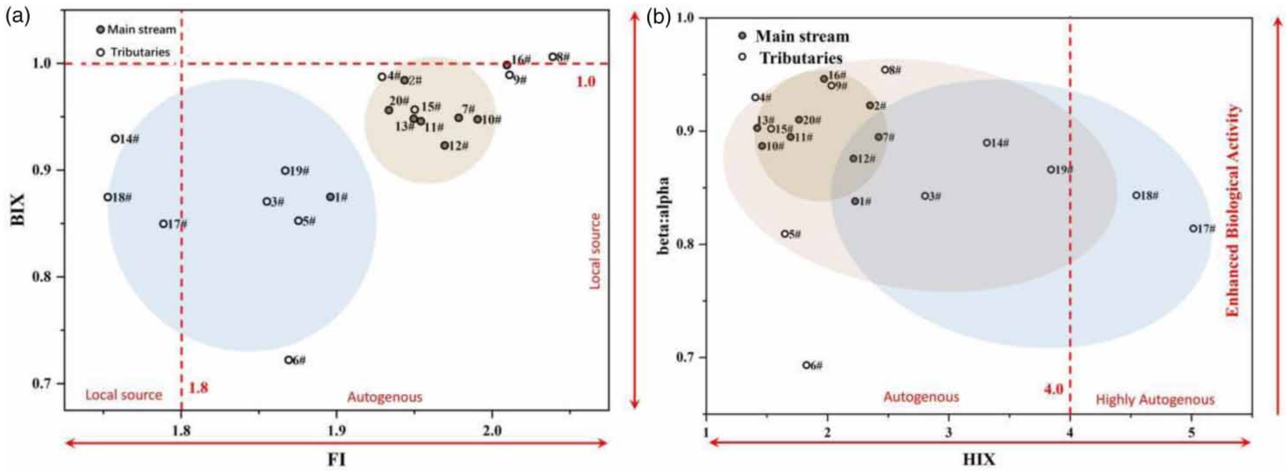


Figure 6 | DOM sources of fluorescence parameters in the Chengdu section of the Tuojiang River Basin and its main tributaries.

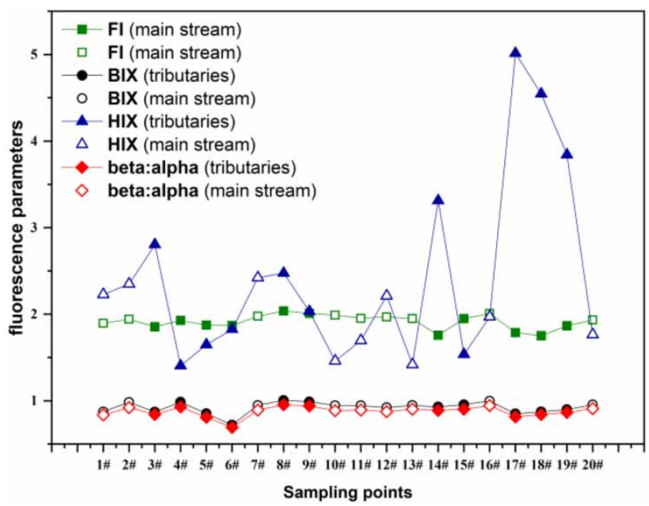


Figure 7 | Spatial variation characteristics of fluorescence parameters in the Chengdu section of the Tuojiang River Basin and its main tributaries.

River Basin in Chengdu, its main stream, and tributaries. The BIX values range from 0.72 to 1.01 (Figure 7), indicating that the DOM sources at different sampling points in the water bodies are influenced by both terrestrial and autochthonous inputs. The autochthonous characteristics of the DOM are stronger in the main stream than in the tributaries. The BIX value at point 6# (0.72) is remarkably lower than those at other sampling points. The DOM at point 6# mainly originates from terrestrial inputs and is strongly influenced by nearby industrial wastewater discharge. The BIX value is highest at point 8# (1.01), where autochthonous input is most prominent, which is consistent with the FI results and those shown in Figure 6(a).

