Effects of land use and nutrients on the characteristics of dissolved organic matter in the Nanchong Section of Jialing River, China in December 2019

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Abstract

Chromophoric dissolved organic matter (CDOM) in aquatic ecosystems can reflect the impacts of human activities on the carbon-cycling process. However, direct evidence of the combined effect of land use and anthropogenic nutrients on CDOM characteristics in river systems is limited. Herein, we collected water samples from 18 sites in the Nanchong section of Jialing River in December 2019 to elucidate how the land use and nutrients affect the source and composition of CDOM through parallel factor (PARAFAC) analysis of excitation–emission matrices (EEMs). First, the absorption coefficient \(a_{254}\) \((r^2 = 0.29, p < 0.01)\) and three fluorescence components (humic-like C1 and C2 and protein-like C3) \((r^2 = 0.31-0.37, p < 0.01)\) significantly increased with increased urban area, and the four parameters were higher in the urban area than in the suburbs \((p < 0.05)\). The correlation between small-size CDOM molecule and cropland was positive \((p < 0.01)\). Second, the increase in nutrient levels increased the \(a_{254}\) \((r^2 = 0.84\) and 0.33, \(p < 0.01)\) and three fluorescence components \((r^2 = 0.30-0.84, p < 0.01\) or \(p < 0.05)\). Third, allochthonous CDOM were prevalent in the Nanchong Section of Jialing River, and the proportions of C1 and C2 were 42 and 41%, respectively. Our findings indicated that the variability of source and composition of CDOM significantly depended on urbanization and increased nutrients in the Nanchong Section of Jialing River.

Key words: chromophoric dissolved organic matter (CDOM), dissolved organic matter (DOM), Jialing River, nutrients, parallel factor (PARAFAC) analysis, urban land

Highlights

- We found that (1) Terrestrial dissolved organic matter (DOM) input was the primary source in the Nanchong Section of Jialing River (2) Nutrients (i.e., TN and TP) and Urban land use enhanced inputs of allochthonous and autochthonous DOM.
- Our study highlights that increased urbanization significantly altered the source and composition of DOM and thus potentially affected the progress of the carbon cycle and aquatic system function of the Jialing River.

1. Introduction

Rivers connect terrestrial and aquatic ecosystems and are thus responsible for the biogeochemical cycles of carbon (Richey 2004). Many rivers are vital sources of drinking water, so they are crucial to human survival and economic development. Dissolved organic matter (DOM), composed of fulvic acid, humus, aliphatic, and aromatic hydrocarbons (Yu et al. 2017; Wu et al. 2019), is a prominent component of biogeochemical cycles (Lambert et al. 2017; Zhuang et al. 2021). In freshwater systems, DOM is derived from terrestrial sources and in-situ primary production (Asmala et al. 2013). Chromophoric DOM (CDOM), the part of DOM that strongly absorbs ultraviolet and visible light, competes with chlorophyll a (Chl a) and thus limits the primary productivity of aquatic ecosystems (Stedmon & Markager 2001; Zhou et al. 2016). The high occurrence of CDOM can produce a sour and pungent odor and carcinogenic by-products, resulting in decreased water quality (Wang & Chen 2018). In recent years, spectrophotometry and three-dimensional excitation–emission matrix spectra (EEMs) have provided useful and timely information about CDOM composition and sources (Song et al. 2019). In particular, the absorption coefficient \(a_{254}\) is a proxy for CDOM abundance (Stedmon & Markager 2001). The spectral slope S and \(a_{250}\), \(a_{365}\) can indicate the molecular weight and relative molecular size of CDOM, respectively (Fichot & Benner 2012; Zhang...
et al. 2018). SUVA_254 can well estimate the aromatic carbon content (Weishaar et al. 2003). Furthermore, fluorescence parameters such as humification index (HIX) and biological index (BIX) are adopted to estimate terrestrial and biological sources, respectively (Stedmon et al. 2011). The fluorescence intensity of components screened by parallel factor (PARAFAC) analysis can effectively reflect the variety of fluorescence DOM (i.e., humic-like and protein-like components) (Stedmon 2003).

