Hydrological isolation affected the chemo-diversity of dissolved organic matter in a large river-connected lake (Poyang Lake, China)

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HIGHLIGHTS

- The impacts of hydrological isolation on DOM chemistry in Poyang Lake (China) were investigated by FT ICR MS and UV-Vis.
- More CHOS formulas were identified in sub-lakes through the Kraft reactions and highly contributed to aliphatic compounds.
- The mainly reaction pathways of DOM in sub-lakes were photodegradation while biodegradation in main lake.
- Hydrological isolation may affect the lability of DOM on the molecular level.

GRAPHICAL ABSTRACT
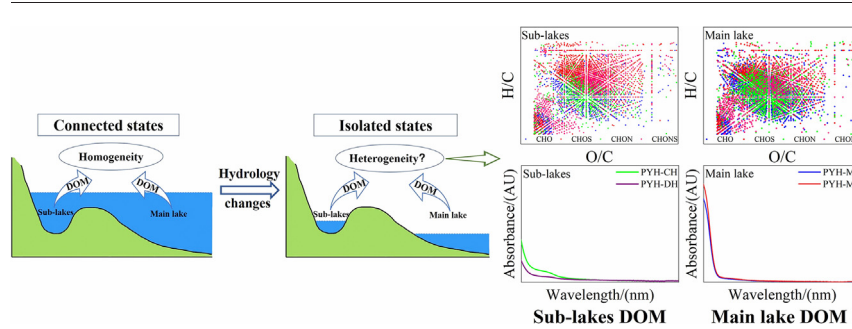
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ABSTRACT

The transportation processes during aquatic systems regulate the ultimate chemistry of dissolved organic matter (DOM), and in recent years, climate changes and human activities have altered the hydrological patterns of many rivers and lakes, which generated some severe issues, such as hydrological isolation. However, how hydrological isolation affects variations of DOM chemistry in large lake systems is still poorly understood. Here, optical properties and molecular compositions of DOM samples derived from a large river-connected lake (Poyang Lake, China) and its nearby seasonal sub-lakes (formed by hydrological isolation) were characterized using Fourier transform ion cyclotron resonance mass spectrometry (FT ICR MS) and ultraviolet-visible (UV–Vis) spectroscopy. The results revealed more abundance of organic matter in sub-lakes than that in the main lake according to high dissolved organic carbon (DOC) concentrations and absorption coefficients (a_{254} and a_{280}). Large proportions of CHOS formulas were identified by FT ICR MS in sub-lakes DOM, which were produced through Kraft reactions (sulfide/bisulfide + lignin CHO → CHOS) in the interface of sediment/water, and greatly contributed to aliphatic compounds. In addition, obvious variations of compounds (such as polyphenols, highly unsaturated and aliphatic compounds) and lability of DOM were observed between sub-lakes and main lakes, which were mainly caused by the different degradation pathways of DOM (photodegradation in sub-lakes while biodegradation in the main lake). Our results demonstrated that hydrological isolation has significant impacts on DOM chemistry, and provides an improved understanding of the DOM biogeochemistry process in Poyang Lake and supports the management of the large lake systems.

1. Introduction

Dissolved organic matter (DOM), as the major form of reactive carbon in aquatic ecosystems, plays a fundamental role in aquatic biogeochemical processes (e.g., nutrients export) and the global carbon cycle (Battin et al.,

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2009; Liu et al., 2022; Lynch et al., 2019). The transportation of DOM through freshwater systems has significant effects on its ultimate chemistry, because associated with its transport there are many biogeochemical processes capable of influencing DOM, such as degradation processes (photodegradation or biodegradation), adsorption-desorption behaviors (i.e., minerals, microplastics), and complexations (e.g., heavy metals) (Kellerman et al., 2014; Ren et al., 2022; Yun and Jin, 2020; Zhang et al., 2021). Hydrology conditions (e.g., flooding period and dry period) and human activities (e.g., dam constructions) alter previous hydrological patterns of rivers and lakes (Huang et al., 2022; Wang et al., 2021), such as hydrological isolation, which could facilitate the formation of small seasonal isolated lakes (sub-lakes) (Maia et al., 2018). Many studies have proposed that although with a small surface area and short hydroperiod, these sub-lakes have high biological diversity and important ecological functions (like retaining water and providing habitat for birds) (Xia et al., 2016; Yu et al., 2015). Hence, the variations of hydrology and ecology may accelerate the changes of DOM chemistry in freshwater systems.

However, the research about the impacts of hydrological isolation on the DOM chemistry of seasonal sub-lakes was still limited. Previous publications have only focused on the impacts of hydrological isolation (or reversely, hydrological connectivity) on water chemistry (i.e., water quality, bacterioplankton diversity) of lakes and their nearby sub-lakes (Cook and Hauer, 2007; Lew et al., 2016; Yu et al., 2015), and significant differences were found between lakes and sub-lakes. For instance, Yu et al. (Yu et al., 2015) investigated the dissolved organic carbon (DOC) and total dissolved nitrogen (TDN) concentrations and compositions of 40 seasonal sub-lakes in Piedmont ecoregion (USA), results showed that surficial hydrological connectivity and surrounding land-use of sub-lakes were the main factors to constrain the variations of DOC and TDN, the DOC and TDN concentrations of isolated lakes were significant higher than those of connected lakes without obvious land-use changes. Generally, the DOC concentration can reflect the abundance of DOM in water. Therefore, we hypothesize that, the DOM chemistry in aquatic systems is related to hydrological isolation.

Poyang Lake, as the typical river-connected lake and one of the six major wetlands in the world, has complicated topographical features (Li et al., 2019). In past years, the wetland area and water level of Poyang Lake have changed considerably, which was affected by the climate change (e.g., rise in temperature) (Huang et al., 2022) and human activity (i.e., construction of the Three Gorges Dam in Yangtze River) (Zhang et al., 2014), and resulting in the formation of >100 seasonal sub-lakes (total surface areas were approximately 800 km²) with varied connectivity (Hu et al., 2015). These sub-lakes are fully or partly connected with the main lake during the flooding period (summer) and isolated in the dry season (winter). Moreover, Li et al. (2019) have pointed out that sub-lakes in Poyang Lake wetlands have highly dynamic hydrological connectivity due to the great fluctuation of water level in the main lake, which provides an ideal study site for investigating the influences of hydrological isolation (or hydrological connectivity).

