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Triclosan in Influents and Effluents from Sewage Treatment Plants Using Chlorine and UV Disinfection

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Abstract: Triclosan is widely used in personal care products, and has been reported to be toxic to aquatic organisms. The purpose of this study was to determine the triclosan levels in influents and effluents from three different sizes of sewage treatment plants using chlorine and ultraviolet (UV) for disinfection (i.e., STPs A, B, and C). Variations of triclosan concentration in influents and effluents (i.e., 9:00 a.m., 11:00 a.m., 13:00 p.m. and 15:00 p.m.) were also observed, respectively. The influent and effluent samples of the three STPs were composite samples collected every 2h during 9:00 a.m.-3:00 p.m by manual grab method, respectively. The samples were analyzed using a solid phase extraction (SPE) procedure, followed by liquid chromatography with tandem mass spectrometry (LC-MS-MS). Results demonstrated all of influent and effluent samples from three STPs contained triclosan. Significantly higher levels of triclosan were observed in effluent samples collected at STPs A (Median: 27.0 ng/L) as compared to in those collected at STPs B (15.0 ng/L), and C (8.0 ng/L) ($p=0.012$). STP A had the most serviced inhabitants and sewage flow. The triclosan levels at STP C with UV disinfection were lowest. The daytime peak of triclosan concentration in the collected influent samples was seen around 9:00 a.m. Notably, sewage discharge is the contamination source of triclosan in water bodies. Moreover, sewage flow and methods of disinfection may be the factors that affect the concentration of triclosan in effluents from sewage treatment plants.

Keywords: Triclosan, Sewage Treatment Plants, Effluent, Sewage Flow, Chlorine and UV Disinfection

1. Introduction

In the 1970s, triclosan (5-chloro-2-(2,4-dichlorophenoxy)phenol) was at first used in hospitals as surgical scrub for its antimicrobial property. Today, triclosan is widely used in personal care products such as toothpastes, shampoos, soaps, deodorants and cosmetics [1-3]. The antimicrobial personal care products commonly contain triclosan at a concentration ranging from 0.1 to 0.3 wt% [4]. However, the triclosan-containing products can enter the sewer system after washing-off by tap water via sinks and are transported to wastewater treatment plants.

Triclosan has been documented to be toxic to aquatic organisms, for example, rainbow trout, Daphnia magna and particularly to algae [5]. In 2008, the European Union has classified triclosan as a hazardous substance to aquatic environment [6]. In addition, triclosan and its metabolite are also considered to have endocrine disrupting effects [7-10].

Wastewater is considered as the major source of triclosan in the aquatic environment, because of the incomplete removal of triclosan by conventional wastewater treatment processes (approximately 58-98%) [11-15]. Triclosan has been detected in the wastewater, sludge, surface water, and sediments [13, 16-18].

The disinfection methods of wastewater may affect the concentration of triclosan in the effluents. Triclosan may react during the disinfection of wastewater with free chlorine to form three chlorinated triclosan derivatives (CTDs) [19]. Buth et al. [20] reported that in the treatment plant that used chlorine disinfection, triclosan concentrations decreased while CTDs were formed during chlorination. The total CTD concentration in the final effluent of the chlorinating treatment plant reached nearly one third of the triclosan concentration. In the treatment plant that used ultraviolet (UV) disinfection, the triclosan concentration in the final effluent

substantially decreased, possibly due to photolysis of triclosan [20]. Previous studies indicated that triclosan is photochemically transformed to 2,8-dibenzodichloro-p-dioxin (2,8-DCDD) under UV light and sunlight irradiation [21, 22].

The purpose of this study was to determine the triclosan levels in influents and effluents from three different sizes of sewage treatment plants using chlorine and ultraviolet (UV) for disinfection. Fluctuations of triclosan concentration in influents and effluents during daytime were monitored, respectively.

2. Materials and Methods

2.1. Chemicals and Materials

Triclosan (>97%) and ammonium formate (>99%) were purchased from Sigma-Aldrich (St Louis, MO, USA). $^{13}C_{12}$-triclosan (>99%) was acquired from Cambridge Isotope Laboratories, Inc. (Andover, MA, USA). HPLC grade methanol was provided by Merck KGaA (Darmstadt, Germany). Ultrapure water was produced by the Milli-Q water purification system (Millipore, USA). The Oasis® HLB cartridges (200 mg, 6 mL) was purchased from Waters (Milford, MA, USA). The stock solutions of triclosan and $^{13}C_{12}$-triclosan were prepared in methanol.

