



Occurrence, spatiotemporal distribution, and ecological risks of steroids in a large shallow Chinese lake, Lake Taihu



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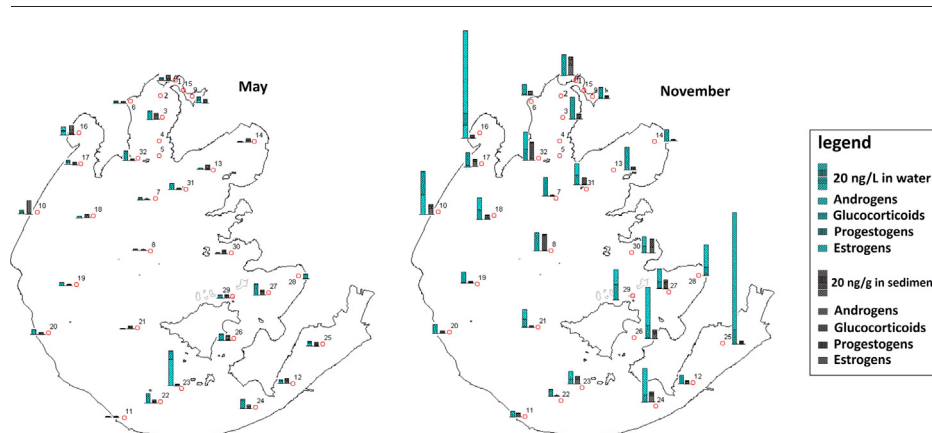
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HIGHLIGHTS

- Twelve and 15 steroids were found in water and sediments of Lake Taihu, respectively.
- Steroids in both surface water and sediments showed obvious temporal variations.
- Steroids were mainly found in bays nearing pollution sources rather than in pelagic.
- Trend of steroids in the lake influenced by pollution sources and lake hydrodynamics.
- Sediments are in general an important sink for steroids in this lake.

GRAPHICAL ABSTRACT



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ABSTRACT

Steroids have been frequently detected in surface waters, and might pose adverse effects on aquatic organisms. However, little information is available regarding the occurrence and spatiotemporal distribution of steroids in lake environments. In addition to pollution sources, the occurrence and spatiotemporal distribution of steroids in lake environments might be related to lake types (shallow or deep), lake hydrodynamics, and sorption-desorption processes in the water-sediment systems. In this study, the occurrence, spatiotemporal distribution, and ecological risks of 36 steroids in a large shallow lake were evaluated by investigating surface water and sediment samples at 32 sites in Lake Taihu over two seasons. Twelve and 15 analytes were detected in aqueous and sedimentary phases, respectively, with total concentrations ranging from 0.86 to 116 ng/L (water) and from 0.82 to 16.2 ng/g (sediment, dry weight). Temporal variations of steroid concentrations in the water and sediments were statistically significant, with higher concentrations in winter. High concentrations of steroids were found in the seriously polluted bays rather than in the pelagic zone of the lake. Strong lake currents might mix pelagic waters, resulting in similar concentrations of steroids in the pelagic zone. Mass balance analysis showed that sediments in shallow lakes are in general an important sink for steroids. Steroids in the surface water and sediments of Lake Taihu might pose potential risks to aquatic organisms. Overall, our study indicated that the concentrations and spatiotemporal distribution of steroids in the large shallow lake are influenced simultaneously by pollution sources and lake hydrodynamics.

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Capsule: Steroids in the large shallow Lake Taihu showed clear temporal and spatial variations and lake sediments may be a potential sink of steroids.

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1. Introduction

Natural and synthetic steroids in the aquatic environment have received particular attention in recent years because they could cause adverse effects on aquatic organisms, even at very low ng/L levels, such as reducing fish fecundity and causing masculinization (Ankley et al., 2003; Jensen et al., 2006; Orlando et al., 2004), impairing reproductive functions in amphibians (Safholm et al., 2014), and increasing proportions of intersex fish (Aris et al., 2014). Endogenous steroids and their metabolites are naturally excreted via the urine and feces of humans, livestock and aquatic organisms. A number of synthetic steroids, particularly glucocorticoids and progestogens, are widely used for human and veterinary therapy, and synthetic androgens such as stanozolol, methyl testosterone, trenbolone, and nandrolone, are also illegally used as growth promoters for cattle and calves in China (Chang et al., 2009; MA, 2002). So far, natural and synthetic steroids have been frequently detected in many pollution sources, such as influents (raw wastewater) and effluents of traditional wastewater treatment plants (WWTPs) (Chang et al., 2007, 2011; Fan et al., 2011; Liu et al., 2012a), livestock farms (Leet et al., 2012; Liu et al., 2012b, 2012c) and aquaculture (Liu et al., 2015a).

