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Direct discharge of sewage to natural water through illicitly connected urban stormwater systems: An overlooked source of dissolved organic matter

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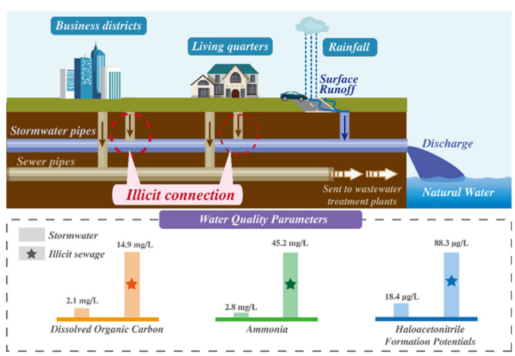
HIGHLIGHTS

- DOM characteristics from stormwater drainage systems were firstly investigated.
- Pipe illicit connections introduced more contents of DOC and DON to stormwater pipes.
- Pipe illicit connections significantly contributed HAL and HAN precursors to stormwater pipes.
- Pipe illicit connections increased tyrosine-like and tryptophan-like aromatic proteins.

GRAPHICAL ABSTRACT



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ABSTRACT

The illicit connection of sewage pipes to stormwater pipes commonly occurs in urban stormwater systems. This brings problems that sewage might be directly discharges into natural water and even drinking water sources without treatment, posing risks to ecological safety. Sewage contains various unknown dissolved organic matter (DOM), which could react with disinfectants and lead to the formation of carcinogenic disinfection byproducts (DBPs). Thus, understanding the impacts of illicit connections on downstream water quality is of significance. This study firstly investigated the characteristics of DOM using fluorescence spectroscopy and the formation of DBPs after chlorination in an urban stormwater drainage system in the case of illicit connections. The results found that the concentrations of dissolved organic carbon and dissolved organic nitrogen ranged from 2.6 to 14.9 mg/L and from 1.8 to 12.6 mg/L, respectively, with the highest levels occurring at the illicit connection points. Concerning DBP precursors, pipe illicit connections introduced considerable precursors of highly toxic haloacetaldehydes and haloacetonitriles into the stormwater pipes. Furthermore, illicit connections introduced more contents of tyrosine-like and tryptophan-like aromatic proteins, which may be related to foods, nutrients, personal care products, etc. in the untreated sewage. This indicated that the urban stormwater drainage system was a significant input source of DOM and DBP precursors to natural water. The results of this study are of great significance for protecting the security of water sources and promoting the sustainability of urban water environment.

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1. Introduction

Dissolved organic matter (DOM), commonly defined as substances that can passthrough the 0.45 μm pore filter (Wei et al., 2008), is a complex mixture of organic compounds originated from both autochthonous and allochthonous sources (Meng et al., 2013; Wen et al., 2022). Autochthonous DOM is mainly generated in the water body via microbial and algal activities, and includes carbohydrates, amino acids, peptides, enzymes, and toxins (Sillanpää et al., 2018; Xiao and Chu, 2020). Allochthonous DOM originates from terrestrial organisms, and reaches the water sources through drainage within watersheds, direct discharge of effluents from urban wastewater treatment plants, indirect leaching into groundwater and aerial dispersal (Hudson et al., 2007). The composition of allochthonous DOM is mainly humic character (Fabris et al., 2008; Sillanpää et al., 2018). DOM has received considerable attention due to its impact on water quality and treatment efficiency. On the one hand, DOM can cause aesthetic concerns such as color, taste, and odor issues (Leenheer and Croué, 2003). On the other hand, DOM may increase the demand of chemical reagents (e.g., coagulants, oxidants, and disinfectants), lead to the membrane fouling, and produce hazardous disinfection byproducts (DBPs) during disinfection process (Chu et al., 2015a; Meng et al., 2013; Phungsai et al., 2018).

In recent years, DOM has been drawing increasing concerns because it acts as the main precursors of hazardous DBPs during chlorination (Chu et al., 2015a; Yang et al., 2023; Zhang et al., 2012). DBPs are formed when chemical disinfectants react with DOM, anthropogenic contaminants, bromide, and iodide, and have become ubiquitous environmental contaminants of emerging concern (Chu et al., 2017; Richardson et al., 2007; Xiao et al., 2022). DBPs have been frequently detected in chlorinated drinking water (Chu et al., 2016b; Li and Mitch, 2018; Wagner and Plewa, 2017). So far, more than 700 DBPs have been identified, and a large number of DBPs have been reported to be cytotoxic, neurotoxic, genotoxic, carcinogenic, teratogenic, and mutagenic (Wagner and Plewa, 2017). Epidemiological studies demonstrated that long-term exposure to DBPs is associated with bladder cancer and other adverse health risks (Li and Mitch, 2018; Plewa and Wagner, 2015). Reducing DBP precursors before disinfection is an effective method to control DBPs. Previous research about the source of DBP precursors focused on autochthonous DOM (e.g., amino acids) or terrestrial organics (e.g., humic substances) (Shah and Mitch, 2012). In fact, DOM in urban stormwater system is originated from a wide variety of sources, including surface runoff, roof erosion, and mixed sewage. Studies have proven that the loads and compositions of DOM in urbanized streams are highly influenced by stormwater discharge (Meng et al., 2013; Wen et al., 2022; Xu et al., 2019b).

Generally, urban drainage systems are composed of urban drainage pipes, pumping stations, and wastewater treatment plants. The urban drainage pipes play the role of transporting sewage (including domestic sewage and pretreated industrial wastewater) and rainwater (Cun et al., 2019; Wang et al., 2021). At present, urban drainage systems in China are divided into two system types based on their construction: combined sewer systems and separate sewer systems (Wang et al., 2021). However, unsynchronized construction of urban drainage systems causes the illegal connection of sewage pipes to stormwater pipes in developing counties (Li et al., 2014), resulting in direct discharge of sewage into natural waters and even into drinking water sources (Xu et al., 2019b; Xu et al., 2014). It was reported in the literature that 26 % of the total amounts of sewage entered stormwater pipes in the southeast of China (Xu et al., 2019a). Moreover, a previous study investigated the conditions of stormwater systems and found that more than half of the 23 stormwater systems existed sewage pipes illicit connection problems (Xu et al., 2019b). In 2018, about 13,000 illicit connection points were found in the 13,000 km pipes in Shanghai (Xu et al., 2021). Therefore, sewage could be easily discharged into natural water without treatment. However, the management of drainage system in developing countries has paid much attention to construction while ignoring maintenance, leading to the issues that pipe illicit connections cannot be repaired timely (Xu et al., 2021). As a result, sewage reaches the aquatic environment through urban stormwater pipes, posing risks to ecological safety and resulting in black and odorous water bodies (Regier et al., 2020; Yin et al., 2017).

Thus, urban stormwater pipes have become the input sources of contaminants (DOM and hazardous DBPs) to natural water due to illicit connection of sewage pipes. Previous studies have focused on the diagnosis of pipe illicit connections (Xu et al., 2021), the location of illicit discharges (Hachad et al., 2022), the runoff quality dynamics (Regier et al., 2020), the nitrogen transport and sources in urban stormwater (Wang et al., 2022) and the management of urban stormwater (Marazuela et al., 2022; Rentachintala et al., 2022). However, little is known about the impacts of illicit connection on the characteristics of DOM and the output level of DOM in urban stormwater systems.

