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Occurrence, removal and risk assessment of pharmaceutical and personal care products (PPCPs) in an advanced drinking water treatment plant (ADWTP) around Taihu Lake in China

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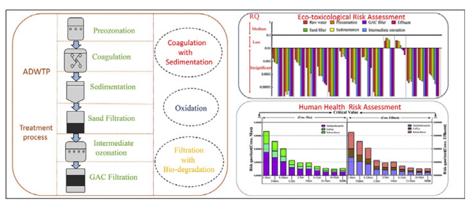
HIGHLIGHTS

- 39PPCPs were monitored at advance drinking water treatment plant near Taihu Lake.
- Three major effects were identified in the advance drinking water treatment plant.
- Roxithromycin and sulfamethoxazole were selected as two "prior indicator PPCPs".
- Uniform removal efficiency may not reflect an equal risk control of each PPCPs.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence and removal of 39 selected pharmaceutical and personal care products (PPCPs) were investigated in an advanced drinking water treatment plant (ADWTP) around Taihu Lake. Fourteen of 39 targeted pharmaceuticals were detected in the raw water. After a series of purification processes, only indomethacin, caffeine and sulfamethoxazole were found in effluent, albeit at concentrations less than 2 ng L^{-1}. The results of principal component analysis suggested that three main purification processes, oxidation, coagulation combined with sedimentation and filtration combined with bio-degradation, influenced the removal performance of PPCPs. The ecotoxicological and human health risk assessment confirmed that drugs detected in effluent posed no potential toxicity and also suggested that two PPCPs (roxithromycin and sulfamethoxazole), especially sulfamethoxazole, should be seriously considered as candidates for regulatory monitoring and prioritization. Finally, the correlation between removal efficiency and risk quotient indicated that uniform removal efficiency for all PPCPs may not reflect an equal risk control in the ADWTP.

1. Introduction

Recently, the occurrence of "emerging contaminants" in the aquatic environment has become a worldwide issue; among these, pharmaceuticals and personal care products (PPCPs) are of great concern (Arp, 2012; Fawell and Ong, 2012). Approximately 12,000 prescription pharmaceuticals are distributed for human

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consumption (FDA, 2013). PPCPs include a diverse collection of chemical substances, including human and veterinary drugs used for preventing or treating human and animal diseases, as well as disinfectants or fragrances used in personal care products (e.g., lotions, body cleaning products and sun-screens) and household chemicals for improving the quality of daily life (Boxall et al., 2012; Bu et al., 2013). Although PPCPs are commonly present in waters at trace concentrations from a few ng L^{-1} to several μg L^{-1}, these are enough to cause great threats to ecosystems or organism exposed (Vulliet et al., 2011). Many PPCPs are persistent in the environment and, once adsorbed, have the potential to bio-accumulate in organisms, which may cause endocrine disruption, antibiotic resistance, inhibition of primary productivity and other effects (Fent et al., 2006; Monteiro and Boxall, 2009; Mackay and Barnthouse, 2010). The PPCPs, due to low concentration, diversity and ecotoxicological potential, not only complicate the associated detection and analysis procedures but also create challenges for water purification.

Taihu, the third largest freshwater lake in China, is located in the Yangtze delta, one of the most developed areas in China. Taihu is a water source of drinking water supply for approximately 36 million people. It also has been used for agricultural and industrial purposes in several important cities such as Shanghai, Suzhou and Wuxi (Wang et al., 2011). With the rapid modernization of Chinese society and great population growth, greater quantities of domestic, industrial and hospital effluents, the main contributors of anthropogenic pollutants, have been discharged into surface waters that feed the lake, leading to trace concentrations of PPCPs (Zhang et al., 2012; Yan et al., 2014). It is essential to know whether a technology applied in the drinking water treatment plants (DWTP) is able to eliminate these refractory compounds. No study of PPCPs occurrence and removal in the DWTP, using Taihu Lake as a water source, yet exists, mainly due to the complicated detection of these anthropogenic pollutants in water treatment processes (Snyder and Benotti, 2010; Yang et al., 2011; Ratola et al., 2012). Previously, conventional water treatment was shown to be relatively ineffective in removing PPCPs (Vieno et al., 2007a). However, there is little research on the performance of the advanced drinking water treatment plant (ADWTP) processes in removing PPCPs from drinking water supply, especially in Taihu Lake.

