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Pharmaceutically active compounds: Their removal during slow sand filtration and their impact on slow sand filtration bacterial removal

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HIGHLIGHTS

- Pharmaceutically active compounds were removed in slow sand filters.
- Removal of caffeine, estrone and 17-beta estradiol was observed.
- Removal of carbamazepine, gemfibrozil, and phenazone was minimal.
- Biologically mature filters removed more than 99% of total coliforms and *E. coli*.
- Caffeine and estrogens may impact bacteria removal and *schmutzdecke* microorganisms.

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ABSTRACT

Slow sand filtration (SSF) has been widely used as a means of providing potable water due to its efficacy, low cost, and minimal maintenance. Advances in analytical instrumentation have revealed the occurrence of pharmaceutically active compounds (PhACs) in surface water as well as in groundwater. It is unclear if the presence of these compounds in the feed water can interfere with the performances of an SSF unit. The aim of this work was to examine i) the ability of two SSF units to remove six PhACs (caffeine, carbamazepine, 17-β estradiol [E2], estrone [E1], gemfibrozil, and phenazone), and ii) the impact of these PhACs on the removal of bacteria by two SSF units. The presence of PhACs in feed water for SSF can occur in surface waters impacted by wastewater or leakage from sewers and septic tanks, as well as in developing countries where unregulated use and improper disposal are prevalent. Two pilot-scale SSF units were used during the study. Unit B1 was fed with stream water with 1% of primary effluent added, while unit B2 was fed with stream water alone. Although limited removal (<10%) of carbamazepine, gemfibrozil, and phenazone occurred, the complete removal of caffeine, and the partial removal (11–92%) of E2 and E1 were observed in the two SSF units. The results of this study suggest that the occurrence of the selected PhACs, probably estrogens and caffeine, in the feed water at 50 μg L^{-1} affected the ability of the *schmutzdecke* to remove total coliform and *Escherichia coli*. The bacterial removal achieved within the *schmutzdecke* dropped from 95% to less than 20% by the end of the study. This decrease in removal may be related to the change in the microbial community within the *schmutzdecke*. A diverse microbial community, including Bacteroidetes and several classes of Proteobacteria, was replaced by a microbial community in which Gammaproteobacteria was the predominant phylum (99%). Despite the low removal achieved within the *schmutzdecke*, removal of total coliform and *E. coli* greater than 99% occurred after both SSF units throughout the study. Bacterial removal occurred in the upper half of the sand filter. This was probably due to a diverse microbial community established in the packing material, in which Bacteroidetes (13–25%), Acidobacteria (7–17%) and several classes of Proteobacteria (35–52%) (Alpha-, Beta-, Delta-, and Gammaproteobacteria) were the predominant phyla.

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1. Introduction

Slow sand filtration (SSF) is a treatment technology that has been widely used due to its low costs and the relative simplicity of construction and maintenance. SSF is accomplished by passing untreated water through a bed of sand having a diameter ranging between of 0.15 and

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0.4 mm (Huisman and Wood, 1974). As water passes through the sand, the *schmutzdecke* forms at the water–sand interface. This layer is gelatinous and typically consists of sand, algae, humus, bacteria and protozoa, and can account for most of the bacterial removal by SSF. With time, biological growth can also occur throughout the sand bed. Physical–chemical mechanisms are significant in the removal of particles between 0.75 and 10 μm (Weber-Shirk and Dick, 1997a), and biological mechanisms, principally bacterivory, are important for the removal of particles smaller than 2 μm (Weber-Shirk and Dick, 1997b).

SSF has been used primarily to remove turbidity and bacteria present in surface waters such as streams (Aydin, 1997; Bellamy et al., 1985; Cleasby et al., 1984) and lakes (Grützmacher et al., 2002; Weber-Shirk and Dick, 1997a,b) as well as secondary (Farooq and Al-Yousef, 1993; Logsdon et al., 1993) and tertiary effluent (Langenback et al., 2009). During the past decade, however, SSF has also been used to treat a broad variety of water such as rain water (Moreira Neto et al., 2012), wastewater from mariculture facilities (Palmer, 2010), and effluent from an up-flow anaerobic sludge blanket (Tyagi et al., 2009). Besides being used to remove turbidity and bacteria, SSF has been used to reduce chemical oxygen demand (COD), total phosphorous (TP), and total nitrogen (TN) from slightly contaminated water (Lwesya and Li, 2010), and iron and manganese from groundwater (Gottinger et al., 2011; Pacini et al., 2005).

Advances in analytical instrumentation that have occurred during the past few decades, revealed the presence of trace organic compounds, which includes personal care products, household chemicals, and pharmaceutically active compounds (PhACs) in environmental waters. Concentrations of PhACs in environmental waters usually range between ng L^{-1} to μg L^{-1} (Luo et al., 2014; Meffe and de Bustamante, 2014; Benotti et al., 2009; Verliefde et al., 2007; Kolpin et al., 2002), but PhACs are capable of disrupting the endocrine systems of many organisms even at low concentrations (Madureira et al., 2011; Vajda et al., 2008). Multiple studies have revealed the limited ability of conventional wastewater treatment to completely remove these compounds (Luo et al., 2014; Verlicchi et al., 2012; Dickenson et al., 2011). Therefore, their presence in surface water is a matter of concern because of the possible impact on the performance of natural filtration units and possible presence in potable water.

