



## Fate of volatile aromatic hydrocarbons in the wastewater from six textile dyeing wastewater treatment plants



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### HIGHLIGHTS

- Eight volatile aromatic hydrocarbons (BTEXSI) were detected in the textile dyeing wastewater.
- Toluene and xylenes predominated in the BTEXSI.
- BTEXSI (76.1–93.1%) was removed from the TDWTPs.
- Aerobic process played a major role in BTEXSI removal.

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### ABSTRACT

The occurrence and removal of benzene, toluene, ethylbenzene, xylenes, styrene and isopropylbenzene (BTEXSI) from 6 textile dyeing wastewater treatment plants (TDWTPs) were investigated in this study. The practical capacities of the 6 representative plants, which used the activated sludge process, ranged from 1200 to 26000 m<sup>3</sup> d<sup>-1</sup>. The results indicated that BTEXSI were ubiquitous in the raw textile dyeing wastewater, except for isopropylbenzene, and that toluene and xylenes were predominant in raw wastewaters (RWs). TDWTP-E was selected to study the residual BTEXSI at different stages. The total BTEXSI reduction on the aerobic process of TDWTP-E accounted for 82.2% of the entire process. The total BTEXSI concentrations from the final effluents (FEs) were observed to be below 1 µg L<sup>-1</sup>, except for TDWTP-F (2.12 µg L<sup>-1</sup>). Volatilization and biodegradation rather than sludge sorption contributed significantly to BTEXSI removal in the treatment system. BTEXSI were not found to be the main contaminants in textile dyeing wastewater.

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## 1. Introduction

Environmental contamination due to the volatile aromatic hydrocarbons quite problematic because of their ubiquity in the environment and their potential impacts on ecosystems and human health (Ruijten et al., 1994; Pelclová et al., 2000; Morata et al., 2002; Collins et al., 2003). Volatile aromatic hydrocarbons commonly contain benzene, toluene, ethylbenzene, xylenes, styrene and isopropylbenzene (BTEXSI). These compounds are widely used in industries, such as the printing, paint, synthetic resin and synthetic rubber industries (Holcomb and Seabrook, 1995; Nolle, 2001); some of these compounds are toxic and are often classified as carcinogens for humans (Pohl et al., 2003; An, 2004; Reineke

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et al., 2006; Farhadian et al., 2008). Some research performed to address these hazards indicated that the main removal mechanisms of BTEXSI from wastewater were biodegradation and volatilization (Farhadian et al., 2008; Mozo et al., 2011). To date, although several investigators have studied the occurrence, removal and fate of BTEXSI from various types of wastewater, they focused on municipal wastewater and petroleum industry wastewater and not on textile dyeing wastewater (Dórea et al., 2007; Fatone et al., 2011; Costa et al., 2012).

The textile sector was an important economic pillar of the industry in China (Ning et al., 2014). The Pearl River Delta, as one of the most highly urbanized and industrialized regions in China, ranked third after Jiangsu and Zhejiang provinces in the textile industry. The textile industry uses a more complicated process compared to other manufacturing sectors; these processes involve washing, scouring, bleaching, mercerizing, dyeing and finishing processes. These processes produced large quantities of highly

toxic wastewater, especially for the washing and finishing processes (Li et al., 2012). According to the China Environment Statistical Yearbook in 2012, the discharge of textile dyeing wastewater was approximately 2.37 billion tons. This type of wastewater is difficult to dispose because it contains a high content of contaminants, such as dyes, heavy metal ions, surfactants, solvents, detergents, auxiliaries and recalcitrant compounds (Wang et al., 2002; Li et al., 2012; Lotito et al., 2012; Liang et al., 2013). In the textile industry, volatile aromatic hydrocarbons are generated from fossil fuels, and a variety of chemicals are used for production (Müezzinoğlu, 1998). In fact, Namkung and Rittmann (1987) indicated that volatile aromatic hydrocarbons typically accounted for over 75% of the load of total volatile organic compounds (VOCs) released from WTPs since the 1980s. To date, the occurrence, removal and fate of BTEXSI from TDWTPs have not been investigated, even though the textile dyeing wastewater has seriously polluted the water environment.

Consequently, the goals of this study were to investigate the occurrence and removal of BTEXSI in textile dyeing wastewater from 6 representative TDWTPs and to evaluate the magnitude of the hazard by focusing on the total contents and the concentration of each compound of BTEXSI in RWs and FEs. Furthermore, the fate of BTEXSI in textile dyeing wastewater from typical TDWTP (i.e., anoxic/aerobic processes) was investigated. Each BTEXSI concentration in the aqueous phase at different stages was compared to determine the ability of each stage to remove the target contaminants.