Here, the impacts of hydrological isolation on DOM chemistry of Poyang Lakes and its nearby sub-lakes were investigated by the combination of Fourier transform ion cyclotron resonance mass spectrometry (FT ICR MS) and ultraviolet-visible (UV–Vis) spectroscopy, which have been successfully used to characterize the DOM chemistry in various aquatic systems and obtained in-depth information about molecular diversity and biogeochemistry process of DOM in aquatic systems (Kellerman et al., 2018; Liu et al., 2022; Melendez-Perez et al., 2016; Minor et al., 2012; Zark and Dittmar, 2018). It is worth noting that little spatial variations were observed in certain areas of natural lakes and rivers in terms of the optical properties and molecular characteristics of DOM, according to previous studies (Liu et al., 2020; Wang et al., 2019). Thus, we assumed the DOM chemistry was similar within a small area in Poyang Lake during the connected states.

The objectives of this paper are: 1) to characterize the optical properties and molecular characteristics of DOM in the main lake and sub-lakes of the Poyang Lake basin; 2) to investigate the variations of DOM diversity and biogeochemistry process between the main lake and sub-lakes; 3) evaluate the relationship between DOM chemistry and hydrological isolation.

2. Materials and methods

2.1. Study area description

Poyang Lake (28°22′–29°45′N, 115°47′–116°45′E), the largest freshwater lake in China (with a watershed area of 16.2×10^4 km²), is located in the middle and lower reach of the Yangtze River (Ni et al., 2020) (Fig. 1). It receives inflow primarily from five tributaries (Ganjiang River, Fuhe River, Xinjiang River, Raohe River, and Xiushui River) in the south and discharged into the Yangtze River at Hukou in the north (Huang et al., 2022). Affected by river inflows and interactions with the Yangtze River, the lake water level fluctuated between 8 and 22 m in one year, resulting in the lake surface area ranged from 907 to 3052 km² in the dry season and flooding season. In addition, the lake basin has a subtropical humid monsoon climate, with average annual temperature and precipitation of 16.6–18.0 °C and 1400–1600 mm, respectively (Li et al., 2019; Tao et al., 2021).

The Poyang Lake National Nature Reserve (PYNNR) is located in the northwest region of Poyang Lake and contained nine sub-lakes (Fig. 1), which are a highly conservative site and considered to be rarely disturbed
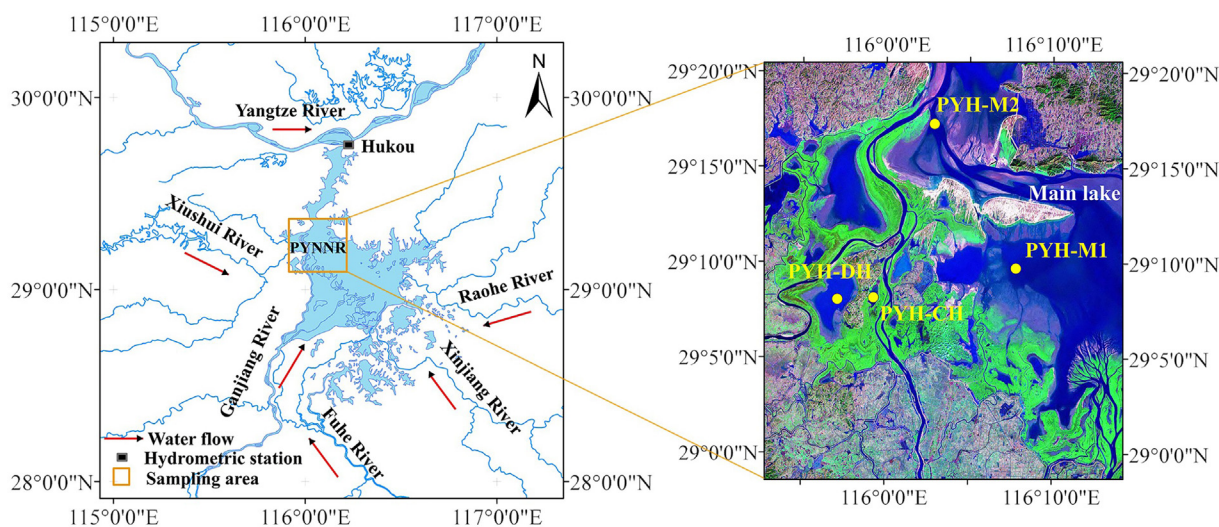

Fig. 1. Location of the Poyang Lake and the sampling sites in this lake.

by human activities, and provides excellent habitats for overwintering birds (Zhang et al., 2012). The sub-lakes were connected with the main lake from April to November, and isolated from December to next March. During the disconnected period, the water level and volume of sub-lakes were relatively stable, which were dominated by precipitation, evaporation, and infiltration. As two of the nine sub-lakes in PYNNR, Changhu lake (surface area of 2.9 km^2) and Dahu lake (surface area of 29.5 km^2) were selected, which have similar geomorphic types and hydroperiod.

2.2. Sample collections and procedure

The sampling time was January 6, 2022, when the sub-lakes were fully separated with the main part of Poyang Lake. Four samples were obtained, two samples in sub-lakes (Changhu lake and Dahu lake), and another two samples in the main lake area near the sub-lakes (Fig. 1). All surface water samples were collected in 1 L acid-cleaned Nalgene bottles and transported back to the lab on the ice within 6 h, samples in the lab were filtered immediately through 0.7 μm and 0.45 μm glass fiber filters (Whatman GF/F, pre-combusted for 5 h at 450 °C), the filtrates were stored at 4 °C in the dark until further analysis.