Human activities have accelerated the export of anthropogenic contaminants derived from point source or/and non-point source, thereby resulting in strong influence on DOM chemistry and even on water quality (Xenopoulos et al. 2021). Substantial evidence indicates that DOM pools today have been altered by humans. Specifically, land use is a direct indicator of human activities, especially the increases in urbanization and agriculture land (Williams et al. 2016). Prior studies have suggested that increasing agricultural land could increase protein-like component with smaller and simpler molecular structures, whereas forest and wetland land use enhances the export of macromolecule humus (Wilson & Xenopoulos 2009). Furthermore, the autochthonous CDOM increase with increased eutrophication in freshwater systems, which is related to algae production or/and microbial degradation (Du et al. 2020). The high level of nitrogen concentrations can stimulate the metabolic activity of microbes and then change the CDOM composition (Kellerman et al. 2014). Land use and nutrients are important factors in mediating the DOM pool, so their effects on DOM chemistry need to be assessed (Xenopoulos et al. 2021).

Jialing River is the largest tributary in the upper reaches of Yangtze River (the third largest river basin in the world with a surface area of ~1.8 million km²), covering an area of 39,200 km² and a total length of 1,345 km, and is an important source of drinking water for riparian inhabitants (Jia & Zhang 2011). To date, anthropogenic activities exert significant effects on Jialing River because of rapid urbanization. Qin et al. (2018) found that human activities and the natural environment greatly affect the water quality of Jialing River in urban Chongqing, including the weathering of rocks, soil erosion, domestic sewage, and agricultural activities. Wu et al. (2012) discovered that the loss of nitrogen and phosphorus through non-point source pollution in Jialing River Watershed is closely related to land-use types and rural residential area. However, studies on the CDOM source and composition and its relationship with land use and nutrient level in Jialing River are rarely conducted. We hypothesized that (1) CDOM had distinct spatial variations in the Nanchong section of Jialing River, and (2) a close and positive correlation existed between CDOM optical properties and land use and nutrients. Accordingly, the present study aimed to (1) recognize the spatial distribution of CDOM source and composition in Jialing River, and (2) estimate the effects of land use and nutrients on CDOM characteristics.

2. MATERIALS AND METHODS

2.1. Study area

Jialing River (102°33’–109°00’E, 29°40’–34°50’N) is the largest tributary with a total length of 1,280 km in the upper reaches of Yangtze River in western China. It flows through Shanxi, Gansu, and Sichuan provinces and eventually flows out from Chongqing City to Yangtze River (Zhang et al. 2014). The climate of Jialing River is subtropical monsoon with an average temperature of 16–18 °C. The average annual rainfall is 1098 mm, and rainfall is concentrated from May to September, accounting for 70–90% of the total annual rainfall (Wu et al. 2012). The Nanchong Section of Jialing River (105°57’–106°23’E, 30°28’–31°37’N) is located in the middle reach of Jialing River Watershed, with a total length of 271 km entering from the Langzhong District in Sichuan and flowing out of Guanyuan District in Sichuan. The Nanchong Section of Jialing River accepts the tributaries of Jingxi River, Luoxi River, Xi River, Nixi River, and Qingshuixi River (Jia & Zhang 2011).

Land use can alter river geochemical processes (Du et al. 2020; Manalilkada Sasidharan et al. 2020), and land use of riparian zones directly reflect the influence of human activities on in-river CDOM (Shao & Wang 2020). In the present study, the land use of the Nanchong Section of Jialing River within 1500 m riparian scales and each sampling site within a circular drainage segment of 1500 m radius upstream of the sampling site (Figure 1(a)–1(c)) were analyzed using the digital elevation model data (50 m resolution, http://data.ess.tsinghua.edu.cn/) and the remote-sensing data in 2017 (Gong et al. 2019) in ArcGIS 10.2. Land-use types were divided into eight groups: cropland, forest, grass, shrub, wetland, water, urban, and unused (Figure 1(a)). Field survey and land-use analysis indicated that the study regions had a clear urbanization gradient (Table S1, Supplementary material). We divided all river segments into two groups, namely, urban and suburb groups (Figure 1(b)), according to the urbanization gradient. In particular, sampling site Nos. 10–15 were classified into the urban group, and the remaining sample sites were classified into the suburb group.
2.2. Sampling collection

