



Occurrence and fate of benzotriazoles UV filters in a typical residential wastewater treatment plant in Harbin, China[☆]



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ABSTRACT

Benzotriazoles (BTs) UV filters are widely used as ultraviolet absorbents for our daily products, which received increasing attention in the past decades. Residential wastewater treatment plant (WWTP) is both an important sink for wastewater and a key pollution source for receiving water for these chemicals. In this study, pretreatment and gas chromatography-tandem mass spectrometry analysis method were developed to determine the occurrence and fate of 9 BTs UV filters in wastewater and sludge from the WWTP with anaerobic-oxic treatment process (A/O) and biological aerated filter treatment process (BAF). Totally, 81 wastewater samples and 11 sludge samples were collected in four seasons. In wastewater, UV-326 and UV-329 were frequently detected, while the highest mean concentrations were detected for UV-234 and UV-329. The concentrations were in the range of <LOQs up to several hundred nanograms per liter. The removal efficiency of BTs UV filters was >85% in A/O process and 60–77% in BAF process except for UV-350, which was more difficult to remove with lower removal efficiencies of 33.3% for both A/O and BAF. All the target chemicals except for UV-320 were detected in sludge samples with the mean concentration ranging from 0.90 ng/g to 303.39 ng/g. There was no significant difference with concentrations and removal efficiency among different seasons. Higher detection frequency and concentration of BTs UV filters in downstream of the receiving water system indicated the contribution of effluent of the WWTP. Compared with other rivers, the lower concentrations in surface water in the Songhua River indicated light pollution status with of BTs UV filters.

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1. Introduction

The benzotriazoles (BTs) UV filters, which have a phenolic group attached to the benzotriazole structure, are known to absorb full spectrum of UV light (Nakata et al., 2009). Therefore, the BTs UV filters are widely used in personal care products (PCPs) and plastic products in our daily life (Farré et al., 2008; Jia et al., 2007; Kiss and Fries, 2009; Liu et al., 2012; Sulej et al., 2013; Switzenbaum et al., 2001; van Leerdam et al., 2009; Weiss et al., 2006). BTs UV filters in PCPs is applied to protect hair and cutaneous membranes from

sun damage (Langford et al., 2015). In 1996, the consumption of BTs UV filters in the North American plastics industry was up to 4 million pounds, and in Europe the consumption reached 2.1 million pounds. In Japan, the consumption of BTs UV filters increased from 2.3 million pounds to 3.1 million pounds between 1985 and 1996 (Crawford, 1999). According to the statistics, the dosage of BTs UV filters in America was 9000 tons/year in 2004, and they were widely used in industry and daily life (Liu et al., 2011b).

In the past decades, BTs UV filters caught more attentions of the scientific community due to their potential hazardous effects. BTs UV filters and benzophenone UV filters had the same action mechanism. Studies found that benzophenone UV filters had obvious estrogenic property (Oishi, 2002; Suzuki et al., 2005), genetic toxicity (Fluck et al., 1976; Mortelmans et al., 1986; Zeiger et al., 1987) and bioaccumulation action (Hany and Nagel, 1995). However, the potential adverse effects, action manners, and toxicological effects with BTs UV filters are still scarce. The first case of

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contact allergic to UV-P was reported as the wrist dermatitis in 1989 (Nikilasson and Björkner, 1989). The derivatives of BTs UV filters were reported with mutagenicity of bacteria and toxicity for plants (Farré et al., 2008). Even though their hazardous effects were not clearly illuminated, many countries have set the consumption limitations of BTs UV filters (Jeon et al., 2006). For example, the Japanese Government banned the production, use, and import of UV-320 since 2007 (Nakata et al., 2009).

BTs UV filters can easily entered into the sewage system directly and/or indirectly via “wash-off” from skin and clothes, and “discharge” from other water activities during our daily life (Liu et al., 2014a). The wastewater in the sewage system is finally transported to the wastewater treatment plants (WWTPs). Therefore, residential WWTP is an important sink for BTs UV filters. It was reported that the average concentration of BTs UV filters in a Berlin WWTP was 12 µg/L and as high as 36.2 µg/L in a WWTP of Barcelona (Matamoros et al., 2010; Weiss et al., 2006). Furthermore, biotransformation products of BTs UV filters had also been detected in the process of aerobic and anaerobic treatment (Liu et al., 2011a). In the view of the effluent, WWTPs can be considered as a key pollution source for receiving water system. For example, higher residue concentrations of BTs UV filters were observed in wild fish in Norway (Langford et al., 2015) and in aquatic organism for UV-329 and UV-326 in Australia (Langford et al., 2015; Liu et al., 2011b; Peng et al., 2015; Tsai et al., 2014). Furthermore, it was found that the removal efficiency of BTs UV filters by biological adsorption and sludge accumulation was not obvious in WWTPs (Asimakopoulos et al., 2013). For example, much higher concentrations of UV-326, UV-327 and UV-329 in municipal sludge than river bottom sludge were observed recently (Domínguez et al., 2012; Liu et al., 2012; Ruan et al., 2012), indicating their high ecological risk with municipal sludge. Generally speaking, the current studies on the fate of BTs UV filters in WWTPs were mainly carried out in Europe and some developed countries. Compared with other worldwide contaminants, more studies should be conducted for BTs UV filters.