Generally, the humification index (HIX) is used to reflect the degree of DOM humification (Bu *et al.* 2019). A higher HIX value indicates a higher degree of DOM humification and a stronger terrestrial origin; a lower HIX value suggests a stronger autochthonous origin of the DOM. The HIX values are basically < 4.0 in the Tuojiang River Basin in Chengdu, its main stream, and tributaries (Figure 6(b)), which indicates a relatively strong autochthonous origin of the DOM in the water bodies. In contrast with other fluorescence parameters, HIX fluctuates more prominently in the range 1.40–5.02 (Figure 7). The degrees of DOM humification at points 17# and 18# are higher than those at other sampling points owing to the strong influence of industrial wastewater and domestic sewage discharge, respectively. This indicates a strong terrestrial origin of the

DOM at the two sampling points (17# and 18#). A previous study has shown that farmland, woodland, and grassland near sampling points can lead to the enhancement of terrestrial origin, aromaticity, and humification degree of DOM in water bodies (Liu 2018). There is a relatively high degree of DOM humification at point 18#, supporting the $SUVA_{254}$ and FI results. The mean HIX values at the other sampling points are <4, suggesting the dominance of autochthonous DOM. Additionally, the HIX values are higher for the tributaries than the main stream. Accordingly, there is a higher humification degree of DOM in the tributaries than in the main stream, which mirrors the FI and BIX results. The autochthonous origin is weaker in the tributaries than in the main stream, similar to the results shown in Figure 6(b).

The freshness index (β:α) reflects the proportion of newly produced DOM in the overall DOM: the relative contribution of endogenous substances to DOM (Huang *et al.* 2016a, 2016b), with its level and variation basically consistent with those of BIX. In this study, the β:α values range from 0.69 to 0.95 (Figure 7), which indicates high biological activity and the high autochthonous characteristics of the DOM, which is in basic agreement with the results of Zhang *et al.* (2022). The β:α value at point 6# is relatively low, suggesting a small proportion of newly produced DOM with the weakest biological activity and autochthonous characteristics, corroborating the BIX results. This sampling point (6#) is close to a commercial area and therefore strongly influenced by terrestrial sources. The highest β:α value is observed for point 8#, reflecting the strongest biological activity and most prominent autochthonous characteristics of the DOM at that point, which mirrors the FI and BIX results.

3.3.2. Fluorescent component characteristics

The composition of the DOM fluorescence in the Tuojiang River Basin in Chengdu as well as its main stream and tributaries were analyzed by PARAFAC analysis. In total, four components of two types were identified in the basin and its tributaries. Comparison of component type with those reported by Coble *et al.* (1990) revealed that they are humic-like (C1, C2, and C4) and protein-like (C3) components. Additionally, three components of two types were identified in the main stream, which are humic-like (C1 and C2) and protein-like (C3) components (Figure 8 and Tables 2–7 of of supplemental materials).

The fluorescent component C1 (255/426 nm) in the basin and its tributaries corresponds to fluorescence peak A, similar to the pattern of a typical UV fulvic acid-like peak. This component is associated with relatively high aromaticity and high molecular weight DOM groups, which are resistant to biodegradation and indicative of terrestrial input (Yan *et al.* 2021). C2 (260, 345/454 nm) in the basin and C2 (260, 345/462 nm) in the tributaries match fluorescence peak F, which resembles the pattern of a typical humic acid-like peak. Such components originate primarily from domestic sewage and represent a typical terrestrial humic-like substance (Garcia *et al.* 2018). C3 (235, 305/338 nm) in the basin and C3 (235, 305/342 nm) in the tributaries correspond to fluorescence peak T, which mirrors the pattern of a typical tryptophan-like peak. Such components are prone to biodegradation, strongly influenced by human activities (Yi *et al.* 2017), and derived mainly from urban domestic

