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Baseline

Occurrence and fate of triclosan and triclocarban in a subtropical river and its estuary

Min Lv [a,b,1], Qian Sun [a,c,1], Haili Xu [a,d], Lifeng Lin [a], Meng Chen [e], Chang-Ping Yu [a,c,*]

[a] Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[b] University of Chinese Academy of Sciences, Beijing 100043, China
[c] Ningbo Urban Environment Observation and Research Station-NUEORS, Chinese Academy of Sciences, Ningbo 351800, China
[d] College of Chemical Engineering, HuaQiao University, Xiamen 361021, China
[e] College of Environment and Ecology, Xiamen University, Xiamen 361005, China

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ABSTRACT

The occurrence of triclosan (TCS) and triclocarban (TCC) in a subtropical river (Jiulong River) and its estuary was investigated for two years. TCS and TCC were ubiquitously detected in the Jiulong River and its estuary. The levels of TCS and TCC ranged from less than the method detection limit to 64 ng/L and from 0.05 to 14.1 ng/L in the river, respectively. The levels of TCS and TCC in the estuary ranged from 2.56 to 27.25 ng/L and 0.38 to 5.76 ng/L, respectively. Temporal and spatial variations of TCS and TCC in the Jiulong River and its estuary were observed during the investigation. The weather conditions did not show significant correlations with TCS and TCC, whereas several water quality parameters showed high correlations with TCS and TCC. The microcosm studies showed that both direct photolysis and biodegradation contributed to TCS removal, whereas indirect photolysis was important for TCC removal in the surface water.

A decade ago, the United States Geological Survey (USGS) national reconnaissance of emerging contaminants survey reported that 80% of the surveyed streams (approximately 108 US streams) were contaminated with trace amounts of organic compounds, including steroidal hormones, antimicrobial agents, stimulants, and many other pharmaceuticals and personal care products (PPCPs) (Kolpin et al., 2002). The detection of these various compounds has gained extensive public attention due to their potential adverse effects on ecological and public health. Two widely used antimicrobial agents on the list, triclosan (TCS) and triclocarban (TCC), have received wide concern. TCS has been suggested to be potentially weakly androgenic (Foran et al., 2000), and TCC has been demonstrated to be an endocrine disruptor by acting as a steroidal hormone amplifier (Chen et al., 2008).

The widespread use of TCS and TCC-containing products has released TCS and TCC into the receiving aquatic environment via treated and untreated sewage due to the imperfect coverage of sewage treatment facilities, particularly in developing countries. Many researchers have reported on their incomplete removal by wastewater treatment plants (Singer et al., 2002; Sun et al., 2014). Therefore, it is necessary to investigate the occurrence of TCS and TCC in the receiving aquatic environment. To date, TCS and TCC have been detected in the aqueous environment of various countries, including the United States (Yu and Chu, 2009), Canada (Hua et al., 2005), United Kingdom (Sabaliunas et al., 2003), Switzerland (Lindstrom et al., 2002) and China (Zhao et al., 2010; Wang et al., 2011, 2012). However, few studies have monitored TCS and TCC in the surface water over an extended period of time, and information about the factors influencing their occurrence and fate is limited.

Natural attenuation, a combination of naturally occurring processes, can reduce concentrations of pollutants during river flow (Gurr and Reinhard, 2006). The possible mechanisms of natural attenuation that could attenuate TCS and TCC in the river include dilution due to wastewater discharges or dilution due to rainfall, sorption, photolysis and biodegradation. Sorption seems to be important for both TCS and TCC due to their high lg *Kow* (Loftsson et al., 2005; Heidler et al., 2006), and sediment was reported to be a sink for TCS and TCC (Zhao et al., 2010). Tixier et al. (2002) combined laboratory studies, field measurements and modeling and concluded that direct photodegradation accounted for 80% of the observed total elimination of TCS in the water column of a Swiss lake. As for biodegradation, although

* Corresponding author at: Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China. Tel.: +86 592 6190768.
E-mail address: cpyu@iue.ac.cn (C.-P. Yu).
[1] First two authors made equal contributions.

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microbial transformation of TCS and TCC were reported (Roh et al., 2009; Miller et al., 2010), information regarding the natural attenuation of TCS and TCC through biodegradation in the surface water is limited.

A two-year study of the occurrence of TCS and TCC in the Jiulong River and its estuary was conducted to address the knowledge gap and to better understand the occurrence of and factors affecting the behavior of TCS and TCC in natural surface water. The Jiulong River is the second largest river in Fujian Province in the southeastern part of China. The study explored the factors that potentially correlated with the occurrence of TCS and TCC, including weather conditions and water quality parameters. Furthermore, the microcosm study was used to assess the existence and relative importance of two attenuation mechanisms: photolysis and biodegradation of TCS and TCC in the surface water.