2.2. Sewage Treatment Plant and Sample Collection

Influent and effluent samples were collected from three civil sewage treatment plants in northern Taiwan (i.e., STPs A and B) and southern Taiwan (STP C), separately. The STPs employed similar conventional treatment processes: primary treatment to remove particles coupled with secondary biological treatment of activated sludge. In addition, the secondary effluent from STPs A, and B were disinfected by chlorination while that from STP C was treated with UV disinfection. Other information on each STP, such as the serviced inhabitants, average daily flow rate, and basic water pollution parameters such as biochemical oxygen demand (BOD_5), and suspended solids (SS) were shown in Table 1.

Table 1. *Main characteristics of sewage treatment plants.*

	STP A	STP B	STP C
Inhabitants serviced (person)	3,120,000	290,000	8,000
Sewage flow (m^3/day)	460,000	120,000	3,600
Effluent			
BOD_5 (mg/L)	7.95	8.2	4.5
SS (mg/L)	8.57	6.8	4.0
Hydraulic retention time (hour)	10	14.6	48
Type of disinfection	chlorination	chlorination	UV

The influent and effluent samples of the three STPs were composite samples collected every 2h during 9:00 a.m.-3:00 p.m. (i.e., 9:00 a.m., 11:00 a.m., 13:00 p.m. and 15:00 p.m.) by manual grab method, respectively. Three sewage samples were poured into a stainless steel bucket to mix one composite sample per sampling time. There was no rain during the sampling. The collected influent and effluent samples were stored in 1L amber glass bottles and then filtered through 0.45 μm glass fiber filters (GF/F, Whatman). After filtration, the samples were extracted within 24 h, and stored in a -20°C freezer until analysis.

2.3. Sample Extraction and Purification

The samples were extracted using an automated solid phase extraction (SPE) system with Oasis HLB cartridges (6 mL, 200 mg, Waters Corporation). The cartridges were conditioned with methanol and Milli-Q water. Samples were loaded onto the cartridges using a vacuum manifold. After loading, the cartridges were washed with Milli-Q water, dried for 30 min under vacuum and eluted with 6 mL of methanol. The eluates were then evaporated to dryness, reconstituted with 1 mL mobile phase (Milli-Q water: methanol, 75:25), vortexed, transferred to vials and stored at 4°C until analysis.

2.4. Instrumental Analysis

The samples were analyzed using an Agilent 6430 triple quadruple mass spectrometry interfaced online with an Agilent 1290 series HPLC system. Separation was performed on a Phenomenex Gemini C18 column (2 mm×100 mm i.d.; 3 μm particle size) and the sample injection volume was 20μL. The mobile phase consisted of a mixture of phase A (5 mM ammonium formate) and phase B (methanol) with the flow rate at 0.2 mL/min. Gradient elution was performed as follows: 0 min, 40% B; 0-8 min, 40-80% B; 8-20 min, 80% B. Quantification was performed through multiple reaction monitoring (MRM) in negative electrospray ionization mode (ESI-), a capillary voltage of 4000 V, and a source temperature of 325°C. Nitrogen was used for desolvation and drying gas at 10 L/min, and for nebulization at 45 psi. The quantitative and qualitative ion transitions of triclosan are 287→35 and 289→37. The quantitative ion transition of $^{13}C_{12}$-triclosan are 299→35. The fragmentor voltage and collision energy were optimized using the Agilent Optimizer software. The limits of quantitation (LOQs) of triclosan were 2.7 ng/L.

2.5. Statistical Analysis

Due to the small sample size, the Kruskal-Wallis test was used to compare the differences of triclosan level in influents and effluents among the three STPs, respectively. Statistical significance for all differences was set at $p \leq 0.05$.

3. Results and Discussion

The analytical results showed that all of the samples collected from three STPs contain triclosan (see Table 2). The triclosan levels of influent samples at three STPs ranged from 13 to 75 ng/L. Significantly higher levels of triclosan were observed in the inffluent samples collected at STPs C (Median: 32.5 ng/L) as compared to in those collected at STPs A (16.5 ng/L), and B (13.5 ng/L) ($p=0.021$), respectively.