The objectives of this study were to 1) provide a general understanding concerning the characteristics of DOM in the urban stormwater pipes, 2) evaluate the formation potentials of DBPs along the urban stormwater pipes, and 3) explore the impact of illicit connections on DOM and DBP precursors in the urban stormwater system. The purpose of this study was to provide useful information about the impacts of illicit connections on the downstream water quality. The results presented here were of great significance for protecting the security of water sources and promoting the sustainability of water environment.

2. Materials and methods

2.1. Chemicals and regents

The standards for trihalomethanes (THMs), haloacetaldehydes (HALs), haloacetonitriles (HANs), halonitromethanes (HNMs), and chlorophenylacetonitriles (CPANs) with the highest purity available were purchased from Sigma-Aldrich (St Louis, Missouri, USA), CanSyn Chemical Corp. (Canada), and Aladdin Industrial Inc. (Shanghai, China). The detailed DBP information is available in Table S1. Sodium hypochlorite (4–5 % active chlorine) and phosphate were purchased from Sigma-Aldrich. Ultrapure water used in this study was produced from a Millipore Milli-Q Gradient water purification system (18 MΩ·cm, Billerica, MA, USA). All other chemicals and reagents were of at least analytical grade and supplied by Sinopharm Chemical Reagent (Shanghai, China), and all tests were undertaken in triplicate. Error bars in figures reflected the standard deviations of triplicates. The abbreviations used in this study were shown in Table 1.

2.2. Sample collection

Water samples were collected from the inspection well of an urban stormwater drainage system in Maanshan city, Anhui province, China. This city was located at the downstream of the Yangtze River, with well-developed hydrological conditions. Furthermore, Maanshan was a representative city that had been reported to have pipe illicit connections (Xu et al., 2021). Although hydrological seasonality had impacts on DOM, one of the most important purposes of this study was to explore the influence of pipe illicit connections on the characteristic and component of DOM. In order to maintain the same experimental conditions, 19 water samples were collected on the same day of October 2021. The sampling day had been sunny for 14 days since the last heavy rainfall. The detailed sampling information was depicted in Table S2 and Fig. 1. Upon collection, 19 water samples were transported to the laboratory and filtered through a 0.45 μm glass fiber filter and stored at 4 °C in the dark until use. As shown in Text S1, the concentrations of dissolved organic carbon (DOC), dissolved organic nitrogen (DON), dissolved inorganic nitrogen (i.e. ammonia, nitrate, and nitrite), ultraviolet absorbance at 254 nm (UV_{254}), and specific ultraviolet absorption (SUVA) were determined immediately under the same experimental condition. Detailed water quality parameters of the water samples were provided in Table S3. Experimental design of this study was provided in Fig. S1.

Table 1
The abbreviations in this study (according to the order in the text).

Abbreviation	Full name
DOM	Dissolved organic matter
DBPs	Disinfection byproducts
THMs	Trihalomethanes
HALs	Haloacetaldehydes
HANs	Haloacetonitriles
HNMs	Halonitromethanes
CPANs	Chlorophenylacetonitriles
DOC	Dissolved organic carbon
DON	Dissolved organic nitrogen
UV254	Ultraviolet absorbance at 254 nm
SUVA	Specific ultraviolet absorption
FP	Formation potential
SEC-OCD	Size-Exclusion Chromatography-Organic Carbon Detector
C-DBPs	Carbonaceous DBPs
TCM	Trichloromethane
BDCM	Bromodichloromethane
DBCM	Dibromochloromethane
TBM	Tribromomethane
DCAL	Dichloroacetaldehyde
TCAL	Trichloroacetaldehyde
TBAL	Tribromoacetaldehyde
N-DBPs	Nitrogenous DBPs
DCAN	Dichloroacetonitrile
TCAN	Trichloroacetonitrile
BCAN	Bromochloroacetonitrile
DBAN	Dibromoacetonitrile
TCNM	Trichloronitromethane
2-CPAN	2-chlorophenylacetonitrile
3-CPAN	3-chlorophenylacetonitrile
4-CPAN	4-chlorophenylacetonitrile
2,3-DCPAN	2,3-dichlorophenylacetonitrile
2,4-DCPAN	2,4-dichlorophenylacetonitrile
2,5-DCPAN	2,5-dichlorophenylacetonitrile
2,6-DCPAN	2,6-dichlorophenylacetonitrile
3,4-DCPAN	3,4-dichlorophenylacetonitrile
CTI	Cytotoxicity index
CHO	Chinese hamster ovary
BIF	Bromine incorporation factor
EEM	Excitation-Emission Matrix
FRI	Fluorescence regional integration
LMW	Low-Molecular Weight

2.3. Formation potential (FP) experiments

DBP FP experiments were conducted in sealed 40 mL amber glass bottles under headspace-free conditions based on the methods developed in previous studies (Zhang et al., 2019c). Free chlorine condition was achieved by adding sufficient chlorine to break free any ammonia. The chlorine dosages for FP tests were calculated based on Eq. (1), which was developed by Krasner et al. (2007), and was commonly used to determine DBP precursors (Chu et al., 2016a; Chu et al., 2010; Zhang et al., 2021a).

$$Cl_2\text{dosage}(\text{mg/L}) = 3 \times \text{DOC}(\text{mg}-\text{C/L}) + 7.6 \times NH_3 - \text{N}(\text{mg}-\text{N/L}) + 10\ (\text{mg/L}) \quad (1)$$

After chlorination for 24 h, water samples were taken for analysis of THMs, HALs, HANs, and HNMs. Similarly, 500 mL water samples were prepared in sealed 500 mL amber glass bottles for the analysis of CPANs (Zhang et al., 2021a). All prepared stormwater samples were kept in the dark at a 25 °C thermostatic incubator for 24 h after the chlorination. After the reaction, DBPs were extracted by adding 2 mL methyl tert-butyl ether to a 10 mL chlorinated sample. Then, this sample was shaken for 5 min using a multi-tube vortex mixer (DMT-2500, Shanghai, China) at 2300 rpm. Afterwards, the extracts were analyzed immediately to determine THMs, HALs, HANs, and HNMs. In addition, the determination of CPANs was conducted by a solid-phase extraction preconcentration procedure according to the previous work (Zhang et al., 2019b).

2.4. Analytical methods

2.4.1. Size-exclusion chromatography-organic carbon detector (SEC-OCD)

The molecular weight distribution of DOM in the water samples from the stormwater system was obtained by size-exclusion chromatography with an organic carbon detector (SEC-OCD). SEC was carried out using a polymethacrylate-packed column (250 mm × 20 mm, 3 μm), and installed on a Waters Alliance high performance liquid chromatography (HPLC, e2695, MA, USA) equipped with a Sievers M9 SEC organic carbon detector. Mobile phase was prepared with 5.6 mM disodium hydrogen phosphate and 14.4 mM sodium dihydrogen phosphate, and then was delivered with a HPLC pump at a flow rate of 1.0 mL/min to an autosampler (MLE, Dresden, Germany), and the injection volume was 250 μL. More information about SEC-OCD is available in a previous study (Zhang et al., 2021a). According to the retention time, DOM in these water samples could be divided into five major fractions, including biopolymers, humic substances, building blocks, low molecular weight (LMW) acids, and LMW-neutrals (Huber et al., 2011).

