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Occurrence and potential human health risks of semi-volatile organic compounds in drinking water from cities along the Chinese coastland of the Yellow Sea

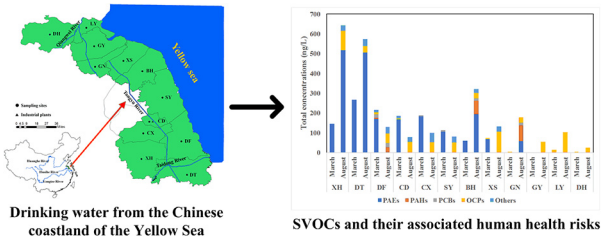
Zekai Li [a, 1], Fangyu Chang [a, 1], Peng Shi [a, *], Xun Chen [a], Feng Yang [b], Qing Zhou [a], Yang Pan [a], Aimin Li [a]

[a] State Key Laboratory of Pollution Control and Resources Reuse, School of the Environment, Nanjing University, Nanjing 210023, China
[b] Nanjing University & Yancheng Academy of Environmental Protection Technology and Engineering, Yancheng 224002, China

HIGHLIGHTS

- SVOC contamination and human health risks of drinking water were investigated.
- Spatial and temporal differences of the SVOCs were observed in drinking water.
- Benzene hexachloride and heptachlor contributed most of carcinogenic risks.
- Drinking water of several cities exhibited high carcinogenic risks.

GRAPHICAL ABSTRACT



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ABSTRACT

Semi-volatile organic compounds (SVOCs) in drinking water have been considered a severe threat to public health worldwide. However, SVOC contamination and the associated human health risks of the drinking water from cities along tributaries of the Yangtze River and Huaihe River in China have been seldom reported. Here, we focused on the occurrence and distribution of a series of SVOCs, mainly including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and phthalate esters (PAEs), in drinking water of this region. In brief, a total of 31 SVOCs were detectable in all of the drinking water samples, and the total concentrations ranged from 0.92 ng/L to 266.16 ng/L in March and from 24.82 ng/L to 643.93 ng/L in August, with the highest concentrations in Dongtai (DT) and Xinghua (XH), respectively. Spatial and temporal differences of the SVOCs were also observed in drinking water samples, demonstrating the linkage between pollutant profiles and geographical locations, as well as exogenous wastewater discharge. Moreover, PAEs occupied 79.17 −100.00% of the total concentrations of SVOCs in drinking water samples collected from the tributaries of the Yangtze River in March, while OCPs were the predominant SVOCs in most of drinking water samples from the tributaries of the Huaihe River. The human health assessment indicated that SVOCs posed negligible non-carcinogenic risks, but residents living in DT, Dafeng (DF), Chengdong (CD), Guanyun (GY) and Lianyungang (LY) may suffer carcinogenic health risks, which could be mainly induced by benzene hexachloride and heptachlor in August.

* Corresponding author. State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, 163 Xianlin Avenue, Nanjing 210023, China.
E-mail address: shipeng@nju.edu.cn (P. Shi).
[1] The two authors contribute equally to this work.

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1. Introduction

Semi-volatile organic compounds (SVOCs) in drinking water that cause deleterious effects on exposed human beings have received great attention in recent years (Fatta et al., 2007; Cai et al., 2012; Tang et al., 2017). The Yangtze River and the Huaihe River are two representative streams that nourish millions of people, and both flow into the Yellow Sea. Their lower reach, especially the Chinese coastland of the Yellow Sea, is one of the most developed area in China (Liu, 2010). However, a booming economy has resulted in severe water pollution in these two rivers through intense human activities such as industrial activities (Gerecke et al., 2002; Cai et al., 2012), agricultural irrigations (Casajuana and Lacorte, 2003) and domestic wastewater discharges (Doong and Lin, 2004), which may pose great human health risks via the consumption of drinking water, inhalation and dermal contact (Shi et al., 2018).