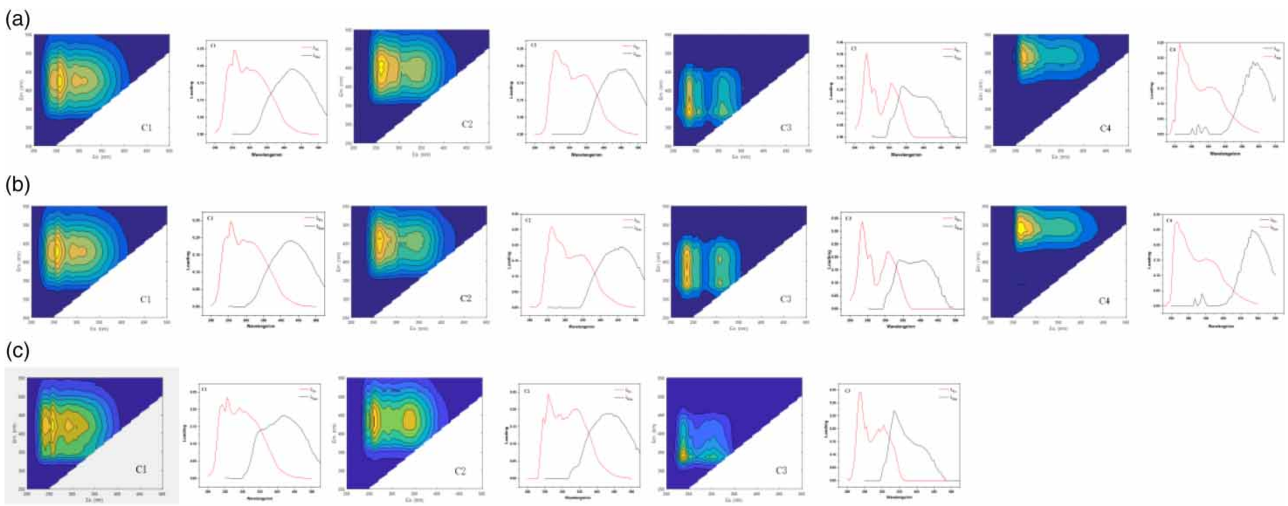


Figure 8 | DOM fluorescence components and their excitation/emission wavelengths ((a) Chengdu area of the Tuojiang River Basin; (b) tributaries; (c) main stream).

sewage and food industry wastewater (Francisco *et al.* 2020). C4 (265/482 nm) in the basin and its tributaries matches fluorescence peak F, similar to the pattern of a typical humic acid-like peak.

In the main stream, the fluorescent component C1 (255/418 nm) corresponds to fluorescence peak A, similar to the pattern of typical UV fulvic acid-like peak. C2 (260, 340/442 nm) corresponds to fluorescence peaks F and C, which mirror the patterns of traditional humic acid-like and visible fulvic acid-like peaks, respectively. The position of the visible fulvic acid-like peak C is unstable, which may be red-shifted or blue-shifted due to differences in sample properties (Zhong *et al.* 2008). The DOM components may be degraded or transformed during water migration in large rivers, with bioavailability playing an important regulating role (Gan 2013). Therefore, there is no component C4 in the main stream. C3 (235, 305/334 nm) matches the fluorescence peak T, which resembles the pattern of a typical tryptophan-like peak.

The maximum fluorescence intensity (FMAX) of the DOM components characterizes the DOM concentration of water bodies. In the Tuojiang River Basin in Chengdu, the fluorescence intensity of the DOM components varies from 3,238.1 to 17,543.1 a.u. (Figure 9(a)). In the main stream and tributaries, the ranges of fluorescence intensity are 2,670.5–10,809.0 a.u. (Figure 9(c)) and 3,088.6–17,162.1 a.u. (Figure 9(b)), respectively. This indicates that the spatial variation in the DOM concentration of the water bodies is large, with significantly higher concentrations in the tributaries than in the main stream. The lowest DOM concentration is shown for sampling point 5# (FMAX = 3,088.6 a.u.), whereas a relatively high DOM concentration is observed at point 17# (FMAX = 17,162.1 a.u.), which is basically consistent with the results of DOC analysis (Figure 9(a)).

