Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/watres

Year-long evaluation on the occurrence and fate of pharmaceuticals, personal care products, and endocrine disrupting chemicals in an urban drinking water treatment plant

Lokesh P. Padhye [a,1], *Hong Yao* [a,b], *Francis T. Kung'u* [c], *Ching-Hua Huang* [a,*]

[a] *School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, GA 30332, United States*
[b] *Department of Municipal and Environmental Engineering, Beijing Jiaotong University, Beijing 100044, People's Republic of China*
[c] *Integrated Circles Technologies, Inc., Atlanta, GA 30303, United States*

ARTICLE INFO

Article history:
Received 28 June 2013
Received in revised form
26 October 2013
Accepted 28 October 2013
Available online 7 November 2013

Keywords:
PPCPs
EDCs
DEET
Nonylphenol
Drinking water treatment
Ozonation
Indicator compounds

ABSTRACT

The occurrence and removal of thirty representative pharmaceutical and personal care products (PPCPs) and endocrine disrupting chemicals (EDCs) in an urban drinking water treatment plant (DWTP) were investigated for a period of one year to evaluate current system's treatment efficacy and assess occurrence of PPCPs and EDCs in finished drinking water. Results showed that the average total PPCPs and EDCs concentration in the surface water source was around 360 ng/L (median concentration = 340 ng/L) with 57% coefficient of variation (CV). The median concentrations of most of the individual PPCPs and EDCs in the surface water were below 15 ng/L except for *N*,*N*-diethyltoluamide (DEET) and nonylphenol, which were at 122 and 83 ng/L, respectively. The compounds DEET, nonylphenol, ibuprofen, triclosan, atrazine, tris(2-chloroethyl)-phosphate (TCEP), bisphenol-A, and caffeine (in the order of decreasing median concentration) were among twenty compounds detected at least once in the surface water, while all of the above detected compounds, except two, were also detected in the finished drinking water. The average total PPCPs and EDCs concentration in the finished drinking water was around 98 ng/L (median concentration = 96 ng/L) with 66% CV. The median concentrations of most detected PPCPs and EDCs in drinking water were below 5 ng/L except for DEET and nonylphenol, which were at 12 and 20 ng/L, respectively. There was a strong correlation ($r = 0.97$) between PPCPs and EDCs' concentrations in the source water and in the drinking water over the one-year study period when data points from two sampling events with unusual removals were excluded. Individual water treatment unit processes showed greater temporal variations of PPCPs and EDCs removal efficiencies than the overall treatment processes. The removal efficiencies also varied greatly among different PPCPs and EDCs. The average removal for total PPCPs and EDCs was 76 ± 18% at the DWTP, with ozonation showing the highest removal efficiency. Based on the similar occurrence and removal trends observed as that of total PPCPs and EDCs in this study, DEET and nonylphenol can be considered as

* *Corresponding author*. Tel.: +1 4048947694; fax: +1 4043857087.
E-mail address: ching-hua.huang@ce.gatech.edu (C.-H. Huang).

[1] Current address: Centre for Environmental Science and Engineering, Indian Institute of Technology Bombay, Powai, Mumbai 400076, India.

0043-1354/$ – see front matter
http://dx.doi.org/10.1016/j.watres.2013.10.070

potential indicator compounds for predicting the occurrence and removal of total PPCPs and EDCs in surface water. No strong correlations could be found between total PPCPs and EDCs removal and the removal of suspended solids, turbidity, or organic carbon.

1. Introduction

In the U.S., approximately 12,000 prescription pharmaceuticals and greater than 100,000 over-the-counter pharmaceuticals are distributed for human consumption (FDA, 2013). These formulations may contain one or more of approximately 3000 active ingredients that are designed to create specific physiological effects (Richardson and Ternes, 2011). Personal care products (fragrances, sunscreens, antimicrobials, etc.) encompass an even wider range of compounds. During the past decade, it has become apparent that many of these compounds are detected in various surface and ground waters (Heberer, 2002; Kolpin et al., 2002; Kummerer, 2001; Richardson, 2007; Focazio et al., 2008; Kuroda et al., 2011). Collectively these compounds are referred to as "pharmaceuticals and personal care products" (PPCPs). Wastewater treatment plants have been identified as a primary source of PPCPs in the aquatic environment (Focazio et al., 2008). Increase in human population, prolonged droughts experienced in many regions, and effects of climate change are putting strain on fresh water resources, and the propensity for source water contamination is expected to increase due to constant or increased loading from wastewater treatment plants and/or agricultural run-off (Snyder and Benotti, 2010). Currently, some communities in the United States draw raw water from sources within relatively short distances downstream of treated wastewater outfalls. Recognizing that most municipal drinking water plants do not have the capability to test drinking water for PPCPs routinely, it is probable that these compounds are present in drinking water in concentrations whose effects to humans and domestic animals are unknown.

