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Bisphenol analogues in surface water and sediment from the shallow Chinese freshwater lakes: Occurrence, distribution, source apportionment, and ecological and human health risk

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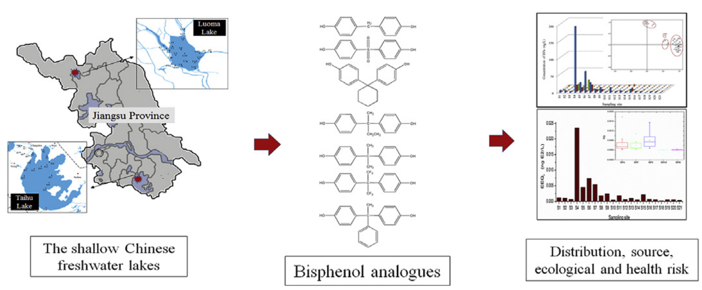
HIGHLIGHTS

- Except BPA, BPF and BPS are predominant in Taihu Lake and Luoma Lake.
- The distribution of BPs in Luoma Lake is more homogeneous than that in Taihu Lake.
- Compared to Taihu Lake, the sources of BPs in Luoma Lake are more simple.
- The distribution of BPs in sediment is positively related with the TOC content.
- No high eco-toxicity or estrogenic risk was found in Taihu Lake and Luoma Lake.

GRAPHICAL ABSTRACT



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ABSTRACT

Compared to Bisphenol A (BPA), current knowledge on the spatial distribution, potential sources and environmental risk assessment of other bisphenol analogues (BPs) remains limited. The occurrence, distribution and sources of seven BPs were investigated in the surface water and sediment from Taihu Lake and Luoma Lake, which are the Chinese shallow freshwater lakes. Because there are many industries and living areas around Taihu Lake, the total concentrations of ∑BPs were much higher than that in Luoma Lake, which is away from the industry-intensive areas. For the two lakes, BPA was still the dominant BPs in both surface water and sediment, followed by BPF and BPS. The spatial distribution and principal component analysis showed that BPs in Luoma Lake was relatively homogeneous and the potential sources were relatively simple than that in Taihu Lake. The spatial distribution of BPs in sediment of Taihu Lake indicated that ∑BPs positively correlated with the TOC content. For both Taihu Lake and Luoma Lake, the risk assessment at the sampling sites showed that no high risk in surface water and sediment (RQ_t < 1.0, and EEQ_t < 1.0 ng E_2/L).

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1. Introduction

Bisphenol analogues (BPs), as a group of anthropogenic chemicals with two hydroxyphenyl functionalities, are extensively used to produce polycarbonates and epoxy resins (Liao et al., 2012a; Ruan et al., 2015; Huang et al., 2012). Because of the estrogenic and anti-androgenic properties, the use of BPA has been one of the most debated examples (Kitamura et al., 2005; Roelofs et al., 2015). Some countries have limited or banned the use of BPA in child care products or thermal receipt papers, such as Canada, Japan and EU (Liao et al., 2012b). Bisphenol AF (BPAF), bisphenol AP (BPAP), bisphenol B (BPB), bisphenol Z (BPZ), bisphenol S (BPS) and bisphenol F (BPF) are developed and synthesized to replace BPA as the intermediates in the manufacturing of polycarbonate plastics, fire-resistant polymers, thermal paper and the resin lining of food and beverage cans (Liao et al., 2012c; Vandenberg et al., 2012; Xue et al., 2015). However, the similar toxicological profiles have been found for several BPA analogues, such as nerve toxicity, developmental toxicity and endocrine disruption (Eladak et al., 2015; Héliès-Toussaint et al., 2014; Kitamura et al., 2005; Svajger et al., 2016). Owing to the widespread uses of essential applications from electronics to food protection, large amount of these chemicals has been discharged into the aquatic environment (Erjavec et al., 2016).

Compared to BPA, data on the occurrence, distribution and environmental risk of BPA alternatives are scarce. BPS and BPF have been reported to occur in surface water (Fromme et al., 2002), sewage sludge (Lee et al., 2015; Yu et al., 2015) and sediments (Fromme et al., 2002) and were the predominant BPs in sediment samples obtained from North America and several Asian countries. BPZ, BPAF and BPAP were detected in the river collected from Japan, China, Korea and India (Liao et al., 2012c; Yamazaki et al., 2015). Chen et al. (2016) has reported that the estrogenic and/or anti-androgenic activities of BPAF, BPB, BPF and BPS were similar or greater than that of BPA. Therefore, it is necessary to monitor the occurrence of BPs in the aquatic environment for their environmental control and management.