Kramer & Herndl (2004) have demonstrated that humic-like substances are less susceptible to microbial degradation than protein-like substances in water bodies. As reported by Hudson *et al.* (2007), humic-like substances account for a relatively large proportion of DOM in natural water bodies, whereas protein-like peaks are enhanced by human activities. The large proportions of humic-like substance (C1, C2, C4) for the DOM in the Tuojang River basin in Chengdu as well as its main stream and tributaries are consistent with the results of Gan (2013) (Figure 10(a)–10(c)). In contrast, the low proportion of protein-like substance (C3) is associated with the dominant terrestrial inputs of DOM pollutants in water across the

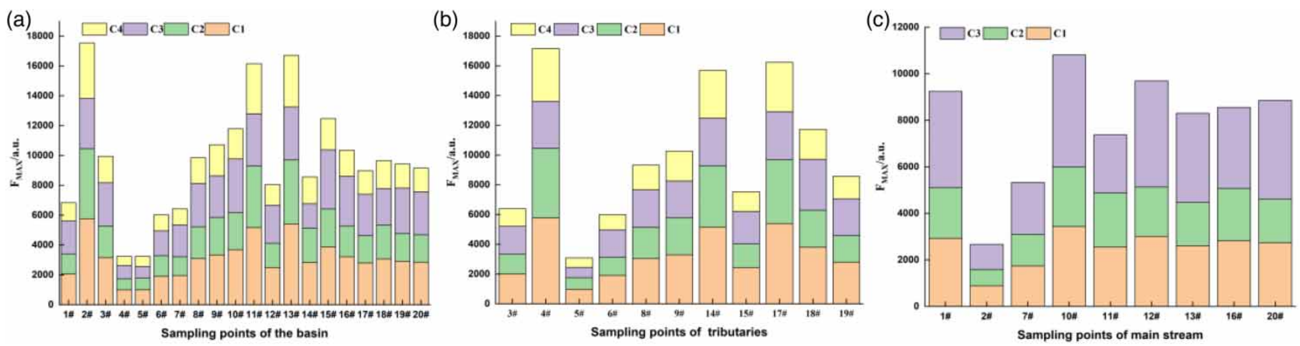


Figure 9 | DOM fluorescence intensity of the Chengdu section and its main tributaries in the Tuojiang River Basin.

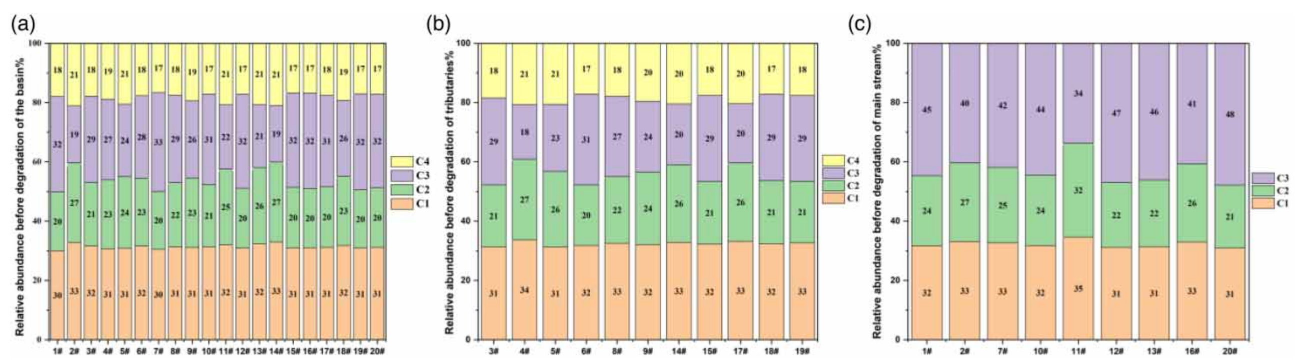


Figure 10 | Proportion of DOM fluorescence components in the Chengdu section of the Tuojiang River Basin and its main tributaries.