2.4.2. Analysis of DBPs

Carbonaceous DBPs (C-DBPs), including trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), tribromomethane (TBM), dichloroacetaldehyde (DCAL), trichloroacetaldehyde (TCAL), as well as tribromoacetaldehyde (TBAL), and nitrogenous DBPs (N-DBPs), including dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN), dibromoacetonitrile (DBAN), and trichloronitromethane (TCNM) were detected using gas chromatography/electron capture detection (QP2010plus, Shimadzu Corporation, Japan). Gas chromatography/mass spectrometry (QP2020, Shimadzu, Japan) operated in the electron ionization (EI) mode was used with an RTX-5MS (30 m × 0.25 mm ID, 0.25 μm film thickness) column for the analysis of CPANs, including 2-chlorophenylacetonitrile (2-CPAN), 3-chlorophenylacetonitrile (3-CPAN), 4-chlorophenylacetonitrile (4-CPAN), 2,3-dichlorophenylacetonitrile (2,3-DCPAN), 2,4-dichlorophenylacetonitrile (2,4-DCPAN), 2,5-dichlorophenylacetonitrile (2,5-DCPAN), 2,6-dichlorophenylacetonitrile (2,6-DCPAN), and 3,4-dichlorophenylacetonitrile (3,4-DCPAN). The details of these DBP analytical methods were available in previous studies (Zhang et al., 2019b; Zhang et al., 2021b).

2.4.3. Calculation of DBP-associated cytotoxicity

Cytotoxicity index (CTI) calculated by Eq. (2), was used to assess DBP-associated cytotoxicity. The cytotoxicity values corresponding to each DBP were derived from the in vitro toxicity test using Chinese hamster ovary (CHO) cells (Plewa et al., 2008; Wagner and Plewa, 2017; Wei et al., 2020).

$$CTI = \Sigma\left[\frac{1}{\%C\frac{1}{2}_x} \times C_x\right] \quad (2)$$

where the %C½ value (M^{-1}) is the molar concentration that induced a 50 % reduction of the CHO cell density compared to the control, C_x is the average molar concentration of each DBP (nM). The %C½ value of each DBP is present in Table S5.

2.4.4. Calculation of bromine incorporation factor (BIF)

BIF indicates the incorporation of bromine ion into organics, and it has been applied by many researchers to evaluate the bromine substitution of DBPs (Chellam and Krasner, 2001; Chu et al., 2014; Fang et al., 2018; Zhang et al., 2019a). The detailed formula for calculating the BIF values was presented in Text S2.

2.4.5. Fluorescence excitation-emission matrix (EEM)

EEM fluorescence spectroscopy has been widely used to characterize DOM in water (Chen et al., 2003). EEM measurement of the 19 water samples from the stormwater system was performed using a three-dimensional

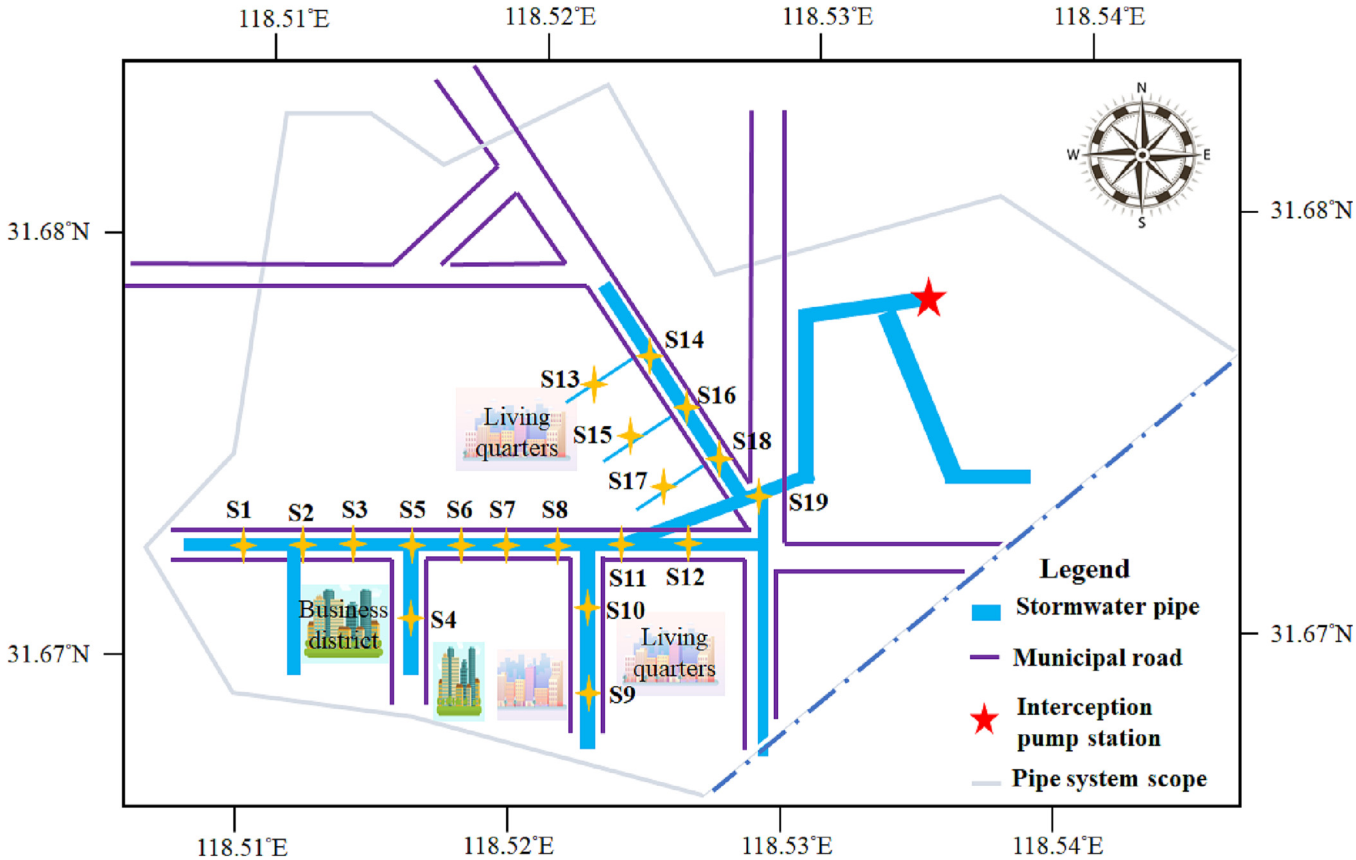

Fig. 1. Map of study area and sampling sites.

spectrofluorometer (F-7100 Fluorescence, HITACHI, Tokyo, Japan). In order to avoid possible detection errors, water samples were filtered through a 0.45 μm membrane and diluted (UV absorbance <0.05 cm^{-1} at 254 nm) to avoid inner-filtering effects prior to EEM testing (Yang and Hur, 2014). Excitation wavelengths in the range of 200–500 nm with 5 nm intervals and emission wavelengths in the range of 250–550 nm with 5 nm intervals were used, and the scan speeding was 12,000 nm/min. Blank EEM spectra of deionized water was also analyzed prior to sample analysis. The obtained EEM data were corrected and standardized based on a previous study (Jutaporn et al., 2021). The final fluorescent intensity was expressed in Raman Units (R.U.). According to Chen et al. (2003), EEM spectra were divided into five excitation-emission regions. The detailed information about the five regions was presented in Text S3. According to the five typical regions, fluorescence regional integration (FRI) of the water samples was calculated using MATLAB R2019a.

