



ELSEVIER

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

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

Sources, distribution and potential risks of pharmaceuticals and personal care products in Qingshan Lake basin, Eastern China

Saichang Zhu, Hong Chen*, Jianan Li

Department of Environmental Engineering, College of Environmental and Resource Sciences, Zhejiang University, Hangzhou 310058, China

ARTICLE INFO

Article history:

Received 9 April 2013

Received in revised form

27 June 2013

Accepted 28 June 2013

Available online 18 July 2013

Keywords:

Pharmaceuticals and personal care products

Seasonal variation

Backup drinking water source

Ecological risk

ABSTRACT

In this study, we examined the occurrence and distribution of 12 selected pharmaceutical and personal care products (PPCPs) in Qingshan Lake basin and evaluated their potential ecological risks. Caffeine was the dominant compound among all selected PPCPs in the aqueous phase (23.8–344.7 ng/L). Sediment release contributed minimally to caffeine levels found in the aqueous phase of Qingshan Lake. Concentrations of selected PPCPs in dry season water samples were generally higher than those in the water samples obtained during the wet or median water seasons. The risks of residual caffeine in water on aqueous organisms was minimal (risk quotient values below 0.01). Sulfamethoxazole and triclocarban posed the highest environmental risks despite their low concentrations (below 25 ng/L in water and 10 µg/kg in sediment) in Qingshan Lake basin. The risks produced by selected PPCPs in sediments were generally higher than in the aqueous phase.

© 2013 Elsevier Inc. All rights reserved.

1. Introduction

Pharmaceuticals and personal care products (PPCPs) are emerging contaminants that have increased the interest of researchers over the last decade. Various PPCP compounds have been detected frequently in effluents from sewage treatment plants, hospitals, livestock farms, pharmaceutical manufacturers, slaughterhouses, and households (Sim et al., 2011; Kosma et al., 2010; Chang et al., 2010). Synergic effects of the continuous introduction of PPCPs might have unwanted effects on the environment (Cleavers, 2003). Residual PPCPs in the environment might disturb the normal metabolism of organisms, thereby producing a toxic effect on organisms or inducing the proliferation of drug-resistant strains (Schnell et al., 2009; Schreurs et al., 2005; Schlumpf et al., 1998). Therefore, the potential environmental risks of residual PPCPs should not be ignored.

PPCPs have been widely detected in the waters of UK, Spain, Sweden, Japan, Korea, and Romania with concentrations varying from ng/L to µg/L and their occurrence in water vary greatly across regions and seasons (Bendz et al., 2005; Kasprzyk-Hordern et al., 2008; Kuster et al., 2008; Moldovan, 2006; Murata et al., 2011; Yoon et al., 2010). External factors such as regional features of land use, proximity to wastewater effluents, and weather conditions (mainly rainfall) could affect PPCP occurrence in the environment (Murata et al., 2011; Kasprzyk-Hordern et al., 2008; Zhao et al., 2010b; Chen et al., 2012). Sediments are considered important sinks for some PPCPs in water (Zhao et al., 2011; Schultz et al., 2010). The hydrophobicity and

chemical structure of organic contaminants might affect their sorption to sediments (Eggleton and Thomas, 2004).

In China, a number of reports have focused on the occurrence and distribution of PPCPs in the Pearl River, Yellow River, Hai River, Liao River, and Huangpu River (Zhao et al., 2010b; Wang et al., 2010; Jiang et al., 2011). However, few studies have concentrated on the distribution and effect of PPCPs in lake basins, especially on drinking water sources. The surface waters of China and the potential environmental impact of PPCPs relative to the large population and huge annual consumption of PPCPs in China should be investigated further to obtain a clear picture of PPCPs (Wang et al., 2010).

Qingshan Lake, which is located west of Linan County, Hangzhou and ultimately drains to the Hangzhou Bay in the East China Sea, is a backup drinking water source. Potential sources include two main rivers, South Tiaoxi River and Jinxi River, upstream of the lake and a municipal sewage plant with a daily treatment volume of 60,000 t (Fig. 1). We have chosen 12 PPCPs as our target analytes (Table S1, Chen et al., 2012; Yu and Chu, 2009; Zhang et al., 2012). The occurrence, distribution, and seasonal variation of 12 PPCPs were studied in South Tiaoxi River, Jinxi River, and effluent from the municipal sewage plant. Furthermore, the ecological risks of residual PPCPs in water and sediment were discussed.

2. Material and methods

2.1. Chemicals

Trimethoprim (99.5 percent), penicillin G (99.0 percent), penicillin V potassium salt (98.8 percent), caffeine (98.5 percent), sulfamethoxazole (99.0 percent),

* Corresponding author. Fax: +86 571 8898 2028.