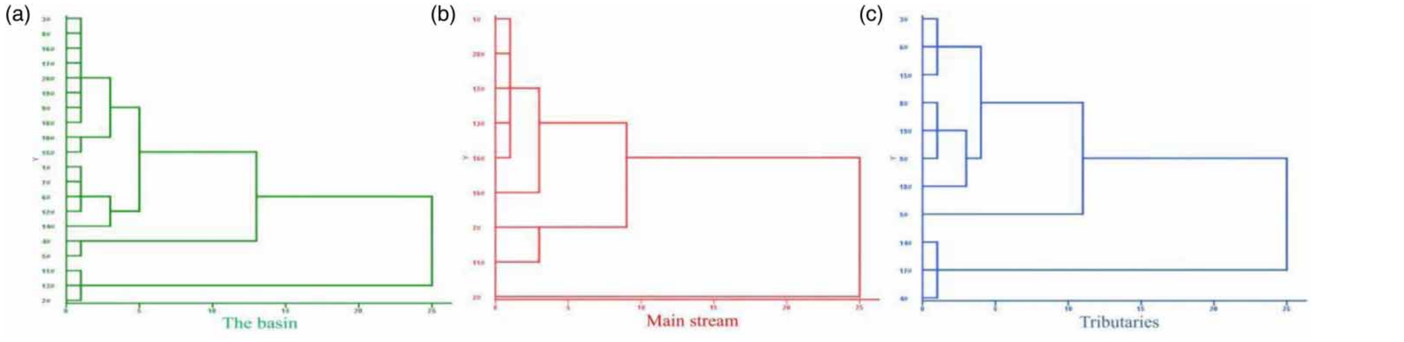


Figure 11 | Cluster analysis of DOM fluorescence components in the Chengdu area of the Tuojiang River Basin and its main tributaries.

sampling points, with a small contribution of wastewater discharge from the food industry and relatively low autochthonous inputs, leading to uniform spatial variation. Additionally, there are large proportions of humic acid-like C2 and C4 in humic-like substances in the Tuojang River Basin in Chengdu and its tributaries, where the terrestrial inputs of pollutants are dominated by domestic sewage (Figure 10(a) and 10(b)). This is in agreement with the results of Huang *et al.* (2010) and are related to the prominent urbanization and domestic sewage discharge in the study area. Furthermore, there is a higher proportion of tryptophan-like substance (C3) in the main stream, with stronger autochthonous DOM characteristics than in the tributaries (Figure 10(c)). Overall, the terrestrial contribution of DOM is higher in the tributaries than in the main stream, mainly because the main stream of Tuojiang River Basin in Chengdu is less urbanized than its tributaries, and the tributary water quality is more influenced by the discharge of domestic sewage and industrial and agricultural wastewater. This situation calls for appropriate control of the amount and standard of wastewater discharge, as well as strict supervision of industrial and agricultural wastewater discharge in the urban areas (Figure 10).

Cluster analysis was conducted on the fluorescence intensity results for the DOM components from PARAFAC analysis. The results show a distance <5 between various sampling points in the Tuojiang River Basin in Chengdu, its main stream, and tributaries (Figure 11(a)–11(c)). This suggests a small variability in the source and type of DOM components across the sampling points. Notably, there are larger differences in these DOM characteristics among the tributaries compared with those in the main stream. The same DOM characteristics are observed at the main stream sampling points 11# and 13#, with high fluorescence intensity and serious pollution, supporting the results of COD_{Mn} analysis (Figure 11(b)). Moderate fluorescence intensity is observed at the tributary sampling points 3#, 8#, 9#, 15#, and 17#–19#. The lowest fluorescence intensity is observed at the tributary sampling points 4# and 5#, consistent with the results of fluorescence intensity analysis (Figure 11(c)).

3.4. Correlation between DOM and water environmental factors

To further investigate the characteristics of the DOM components in the Tuojiang River Basin in Chengdu, its main stream, and tributaries, Pearson correlation analysis was conducted between the optical parameters of the DOM components and the