Taihu Lake is a typical water source around the industrial area, which is surrounded by the cities of Changzhou, Wuxi, Suzhou and Huzhou, with an area of 2338 km^2 and an average depth of 1.9 m (Tao et al., 2013). It provides the main source of freshwater to support human, fisheries, irrigation and industrial consumption in the surrounding area (Xie et al., 2015). However, the pollution of Taihu Lake by anthropogenic organic pollutants has received growing attention (Lin et al., 2016; Liu et al., 2016). Jin and Zhu (2016) has reported that nine bisphenol analogues were measured in water and sediment samples from Taihu Lake and this has become a not allow to neglect problem.

Meanwhile, perspectives of recent studies heavily focused on the occurrence and distribution of contaminants in the water samples collected from lake and/or river surrounded by industrial districts, while ignored the possibility of pollution in the water bodies away from the industry-intensive areas. However, pollutant molecules will be transmitted to the so-called clean areas via waterfall and air movement (Jie et al., 2016; Ter Schure et al., 2004; Zakari et al., 2015). Luoma Lake, which is the one of the four largest lakes in Jiangsu province, located in the north section of Jiangsu province, China (Huang et al., 2011), with an average water depth of 3.3 m and a surface water area of 260 km^2. Luoma Lake is the 4A scenery spot in Jiangsu, a man-controlled, lightly eutrophic reservoir and also part of the transfer stations of South-to-North Water Transfer Project (Feng et al., 2007). Because of few factories around this water, Luoma Lake was once widely thought of “clean water”. Recently, most research focused on the water eutrophication and the microbial communities in Luoma Lake (Ren et al., 2015; Zhao et al., 2010). To our knowledge, few data have dealt comprehensively with the degree of organic pollution in the waters.

Thus, it is extremely urgent to monitor the occurrence of BPs in the freshwaters to systematically evaluate their ecological risk and provide systemic guide for risk assessment of them. The aims of the present study were (i) to study the concentration levels and distribution characteristics of target BPs in the surface water and sediment in Taihu Lake and Luoma Lake and compare the difference of the distribution characters between the two lakes; (ii) to provide new evidence and more comprehensive fundamental data for the potential risk of BPs. To our best knowledge, this is the first study to report the comparison data of bisphenol analogues in waters and sediments collected from the watersheds in the industrial zone and non-industrial zone.

2. Materials and methods

2.1. Chemicals and reagents

Standards for all chemicals including BPA, BPAF, BPAP, BPB, BPZ, BPS and BPF were purchased from J&K Chemical, Ltd. (St. Louis, MO, USA). Detailed substance information for all the target chemicals is listed in Table S1. All the solvents used in this study were of high-performance liquid chromatography (HPLC) grade.

2.2. Sampling sites and sample collection

The study areas and sampling sites were shown in Fig. 1 and the details for the sampling sites were shown in Table S2. Taihu Lake (30°55′-31°32′N; 119°52′-120°36′E) is located in southern Jiangsu Province, while Luoma Lake (34̊00′−34̊11′N, 118̊06′−118̊18′E) is located in northern Jiangsu province, near the city of Suqian. In consideration of anthropogenic activities, 21 sampling sites were chosen around Taihu Lake and its tributaries. For Luoma Lake, the surface water samples (0–1 m) were collected from 22 sites, including 14 sites from Luoma Lake (L1-14), 4 sites from two inflowing rivers, i.e., Yi River and Fangting River (Y1/2, and F1/2) and 4 sites from outflowing river, i.e., Zhongyun River and Zhangshan Gate (ZY1/2 and Z1/2). Due to many sands and gravels in Luoma Lake, the sediment samples (0–0.1 m) were collected only from 6 sites, i.e., L1, 3, 9, 10, 11 and 12, which distributed in the estuary, the center and the outlet of the lake. Water and sediment samples were collected from Luoma Lake in November 2015, while them from Taihu Lake in April 2016. 2 L pre-cleaned brown glass bottles were used to preserve water samples and transport to the laboratory. The suspended particulate matter was removed immediately by filtered through 0.45 μm × 50 mm glass fiber membranes (0.45 μm) (Membrane Solutions LLC., America). Sediment samples were collected with a Peterson grab sampler and wrapped into stainless steel containers, freeze dried, ground, and sieved. Then the water samples were stored at 4 °C, while the sediment samples were frozen at −20 °C for further extraction.

2.3. Sample pre-treatment and analyses

Water samples (1 L) were extracted by solid phase extraction (SPE). All Oasis HLB cartridges (500 mg, 6 mL, Waters, USA) were conditioned with 5 mL methanol and 5 mL ultrapure water. Then, filtrates were passed through the cartridges at a flow rate of about 5 mL/min. After they were extracted, the analytes were eluted with 10 mL methanol/ultrapure water (99/1, v/v) after 30 min dryness under a gentle stream of N_2, and then the solution was concentrated to 1 mL methanol.

Sediment samples were extracted by ultrasonic extraction (30 min) with a mixture solvent of 50 mL methanol/acetone (50:50