Many PPCPs have been detected quite frequently in the low ng/L range to greater than 1 μg/L levels in the surface/source waters (e.g., Watkinson et al., 2009; Vulliet et al., 2011; Valcárcel et al., 2011). Although most PPCPs found in the drinking water are at trace levels (i.e., <1 μg/L) (Vulliet et al., 2011; Huerta-Fontela et al., 2011), and are not expected to cause adverse health effects, there may, however, be occasions when a number of contaminants with similar toxicological effects are present at levels near their respective guideline values (Bruce et al., 2010). Some of these PPCPs have been linked to adverse health and ecological effects including endocrine disruption, antibiotic resistance, inhibition of primary productivity, alteration of chemical communication, and others (Daughton and Ternes, 1999; Fent et al., 2006).

There are currently no federal regulations for PPCPs and endocrine disrupting chemicals (EDCs) in drinking or natural waters. However, in the state of California, monitoring pharmaceuticals and EDCs in recycled water is being conducted when municipal wastewater effluent is used for indirect potable reuse (Snyder et al., 2003). Pharmaceuticals and hormones were included on the U.S. EPA's Contaminant Candidate List 3 (CCL 3) for safe drinking water (USEPA, 2009). While studies have shown that conventional water and wastewater treatment processes are relatively ineffective in removing PPCPs and EDCs, advanced treatment technologies such as activated carbon, reverse osmosis and advanced oxidation may be viable for the removal of many trace PPCPs (Snyder et al., 2003; Vieno et al., 2007; Westerhoff et al., 2005).

Many studies have reported monitoring of PPCPs and EDCs in water and wastewater treatment systems in the recent decade. In general, considerably more research has been conducted for wastewater than for drinking water. For studies that evaluated PPCPs in drinking water, some emphasized on developing advanced analytical techniques to monitor a large suite of PPCPs for their occurrence (Stackelberg et al., 2004), some compared the levels of representative PPCPs in waters from drinking water treatment plants (DWTPs) with different treatment processes (e.g. Benotti et al., 2009; Wang et al., 2011), and some tracked select PPCPs throughout the treatment processes within a DWTP to evaluate treatment efficacy (e.g., Huerta-Fontela et al., 2011; Boleda et al., 2011). In this study, we employed composite samples to monitor the occurrence, removal and seasonal variation of PPCPs and EDCs at an urban DWTP from source to finish for the period of one year. Compared to previous studies, the significance of this study is that not only a wide range of PPCPs and EDCs (30 compounds) were analyzed, but also the occurrence and removal efficacy of PPCPs and EDCs were evaluated for seasonal patterns. The DWTP focused on in this study was a large facility located in a metropolitan area in the Southeastern U.S. which used river water as the source, and consisted of both conventional and advanced treatment processes common in many DWTPs in the U.S. and other countries. Hence, it could be considered as a representative for many urban drinking water treatment facilities.

The objective of this study was to evaluate a major urban DWTP's treatment efficacy for PPCPs and EDCs and to obtain a comprehensive understanding of the seasonal variation of the occurrence and removal of PPCPs and EDCs in source and finished drinking water. Based on the data obtained, evaluations were conducted to assess the impact of chemical properties and water quality parameters on the removal of PPCPs and EDCs, and to identify potential useful indicators that could be linked to the occurrence and fate of these contaminants.

2. Approach and experimental methods

2.1. Selection of PPCPs and EDCs

On the basis of extensive literature review and selection criteria (see Text S1), 30 PPCPs and EDCs consisting of 24