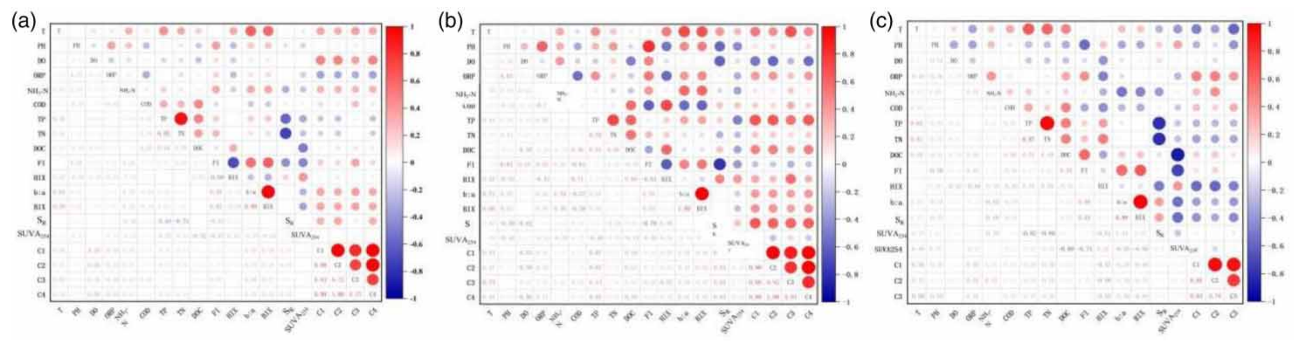


Figure 12 | Correlation coefficient between DOM components and main tributaries in the Chengdu section of the Tuojiang River Basin and its main tributaries.

water environmental parameters (Figure 12(a) and 12(c)). The four components C1, C2, C3, and C4 are significantly positively correlated with each other in the basin and its tributaries ($P < 0.01$), suggesting a high homology for the four components (Figure 12(a) and 12(b)). In the main stream (Figure 12(c)), C1 is significantly positively correlated with C2 and C3, indicating the same pollutant sources for C1 along with C2 and C3. However, there is a low correlation between C2 and C3, indicating a remarkable difference in their pollutant sources. Overall, the correlations between various DOM components and water environmental parameters are relatively low, suggesting minimal environmental influence. BIX and β:α are highly positively correlated with temperature and FI, respectively, in the river basin, its main stream, and tributaries ($P < 0.01$). This is consistent with the results of Zhou *et al.* (2020), indicating that appropriate temperature rise can promote autochthonous inputs for the DOM in water bodies.

4. CONCLUSIONS

- Based on PARAFAC analysis, four fluorescent components of the DOM in the Tuojiang River Basin in Chengdu and its tributaries were identified, i.e., humus-like components C1, C2, and C4 and protein-like component C3. Additionally, three fluorescent components were identified in the main stream, i.e., humus-like components C1 and C2 and protein-like component C3. The spatial variation of DOM concentration in water bodies is large. Correlation analysis revealed a homology for the four DOM fluorescent components in the basin and its tributaries. In the main stream, DOM component C1 is derived from the same sources as C2 and C3, although the latter two components show different sources.
- The UV absorbance parameters ($SUVA_{254}$, S_R) and fluorescence parameters (FI, BIX, HIX, β:α) indicate that both autochthonous (mainly from soil, human activities, surface runoff and decomposition of plant and animal residues) and allochthonous (formed by biological activity in water bodies) inputs contribute to the sources of DOM in the Tuojiang River Basin in Chengdu, with a greater autochthonous contribution. This can be used to judge the source of DOM and provide a basis for the treatment of Tuojiang River Basin. Based on FI, HIX, and component proportions, the DOM shows stronger autochthonous characteristics in the main stream than in the tributaries, which is primarily due to the strong influence of urbanization on tributary waters.
- In the Tuojiang River Basin in Chengdu, the population is dense, the degree of urbanization is large, the industry is developed, and the agricultural non-point source pollution is serious. The analysis results show that the BIX, FI, HIX, $SUVA_{254}$ values of the water DOM in the sampling points (6#, 17#, 18#) affected by the discharge of domestic sewage and industrial and agricultural wastewater are relatively large and have relatively prominent terrestrial origin. Therefore, we recommend that pollution source control standards be quantified by region and type, and that point source control standards for enterprises in key industries, urban non-point source pollution control standards, and agricultural non-point source pollution control standards are developed. Additionally, we suggest that decentralized harmless utilization of rural wastewater and rational desilting of urban rivers be promoted.

AUTHOR CONTRIBUTIONS

H.T.L., B.B.C., and B.C. conceptualized the study, did formal analysis, and carried out investigation; X.X.L. prepared and wrote the original draft; B.B.C., X.H.L., Y.W., L.X.H., and C.W. wrote, reviewed, edited, and supervised the study.

CONSENT TO PUBLISH

All the authors have approved the submission and consented for publication.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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