Previous studies have revealed that the drinking water of main cities along the Yangtze River and the Huaihe River were contaminated by a series of SVOCs, including polychlorinated biphenyls (PCBs) (Xing et al., 2005; Bao et al., 2012), organochlorine pesticides (OCPs) (Wang et al., 2009a; Feng et al., 2011; Zhao et al., 2016), substituted benzenes (Ma et al., 2005), polycyclic aromatic hydrocarbons (PAHs) (Huang et al., 2004; Wu et al., 2009a, 2009b; Liu et al., 2016), phthalic ester (PAEs) (Zhang et al., 2011a; Li et al., 2012) and environmental endocrine disruptors (Zhang et al., 2011a; Li et al., 2012). The existence of these contaminants in drinking water could potentially bring adverse effects to local residents, e.g., reproductive disorders (Axmon et al., 2008), behavioral abnormalities, genetic mutations (Huang et al., 2004), and even cancers (Yoon et al., 2007; Yang et al., 2017). It was reported that the esophageal cancer incidence of the northern Jiangsu Province of China was over 80/100,000 individuals, which was six times greater than the national average rate (13/100,000) (Wang et al., 2006). Approximately 80% of the cancer occurrence was supposed to relate to environmental factors, of which water pollution was the most important one (Wu et al., 2010). However, previous studies have mainly focused on the occurrence and fates of SVOCs in drinking water from the main streams of these two rivers (Casajuana and Lacorte, 2003; Tang et al., 2008; Wang et al., 2009a; Bao et al., 2012), but the pollutant profile of drinking water from the tributaries has usually been ignored. Unfortunately, the tributaries of both these rivers join together at the Chinese coastland of the Yellow Sea, and the pollutant concentrations in the river tributaries may be higher than those in the main streams due to the low volume of water and the upstream influence. Moreover, the river tributaries in this region also receive pollutants from the flourishing local industrial and agricultural activities (Wang et al., 2009b), which may also exert negative impacts on the water quality. Importantly, more than 30 million inhabitants in this region are feed with drinking water from the river tributaries, so it is imperative to systematically investigate the occurrence, as well as spatial and temporal variations of the SVOCs in drinking water of this region. Moreover, increasing number of pollutants were found in aquatic environments with the development of analytical instruments, but it is hard for us to evaluate whether the pollutants may pose health risks to human only according to the type and concentration. Human health risk assessment models by the Environmental Protection Agency of the United States (USEPA), which are designed to estimate the likelihood and extent of adverse effects, have been applied to determine the potential health risks of organic pollutants in drinking water worldwide (Tchounwou et al., 2002; Muhammad et al., 2011; Dominguez-Morueco et al., 2014). Several studies surveyed the levels of organic contaminants and their associated human health risks of drinking water in large cities such as Nanjing (Wu et al., 2010), Wuxi (Hu et al., 2015), Changzhou (Zhang et al., 2011a), and Huaian (Wang et al., 2009a) along the main streams of the Yangtze River and the Huaihe River. However, human health risks in drinking water from cities along the Chinese coastland of the Yellow Sea are still unknown.

This study aimed to determine the occurrence and distribution of a series of SVOCs, mainly including PAHs, PCBs, PAEs and OCPs, in drinking water from cities along the Chinese coastland of the Yellow Sea. The high-risk points and major contributing pollutants were also identified by calculating the non-carcinogenic and carcinogenic health risks of all of the detected SVOCs. This study may provide useful information for selecting proper advanced drinking water treatment processes, and serve as a benchmark for policy makers to reduce health risks arising from SVOCs.

2. Materials and methods

2.1. Standards and reagents

A total of 74 kinds of SVOCs, including 16 PAEs, 16 PAHs, 15 PCBs, 23 OCPs and 4 types of other SVOCs, were all purchased from J&K Scientific LTD., Co. (Shanghai, China). Methyl alcohol, methylene chloride and ethyl acetate were of HPLC grade and were supplied by Merck Company (Darmstadt, Germany). The full and abbreviated name, purity, CAS number and supplier of each compound are provided in Supplementary Material Table S1.

2.2. Sample collection and pretreatment

We sampled drinking water from 12 cities including Xinghua (XH), Dongtai (DT), Dafeng (DF), Chengdong (CD), Chengxi (CX), Sheyang (SY), Binghai (BH), Xiangshui (XS), Guannan (GN), Guanyun (GY), Lianyungang (LY) and Donghai (DH) along the Chinese coastland of the Yellow Sea in March and August of 2014. These 12 cities obtain source water from the Taidong River (XH and DT), Tongyu River (DF, CD, CX, SY, BH, and XS) and Qiangwei River (GN, GY, LY, and DH), which are the three main tributaries of the Yangtze River and the Huaihe River (Fig. 1). Drinking water samples were collected from the waterworks of each city. The water taps were kept flowing for at least 10 min before collection to minimize the pipeline effects. Residual chlorine was eliminated by adding 500 mg of ascorbic acid, and the pH was adjusted to be below 2.0 by adding 5 mL of concentrated hydrochloric acid. Water samples were sealed and then transported to the lab within 4 h before filtration with 0.45-μm glass fiber filters. The filtered water was stored in 1-L brown glass containers to avoid light at 4 °C before further treatments.

2.3. Sample extraction and instrumental analysis

A solid phase extraction (SPE) was performed to concentrate the 1 L filtered water samples by 1000-fold using 500 mg C_{18} cartridges (CNWBONND HC-C_{18}, ANPEL, China). The cartridge was sequentially washed and conditioned with 5 mL each of methylene chloride, ethyl acetate and a mixture of the two (1:1 of v/v) and with 10 mL each of methanol and milli-Q water. A vacuum pump was used to control the water flow at a constant rate of 10 mL/min. After extraction, the cartridge was washed with 10 mL of milli-Q water and dried with a gentle stream of nitrogen gas (99.999% pure) for 5 min. Analytes were eluted with the same solvents used in cartridge activation and conditioning. The eluent was first concentrated to 2–3 mL using a rotary evaporator and then further blown to dryness under a gentle nitrogen flow. Finally, the analytes were re-dissolved with 1 mL of methylene chloride and then transferred to a 2-mL sample vial before analysis.