2.3. Dissolved organic matter analysis

The concentrations of dissolved organic matter (DOC) were determined by high-temperature catalytic oxidation on a TOC analyzer (TOC-L CPN, Shimadzu), following a method reported previously (Benner and Hedges, 1993), and the relative standard deviation of all tests was <2 %.

2.4. Ultraviolet-visible absorption spectrophotometry analysis

The Ultraviolet-visible (UV–Vis) absorption spectrum of DOM samples was measured using an Agilent 8543 UV–Vis spectrophotometer with a 1 cm quartz cuvette. The spectrum ranged from 200 nm to 600 nm with a 1 nm interval. The ultrapure (Milli-Q) water was used as reference, absorbance measurements at each wavelength were subtracted the absorbance at 700 nm for baseline correction in each sample (Liu et al., 2022), and the corresponding absorption coefficient was calculated by the following equation:

$$a\,(\lambda) = 2.303D\,(\lambda)/L$$

where a (λ) is the measured absorbance coefficient at a specific wavelength λ (m^{-1}), D (λ) is the absorbance value of wavelength λ, L is the path length of quart cuvette, 2.303 is the common-to-natural logarithm transformation (Feng et al., 2018).

The absorption coefficient at the wavelength of 254 nm or 350 nm represented the chromophoric DOM (CDOM) abundance (Liu et al., 2022). The specific ultraviolet absorption of DOC concentration at the wavelength of 254 nm (SUVA$_{254}$) or 280 nm (SUVA$_{280}$), was positively correlated with the aromaticity of CDOM (Zhang et al., 2020). Spectra slope ratio (S_R) was defined as the ratio of spectra slope $S_{275-295}$ and $S_{350-400}$, which have been proposed to characterize the source and structural characteristics of CDOM (Helms et al., 2008; Li and Hur, 2017).

2.5. Solid-phase extraction of DOM and FT-ICR MS analysis

DOM of all water samples (0.3–0.5 L, depending on the concentration of DOC) were extracted via an established solid-phase extraction method using 500 mg, 6 mL Agilent Bond Elut PPL cartridges, following procedures detailed in (Dittmar et al., 2008). Ultrahigh-resolution mass spectra of SPE-DOM samples (with the DOC concentration of 100 mg/L) were acquired using a 15 T Fourier transform ion cyclotron resonance mass spectrometer (FT-ICR MS; Brucker Daltonics) equipped with an Apollo II electrospray ion source under the negative ion mode. The measurement procedures and instrument parameter settings were both followed by previous studies (Cao et al., 2015; Lv et al., 2016). Briefly, the methanol samples were

continuously injected into the electrospray source at a rate of 120 μL h^{-1} through a syringe infusion. For each spectrum, >300 time-domain transients were accumulated in the mass spectra range from 100 to 800 Da with an ion accumulation time of 0.1 s, and the data acquisition size was set to 4 M (32 Mbyte). All spectra were first externally calibrated using a 10 mM of sodium formate solution in 50 % isopropyl alcohol and then internally calibrated with a reference mass list of natural organic matter molecules, which ensure the mass error was lower than 500 ppb for singly charged molecular ions over the entire mass range. Peaks identification were performed with Bruker Data Analysis software.

Data processing were calculated using a homemade software, established based on previous publications (Fu et al., 2020; Lv et al., 2016) with modifications as the following content. Molecular formulas were assigned to exact mass peaks according to the stringent criteria with elemental combinations of $^{12}C_{1-100}^{1}H_{1-200}^{16}O_{0-50}^{14}N_{0-3}^{32}S_{0-1}$ and only the peaks with signal to noise ratio (S/N) greater than four were considered (Koch et al., 2007; Osterholz et al., 2015). Formula with the lowest number of non-oxygen heteroatoms (N + S) were selected when the measured m/z value correspond to non-unique possible formulas (Kujawinski et al., 2009) and mass measurement error was <1 ppm. Additionally, the elemental ratio limits of 0.2 < H/C < 2.2, 0.1 < O/C < 1.2 were further restricted for formula calculation. Relative peak intensity was normalized to the total intensity of all assigned molecular formula peaks in each sample. The molecular formula classes (CHO, CHOS, CHON, and CHONS), the mass-to-charge ratio (m/z), and atomic ratios (H/C and O/C) were calculated based on the magnitude-weighted average for each sample. Double bond equivalent (DBE), modified aromaticity index (AI$_{mod}$) (Koch and Dittmar, 2006), molecular lability index (MLB$_L$) (D'Andrilli et al., 2015) and an island of stability index (IOS) (Lechtenfeld et al., 2014) were obtained to describe the characteristics of DOM. Moreover, the inter sample ranking analysis was used to reveal principal differences in DOM quality and detailed descriptions can be found in supplementary information 1.

Various molecular categories were applied to deduce the biogeochemistry transformation and transport process of DOM based on the DBE, AI$_{mod}$ and atomic ratios (H/C and O/C) of identified molecules (Kellerman et al., 2018; Liu et al., 2022; Seidel et al., 2014), these molecular categories are: 1) polycyclic aromatics (AI$_{mod}$ > 0.66); 2) polyphenolics (0.66 ≥ AI$_{mod}$ > 0.50); 3) highly unsaturated and phenolic compounds (AI$_{mod}$ ≤ 0.50, H/C < 1.5); 4) aliphatic compounds, including unsaturated aliphatic compounds (2.0 > H/C ≥ 1.5, $N = 0$) and peptide-like molecular formulae (2.0 > H/C ≥ 1.5, $N > 0$); 5) saturated compounds (H/C ≥ 2.0, or O/C ≥ 0.9); 6) carboxylic-rich alicyclic molecules (CRAMs; DBE/C = 0.30–0.68, DBE/H = 0.20–0.95, DBE/O = 0.77–1.75) (Hertkorn et al., 2006).