The effect of trace organic compounds on the performance of SSF units has been investigating in few studies (Oszako et al., 2013; Woudneh et al., 1997). For example, microbial removal achieved in an SSF unit was not impacted by the presence of 2,4-D herbicide, with feed water concentrations ranging between 6.5 and 10 μg L^{-1} (Woudneh et al., 1997), while reduced removal of bacteria was observed in the presence of feed water polluted with a mineral fertilizer and the fungicide pentachloronitrobenzene (PCNB) with a feed water concentration of 20 μg L^{-1} (Oszako et al., 2013).

A few studies examine the ability of SSF to remove trace organic compounds (Coelho and Di Bernardo, 2012; Hsieh et al., 2010; Woudneh et al., 1997); more specifically, PhACs (Rooklidge et al., 2005; Ternes et al., 2002). Coelho and Di Bernardo (2012) shows that limited removal of atrazine and its metabolites deethylatrazine, deisopropylatrazine, and diethylhydroxylamine was achieved using SSF alone, while higher removal can be achieved using an SSF unit with an intermediate layer of granular activated carbon. Bezafibrate, clofibric acid, diclofenac, carbamazepine, and primidone are not removed by a municipal SSF unit (Ternes et al., 2002). According to Rooklidge et al. (2005), at the end of a 14-day simulation period, 100% removal of tylosin, 99% removal of trimethoprim, <25% removal of lincomycin, and <4% removal of a sulfonamide class of antimicrobial can be achieved with an SSF unit.

We hypothesize that the presence of PhACs in surface water can affect the efficiency of an SSF unit. The occurrence of high concentrations of PhACs in surface water may be due to discharges from wastewater treatment plants, leakage from sewer pipes and cesspools, or the lack of regulations and enforcement for the disposal of these compounds. These conditions can occur in developing countries where more than 70% of industrial wastes are dumped untreated into waters where they can pollute the usable water supply (WWAP, 2009). Many cities in developing countries have inadequate or non-existing sewer infrastructure, are unable to keep up with rising population (Corcoran et al., 2010), and discharge an estimated 80 to 90% of all wastewater generated untreated directly into surface water bodies (UN Water, 2008). The relocation of global pharmaceutical industries from the West to developing Asian countries (i.e., Bangladesh, China, India, and Pakistan) has also posed serious threats to the environment, especially to the surface water, since most of the industrial units discharge wastewater into the domestic sewage network without any treatment (Rehman et al., 2013).

One objective of this study was to examine the ability of two SSF units to remove a variety of PhACs occurring in the feed water. Another objective was to examine the impact of selected PhACs on bacterial removal by two SSF units.

2. Materials and methods

2.1. Packing materials

Silica sand (0.015–0.030 cm, Orange County # 60) with a uniformity coefficient of 1.6 was used as the main packing material for the sand filters. Typical uniformity coefficients should range between 1.5 and 3.6 (LeChevalieer and Au, 2004; Cleasby and Logsdon, 1999; Letterman, 1991). Pea gravel with an effective size of 0.95 cm was used as the underlying supporting material for the sand in the SSF units.

2.2. Feed water

Two types of feed water were used: Manoa Stream with 1% primary effluent added and Manoa Stream alone. The 1% primary effluent was added to Manoa Stream water to increase the amount of naturally occurring bacteria in the feed water.

Manoa Stream (21°17′35.6″ N, 157°48′45.8″ W; Honolulu, HI, USA) runs by the University of Hawaii's main campus and is subject to urban pollution. Primary effluent was collected from the Honouliuli Wastewater Treatment Facility in Ewa Beach, Oahu, Hawaii, and stored at 4 °C prior to being used.

2.3. Chemicals

The PhACs used in this study were caffeine, carbamazepine, 17-β estradiol (E2), estrone (E1), gemfibrozil, and phenazone. The selected PhACs have been observed in surface waters and they represent different pharmaceutical classes: caffeine is a stimulant, carbamazepine is an anticonvulsant, E1 and E2 are estrogens, gemfibrozil is an antilipemic, and phenazone is an analgesic. E1 is the breakdown product of E2 (Lee and Liu, 2002). Caffeine and phenazone are hydrophilic ($\log K_{ow} < 3$), while the remaining PhACs are hydrophobic ($\log K_{ow} > 3$). The availability of analytical standards and adequate instrumentation also played a role in the selection of the six PhACs. Additional information related to the selected PhACs is given elsewhere (D'Alessio et al., 2015). Table SM-1 (Supplementary Material) provides the physico-chemical properties of the selected PhACs.

Caffeine (100% pure), carbamazepine (100% pure), E2 (100% pure), E1 (99% pure), gemfibrozil (99.9% pure), phenazone (99.9% pure), and potassium chloride (KCl) were purchased from Sigma-Aldrich (St. Louis, MO, USA); HPLC-grade methanol and acetonitrile were purchased from EMD Chemicals Inc. (Gibbstown, NJ, USA). The selected PhACs were added to the feed water at higher concentrations (50 μg L^{-1}) than normally found in surface waters in western countries (Meffe and de Bustamante, 2014; Lapworth et al., 2012; Hoppe-Jones et al., 2010; Kolpin et al., 2002) to mimic the high concentrations that may occur during discharges from wastewater treatment plants, or sewage

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