3. Results and discussion

3.1. DOM from the urban stormwater drainage system

3.1.1. Water quality parameters

The 19 water samples were collected on sunny days from the inspection wells, but significant water flow was observed in the inspection wells at S4, S9, and S10. The three points were presumed to be illicit connection points, but further data confirmation was needed. Meanwhile, it could be seen from Fig. 2c that much higher concentrations of ammonia were found at S4 (25.5 mg/L), S9 (45.3 mg/L), and S10 (44.7 mg/L) compared with S3 (0.04 mg/L) and S8 (2.8 mg/L). Usually, ammonia level in rainwater was a few tens to several mg/L, but the ammonia level in sewage was particularly high, reaching tens even hundreds of mg/L (He et al., 2020; Hou et al., 2018; Hu et al., 2023). For instance, the ammonia value of S1-S3 was 0.04, 0.03, and 0.04 mg/L, respectively. This was comparable to the value of ammonia in rainwater (0.03–0.88 mg/L) reported by Hou et al. (2018), while the ammonia value in sewage was reported to be 46.6 mg/L (Hu et al., 2023). Thus, S4, S9, and S10 could be determined to be illicit connection points of sewage pipes to stormwater pipes. In general, DOC concentrations of 19 water samples were in the range of 2.6–14.9 mg/L (Fig. 2a). The elevated DOC levels of S5 (5.3 mg/L) compared with S3 (3.1 mg/L), as well as the surged DOC levels of S11 (12.7 mg/L) compared with S8 (3.1 mg/L) can be explained by illicit connection of sewage pipes to stormwater pipes from the business districts (S4, 10.1 mg/L) and the living quarters (S9, 14.9 mg/L; S10, 14.1 mg/L). Besides, there was a good linear relationship between DOC and UV_{254} for all water samples (Fig. S2, $UV_{254} = 0.019 \times$ DOC-0.097, $R^2 = 0.97$). This indicate that DOM in the 19 water samples were mainly aromatic organic matter. As is shown in Fig. 3b, DON concentrations ranged from 1.8 to 12.6 mg/L. Similarly, it could be found that the DON levels in the water samples at the illicit connection points, S4 with 8.3 mg/L, S9 with 10.7 mg/L, and S10 with 12.6 mg/L, were extremely high. Although there was dilution with the flow of water, it still resulted in a higher DON level for S11 (8.9 mg/L) than S8 (2.3 mg/L) due to the illicit connections. Even at the end sample point of the stormwater drainage system (S19), which was the last site before stormwater entering receiving waters, the DON level (7.3 mg/L) was 10 times higher than the source water of Lake Tai (average concentration was 0.7 mg/L) (Zhang et al., 2021b), and was 2–20 times higher than the Yangtze River (0.26 to 3.23 mg/L) (Fang et al., 2021). Given that the sampling sites in this study were residential and commercial areas, it was speculated that the higher concentrations of DON observed in this study may be related to the sewage quality in this area, which contained more organic nitrogenous substances as a result of human activities. In addition, sprinklers would flush DON out of the soil, causing DON to leach from particles in the catchment. A previous study has reported that DON accounted for an average of up to 82 % of TN loads in the stormwater samples over the study period (Jani et al., 2020).

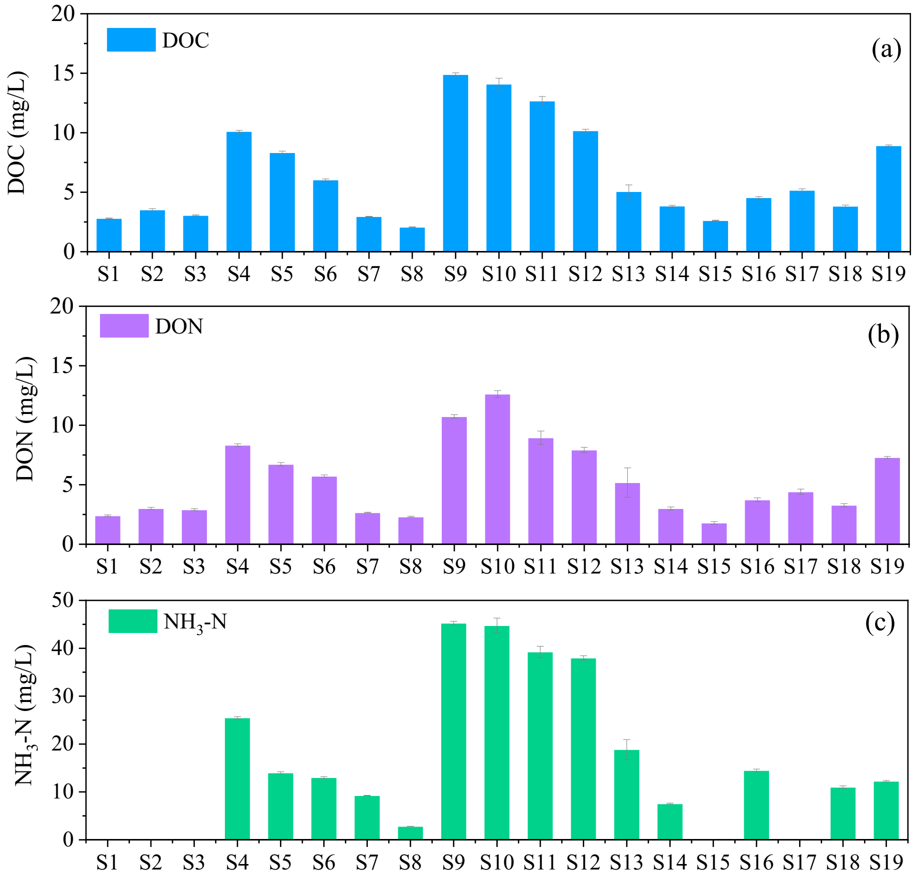

Fig. 2. (a) DOC, (b) DON, and (c) NH_3-N levels of water samples from the urban storm water drainage system.

This indicated that the direct discharge of stormwater would affect the quality of the receiving water body. Regier et al. (2020) has also reported that urban stormwater has impacts on water quality dynamics of downstream ecosystems.

3.1.2. Molecular weight distribution

Determining the molecular weight distribution of DOM was beneficial to identify key DBP precursors. SEC-OCD was commonly used to analyze the molecular weight distribution of DOM (Fang et al., 2021; Ren et al., 2022; Zhang et al., 2021a). Thus, the molecular weight distribution at illicit connection points and the adjacent points was also analyzed. Fig. 3a and c show SEC-OCD chromatogram of water samples with response for OCD. Fig. 3b and d illustrate the proportions of five fractions in the molecular weight distribution of the selected water samples.