3. Results and discussions

3.1. DOC concentrations and UV–Vis absorption properties

As listed in Table 1, the DOC concentrations in the main lake were 4.69 mg/L (PYH-M1) and 4.30 mg/L (PYH-M2), and in sub-lakes were 18.83 mg/L (PYH-CH) and 11.19 mg/L (PYH-DH), indicating that the DOC concentrations in sub-lakes were greatly higher than those in the main part of Poyang Lake in the dry season. Consistently, Lynch et al., (Lynch et al., 2019) found that the DOC concentration of a disconnected pond (9.90 mg/L) was also much higher than that in the nearby mainstream (2.70–4.27 mg/L) in a fluvial network. In general, the DOC

Table 1
DOC concentrations and parameters of ultraviolet-visible absorption spectrum.

Samples	DOC (mg/L)	$a_{(254)}$ (m^{-1})	$a_{(350)}$ (m^{-1})	SUVA$_{254}$ (L/mg·m)	SUVA$_{280}$ (L/mg·m)	S_R
PYH-M1	4.692	11.01	3.31	2.35	1.79	2.80
PYH-M2	4.304	12.99	4.12	3.02	2.35	2.33
PYH-CH	18.83	33.74	8.87	1.79	1.32	4.40
PYH-DH	11.19	17.56	7.65	1.57	1.30	7.50

concentration representing the abundance of DOM in water was easily affected by many factors, including the sources and potential biogeochemical processes. The sub-lakes were the areas associated with the largest plant biomass and the highest species richness in the Poyang Lake wetland, and also the superior habitats for overwintering birds (Hu et al., 2015). In addition, the water levels of sub-lakes were only 0.5–0.8 m during sampling time, the low overlying water column may facilitate the resuspension of sediment, potentially leading to the release of DOM from the sediments. Thus, the apoptosis of aquatic plants, the activities of overwintering birds, and sediment releasing may be the reasons for the higher DOC concentration in sub-lakes. Moreover, we noticed that the DOC concentrations were more variable in the two sub-lakes than in main lake samples. Because PYH-DH has substantial higher water volume than PYH-CH, and high volume may dilute DOC to lower concentrations, explaining the variations of DOC between the two sub-lakes.

UV-Vis spectra could reflect molecular structures and complexity of organic matters. The UV–Vis spectra of four DOM samples were presented in Fig. 2, spectra curves of PYH-M1, PYH-M2, PYH-CH and PYH-DH all decrease exponentially with wavelength increments, and no obvious absorption peak after 500 nm was detected. The curves show significant differences in the range from 200 to 350 nm between the main lake and the sub-lakes, with the shapes of the curve being identical for main lake sites but not for the sub-lakes. Absorption peaks of the spectral curves in the region of 226–250 nm were mainly caused by inorganic ions (bromide and nitrate) (Khan et al., 2014), while absorption peaks in the region of 250–280 nm were related to lignosulfonate acid and its derivative components (Peuravuori and Pihlaja, 1997). Therefore, the notable distribution differences in spectra curves between the main lake and the sub-lakes have indicated the different structures and compounds of chromophoric DOM (CDOM).

The absorption coefficients $a_{(254)}$ and $a_{(350)}$ can be used to characterize the relative abundance of CDOM (Liu et al., 2022). The $a_{(254)}$ and $a_{(350)}$ were respectively 33.74 m^{-1} and 8.87 m^{-1} for PYH-CH, and 17.56 m^{-1} and 7.65 m^{-1} for PYH-DH, both of which were higher than those for PYH-M1 (11.01 m^{-1}, 3.01 m^{-1}) and PYH-M2 (12.99 m^{-1}, 4.12 m^{-1}) (Table 1). These results suggest the abundance of CDOM in sub-lakes was higher than in main lake, consistent with the results of DOC concentration. The lower values of SUVA$_{254}$ and SUVA$_{280}$ in PYH-CH and PYH-DH were observed than in PYH-M1 and PYH-M2, indicating the aromaticity of DOM were lower in the sub-lakes than in main lake. The lower aromatic degree of DOM may be attributed to weaker activities of microorganisms and stronger photobleaching in sub-lakes (Helms et al., 2008), which will be discussed further later along with mass spectrometry results. The S_R values, which could indicate the origins of DOM (>1 means endogenous origins)

(Li and Hur, 2017). In the present study, the S_R values of sub-lakes (7.50 and 4.40) and the main lake (2.80 and 2.33) are both >1, indicating that the source of DOM is mainly endogenous in Poyang Lake. Sub-lakes are associated with higher abundance of aquatic plants than main lake area, may explaining larger S_R in sub-lakes.

3.2. Molecular characteristics of DOM

Fig. 3 shows the broadband ESI FT ICR MS spectra of PYH-M1, PYH-M2, PYH-CH, and PYH-DH DOM samples, thousands of peaks were detected in the mass range of 100–800 Da for each sample, and all the spectra approximately match the Gaussian distribution, as lake DOM in other studies (Flerus et al., 2012; Zark and Dittmar, 2018). Intensity-weighted average parameters of FT ICR MS were presented in Table 2. Fewer molecules were identified at PYH-CH (4773) and PYH-DH (4393) in sub-lakes compared to PYH-M1 (5506) and PYH-M2 (5916) in main lake, suggesting the compositions of DOM were less diverse in sub-lakes. Alternatively, the result suggests more molecules were vulnerable for extracting or ionization in DOM of main lake (Liu et al., 2020). The average m/z of PYH-CH and PYH-DH were 378.21 Da and 378.35 Da respectively, lower than those of PYH-M1 (396.54 Da) and PYH-M2 (399.91 Da). Previous studies have reported that low molecular weight DOM has a stronger diagenetic process and lower bioreactivity than high molecular weight DOM (Benner, 1996; Flerus et al., 2012). Additionally, the average DBE values of PYH-M1, PYH-M2, PYH-CH, and PYH-DH were respectively 8.42, 8.62, 7.12, and 8.13, and AI$_{mod}$ values were respectively 0.25, 0.26, 0.13, and 0.21, indicating that sub-lakes have relative smaller unsaturation degree and aromaticity, consistent with previous UV–Vis results.