Field campaign was performed for a total of 18 sampling points along the Nanchong Section of Jialing River on 15–19 December 2019. Specifically, No. 1 was the inflowing river mouth of the Nanchong Section and No. 18 was the outflowing river mouth. Nos. 2, 6, 11, and 14 were four tributaries, i.e., Dong River, Xi River, Luoxi River, and Xixi River. Nos. 10, 12, 13, and 15 were the urban area, and the remaining sampling points were located in small villages or towns (Figure 1). Samples were collected at about 0.5 m below the water surface from the river shore by using a 2.5 L organic glass hydrophore. All water samples were passed through Whatman GF/F 0.7 mm filters preheated at 450 °C. The water samples were stored in a refrigerated box (4 °C) and transported to the laboratory in a frozen state prior to further processing. CDOM optical measurements were made within 1 d, and all laboratory measurements were finished within 5 d (Figure 2).

2.3. Water quality parameters

The water samples for DOC were passed through 0.7 μm Whatman GF/F filters by using a TOC-V CPN (Shimadzu, Tokyo, Japan) analyzer with combustion at ∼680 °C (Wang et al. 2014). The water samples for Chla (μg·L⁻¹), was passed through 0.45 μm Whatman GF/F filters. Chla was extracted with acetone solution and analyzed with an ultraviolet (UV) spectrophotometer (China, TU-1810) at 750 and 665 nm. TN and TP (mg·L⁻¹) were determined by spectrophotometry after digesting the samples with alkaline potassium persulfate at wavelengths of 210 and 700 nm, respectively, according to the ‘Standard Methods for the Examination of Water and Wastewater’ (Eaton et al. 1966; Zhu et al. 2013).

Figure 1 | (a) Location of sampling sites and land use types in the Nanchong Section of Jialing River, and (b) distribution of sampling sites in Nanchong City. (c) Land use types of each sampling point within a radius of 1,500 m circle buffer. ‘Tributary,’ ‘Urban,’ and ‘Other’ indicate that these sampling sites were located in a tributary of Jialing River, urban area, and suburb area, respectively.
2.4. CDOM spectral measurements

The water samples were passed through Millipore membrane cellulose filters (0.22 μm) to obtain the absorption and fluorescence spectra of CDOM. CDOM absorption spectrum was determined with a UV-Vis spectrophotometer (Shimadzu, Tokyo, Japan, UV-2550) from 200 to 800 nm at an interval of 1 nm by using a 5 cm cuvette. Milli-Q water served as a blank control, and 700 nm absorbance was subtracted to eliminate potential scattering effects. The absorption coefficient of the corresponding wavelength was then calculated according to the following equation:

\[
a(\lambda) = 2.303D(\lambda)/r
\]

where \(a(\lambda), D(\lambda), \) and \(r\) are the CDOM absorption coefficient (m\(^{-1}\)) corresponding to the wavelength \(\lambda\), the absorbance at wavelength \(\lambda\) after subtracting the absorbance at 700 nm, and the optical path (m), respectively (Kowalczuk et al. 2005; Stedmon & Markager 2005; Stedmon et al. 2011).

The spectral slope \(S\) has been proposed as a proxy of molecular weight and source of CDOM, and it was calculated from the absorption spectrum between 275 and 295 nm (\(S_{275-295}\)) through the nonlinear regression of the Equation exponential function. A smaller value meant a larger-molecule CDOM and a higher degree of humification. The slope of the spectrum was calculated according to the equation:

\[
a(\lambda) = a(\lambda_0) \times \exp \left(S(\lambda_0 - \lambda)\right)
\]

where \(\lambda_0\) and \(S\) are the reference wavelength of 440 nm and the spectral slope, respectively (Fichot & Benner 2012).

The relative molecular size (i.e., the average molecular weight) of the humic solute molecules was estimated from the ratio of the absorption coefficients at 250 and 365 nm (\(a_{250}/a_{365}\)), and \(a_{250}/a_{365}\) decreased with increased molecular size (Peura-vuori & Pihlaja 1997). The ratio of specific UV absorption at 254 nm to DOC concentration (SUVA\(_{254}\)) was positively associated with aromatic content and degree of humification (Weishaar et al. 2003).