As depicted in Fig. 3a, five peaks were observed in all the chromatograms, and there was little difference between S3, S4, and S5 in the five responses, except for a slightly higher response in building blocks and LMW-neutrals at S5 compared with S3. From Fig. 4b, compared with S3, a higher proportion of biopolymers, building blocks, and LMW-neutrals could be found at S4 in SEC-OCD, indicating that there was input of low-molecular-weight DOM and high-molecular-weight DOM at S4 resulting from the illicit connections of sewage pipes and stormwater pipes in business districts. The decrease of biopolymer fractions at S5 compared with S3 implied the possible degradation of biopolymers in the stormwater pipes. Building blocks reflect humic substances-like material of lower molecular weight DOM, which contains the bulk of DBP precursors (Bond et al., 2009). The proportion of this fraction increased at S4, which might play a role in DBP formation during chlorination. Furthermore, the properties of LMW-neutrals point to LMW alcohols, aldehydes, ketones, sugars, as well as amino acids (Huber et al., 2011). These are commonly known as typical DBP precursors (Chu et al., 2015b; Chu et al., 2016c; Chu et al., 2015c).

Importantly, higher response of biopolymer fractions was observed at S9, S10, and S11 compared to S8 (Fig. 3c), and the proportion of biopolymer fractions at S9, S10, and S11 was higher than that at S8 (Fig. 3d). Biopolymers with the molecular weight of 10 kDa or higher nitrogen-containing materials such as proteins, amino sugars, and polysaccharides (Huber et al., 2011). These substances are acknowledged to be important N-DBP precursors (Fang et al., 2019; Zhang et al., 2021a). In addition, the DOM of water samples from the urban stormwater drainage system was dominated by humic substances and LMW-neutrals. Overall, pipe illicit connections enhanced the content of biopolymers such as proteins, amino sugars, and polysaccharides, which were important DBP precursors. In order to identify the key DBP precursors, it was necessary to explore the DBP precursor levels and the impact of pipe illicit connections on DOM and DBP precursors in the urban stormwater drainage system.

3.2. Formation potentials of DBPs from the urban stormwater drainage system

3.2.1. Formation potentials (FPs) of DBPs

Fig. 4 provided the concentration of DBPs formed after chlorination of 19 water samples collected from the urban stormwater drainage system. THM precursors widely occurred in all water samples, as FP tests showed that the total THM FPs were in the range of 63.5–695.4 μg/L, with TCM as the dominant class ranging from 54.6 to 690.5 μg/L. This was much higher than the standard limit of 60 μg/L for TCM regulated by specific projects of centralized surface water source for drinking water in Environmental Quality Standards for Surface Water in China (GB3838-2002, PR, 2002). As for HAL FPs, the total concentrations ranged from 28.6 to 181.3 μg/L, with TCAL possess a higher concentration in its analogs. Obviously, the highest HAL concentration appeared at S9, which was a branch of the stormwater pipe in living quarters. Overall, at S19, the end sample point of the stormwater pipelines in this study, THM and HAL FPs were 121.1

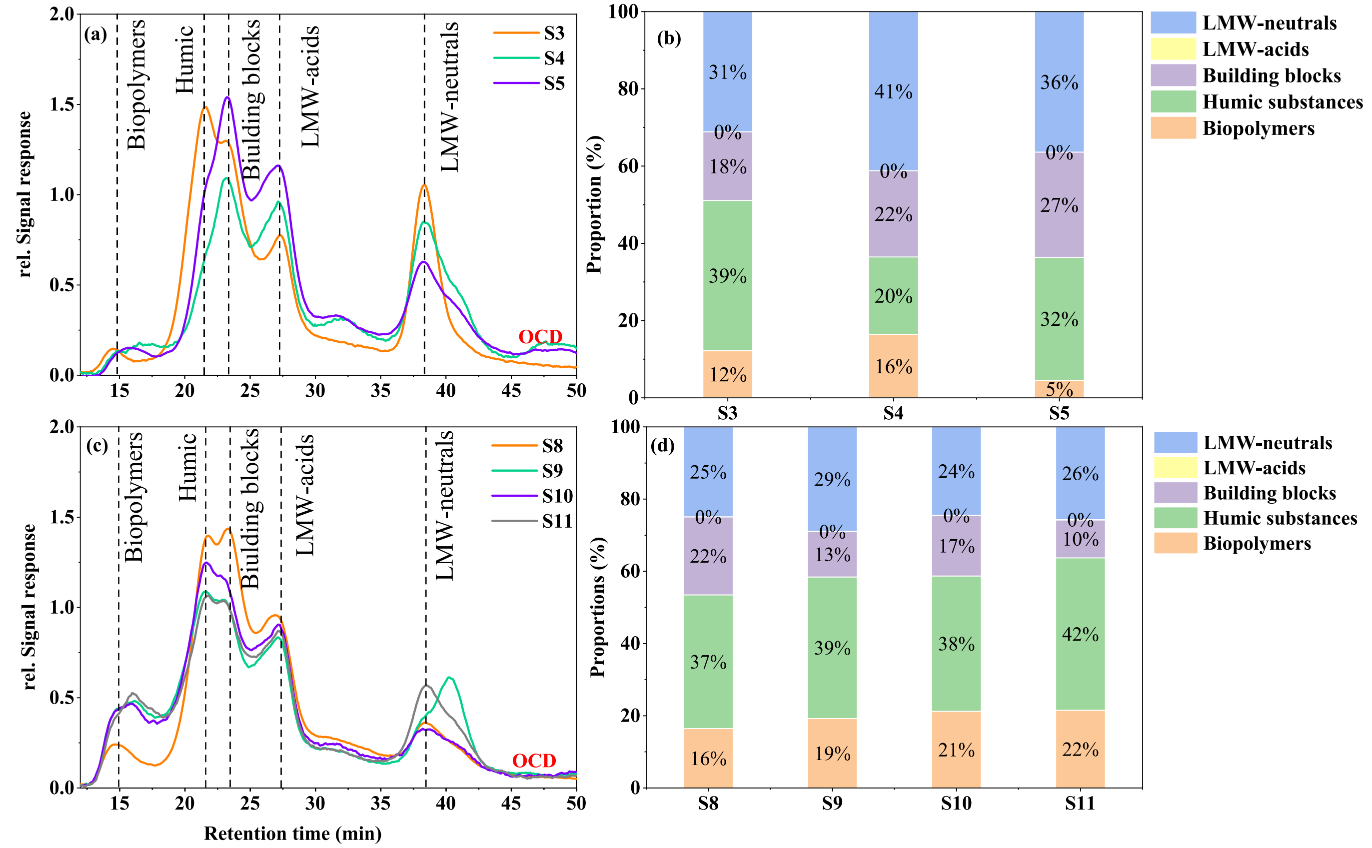

Fig. 3. SEC-OCD chromatogram of water samples with response for OCD at the illicit connection points and their adjacent points from the urban storm water drainage system. LMW = Low-Molecular Weight.