The assigned formulas in all DOM samples were divided into four formulas categories according to the elements contained: CHO, CHOS, CHON, and CHONS. As shown in Fig. 3, the proportions of each formulas were similar in PYH-M1 and PYH-M2, which were dominated by CHO formulas, accounting for 59.67 % and 63.60 %, respectively. Little differences were observed in the proportions of CHOS and CHON formulas, both accounting for 15 %–20 %. The CHONS formulas were least of them, only 4 %–5 %. However, the percentages of each category in PYH-CH and PYH-DH were notably different. Compared to the main lake samples, the proportions of CHO and CHON formulas were lower in PYH-CH (37.13 % and 8.84 %) and PYH-DH (49.96 % and 11.78 %), and more CHOS formulas were identified in PYH-CH (46.30 %) and PYH-DH (30.11 %), particularly in PYH-CH. Most publications have reported that proportions of CHOS formulas were 5 %–20 % of natural water DOM (Valle et al., 2019; Wagner et al., 2015), but Pang et al. (2020) found higher proportions of CHOS formulas (23.8 % - 39.2 %) in DOM of Lake Taihu. Another study also showed that the proportions of CHOS formulas were in a wide range from 3.6 % to 41.2 % among 11 main lakes in China (Liu et al., 2022). In lake DOM, CHOS compounds were mainly derived from two origins. The first one being human activities, the O$_3$S and O$_5$S classes in CHOS formulas were likely associated with linear alkylbenzene sulfonates (LAS) and its degradation products, called sulfophenyl carboxylic acids (SPC) (He et al., 2022; Wang et al., 2019). LAS is a kind of synthetic surfactants used in detergents and personal care products, and has been widely detected in wastewater and other inland water with severe anthropogenic impacts (Gonsior et al., 2011; He et al., 2019; Melendez-Perez et al., 2016). The second origin is that CHOS compounds were formed by the addition of reduced sulfur anions (e.g. sulfide, bisulfide) to lignin CHO compounds via Kraft reactions under anaerobic environments (sulfide/bisulfide + CHO → CHOS) (Melendez-Perez et al., 2016; Sleighter et al., 2014). In this study, the abundance of O$_3$S and O$_5$S classes in the main lake and sub-lakes were relatively similar (Fig.S1), indicating that human activities were not the main reason for the differences in CHOS formulas abundance between DOM of sub-lakes and main lakes. Some previous studies have proved that CHOS compounds were naturally generated at the interface of sediment and water, and sediments can be the heterogeneous catalysts to promote the formation of CHOS compounds (Melendez-Perez et al., 2018; Yuthawong et al., 2020). Thus, due to a really low water level (close to the sediment-water interface)
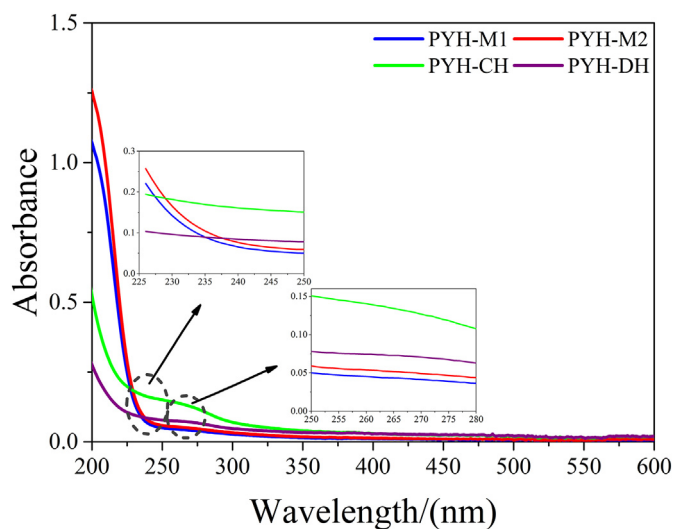

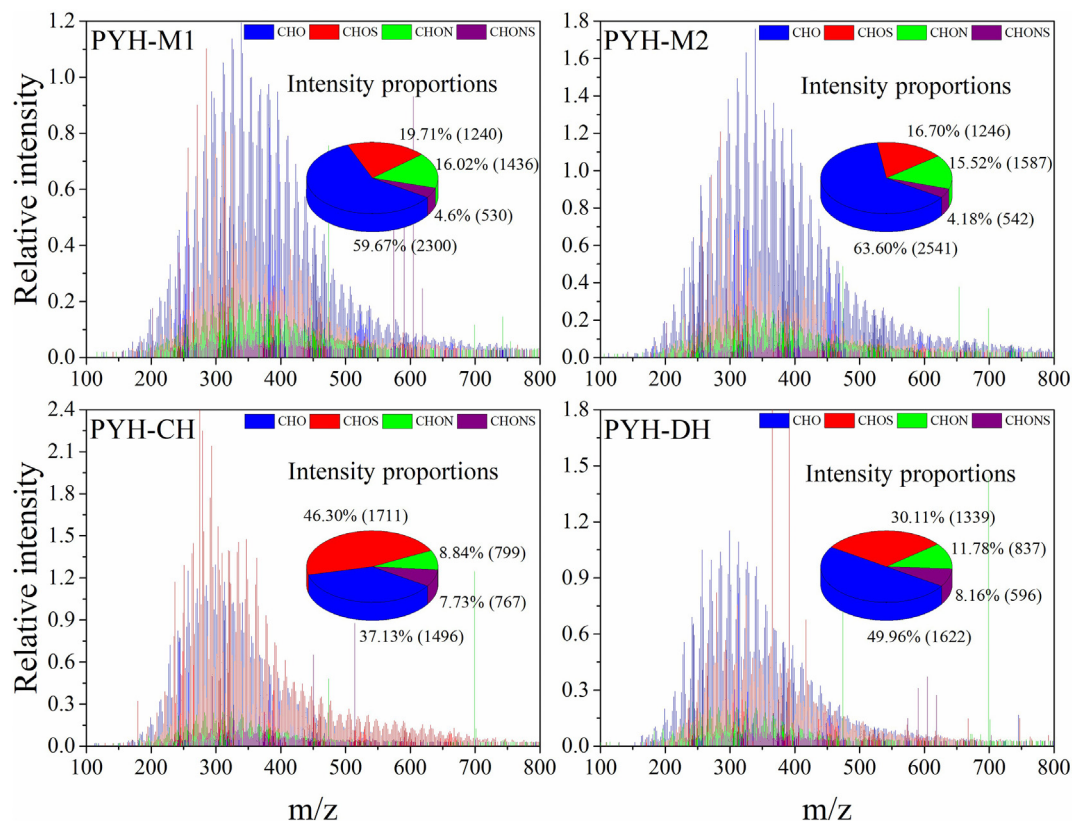**Fig. 2.** Uv-vis absorption spectra of DOM samples.

