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Presence and transport of the antimicrobials triclocarban and triclosan in a wastewater-dominated stream and freshwater environment

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ABSTRACT

The presence of the antimicrobials triclocarban (TCC) and triclosan (TCS)in Fountain Creek, a wastewater-dominated stream, and the Arkansas River, Colorado, USA was measured in the surface water, suspended sediments, and bed sediments during spring runoff (high flow) and summer base flow (low flow) conditions. Fountain Creak is a tributary of the Arkansas River. Passive polar organic chemical integrative samplers (POCIS) were used along with active sampling (water grab samples) to measure and TCS concentrations in these surface waters. The concentration of TCC and TCS, based on POCIS measurements, ranged from 4.5 to 47.3 ng/L and 3.9 to 28.3 ng/L, respectively, at the five sample sites monitored in this study under both flow conditions. The range of concentrations of TCC and TCS in suspended sediments was 0.7–57.3 ng/g and 0.7–13.3 ng/g, respectively, and was closely tied to the quantity of organic carbon in the suspended sediment, which ranged from 1.6 to 14.5%. The quantity of organic carbon in suspended sediment during the summer base flow was influenced by runoff from the burn area of a large forest fire that occurred between the two sampling periods. The primary transport mechanism of TCC and TCS in these surface waters was in the dissolved phase, with 64–99% of TCC and 68–99% of TCS transported in the dissolved phase. The total amount of TCS and TCC in bed-sediments was relatively low, with the maximum amount at any one site being 0.38 ± 0.15 ng/g TCS and 4.09 ± 5.26 ng/g TCC. Fountain Creek contributed up to 76% and 69% of the TCC and TCS, respectively, that is transported directly below its confluence with Arkansas River. Fountain Creek drained approximately 3.0 g/day TCS (in spring), 2.9 g/day TCS (in summer) and 1.9 g/day TCC (in spring), 2.0 g/day TCC (in–summer) into the Arkansas River, which suggests consistent input of TCC and TCS into Fountain Creek, such as in discharge of treated wastewater that is independent of changing creek flow conditions.

1. Introduction

Two broad spectrum antimicrobials, Triclosan (5-chloro-2-2,4-dichlorophenoxy) phenol; TCS) and triclocarban (3,4,4′-trichlorocarbanilide; TCC), are commonly found in a number of consumer and personal care products including soaps, shampoos, detergents, toothpastes, medical disinfectants, and kitchen utensils. It is estimated that up to 0.3% (w/w) and

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5% of TCS and TCC, respectively, are added to a variety of consumer products in the US (Chalew and Halden, 2009; Halden and Paull, 2005; TCC Consortium, 2002). Due to the widespread use of these two antimicrobials in personal care products, humans are commonly exposed through ingestion (e.g. toothpaste) or dermal absorption (e.g. soaps) (Sandborgh-Englund et al., 2006; Calafat et al., 2008). Both antimicrobials have been detected in human breast milk, blood plasma, and urine samples (Allmyr et al., 2006; Calafat et al., 2008; Schebb et al., 2011). A recent report shows that TCS has the potential to disrupt excitation-contraction coupling in skeletal and cardiac muscle in humans (Cherednichenko et al., 2012). TCS was also found to cause an allergy or hay fever especially among those below 18 years of age (Clayton et al., 2011).

Both TCC and TCS are commonly rinsed down the drain following use of everyday consumer products, and thus, enter municipal wastewater treatment plants (WWTP) (Halden and Paull, 2005; Chalew and Halden, 2009). Like many anthropogenic organic contaminants (AOCs), such as detergent metabolites, pharmaceuticals, and plasticizers, TCC and TCS are incompletely removed during wastewater treatment (Chalew and Halden, 2009; Kolpin et al., 2002) The removal rate of TCC and TCS from the liquid phase during wastewater treatment has been reported to be between about 75 and 99%, however much of the removal can be accounted for by partitioning to the organic carbon rich sludge phase during treatment (Halden and Paull, 2005; Heidler et al., 2006; Kumar et al., 2010; USEPA, 2002; Ying and Kookana, 2007). As a result of incomplete removal during wastewater treatment TCC and TCS commonly enter the environment through the discharge of treated wastewater effluent to surface waters or land application of biosolids (sewage sludge) (Heidler and Halden, 2008; Kinney et al., 2006). TCS and TCC have been reported up to 2.3 μg/L and 6.8 μg/L, respectively, in US streams (Halden and Paull, 2005; Kolpin et al., 2002). These two antimicrobials are among the most commonly detected AOCs in US water resources impacted by discharge of treated wastewater (Halden and Paull, 2005; Kolpin et al., 2002).