E-mail address: chen_hong@zju.edu.cn (H. Chen).

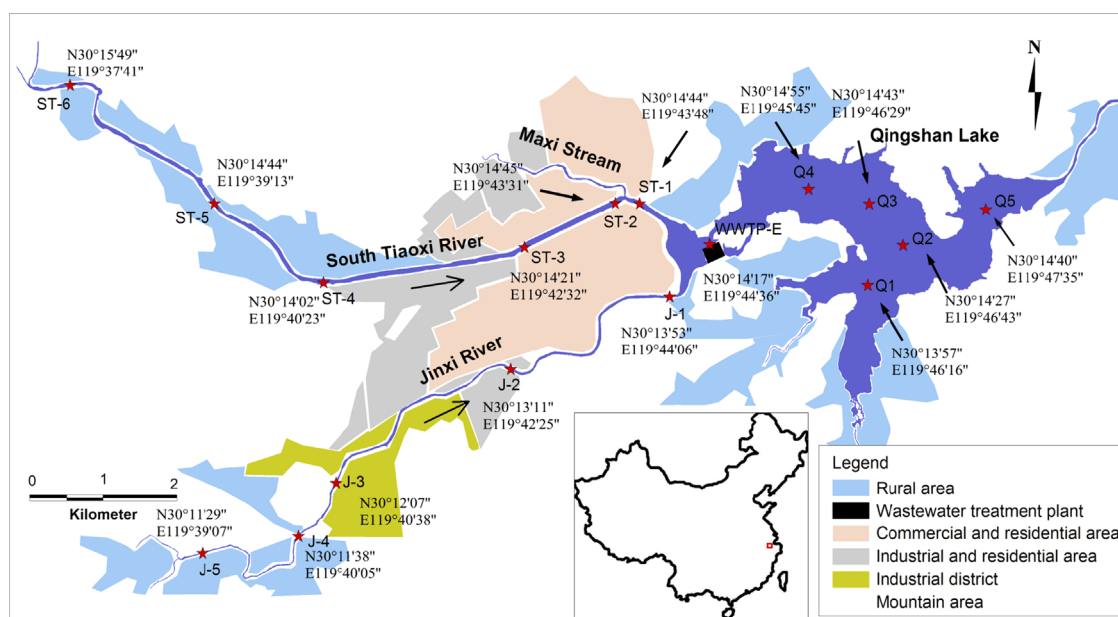


Fig. 1. Sampling sites of Qingshan Lake basin.

carbamazepine (99.5 percent), N,N-diethyl-3-methylbenzoylamide (98.0 percent), ibuprofen (99.0 percent), triclosan (99.5 percent), and triclocarban (99.5 percent) were purchased from Dr. Ehrenstorfer GmbH (Germany); and diclofenac sodium salt and naproxen of analytical grade with a purity of 98 percent or higher were purchased from Sigma Aldrich (USA). Detailed information about all the compounds is listed in Supplementary materials Table S1. Methanol (LC/MS grade) and acetonitrile (LC grade) were purchased from Fisher (USA); formic acid (LC grade) was purchased from Tedia Company; and ammonium acetate was purchased from Merck KGa (Germany). Na₂EDTA (analytical standard) was purchased from Sino-pharm (China). Ultra-pure water was produced by a Milli-Q unit (Millipore, Japan). Stock solutions of individual compounds were respectively prepared at 1 mg/mL in methanol and stored in dark at -20°C . New stock solutions were made every 6 months. Standard solutions were prepared by diluting stock solutions with methanol.

2.2. Sampling

Samples were collected from Qingshan Lake basin. Qingshan Lake is spread across 10 km² and an average water stage of 25 m throughout the year. Two main rivers, namely, South Tiaoxi River and Jinxi River, drain into Qingshan Lake. South Tiaoxi River is a mountain stream (the total length is 63 km) with a drainage spread across 620.8 km² and an average annual flow of 14.5 m³ s⁻¹. About 87 percent of the water area of South Tiaoxi River is distributed over Linan County. South Tiaoxi River is joined by its tributary Maxi stream in the downstream area and drains into Qingshan Lake after flowing with Jinxi River. Jinxi River, which has an overall length of 25.5 km and a catchment area of 84.6 km², is one of the biggest tributaries of South Tiaoxi River. It flows through rural areas, industrial districts, industrial-residential areas, and commercial-residential areas from upstream to downstream. The study area is a humid subtropical monsoon climate zone characterized by abundant sunshine and adequate rainfall. The wet season is from May to September, and the dry season is from December to February. The median water season occurs at October–November and March–April.