Fig. 3. Broadband negative ion ESI FT-ICR mass spectra and intensity proportions of CHO, CHOS, CHON, and CHONS formulas in DOM samples.

in the dry season compared with the main lake area, this formation process may be the main reason for higher proportions of CHOS formulas in sub-lakes. As for CHON formulas, the proportions of CHON formulas were lower in sub-lakes than in main lake. Farmland in the lake basin may contribute sources of organic N to main lake, but not to sub-lakes when isolated (Mattsson et al., 2009; Wilson and Xenopoulos, 2009). Relative abundance of classes species of DOM samples was presented in Fig.S2, 148 (PYH-M1), 144 (PYH-M2), 132 (PYH-CH), and 138 (PYH-DH) heteroatom classes were assigned to four samples, respectively. Referred to the detected classes of species (Fig.S2), main lake samples likely had a higher diversity of DOM compositions than sub-lakes. Moreover, the class of the highest abundance in each category was relatively consistent, for example, the O_8 class was the highest abundance in CHO formulas among all DOM samples.

The inter samples ranking analysis have been widely employed to understand differences in DOM quality between two or more samples. As shown in Fig.S4, the distributions of common molecular formulars (MFs)

in main lake and sub-lakes DOM were displayed. The first- and second-rank molecular formulas (relatively high molecular relative intensities) of main lake DOM mainly occupied the region of $0.5 < H/C < 1$, and the region of $1 < H/C < 1.5$ and $O/C < 0.5$. Whereas the first- and second-rank MFs of sub-lakes DOM mainly distributed in the region of $H/C > 1$ and $O/C > 0.5$ (with relative low m/z values) and in the region of $H/C < 0.5$ and $O/C < 0.5$, with m/z values ranging from 350 to 550 Da. All these results confirm the composition differences between main lake and sub-lakes DOM as well.

Complicated FT ICR MS data can be visualized by van-Krevelen (V—K) diagrams, the proportions of five compounds (polycyclic aromatics, polyphenolics, highly unsaturated and phenolic compounds, aliphatics and saturated compounds) were presented in Fig. 4, and distributions of CHO, CHOS, CHON, and CHONS formulas were shown in Fig. 5. The relative abundance of polyphenols and highly unsaturated and phenolic compounds in PYH-CH and PYH-DH were lower than in PYH-M1 and PYH-M2, while the aliphatic compounds were greatly higher (Fig. 4). Previous studies have shown that highly aromatic substances (such as polyphenols and highly unsaturated and phenolic compounds) in DOM were susceptible to photodegradation and converted into smaller molecules (photoproducts, like aliphatics) (Medeiros et al., 2015; Mesfioui et al., 2015; Wilske et al., 2020). The aliphatics (including unsaturated aliphatic and peptide-like compounds), is broadly considered to be bio-labile and photo-resistant, and readily vulnerable to microbial degradation (e.g., decarboxylated) (Liu et al., 2022; Medeiros et al., 2017; Stubbins et al., 2010). Therefore, the high proportions of polyphenols and highly unsaturated compounds in the main lake DOM may be due to the greater degree of microbial degradation, while the high abundance of aliphatic compounds in sub-lakes was mainly affected by photodegradation. It is easy to understand that Poyang Lake is a typical river-connected lake, the main lake has a high flow speed and therefore higher oxygen contents favoring microbial activities, compared to sub-lakes with stable water levels and longer water retention time, stimulating photodegradation. Interestingly, it is worth noting that

Table 2
Intensity-weighted average parameters based on ESI FT ICR MS in DOM samples.