Due to the frequent reports of PPCPs detection in Taihu Lake, the ecotoxicological and human health risk assessments to assess the collective potential environmental risk of multiple contaminants should be performed to guarantee safety of drinking water (He et al., 2013; Liu and Wong, 2013). However, there is a lack of research using such risk assessments to evaluate the performance of an ADWTP around Taihu Lake.

The objective of this work was to obtain a comprehensive understanding of the occurrence and removal of PPCPs in an ADWTP. The principal component analysis (PCA), usually used for reducing an original large data set to a small set of latent variables, was applied to investigate the major removal effects of PPCPs in the ADWTP with mathematical statistics. The chosen ADWTP was located in the Taihu region in eastern China, and was representative of many DWTPs around Taihu Lake. To avoid problems with seasonal variations in PPCP concentrations due to different rainfalls, as well as the effects of temperature variation on treatment processes (Loraine and Pettigrove, 2006; Sun et al., 2014), we performed our investigation in October and November when the rainfall and temperature were “average” (Fig. S1) and removal efficacy in the ADWTP was believed to be stable. We used the ecotoxicological and human health risk assessments to assess the potential environmental risk of a collection of PPCPs and to evaluate toxicity variations in individual steps within the treatment process.

2. Methods and materials

2.1. Chemicals and selected PPCPs

As seen in Table S1, 39 PPCPs were selected for monitoring. The selected PPCPs included antibiotics, β-blocking drug, blood lipid regulator, blood circulation drug, anti-arrhythmia, cardiovascular drug, anti-convulsant drug, scabicide, insecticide, lean meat powder, calcium antagonist, anticholinergic drug, antipsychotic drug and emictory. In each classes, compounds that are frequently detected were considered. Drugs such as sulfamethoxazole, erythromycin, trimethoprim, lincomycin, caffeine, N,N-Diethyl-3-methylbenzoylamide (DEET), acetaminophen, roxithromycin, indomethacin and atenolol were among the 30 most frequently detected PPCPs as reported by the US Geological Survey (Esplugas et al., 2007). Erythromycin, sulfamethoxazole, carbamazepine, ibuprofen, and diclofenac were among the top 10 high priority pharmaceuticals identified in a European assessment of PPCPs (Voogt et al., 2008). The drugs such as clarithromycin, lincomycin, sulfamethoxazole, roxithromycin, tiamulin, antipyrine, caffeine, propyphenazone, sulpiride, DEET, sulfapyridine and naproxen were frequent detected and studied in surface water especially in Taihu Lake (Ikehata et al., 2006; Liu et al., 2014; Yan et al., 2014). The other drugs, such as azithromycin, 2-Quinoxaline carboxylic acid (QCA), primidone, etc, were not frequently detected in Taihu Lake but were warned to be cautioned by the Chinese Food and Drug Administration (CFDA) (Luo et al., 2011; Liu and Wong, 2013).

Analytical-standard pharmaceuticals were purchased from Toronto Research Chemicals (Toronto, Canada), Tokyo Chemicals Industry (Tokyo, Japan) and Dr. Ehrenstorfer GmbH (Augsburg, Germany). Reagent-grade HCl, methanol, and other reagents were obtained from Sigma–Aldrich (Steinheim, Germany). All deionized water used in the experiments was produced by a Milli-Q unit (Millipore, MA, USA). Stock solutions of individual PPCPs were prepared in methanol and standard mixtures were prepared by diluting the stock solution. All the standard solutions were stored at −20 °C in the dark.

2.2. Sampling sites and sample collection

The ADWTP examined in this study receives raw water from Gong Bay, a vital surface water source in the Taihu region, and supplies a population exceeding ten million. The treatment process of the ADWTP is displayed in Fig. S2.

Grab samples were collected in 2 L amber glass bottles from each unit treatment process; the timing of sample collections from the influent to, and effluent from, each unit process took consideration of the hydraulic retention time of each process to assess the removal efficiency on the same theoretical unit of water as it moved through the plant. Sodium thiosulfate (250 mg L^{-1}) was added to quench any residual oxidant. Samples were collected once a week. The collection was performed in five consecutive weeks. The samples were refrigerated soon after collection and subsequently analyzed within 48 h. Each sample was analyzed in five parallels. Characteristics of the collected samples are reported in Table S2. The parameters in Table S2 were the mean values. Based on the study by Padhye et al. (2014), we also defined those PPCPs with total detected frequency below 50% as compounds with no expressed mean concentration, noted as BL signs.

2.3. Sample extraction and analysis

In this study, the raw water and final effluent of the ADWTP were sampled and, in order to study the removal efficiency of each treatment units in ADWTP, the influent and effluent of each