2.5. PARAFAC analysis

CDOM fluorescence EEMs were measured using a fluorescence photometer (Hitachi, F-7000). The scanning ranges were set to 200–450 nm for excitation and 250–600 nm for emission. Readings were collected in ratio mode at 5 nm intervals for excitation and at 1 nm intervals for emission. PARAFAC analysis uses an iterative three-dimensional array decomposition algorithm based on the principle of alternating least squares to analyze three-dimensional fluorescence data and divide them into several fluorescent components with unique corresponding emission-wavelength extremes. The DOMFluor toolbox of MATLAB R2015b was used for PARAFAC analysis. Results showed that the three-component models passed split-half analysis, random-initialization analysis, and residual analysis (Stedmon & Markager 2005).

HIX is defined as the area under the emission spectra from 435 to 480 nm divided by that from 300 to 345 nm by using an excitation wavelength of 254 nm. This index is a proxy for the degree of humification and sources, and a value over 10 indicates that CDOM is characterized by significant humification and is dominated by exogenous input, whereas a value be low
The values of a254, SUVA254, and S275-295 ranged within 6.02–17.85 m\(^{-1}\), 4.53–5.90 L·m\(^{-1}\)·mg\(^{-1}\), and 16.94–18.83 μm\(^{-1}\), respectively (Table S2). a254 was significantly higher in the urban group than in the suburb group (p < 0.05), whereas a250: a365 was higher in the suburb group (p < 0.05) (Table 1). The increasing trend of a254 was consistent with that of DOC (Figure 4(a)). SUVA254 was higher in the inflowing river mouth area (sampling points 1–3) and urban area (sampling points 10–15) than in the other areas, indicating that higher aromaticity CDOM exited in these areas above (Figure 4(b)). Correspondingly, S275-295 was lower in the inflowing river mouth and urban area, further demonstrating the larger molecule CDOM input (Figure 4(c)).

The means of HIX and BIX were 4.66 ± 0.60 and 0.85 ± 0.04, respectively, suggesting significant allochthonous CDOM in the Nanchong Section of Jialing River (Table S2). The highest value of HIX was in the suburb group (Figure 4(d)), whereas the two indices had no significant difference between the urban and suburb groups (Table 1).

### Table 1 | Summary of water chemistry variables and CDOM optical parameters (mean ± SD) in urban and suburb groups in December 2019 and significance level of difference by using the Mann–Whitney U test

| Class   | DOC (mg·L\(^{-1}\)) | TN (mg·L\(^{-1}\)) | TP (mg·L\(^{-1}\)) | Chla (μg·L\(^{-1}\)) | a254 (m\(^{-1}\)) | SUVA254 (L·m\(^{-1}\)·mg\(^{-1}\)) | S275-295 (μm\(^{-1}\)) |
|---------|---------------------|---------------------|---------------------|----------------------|------------------|-------------------------------|-----------------------|
| Urban   | 1.92 ± 0.56         | 2.17 ± 0.43         | 0.08 ± 0.02         | 7.58 ± 5.76          | 10.94 ± 3.39     | 5.67 ± 0.18                   | 17.81 ± 0.59          |
| Suburb  | 1.61 ± 0.28         | 1.68 ± 0.26         | 0.07 ± 0.01         | 5.34 ± 2.67          | 8.68 ± 1.37      | 5.40 ± 0.43                   | 17.93 ± 0.43          |
| p       | >0.05               | <0.05               | >0.05               | >0.05                | <0.05            | >0.05                         | >0.05                 |

| Class   | a250:a365 | BIX  | HIX  | C1 (R.U.) | C2 (R.U.) | C3 (R.U.) |
|---------|-----------|------|------|-----------|-----------|-----------|
| Urban   | 7.40 ± 0.16 | 0.88 ± 0.04 | 4.81 ± 0.61 | 0.48 ± 0.19 | 0.48 ± 0.22 | 0.21 ± 0.10 |
| Suburb  | 7.63 ± 0.21 | 0.84 ± 0.03 | 4.46 ± 0.56 | 0.35 ± 0.07 | 0.33 ± 0.07 | 0.13 ± 0.04 |
| p       | <0.05     | >0.05 | >0.05 | <0.05     | <0.05     | <0.05     |
3.3. Spatial variation of fluorescent components

A three-component model was well validated by the split-half procedure, including two humic-like (C1 and C2) and protein-like (C3) components (Figure 5). C1 was categorized as a traditional terrestrial humic-like component (Ex/Em = 260/450 nm) (Stedmon 2003), and C2 was correlated with CDOM exported from agricultural and animal waste (Ex/Em = 240/396 nm) (Stedmon & Markager 2005). C3 had similar characteristics as the traditional tryptophan-like T peak, which has been linked to biological production (Ex/Em = 275/332 nm) (Stedmon 2003).