## 2. Materials and methods

### 2.1. Chemicals and materials

The standard solution of BTEXSI in methanol contains 8 compounds, each at 1000  $\mu\text{g mL}^{-1}$ , i.e., benzene, toluene, ethylbenzene, m-xylene, o-xylene, p-xylene, styrene and isopropylbenzene. The 4-bromofluorobenzene (at 100  $\mu\text{g mL}^{-1}$ ) was used as surrogate standard. All standard solution were provided by O2si (Charleston, SC, USA) and stored at  $-18\text{ }^{\circ}\text{C}$ . The purities of the standards ranged from 98.0% to 99.9%. Intermediate solutions were prepared by diluting the standard mixture at concentrations of 10  $\mu\text{g mL}^{-1}$  and 100  $\mu\text{g mL}^{-1}$  in methanol. The acetone, carbon disulfide and methanol (HPLC grade) used for sample processing and analysis were supplied by CNW Technologies GmbH (Düsseldorf, Germany). The millipore polyether sulfone filters (0.45  $\mu\text{m}$ , product code: 0256662) were purchased from Jinteng (Tianjin, CHN). Ultrapure water was obtained from a Milli-Q water purification system (Millipore, Paris, France).

### 2.2. The analyzed TDWTPs and the sampling process

To account for the heterogeneity of the TDWTPs in the Pearl River Delta, 6 representative TDWTPs located in three cities (Guangzhou, Dongguan and Zhongshan) were selected for study.

The information of the 6 plants is shown in Table 1. Sampling campaigns were performed from January 3 to 25, 2014. To eliminate the variability of the influents and effluents, all of the samples were obtained when the plants ran normally. Three samples were taken from each sampling point every day during three days. Raw wastewater (RW) containing suspended particles, anoxic effluent (AE), aerobic effluent (OE), final effluent (FE) and dewatered sludge were collected from 6 TDWTPs using the grab sampling method. One-hundred milliliter bottles were filled with wastewater to capacity to avoid any headspace. After filling with water, the bottles were sealed with Teflon-lined screw caps and then transported on the same day to the laboratory under refrigerated conditions. The sludge samples were freeze dried and kept at  $-20\text{ }^{\circ}\text{C}$  before analysis. In the laboratory, the samples were preserved at a temperature of  $4\text{ }^{\circ}\text{C}$ .

### 2.3. Sample preparation

Volatile aromatic hydrocarbons were analyzed by means of gas chromatography–mass spectrometry according to the U.S. EPA methods (EPA 5030b/96 and EPA 8260b/96).

The aqueous samples for the BTEXSI analyses were filtered through a 0.45- $\mu\text{m}$  millipore filter. Next, an aliquot of 5 mL of filtered water was transferred to the purge and trap sample concentrator without air bubbles. At the same time, the RW was filtered to partition liquid and particle phase. The suspended particles were freeze dried and kept at  $-20\text{ }^{\circ}\text{C}$  before analysis.

The sludge samples were extracted using ultrasound extraction followed by dispersive liquid–liquid microextraction (USE–DLLME). Two grams of freeze-dried sludge was spiked with 10  $\mu\text{L}$  of surrogate standards (100  $\mu\text{g mL}^{-1}$ ) and then ultrasound extracted with 5 mL of acetone under 53 kHz in a ultrasound bath. USE was outperformed for 30 min. Next, the mixture was centrifuged at 3000 rpm for 10 min. One milliliter of acetone extract filtered by Millipore filters (0.45  $\mu\text{m}$ ) and 80  $\mu\text{L}$  of carbon disulfide were added to 5 mL of ultrapure water placed in a 15-mL glass-centrifuge tube and mixed for 10 s using handshaking. After centrifugation at 4000 rpm for 10 min, 20  $\mu\text{L}$  of the sediment (carbon disulfide) was analyzed using GC–MS equipped with an automatic injector.

### 2.4. Instrumental measurement

#### 2.4.1. Purge and trap (P&T)

A Teledyne Tekmar Stratum Purge and Trap Sample Concentrator apparatus equipped with a 5 mL purge tube was used to preconcentrate the BTEXSI in the aqueous phase. The P&T apparatus was programmed as follows: a 5-mL water sample was drawn by a 5-mL air tight syringe and then transferred to the purge tube. The water sample was purged with nitrogen (99.995% purity) at 40 mL  $\text{min}^{-1}$  under ambient temperature. The BTEXSI were purged and absorbed onto the trap for 11 min. Subsequently, the trap was heated, causing the trapped components to be desorbed

**Table 1**  
The information of the six TDWTPs.

TDWTP	Location	Average influent flowrate ( $\text{m}^3\text{ d}^{-1}$ )	Type of wastewater treatment system	Sludge ( $\text{ton d}^{-1}$ )	Main dyes
TDWTP-A	Dongguan	8500	Equalization + AO <sup>2</sup> + flocculation	6	ID + AD
TDWTP-B	Dongguan	9000	Equalization + flocculation + AO	7	MD
TDWTP-C	Dongguan	1200	Equalization + flocculation + hydrolysis acidification + aerobic	1	RD
TDWTP-D	Dongguan	17000	Equalization + anaerobic + flotation + aerobic + BAF	3	ID + DD
TDWTP-E	Guangzhou	26000	Equalization + Flocculation + AO	10	RD
TDWTP-F	Zhongshan	20000	Equalization + hydrolysis acidification + activated sludge	20	BD

AO<sup>2</sup>: anaerobic/aerobic/aerobic system, AO: anoxic/aerobic system, MD: mix dyes, ID: ionic dye, RD: reactive dye, DD: disperse dye, AD: acid dye, BD: basic dye, BAF: biological aerated filter.

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