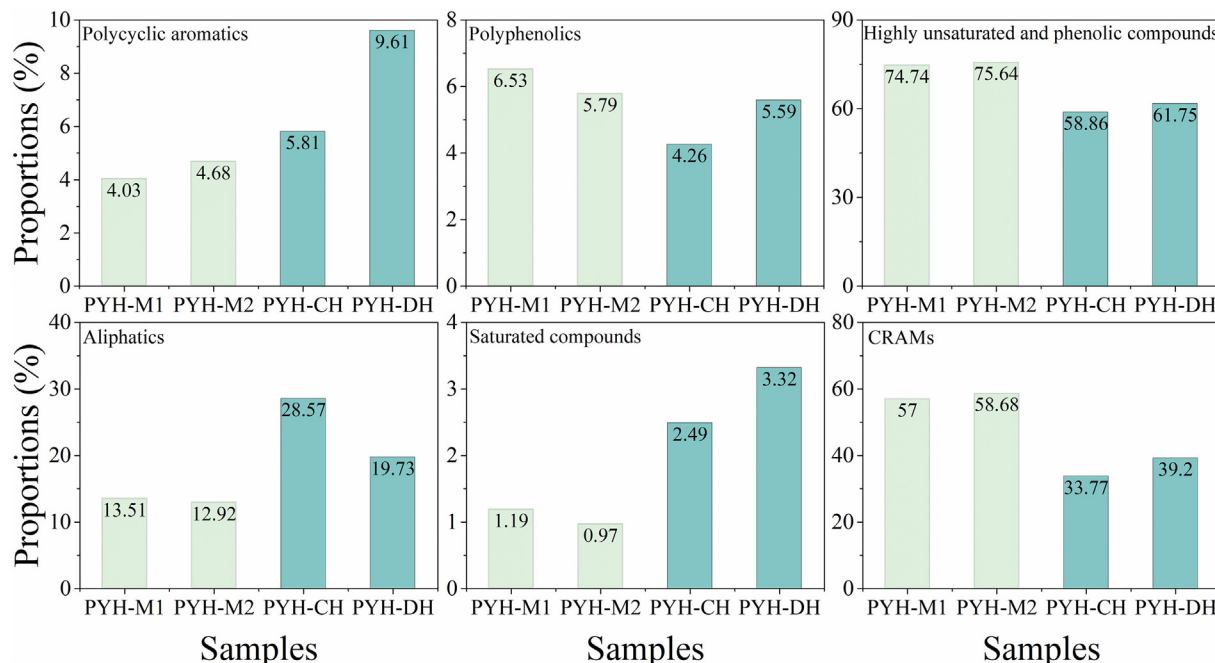| Parameters	PYH-M1	PYH-M2	PYH-CH	PYH-DH
Number	5506	5916	4773	4393
m/z	396.54	399.91	378.21	378.35
C	18.44	18.92	16.64	17.40
H	22.36	22.91	21.27	20.84
O	8.86	8.74	8.60	8.33
N	0.30	0.30	0.23	0.30
S	0.24	0.21	0.54	0.38
H/C	1.22	1.22	1.31	1.23
O/C	0.50	0.48	0.55	0.52
DBE	8.42	8.62	7.12	8.13
AI_{mod}	0.25	0.26	0.13	0.21
MLB_L (%)	14.70 %	13.88 %	31.07 %	23.05 %
IOS (%)	9.87 %	9.87 %	6.08 %	7.21 %

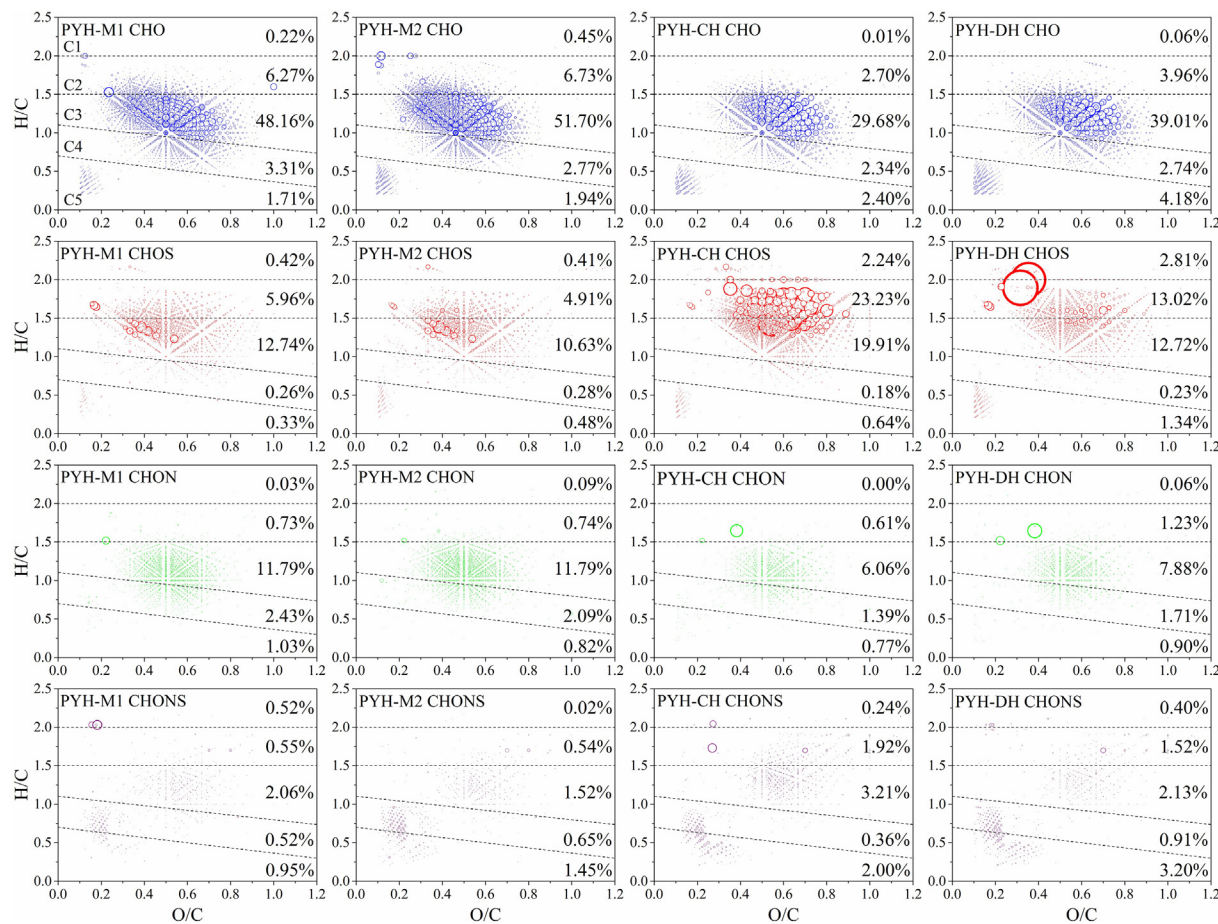**Fig. 4.** Proportions of each compound of four DOM samples.