The fluorescence intensity of the three components was significantly higher in the urban than in the suburb group (p < 0.05) (Table 1), and the three components increased along the river flows (i.e., from sampling point 1 to sampling point 18) (Figure 6). Three fluorescence components of Xixi River (sampling of no. 14) were the highest, whereas No. 4 (located in a small town) had the lowest fluorescence intensity of three components (Figure 6(a)). Figure 6(b) shows that the percentage of humic-like C1 and C2 was higher than that of tryptophan-like C3, ranging within 38–46%, 38–43%, and 13–20%, respectively.

3.4. PCA of CDOM optical parameters, nutrients, and land use

For all samples of Jialing River, PC1, and PC2 explained 40.7% and 16.0% of the whole variance in the dataset, respectively (Figure 7). The DOC concentration, a_{254}, BIX, three fluorescent components, TN concentration, and percentage of urban land use (%Urban) displayed distinct positive correlation with PC1 loading, whereas HIX and percentage of water land use (%Water) had slightly negative correlation with PC1 loading. Thus, PC1 represented DOC, TN, and %Urban had dominant correlation with CDOM abundance and autochthonous signal. Meanwhile, a_{250}, a_{365}, S_{275-295}, percentage of cropland...
land use (%Cropland), and percentage of forest land use (%Forest) displayed strong positive correlation with PC2 loading, whereas %Water and percentage of wetland land use (%Wetland) displayed strong negative PC2 loading. %Cropland and %Forest were associated with the molecular size of CDOM.

3.5. Linkages between CDOM and nutrients and land use

Correlation analysis showed that $a_{254}$ had a close and positive correlation with TN and TP ($r^2 = 0.84$ and $r^2 = 0.33$, respectively, $p < 0.01$) (Figure 8). BIX had a significant positive correlation with DOC and TN ($p < 0.01$) (Table S3). The fluorescence intensities of three components were positively correlated with TN and TP ($r^2 = 0.30–0.84$, $p < 0.01$ or $p < 0.05$). Furthermore, the %Urban had a distinct effect on CDOM compared with the other six types of land use; that is, cropland, forest, grass, wetland, water, and unused (Table S3). $a_{254}$ and BIX had a positive correlation with %Urban ($r^2 = 0.29$, $p < 0.01$; $r^2 = 0.22$, $p < 0.01$), whereas $a_{250-236}$ had a negative correlation with %Urban ($r^2 = 0.34$, $p < 0.01$). The correlation between the three fluorescence components and %Urban was close and positive ($r^2 = 0.31–0.37$, $p < 0.01$) (Figure 9).

4. DISCUSSION

Our results indicated that elevated urban area resulted in the proportionate accumulation of CDOM, including the increase in humic-like and protein-like components and relative molecular size of CDOM in the Nanchong Section of Jialing River in December 2019. Meanwhile, increased cropland land use preferred to export simpler and smaller size of CDOM molecules into the river. First, CDOM absorption $a_{254}$, humic-like C1 and C2, and tryptophan-like C3 increased along the river flows,
which was significantly higher in the urban than in the suburb area. Moreover, the correlations of %Urban with $a_{254}$ and the three fluorescent components were close and positive. These results indicated that human activities could contribute to inputs of autochthonous and allochthonous CDOM. Moreover, the allochthonous sources were dominant in the river, as evidenced by the higher proportions of humic-like components and HIX has a mean of $>4$ and BIX has a mean of $<1$ (Salve et al. 2012). Previous studies indicated that with increased urban gradient, the percentage of allochthonous humic-like DOM component decreases whereas bioavailable autochthonous DOM increases (Parr et al. 2015). Conversely, the proportion of the two humic-like components was higher than that of the protein-like component in our study. This result was consistent with the finding that in Lake Tianmu, intensive anthropogenic land use (urban and cropland) enhances terrestrial humic-rich DOM inputs and accumulation (Shi et al. 2020). Second, the correlations of $S_{275-295}$ and $a_{250}a_{365}$ with %Cropland were
close and positive, whereas the correlation between $a_{250}/a_{365}$ and %Urban was negative. Thus, cropland land use significantly contributed to the increase in small-molecule CDOM, whereas urban land use resulted in larger-molecule CDOM. This can be supported by the positive correlation between humic-like component and %Urban. Furthermore, phytoplankton growth was inhibited in winter with low temperatures, resulting in decreased alga production, which can be supported by the relatively low Chl$\alpha$ concentration and low proportion of tryptophan-like component in the Nanchong section of Jialing River...
Previous studies have suggested that phytoplankton degradation in eutrophic systems is an important autochthonous source of CDOM (Coelho et al. 2017). Thus, human activities significantly affect the source and composition of CDOM in river systems.