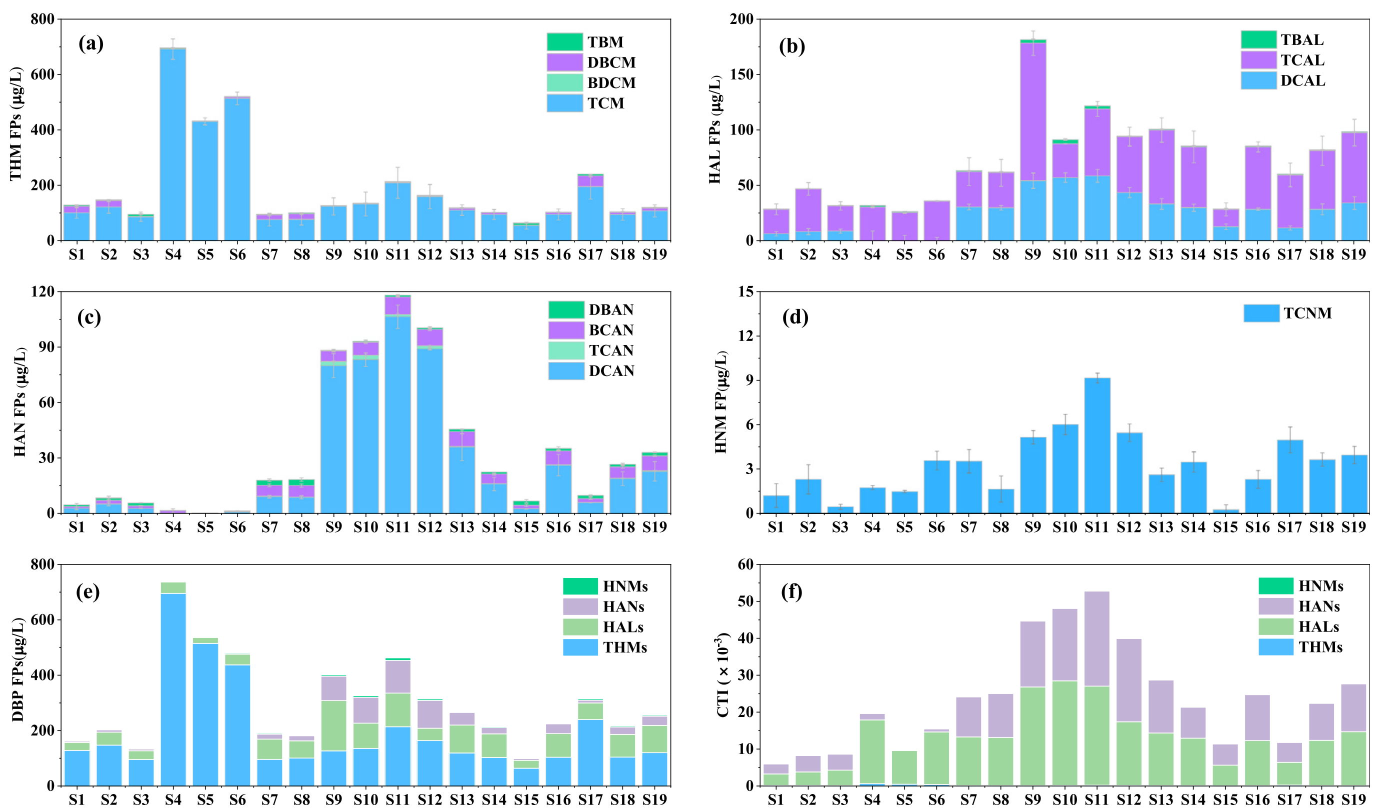

Fig. 4. (a) Formation potentials (FPs) of THMs, (b) FPs of HALs, (c) FPs of HANs, (d) FP of HNM, (e) FPs of total DBPs, and (f) DBP-associated cytotoxicity index (CTI) after 24 h chlorination of the 19 water samples. THMs = trihalomethanes; HALs = haloacetaldehydes; HANs = haloacetonitriles; HNMs = halonitromethanes; DBPs = disinfection by products.

and 98.4 μg/L, respectively, which were approximately 1.2 and 8.9 times higher than the reported THM and HAL FPs in source water of Lake Tai (THM FPs was 98.1 μg/L, HAL FPs was 11.0 μg/L) (Zhang et al., 2021b).

Moreover, FP tests (Fig. 4c) showed that the total concentrations of HANs ranged from 0.2 to 118.1 μg/L, with DCAN as the dominant class. The highest HAN FPs was at S11 (118.1 μg/L), which was the subsequent rendezvous point of stormwater from the living quarters. Domestic sewage contains pharmaceuticals and personal care products, faeces, biopolymers, and amino acids. Without treatment, these waters feature higher organic nitrogen content and can serve as important sources of N-DBP precursors (Ding et al., 2018; Shah and Mitch, 2012). Meanwhile, the highest DCAN FP was also appeared at S11 (106.5 μg/L), this was well above than the provisional guideline value of DCAN (20 μg/L) specified in World Health Organization (WHO) guidelines for drinking water (WHO, 2011). Besides, the level of TCNM was in the range of 0.5–9.2 μg/L, and the highest TCNM FP was also at S11, followed by S10 and S9 (6.0 and 5.2 μg/L, respectively). Overall, higher levels of DBP FPs were found at S9, S10 and S11. Importantly, at the end point of the pipelines (S19), HAN and TCNM levels were 33.1 and 4.0 μg/L, respectively. However, a previous study reported that the concentration of HANs ranged from 2.6 to 6.0 μg/L and the average concentration of TCNM was 1.1 μg/L in finished waters from drinking water treatment plants (Zhang et al., 2021b). This showed that the level of HANs at this point, where waters would enter the receiving water body directly, was 5 to 12 times higher than the reported HANs in finished waters from drinking water treatment plants. Notably, HANs was reported to be the toxicity driver in drinking water and suggested to be prioritized in future research for potential regulation consideration (Allen et al., 2022; Zhang et al., 2021b). Thus, it was quite important to identify the key DBPs as well as its precursors that contributed to the formation of DBPs in urban stormwater drainage system.

3.2.2. DBP-associated cytotoxicity index (CTI)

As depicted in Fig. 4e, the total DBP levels ranged from 99.2 to 740.2 μg/L along the urban stormwater drainage system. The total DBP FPs was 255.5 μg/L at the end sampling point (S19) of the stormwater drainage system in this study, which was higher than that in upstream (137.4 μg/L), midstream (196.1 μg/L), and downstream (208.4 μg/L) waters along the Yangtze River (Fang et al., 2021). For most of the 19 chlorinated water samples, THMs was the dominant DBP classes, accounting for 50 % ~ 95 % of the total DBP concentrations of all measured individual DBPs. However, the calculated DBP-associated CTI indicated that HANs and HALs made great contributions to CTI, rather than THMs (Fig. 4f). Therefore, HANs and HALs were key DBPs and therefore need more attention.

Furthermore, during data exploration, it was found that DOM characteristics have relationships with HANs and CTI for all water samples. A recent study suggested that DOC has good correlations with C-DBPs (Golea et al., 2017), while also in this study a positive correlation between DOC and HANs was found. As is illustrated in Figs. S3a-S3b, DOC has correlations with HAN FPs ($R^2 = 0.62$) and CTI ($R^2 = 0.65$). In addition, as HAN made great contribution to CTI, the good correlation between DON and HAN FPs ($R^2 = 0.67$) also resulted in positive correlation between DON and CTI ($R^2 = 0.67$).

Notably, the CTI value at S11(53.2) was 2 times higher than that at S8 (24.9) and 8 times higher than that at S1 (6.0), which may be due to the contribution of S4, S9, and S10 derived from the stormwater in business districts and living quarters. At S19, the CTI value (27.7) was approximately 10 times higher than that in finished water samples from drinking water treatment plants (Zhang et al., 2021b). Overall, water samples from business districts and residential areas appear to contribute more to DBP FPs and CTI. This indicated that it was time to focus on the direct discharge of stormwater in case of illicit connections. In addition, the impact of pipe illicit connections needs to be further investigated.