There are numbers of reports describing the potential impact of TCC and TCS on the biota due to their active presence in the total environment. For example, bioaccumulation of TCC and TCS in aquatic organisms (e.g. fish, algae, water fleas, and snails) and terrestrial organisms (e.g. earthworms) have been reported (Orvos et al., 2002; Coogan et al., 2007; Chalew and Halden, 2009; Kinney et al., 2008; TCC Consortium, 2002). TCS has been shown to be an endocrine disruptor in North American Bullfrogs causing interference in thyroid hormone metamorphosis process (Veldhoen et al., 2006). Both compounds have also been found to accumulate in the crops grown in soil irrigated with reclaimed wastewater or amended with biosolids (Holling et al., 2012; Wu et al., 2010, 2012). Other environmental concerns due to TCC and TCS include formation of toxic degradation byproducts and the development of microbial resistance (Aranami and Readman, 2007; Gledhill, 1975; McMurry et al., 1998). To protect aquatic and terrestrial organisms from contamination from these compounds and their toxic byproducts, more information on their environmental fate, transport, and potential effects is needed.

TCC and TCS have relatively low water solubility (TCC 0.7–1.6 mg/L, TCS = 2.0–4.6 mg/L at 20 °C), relatively high log K_{ow} (TCC = 4.9, TCS = 4.8 at pH 7.0), and are generally found to be relatively persistent in soils and aquatic sediments (Halden and Paull, 2005; Miller et al., 2008; Higgins et al., 2011). Due to their hydrophobic nature, TCC and TCS are likely to sorb with sediment containing high organic carbon content (Lei et al., 2009; Higgins et al., 2011). Miller et al. (2008) reported substantial amounts of TCS and TCC in dated estuarine sediment collected from Chesapeake Bay, Maryland, and Jamaica Bay, New York with their data suggesting that TCC may be more persistent than TCS in sediments. Although hydrophobic in nature both TCC and TCS have been reported in natural surface waters in the low to mid ng/L concentrations (Coogan et al., 2007; Kolpin et al., 2002; Kumar et al., 2010).

For several years, aquatic environment monitoring studies have primarily utilized active (grab, spot, or bottle) sampling techniques for qualitative and quantitative analysis. Active (water grab) sampling can require large sample volumes and it generally captures information at a very specific point in time, sometimes referred to as taking a 'snapshot'. Active sampling techniques may miss key events or transient inputs such as agricultural runoff, rain events, occasional industrial discharge, and changes in flow regime which can have a substantial effect on the concentration of pollutants in water over time (MacLeod et al., 2007; Greenwood et al., 2009). Other key drawbacks of active sampling that limit its utility include collection, transport, and storage of collected samples and the potential for chemical or microbial degradation prior to sample preparation or analysis (Greenwood et al., 2009).

Recently the passive sampler POCIS (polar organic chemical integrative sampler) has been developed to sequester and concentrate a wide variety of AOCs including pesticides, pharmaceuticals, plasticizers, preservatives, etc. in aquatic systems. POCIS has been widely used to monitor a variety of AOCs in the water due to its simplicity, ease of use, detection of low level concentrations, higher recovery rates, and resistance to biofouling (Alvarez et al., 2004, 2005). POCIS consists of a sequestration media, which is a solid-phase sorbent or sorbent mixture inside a microporous polyethersulfone (PES) membrane sandwiched between two stainless steel compression rings. A typical POCIS contains a water boundary layer, the diffusive membrane, and the sorbent (Greenwood et al., 2009; Alvarez et al., 2004).

To help address the continued need to better understand the fate of TCC and TCS in wastewater impacted streams and freshwater sediments, this study of the presence and transport of TCC and TCS in a wastewater dominated stream system was conducted. The key objectives addressed in the study presented here are (1) to determine and compare the concentration of TCC and TCS in surface water determined from water grab and POCIS sampling and (2) to conclude the primary transport mechanism of TCS and TCC in a wastewater-dominated stream (dissolved vs. suspended sediment).

2. Materials and methods

2.1. Chemicals and materials

Analytical standards of triclosan (97% pure) and triclocarban (99% pure) (Sigma–Aldrich Inc, St. Louis, MO 63103 USA) were