Six sampling sites were selected from both the North River (NR, N1-N6) and the West River (WR, W1-W6), the two major prongs of the Jiulong River (Fig. 1). Samples were collected on October 21–22, 2011; March 27–28, 2012; September 5–6, 2012; January 15–17, 2013; and June 7–9, 2013. Detailed information about the weather conditions, including rainfall, sunshine duration and temperature is provided in the Supporting Information (SI) Table S1. The Jiulong River Estuary is a typical subtropical estuary on the southwest coast of the Taiwan Strait. The surface water of the Jiulong River Estuary was collected on September 8, 2012; January 16, 2013; and June 9, 2013. The estuary water sampling sites (E1–E12) are described in Fig. 1. The salinities and pH are shown in Fig. S1.

For the TCS and TCC analysis, grab samples (1 L) were collected from each site using amber glass bottles. All of the samples were transported in ice-packed coolers to the laboratory and stored at 4 °C. Samples were processed within 24 h. Detailed information about the sample preparation and analytical methods of TCS and TCC in the present study is listed in SI. The SI includes sampling and analysis information for water quality parameters, including pH; electronic conductivity (EC); dissolved oxygen (DO); water temperature (Tem); salinity (SAL); ammonium (NH_4^+); nitrate (NO_3^-); nitrite (NO_2^-); phosphate (PO_4^{3-}); total dissolved phosphorus (TDP); and total dissolved carbon (TDC).

More than 10 L of surface water samples were collected from N1, W2, and E8 on June 7–9, 2013 for the microcosm study. The microcosm study was set up in duplicate according to the previous study (Fono et al., 2006), with slight modifications. The first set of experiments (referred to as Dark surface water) investigated biodegradation and used 1050 mL water samples in Pyrex Beakers that were covered with aluminum foil. The second set of experiments (referred to as Light surface water) investigated the effect of the combined photolysis–biodegradation. These experiments used 1050 mL water samples that were covered with quartz lids. The third set of experiments (referred to as Light killed surface water) investigated the effect of photolysis and used 1050 mL autoclaved water samples that were covered with quartz lids. The fourth set of experiments (referred to as Light Milli-Q water) investigated the effect of photolysis in Milli-Q water control. These experiments used 1050 mL autoclaved Milli-Q water samples that were covered with quartz lids. The fifth set of experiments (referred to as Dark killed surface water) served as the kill control and used 1050 mL autoclaved water samples that were covered with aluminum foil. TCS and TCC were spiked into all of the beakers at an initial concentration of 2 μg/L. The beakers were placed on the roof of the Environmental Technology Building at the

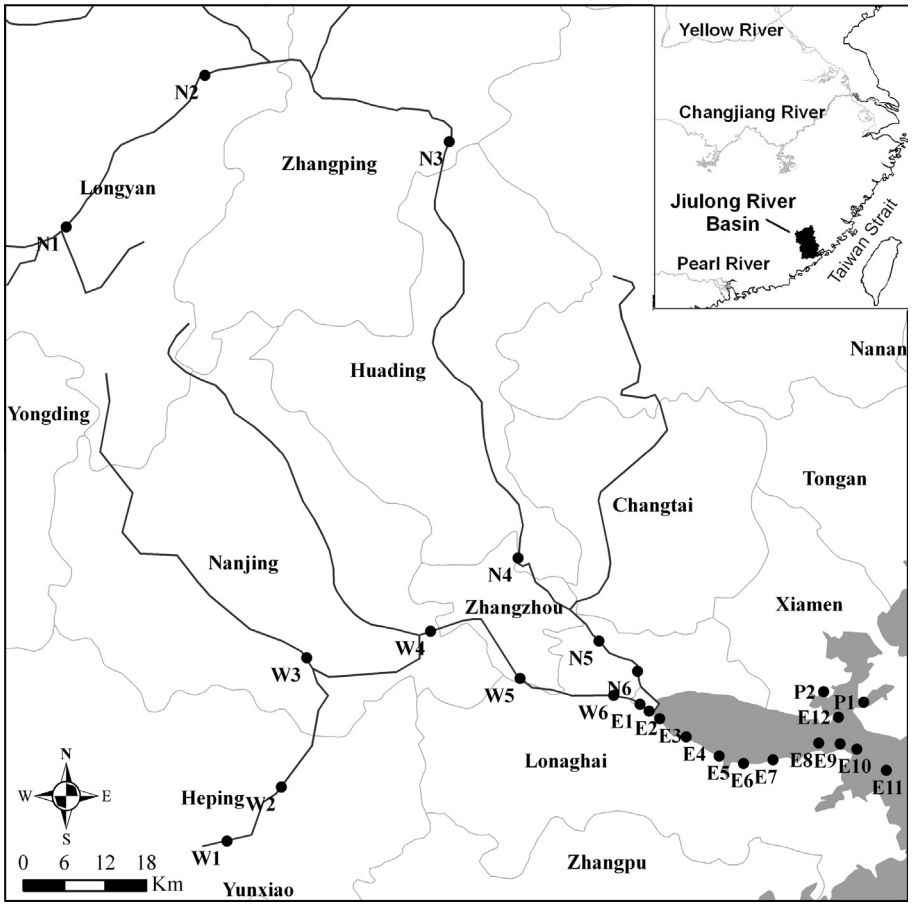

Fig. 1. Area maps and sampling sites in Jiulong River and its estuary (Southeast China).