3.3. Impact of pipe illicit connections on DBP FPs

As depicted in Fig. 5a, there was a significant rise of THMs at S5 (514.7 μg/L) compared with S3 (95.2 μg/L). This resulted from the illicit

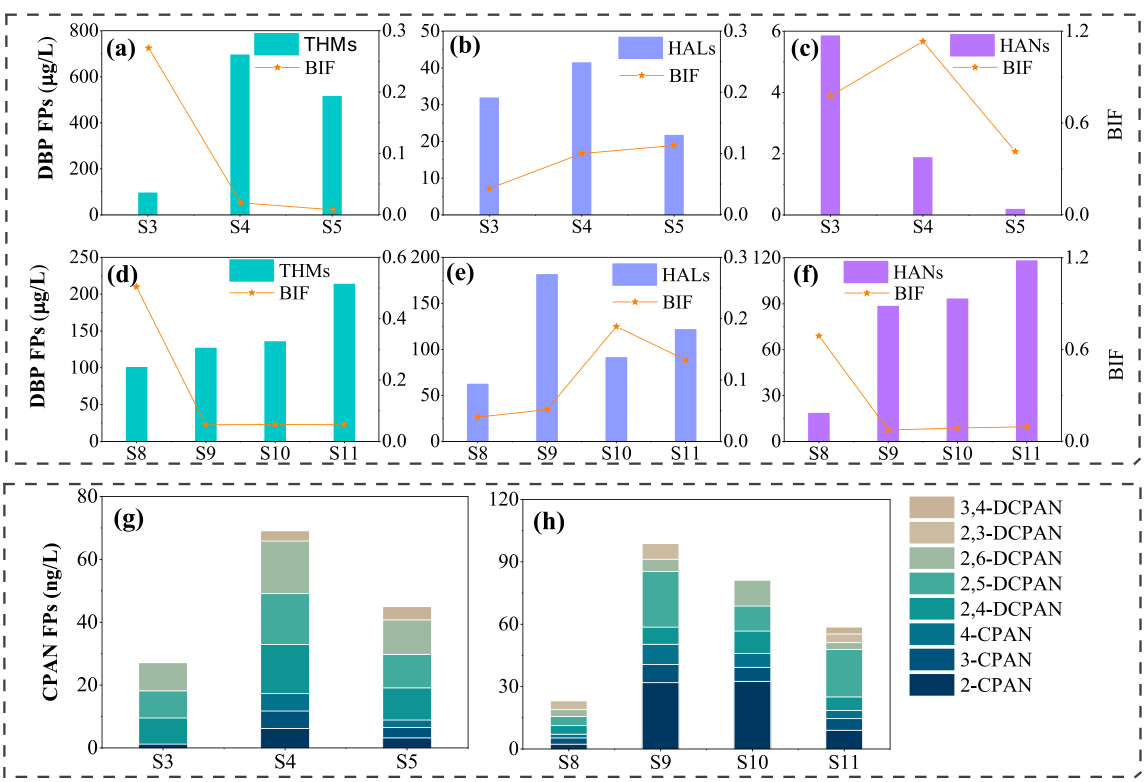

Fig. 5. (a, d) THMs, (b, e) HALs, (c, f) HANs, and (g-h) CPAN FPs and bromine incorporation factor (BIF) at the illicit connection points and their adjacent points from the urban storm water drainage system.

connections of sewage pipes to stormwater pipes from business districts (S4, 695.4 μg/L). However, the bromine incorporation factor (BIF) value of THMs was greatly decreased at S4, which suggested that the sewage in business districts mainly contributed to the formation of chlorinated THMs. Additionally, compared with S8, the introduction of domestic sewage at S9 and S10 from living quarters increased the formation of THM, HAL, and HAN at S11 (Fig. 5d-f). The increased DBP FPs at S9, S10, and S11 was possibly related to the higher responses of biopolymer (Fig. 3c). It was noteworthy that the high levels of HALs and HANs at S9, S10, and S11 resulted in the high values of CTI (Fig. 4f). Besides, the BIF value of HALs at S9 and S10 was higher than that of S8, which showed that incorporation of bromine into HALs at S9 and S10 was much easier. These results indicated that the domestic sewage from living quarters was the main contributor to the formation of HALs and HANs, which were identified as key DBPs in the urban stormwater drainage system. Interestingly, it was found that THM and HAL FPs increased at the illicit connection point of S4, while at S9 and S10, the FPs of THMs, HALs, and HANs increased. This indicated that domestic sewage in living quarters contains more HAN precursors. Notably, various unknown organic matters, chemicals, as well as DBPs may enter natural water through urban stormwater drainage system. This might explain the severe pollution and deterioration of water quality in receiving waters during rainy day (Xu et al., 2019b).

Furthermore, the FPs of CPANs, another emerging aromatic N-DBP class, were also determined at the illicit connection points and their adjacent points. A previous study has reported that the cytotoxicity of CPANs was approximately one order of magnitude more toxic compared with their aliphatic counterparts like HANs (Zhang et al., 2018). As shown in Fig. 5g, CPAN FPs were 69.1 ng/L at S4, this probably resulted in the increase of CPAN FPs at S5 compared with S3. In addition, a higher concentration of CPANs also can be found at S11 (58.6 ng/L) compared with S8 (23.1 ng/L). It was noteworthy that CPAN levels were 98.8 and 81.1 ng/L at S9 and S10, respectively (Fig. 5h). This indicated that many CPAN precursors were introduced into the stormwater drainage system due to the illicit connection of sewage conduits. These phenomena indicated that domestic sewage was an important source of aromatic N-DBP precursors. Ultimately, these waters containing large number of DBP precursors directly discharged to the receiving waters may bring potential hazards to the aquatic environment. Thus, it was vital to identify the characteristics of key DBP precursors.

3.4. Impact of pipe illicit connections on DOM characteristics

Fig. 6a and b presented the region EEM intensity and the fluorescence region integration (FRI) distribution of the five regions in 19 water samples along the urban stormwater drainage system. Tryptophan-like aromatic proteins (region II) accounted for 39–52 % of the total EEM intensities, making them the dominant DOM components in all water samples. This indicated that tryptophan-like aromatic proteins were main DBP precursors from urban stormwater drainage system. It is partially concurred with the findings of Hou et al. (2018), who reported that aromatic proteins as well as SMP-like substances were found to be main compounds in fresh rainwater and were also reported to be DBP precursors. Differently, another study found that fulvic acid-like and humic acid-like substances were the main DOM components in the Yangtze River, while the percentage of tryptophan-like aromatic proteins ranged from 11 % to 16 % (Fang et al., 2021). The results obtained in this study showed that the DOM characteristics of waters from urban stormwater drainage system were similar but not exactly the same as those of fresh rainwaters. Moreover, the DOM characteristics of water samples from urban stormwater drainage system were different from natural waters.