Table 2. *Levels of triclosan in influents and effluents at STPs A, B, and C.*

	STP A	STP B	STP C
Influent (ng/L)			
Mean	16.5	13.5	32.5
Range	15-26	13-20	24-41
Effluent (ng/L)			
Mean	27.0	15.0	8.0
Range	23-30	9-22	5-11

The triclosan levels of effluent samples at three STPs ranged from 5 to 39 ng/L. Significantly higher levels of triclosan were observed in the effluent samples collected at STPs A (Median: 27.0 ng/L) as compared to in those collected at STPs B (15.0 ng/L), and C (8.0 ng/L) (p=0.012), respectively. In the present study, STP A had the most serviced inhabitants and sewage flow. Moreover, the effluents at STP C were disinfected with UV light before discharging into the river. Maybe these were the causes of lower triclosan levels in effluents at the STP C. Under UV irradiation, triclosan in water can be converted to 2,7/2,8-dibenzodichloro-p-dioxin (2,7/2,8-DCDD) [21, 22]. Mezua et al.(2004) reported that triclosan and 2,7/2,8-DCDD were found in 100% and 80% of the effluent samples, respectively [22]. Buth et al. (2011) also reported that in the treatment plant that used UV disinfection, the triclosan concentration in the final effluent substantially decreased, possibly due to photolysis of triclosan [20]. Thus, photolysis of triclosan might occur in the effluents at STP C with UV disinfection in the present study.

The variations of triclosan level in influents at STPs A, B, and C during daytime were shown on Figure 1. The levels of triclosan usually peaked around 9:00 a.m. and then slowly leveled off around 3:00 p.m. Apparently, before 9:00 a.m. more triclosan-containing products (e.g., triclosan-containing toothpaste) were used and washed down in the sewer lines, which were connected with STPs. No obvious variations of triclosan concentration were observed in effluents at STPs A, B, and C. Nishi et al. [23] investigated daily variation of triclosan concentration in the surface water that domestic wastewater inflow occurred [23]. And the peak in the loading amounts of triclosan was observed from 10:00 a.m to 12:00 noon. The daily peak of triclosan concentration in the influent samples in the present study was similar to the results reported by Nishi et al [23].

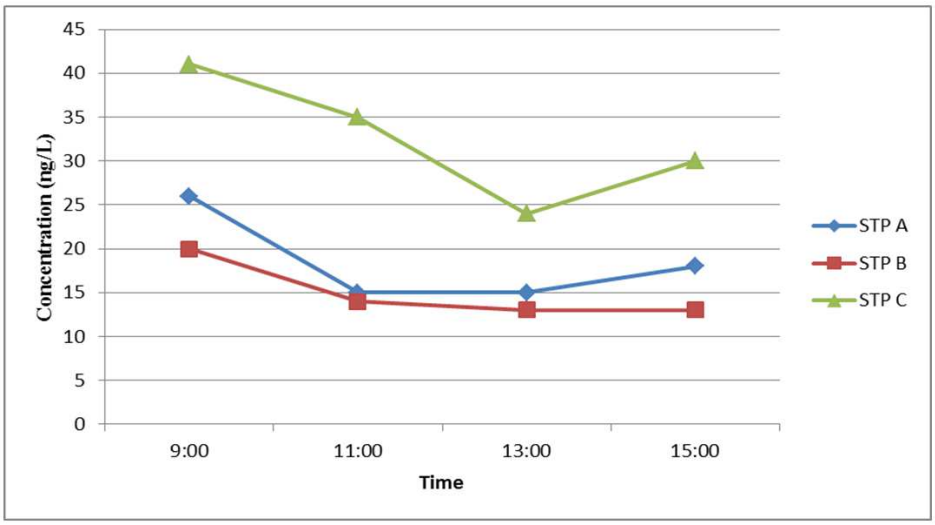

Figure 1. *Daytime variations of triclosan levels in influent samples at STPs A, B, and C.*

4. Conclusions

In summary, the serviced population in the coverage area of STP was directly resulted to the levels of triclosan in the effluents. The daytime peak of the triclosan concentration in the collected influent samples was observed around 9:00 a.m. This means household consumer products are major source of triclosan contamination. In addition, for three STPs selected in this work, triclosan was found in all the samples of effluents discharging into various receiving rivers. Methods of disinfection may be also a factor that affect the concentration of triclosan in effluents. Since triclosan is very toxic to aquatic organisms, it is important to improve and modernize the current civil sewage treatment plants. Further research needed to investigate the distribution, fate and ecological effects of photoproducts of triclosan in the aquatic environment.

Conflicts of Interest

The authors declare that they have no competing interests.

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