In accordance with a functional land use map, six and five sampling sites were established along the banks of South Tiaoxi River and Jinxi River, respectively (Fig. 1). Five sampling sites were also established based on the shape and area of Qingshan Lake. A municipal wastewater treatment plant (WWTP-E), which has a daily treatment volume of 60,000 t, is located near the influx of South Tiaoxi River and Jinxi River into Qingshan Lake. Samplings were conducted thrice, i.e., during the wet season (August), the median water season (October), and the dry season (December). The average daily temperatures of the study area during August, October, and December ranged from 25 °C to 33 °C, 15 °C to 2 °C, and 3 °C to 11 °C, respectively, and the average precipitation was 148, 78, and 51 mm, respectively. Grab water samples were collected by a water sampler from the South Tiaoxi River, Jinxi River, Qingshan Lake, and effluents of the WWTP-E during each study period. All the lake and river samples were collected approximately 50 cm below the water surface in a boat or from the bridges over the rivers (Zhang et al., 2012). Five sediment samples were also collected from Qingshan Lake by a sediment

sampler. All samples were stored in amber glass bottles in dark at 4 °C for no longer than 2 days.

2.3. Sample treatment and instrumental analysis

Aqueous samples were filtered through 0.45 μm glass fiber filters after sampling and later extracted by solid phase extraction (SPE) within 48 h. 1 L filtered aqueous samples were pretreated as follows: adjust pH to 6.5 ± 0.5 with 5 M HCl and NaOH; add 2 g/L Na₂EDTA and react for 1 h with a shake every 30 min. Oasis HLB cartridges (6 mL/500 mg, Waters, USA) were successively activated with 10.0 mL of methanol and 10.0 mL of ultra-pure water. Samples were passed through cartridges at a flow rate of 2 mL/min. After that, cartridges were rinsed with 20 mL ultra-pure water and later dried under gentle nitrogen gas for 15 min. Then cartridges were eluted with 4 mL mixture of methanol:acetonitrile (1:1, v/v) for three times. Elutes were concentrated to 1 mL under gentle nitrogen gas at 35 °C and later brought to a final volume of 2 mL with methanol. Treated samples were analyzed by Ultra-high Performance Liquid Chromatography tandem Mass Spectrometry (UPLC-MS/MS) within 40 days and stored under -20°C in dark until analysis.

Solid samples were firstly extracted by Pressurized Fluid Extraction (PFE) using an ASE 300 system (Dionex, Sunnyvale, CA, USA) equipped with a solvent controller. Wet sludge samples were lyophilized, ground and sieved (80 mesh). A 0.2 μm filter was placed at the bottom of the extraction cell to avoid the obstruction of the end caps by the solid particles. An aliquot of 1 g dried sediment sample was accurately weighed. Samples were transferred into 33 mL extraction cells partly filled with quartz sand. During mixing, more sand was added until the cell was completely filled. After preheating the ASE 300 system for 5 min, samples were heated to 80 °C and extracted by a flush volume of 100 percent in three cycles using mixture of methanol:acetonitrile:water (1:1:2, v/v/v). The pressure was set to 1500 psi and a purge time of 60 s. The final extraction volume ranged from 70 to 75 mL. The PLE extracts were evaporated to reduce the organic phase to about 10 mL by a rotary evaporator at 60 °C. Then extracts were diluted by 800 mL ultra-pure water to control the organic phase to below two percent. Then the diluted extracts were cleaned up by the SPE procedure following the method mentioned above.

The final extracts were analyzed by the Ultra-high Performance Liquid Chromatography tandem Mass Spectrometry (UPLC-MS/MS) system equipped with an Acquity™ UPLC and a Quattro Premier Micromass[®] MS (Waters/Micromass, Milford, MA). All target compounds were firstly separated by a BEH C₁₈ column (Waters Corp., 50 mm × 2.1 mm length, 1.7 μm) and later identified and quantified by the MS/MS system using the multi-reaction monitoring (MRM) mode. Instrumental conditions varied with analytical groups and the MS/MS analysis was performed in the positive or negative electrospray ionization (ESI) mode. The combination of standard addition method and the single-point method was employed to determine target compounds in samples. Briefly, the sample was directly injected into the system to produce response values. Then certain amounts of target compounds were spiked, after which the sample was injected again to produce another values. The target compounds in the sample were determined through

Download English Version:

<https://daneshyari.com/en/article/6312357>

Download Persian Version:

<https://daneshyari.com/article/6312357>

[Daneshyari.com](https://daneshyari.com)