Our results suggested that increased nutrient loading was an important factor for increasing CDOM abundance and the three fluorescence components, as well as for increasing the autochthonous signals. The close and positive relationship was reported between $a_{254}$ and nutrients, and $a_{254}$ had a similar spatial variation with TN concentration, indicating that increased nutrient loading stimulated the accumulation of CDOM abundance. Previous studies have reported that Jialing River is the main water source of agricultural irrigation and increases the release of nitrogen from agricultural land (Jia & Zhang 2011; Zhu et al. 2019a). A positive correlation existed between TN concentration and %Urban (Table S3), indicating that anthropogenic activities fueled TN input to indirectly alter in-river CDOM. BIX and tryptophan-like component had strong and positive correlation with TN concentration, which was related to the increased organic matter discharged from the domestic wastewater (Shi et al. 2020). The percentage of the three fluorescent components had strong and positive correlation with TN and TP, further confirming that nutrients are the important driver to CDOM. Das et al. (2017) showed a similar result for the coastal waters of the northern Bay of Bengal. However, no significant correlation existed between Chl$\alpha$ and CDOM optical parameters, suggesting that phytoplankton was likely a secondary factor affecting the source and composition of DOM (Yao et al. 2018).

We also found a high spatial variability of CDOM optical property and water quality parameters along river flows, especially the higher value of CDOM abundance, three fluorescence components, and TN in the urban area. Notably, Xixi River had the highest fluorescence components, DOC concentration, nutrients, and Chl$\alpha$ concentration. This finding was likely due to the fact that Xixi River is the closest to the urban area, and a dam has been built in its upper part, resulting in decreased self-purification capacity for urban sewage (Zhu et al. 2019b; Wang et al. 2020). Thus, the water quality of Xixi River requires improvement.

Human activities alter land-use types, thereby affecting the variations in sources and composition of CDOM in river systems, i.e., size and aromatic, terrestrial, and endogenous components (Xenopoulos et al. 2021). Hence, investigating the composition of land use in riparian zones is important to understand river-ecosystem protection and the carbon-cycle progress. Based on the short-term observations in the study, rapid urbanization and increased human-induced nutrients resulted in increased CDOM abundance. Therefore, water management should be enhanced to protect river systems with rapid urbanization.
5. CONCLUSION

Urbanization and nutrient loading are important factors driving the spatial variations of sources and composition structure of CDOM in the Nanchong Section of Jailing River in December 2019. DOC concentration, TN concentration, CDOM abundance and fluorescence components were higher in the urban area. Terrestrial humic-like and agriculture humic-like components were dominant parts of CDOM in the Nanchong Section of Jialing River, whereas the lower proportion of tryptophan-like component may be likely due to the inhibition of algal growth in December 2019. Increased urban land use and nitrogen loading in Jialing River contributed to the increase in inputs of allochthonous CDOM, and cropland land use stimulated the smaller-molecule CDOM inputs. Therefore, rapid urbanization and elevated nutrient levels can significantly drive the variations in CDOM and the study provides critical information to understand how land use affects the carbon cycle in aquatic ecosystems, thereby guiding sustainable strategies to protect drinking water.

ETHICAL APPROVAL AND CONSENT TO PARTICIPATE

Not applicable.

CONSENT TO PUBLISH

Not applicable.

AVAILABILITY OF DATA AND MATERIALS

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

CONFLICTS OF INTEREST

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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AUTHOR CONTRIBUTIONS

LQZ wrote the initial draft and prepared the published work, as well as analyzed data. XHZ contributed to the revision of the initial draft and validation of the data. XH and CRL completed the investigation of the study area, as well as the collection, transportation, and preservation of samples. YY formulated the overarching research goals and commented on the initial draft.

DATA AVAILABILITY STATEMENT

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

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