Fig. 5. Van Krevelen diagrams of four formulas (CHO, CHOS, CHON, and CHONS) in DOM samples. C1, C2, C3, C4, and C5 were saturated compounds, aliphatics, highly unsaturated and phenolic compounds, polyphenolics and polycyclic aromatics, respectively. (Point size reflects molecular abundance; Numbers in the diagram represents the proportions of each corresponding compounds).

the contributions of CHOS formulas to aliphatic compounds were great in sub-lakes, which account for 23.3 % and 13.02 % in PYH-CH and PYH-DH, and as for the CHO formulas in highly unsaturated and phenolic compounds, the contribution pattern was opposite, higher proportions of 48.16 % and 51.70 % were distributed in PYH-M1 and PYH-M2 (Fig. 4).

Carboxyl-rich alicyclic molecules (CRAMs) were proposed to reflect the abundance of refractory components in DOM, and have been identified widely in natural water DOM (Hertkorn et al., 2006; Zhou et al., 2022). As shown in Fig. 4, the CRAMs in PYH-M1, PYH-M2, PYH-CH, and PYH-DH were 57.00 %, 58.68 %, 33.77 %, and 39.20 %, respectively, demonstrating that sub-lakes DOM contained less refractory components. $MLB_L\%$ and IOS% evaluate the overall lability of DOM (D'Andrilli et al., 2015; Lechtenfeld et al., 2014). The value of $MLB_L\%$ were higher in PYH-CH (31.07 %) and PYH-DH (23.05 %) than in PYH-M1 (14.70 %) and PYH-M2 (13.88 %), and IOS% were lower in PYH-CH (6.08 %) and PYH-DH (7.21 %) than in PYH-M1 (9.87 %) and PYH-M2 (9.87 %). Both two indices indicated that the DOM of sub-lakes were more labile, consistent with previous results. Previous studies have shown that the main lake DOM has a higher degree of microbial degradation, which could convert organic matter into the refractory organic matter with strong aromaticity (Minor et al., 2012; Nebbioso and Piccolo, 2013).

3.3. Implications for DOM biogeochemistry and further considerations

Obvious differences in DOM chemistry between the main lake and sub-lake influence by the hydrological isolation have important implications for DOM biogeochemistry. DOM is the largest pool of organic matters in natural waters (Liu et al., 2022), the sources and diverse biogeochemical processes of DOM determine its quality and quantity. In our study, hydrological isolation restructures the quality and quantity of DOM in seasonally formed lakes. Firstly, the higher DOC concentrations observed in sub-lakes reveal more organic matters were existing in the water column due to hydrological isolations. Secondly, the relatively high proportions of S-containing components in water DOM of sub-lakes suggests the hydrological isolation may facilitate the release of sulfur (sulfide/bisulfide) from sediments. This could indicate a potential pathway for the sulfur cycle between water column and sediments in aquatic systems, and another study had reported sulfur additions could change the optical properties of DOM (Powers et al., 2021). Thirdly, different degradation processes occurred in main lake and sub-lakes and had great impacts on DOM compounds in terms of their photo-reactivity (i.e., aromatic substances) and bioreactivity (i.e., aliphatics). This must affect the migration and transformation of DOM and its environmental behaviors in aquatic systems, for instance, the organic matter burial (Wang et al., 2021). Moreover, previous studies have proposed that the intensity and frequency of extreme weather and flooding are increasing globally (Lee et al., 2017; Pang et al., 2021), which could potentially lead to higher chances of hydrologically isolated lakes in large river-connected systems, and could have a crucial impact on the regional DOM biogeochemistry.

In the natural environment, the chemistry of DOM in lakes is affected by various factors. Here, it is important to realize that the number of samples was limited for each group (the main lake and isolated lakes). However, the results within each group agree with each other well such that the optical properties and molecular characteristics of DOM were very close between both the two main lake sites and between the two sub-lake sites. Therefore, limited samples still revealed large differences in optical properties and molecular characteristics of DOM between main lake and sub-lakes, which provides a novel insight into the impacts of hydrological isolations on the DOM chemistry in river-connected lake ecosystems. For a better understanding of the regional DOM biogeochemistry, more future investigations should be carried out. Firstly, this work mainly focused on the differences of DOM molecular characteristics in surface water, the vertical space resolution (depth profile) of DOM chemistry should be taken into account; secondly, the effects of seasonal changes, as the main feature of river-connected lakes, on DOM chemistry in main lake and sub-lakes should be fully explored.

4. Conclusions

The optical properties and molecular composition of DOM in a large river-connected lake (Poyang Lake, China) were characterized using UV–Vis and FT ICR MS analysis to comprehensively explore the influences of hydrological isolation on DOM chemistry. Significant differences in DOM chemistry were observed between the main lake and sub-lakes, DOC concentrations and UV–Vis properties have shown the same patterns that higher abundance of organic matter existed in sub-lakes, and relative more refractory components of DOM with higher aromaticity and molecular weight in the main lake than sub-lakes, which were evidenced by a variety of parameters in UV–Vis and molecular compounds in FT ICR MS. These significant variations in DOM chemistry between main lake and sub-lakes were mainly attributed to the biodegradation processes in the main lake while photodegradation processes in sub-lakes. And sediments greatly facilitated the formation of CHOSs in sub-lakes DOM as well. In particular, the DOM characteristics in the two main lake samples were really similar, demonstrating that hydrological homogeneity existed within the certain region of large lakes. Further investigations should focus on the variations of depth profile and seasonal changes of DOM chemistry in main lake and sub-lakes, which would better improve the understanding of the biogeochemistry of DOM and the global carbon cycle processes in large aquatic systems.

CRediT authorship contribution statement

Lei Xu: Investigation, Methodology, Data curation, Writing – review & editing. **Qian Hu:** Data curation, Writing – review & editing. **Libing Liao:** Writing – review & editing. **Zhongxin Duan:** Investigation. **Songping Liu:** Investigation. **Ludan Chen:** Data curation. **Qiuping Zhu:** Data curation. **Aiwen Zhong:** Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests that could have appeared to influence the work reported in this paper.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.158047.

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