It could be observed from Fig. 4e that while DBP levels for most sites were comparable, high levels of DBP FPs were found in samples at the illicit connection points and the subsequent points (S4, S9, S10, and S11). Notably, compared with other water samples, higher intensities were also found at the illicit connection points and the subsequent point (S4, S9, S110, and S11) in EEM regions, with tryptophan-like aromatic proteins as

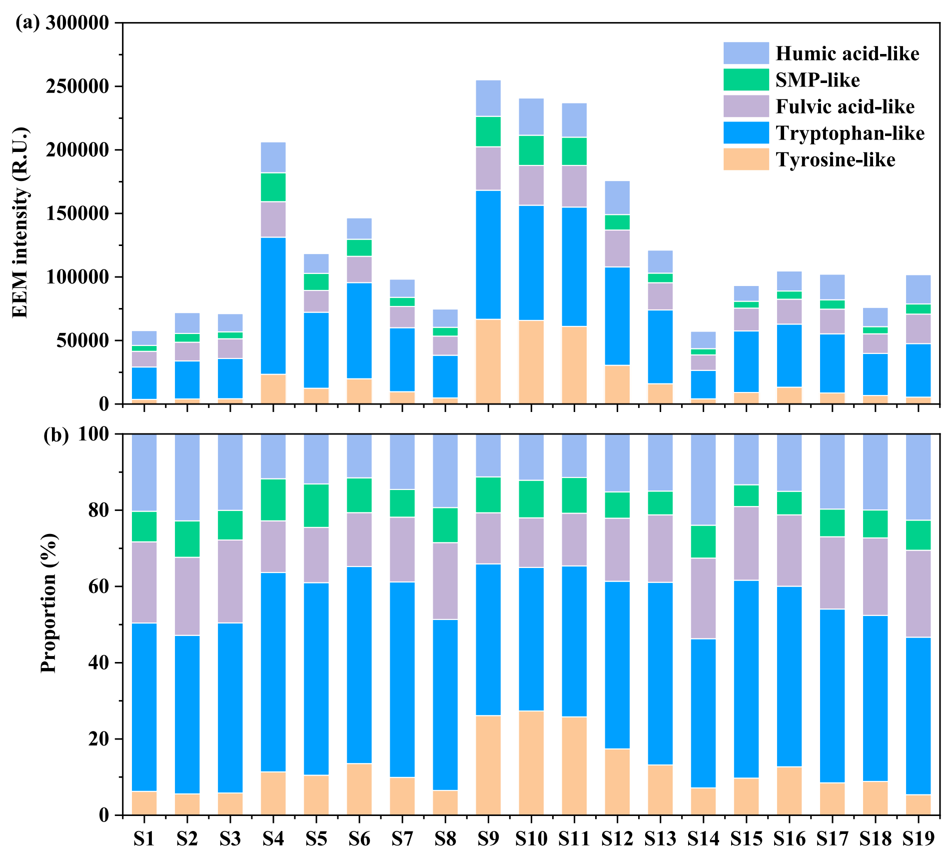

Fig. 6. (a) Region EEM intensity and (b) fluorescence region integration (FRI) distribution of EEM in DOM of the 19 water samples along the urban storm water drainage system.

dominant components, followed by tyrosine-like aromatic proteins (region I), fulvic acid-like organics (region III), humic acid-like organics (region V), and SMP-like materials (region IV). It is well known that aromatic proteins (e.g., tyrosine and tryptophan) are important DBP precursors (Chu et al., 2010; Zhang et al., 2019c). Additionally, studies have identified that fulvic acid-like and humic-like substances are DBP precursors (Hao et al., 2012; Zhang et al., 2008). Importantly, EEM intensity as well as the FRI distribution of tryptophan-like aromatic protein and tyrosine-like aromatic proteins at S4, S9, and S10 was much higher than that of their front connection sampling point like S3 and S8, which was consistent with the results of high response in biopolymers at S9 and S10 (Fig. 3c). This probably resulted in the higher DBP FPs at S4, S9, and S10, especially for the increase of HAN and CPAN FPs at S9 and S10, and thus contributed to substantial increase in CTI values at S9 and S10. Previous studies also suggested that there was high HAN FPs during chlorination of amino acids (Chu et al., 2015b; Zhang et al., 2019c). Considering that the sampling locations belonged to living quarters and business districts, it was speculated that the higher intensities of EEM at the illicit connection points might be related to foods, nutrients, personal care products, etc. as a result of human activities, and these compounds in sewage may enter stormwater pipes due to illicit connection. Moreover, a previous study reported that amino acid (tryptophan) could produce 3-methylindole, which was one of the most notorious compounds (Ma et al., 2021). This indicated that water deterioration and black-odor water in many urban rivers may be related with direct drainage of stormwater involving illicit connections. Anyway, pipe illicit connections did lead to the increase of DBP precursors in the urban stormwater drainage system.

In addition, the observed variation trend of EEM intensity was consistent with that of DOC (Fig. 2a), DON (Fig. 2b), DBP FPs (Fig. 4e), and cytotoxicity index (Fig. 4f) along the urban stormwater drainage system. Thus, the relationships between EEM intensity and DBP FPs as well as CTI were further investigated. Fig. S4 shows that there were strong linear correlations between EEM intensity and DOC (coefficient of determination (R^2) = 0.94) as well as DON (R^2 = 0.73). More importantly, there was a positive linear relationship between EEM intensity and HAN FPs. Previous studies also observed that N-DBPs had close relationship with EEM intensities such as tyrosine-like, tryptophan-like aromatic proteins and SMP-like species (Chu et al., 2010; Zhang et al., 2021b). Additionally, a positive linear correlation between EEM intensity and CTI could also be found (R^2 = 0.60). This suggests that EEM intensity based on FRI may be used to reflect DBP-associated cytotoxicity in waters.

4. Conclusions

This study firstly surveyed the characteristics and output levels of DOM and DBP precursors from an urban stormwater drainage system in the case of pipe illicit connections. The results found that the DOC and DON levels in water samples at the illicit connection points were dramatically increased. FP tests showed that THM precursors were dominant along the urban stormwater pipes. However, the calculated results of CTI indicated that HANs and HALs made great contributions to DBP-associated cytotoxicity. Thus, HAN and HAL precursors deserved more attention. Notably, pipe illicit connections in the urban stormwater drainage system dramatically increased the precursors of HALs and HANs. Furthermore, pipe illicit connections introduced more contents of tyrosine-like and tryptophan-like aromatic proteins, which served as typical precursors of highly toxic N-DBPs. Overall, the results of this study suggested that the urban stormwater drainage system was a significant input source of DOM and DBP precursors to natural water.

Currently, there is a lack of research on urban stormwater drainage systems in the presence of illicit connections. More attention should be paid to the pollutants discharged into the water environment through stormwater pipes. Therefore, it is necessary to further investigate the pollution characteristics of illicit sewage in different regions and seasons, as well as the impact of illicit connections on downstream water quality. Meanwhile, there is an urgent need to develop the feasible diagnostic methodology and control technology of pipe illicit connections to mitigate water pollution, protect water sources and further promote the sustainability of water environment.

CRediT authorship contribution statement

Ruihua Zhang: Performing the experiment, Writing, Data curation.
Rong Xiao: Performing the experiment, Writing-reviewing and Editing.
Feifei Wang: Supervision, Writing-reviewing and Editing.
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Jinglong Hu: Performing the experiment, Writing-reviewing and Editing.
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Wei Jin: Supervision, Writing-reviewing and Editing.
Jan Peter van der Hoek: Supervision, Writing-reviewing and Editing.
Zuxin Xu: Supervision, Writing-reviewing and Editing.

Data availability

Data will be made available on request.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.164248.

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