In this study, 9 BTs UV filters were analyzed with wastewater and sludge samples collected from a typical residential WWTP in Harbin City, Northeastern China. Furthermore, the surface water of the receiving water system (the Songhua River) was also collected. The objectives of the study were: (1) to investigate their occurrence and removal efficiency in the WWTP; (2) to compare the difference between the two typical wastewater treatment processes; (3) to figure out the seasonal variation of their fate; and (4) to assess the influence of effluent to receiving water system.

2. Experimental

2.1. Reagents and standards

All organic solvents were HPLC-grade and purchased from J.T.Baker (Phillisburg, NJ). Standard solution (100 µg/mL) of 9 BTs UV filters, including UV-234, UV-320, UV-326, UV-327, UV-328, UV-329, UV-350, UV-P and UV-PS, was purchased from Sigma-Aldrich, Inc (purity > 97%, St. Louis, MO). The molecular structure and basic information for the 9 BTs UV filters are presented in Table S1 (Supporting Information). Ultrapure water was prepared with a Milli-Q ultrapure system. All standards and stock solutions were stored at –20 °C before analysis.

2.2. Sample collection

The studied WWTP is located in the downstream of the Songhua River in Harbin Section, Northeastern China. The wastewater in the WWTP is mainly originated from residential discharge with the

contribution up to 80%. The biological aerated filter treatment process (BAF, primary + secondary biological treatment by trickling filter) and the anaerobic-oxic treatment process (A/O, primary + secondary biological treatment by activated sludge process) were applied in the WWTP. For each season, three consecutive days were selected for sampling. The wastewater samples were collected using 4 L brown glass bottles in the period of 10 a.m., 13 p.m. and 16 p.m. each day, then they were uniformly mixed for the sample as the day. Totally, 81 wastewater samples were collected for different process units of the two processes for 4 seasons. All the collected wastewater samples were added dichloromethane to prevent biodegradation and stored in a refrigerator at 4 °C until extraction. Detailed information regarding the wastewater sampling is presented in Fig. S1 and Table S2. Sludge samples were collected from A/O biological tank (n = 6) and sludge dewatering tank from the WWTP (n = 5), respectively. The sludge samples were collected with a stainless steel jar. Sludge samples were sealed in aluminum bottle and kept at –20 °C until extraction. For the receiving water system, 6 sampling sites were selected in the Songhua River of Harbin Section. 4 sites (S1-S4) were located in the upstream and 2 sites (X1-X2) in the downstream of the WWTP discharge area in order to study the influence of effluent to the receiving water. The surface river water samples were collected using 4 L brown glass bottles and kept in a refrigerator at 4 °C with dichloromethane to prevent biodegradation until extraction.

2.3. Sample treatment

Liquid-liquid extraction method and liquid-solid shaking extraction method were applied for wastewater (surface water) samples and sludge samples, respectively (Fig. S2).

For wastewater and surface water samples, 1 L water was transferred into a Teflon separatory funnel containing 100 mL dichloromethane and shaken violently for 3 min. After stewing for 15 min, the bottom dichloromethane layer was filtered through anhydrous sodium sulfate, then transferred into a clean 500 mL flat bottom flask. Totally, the sample was extracted three times in the same way, and the dichloromethane extract was combined together. After being added 5 mL isooctane, the extract was concentrated to 2–3 mL using a rotary evaporator. Finally, the volume was concentrated to 1 mL under a gentle nitrogen stream for further treatment.

For sludge samples, 0.5 g of sample was vortex-mixed with 15 g sodium sulfate and extracted with 25 mL acetone/hexane mixture (1:1, v/v) by shaking in an orbital shaker for 30 min. Supernatant was transferred into a 250 mL flat bottom flask, and the extraction was repeated twice again. The supernatants were combined together and concentrated to 2–3 mL in isooctane under a rotary evaporator, then the extract was concentrated to 1 mL under a gentle stream of nitrogen for further treatment.

Extracts of water and sludge samples were purified with a purification column containing 5 g 5% water de-activated silica gel and 1.5 g sodium sulfate on top. After the column was pre-cleaned with 100 mL hexane, the 1 mL extract was loaded into the column. Then, the column was eluted with 50 mL hexane/diethyl ether mixture (1:1, v/v), and the eluent was collected in a 250 mL flat bottom flask. After adding 5 mL isooctane, the eluent was concentrated to 2–3 mL under a rotary evaporator. Finally, the eluent was concentrated to 1 mL under a gentle stream of nitrogen and transferred into a gas chromatographic vial at –20 °C until analysis.

2.4. Chemical analysis

Analysis of the 9 BTs UV filters was carried out using a gas

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