Because of incomplete removal or limited treatment, natural and synthetic steroids can enter surface waters via the effluents of WWTPs (Liu et al., 2012a), wastewater from livestock farms and aquaculture (Liu et al., 2012b, 2012c, 2015a; Kolodziej et al., 2004), and runoff from manure-fertilized fields (Bartelt-Hunt et al., 2011; Jones et al., 2014). These steroids have been frequently detected in rivers (Chang et al., 2009; Shi et al., 2014; Ying et al., 2002). Because the outflow from rivers and wastewaters feeds into lakes, some steroids such as estrogens have also been detected in a few lakes (Huang et al., 2013). However, little information is available regarding the occurrence and distribution of natural and synthetic steroids (including estrogens, androgens, glucocorticoids and progestogens) in lake environments. The spatial distribution of steroids in lake environments might be influenced by both pollution sources and lake hydrodynamics. For example, strong water currents may result in homogeneous distribution of steroids in the pelagic zone, while limited water exchange and terrestrial pollution sources may lead to different steroid concentrations in bays of the lake (Qin et al., 2007). Furthermore, adsorption and desorption processes of steroids to reach equilibrium in the water-sediment systems might change the transition of sink/source of steroids, and subsequently influence their spatiotemporal distribution (Das et al., 2004; Kim et al., 2007; Wang et al., 2015; Wiberg and Harris, 2002).

Lake Taihu is the third largest freshwater lake in China. It is located in the Yangtze Delta, which is the most industrialized area in China, and it has a high population density and urbanization. There are 38 cities and a reported 34.2 million people (Li et al., 2009) and plenty of livestock farms in proximity to the lake (Table 1). Moreover, the aquaculture in Taihu Basin is greatly developed (Table 1). In recent decades, rapid economic growth and urban development have caused the lake and its main inflow rivers to become polluted by wastewaters and waste from industry, agriculture and households. For example, many antibiotics have been frequently detected in sediments (4.1–731 ng/g), overlying water (127–1210 ng/L) and pore water (1.5–216 ng/L) from Lake Taihu (Xu et al., 2014). Estrone (E1) and bisphenol A (BPA) were the dominant compounds detected in the surface water of the Yangtze River Estuary which is connected with Lake Taihu (Shi et al., 2014). Considering these plenty of potential pollutant sources (domestic wastewater, aquaculture, and livestock) (Table 1), high concentrations of steroids are expected in Lake Taihu. Meanwhile, Lake Taihu serves as

the drinking water source for several cities, including Shanghai, Suzhou, Wuxi, and Huzhou, and the eastern coast of Lake Taihu is also an important ecological preservation area (Qin et al., 2007). It is therefore urgent to understand the pollution level, spatiotemporal distribution and ecological risks of steroids in Lake Taihu.

The major objectives of this study were to investigate the occurrence, spatiotemporal distribution, and ecological risks of multi-classes of steroids in Lake Taihu. We also tried to understand the mechanisms underlining these distribution patterns. To our best knowledge, this is the first study about steroids in lake environments. Our study might facilitate the ecological risk assessment of steroids in lake environments and have implications for effective control measures.

2. Materials and methods

2.1. Site description and sample collection

Lake Taihu (119°08′–121°55′E and 30°05′–32°08′N) is characterized by its shallowness (mean depth = 1.9 m) and large surface area (2338 km²) (Qin et al., 2007). Lake Taihu has a complicated inflow and outflow system, with the water retention time of approximately 5 months. Most of the runoff sources are from the mountainous west and southwest, and the outflows are located throughout East Taihu (Table 1). Urban pollutants and livestock wastewater discharge into northern Taihu. This inflow/outflow system results in shorter water retention times and better water quality in the south or southeast areas, and longer retention times and poorer water quality in the north or northeast areas (Qin et al., 2007). Detailed information on Lake Taihu (e.g., inflow/outflow rivers, catchment areas, land use, population, and livestock number) is provided in Table 1.

Surface water and sediment samples were collected in November 2013 and May 2014 from 32 national surface water quality sites in Lake Taihu (Fig. 1). The basic information for each sampling site is presented in Table S1, Supplementary. The surface water samples were collected by using a transparent organic glass water sampler, and the surface sediment samples (0–20 cm) were collected with a stainless grab sampler and then stored in 250 mL glass bottles. The suspended particle samples were collected by passing the water samples through pre-baked and pre-weighted 0.7 µm glass fiber filters and then extracted with the same method as extracting the sediment samples. Water samples were adjusted to pH 3 using 4 M H₂SO₄, added with methanol (5% v/v) to inhibit microbial activity, and then the samples were transported to the laboratory in a cooler with ice packs. The solid samples were preserved by adding 0.2 g sodium azide to inhibit microbial activity. Upon arrival in the laboratory, the samples were immediately stored at 4 °C to minimize the degradation of target compounds during the preservation. Prior to the analysis, the solid samples were freeze-dried, ground, sieved through a 0.5 mm pore size filter and then maintained at −18 °C in the dark until extraction. The following water quality properties were assessed according to standard methods: chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), total phosphorus (TP), total nitrogen (TN), and ammonia-nitrogen (NH₄–N) (Jin and Tu, 1990). The pH and dissolved oxygen (DO) levels were monitored in the field using the YSI 6600 multi-sensor sonde (Yellow Springs Instruments Inc., OH, USA). The total organic carbon (TOC) content was measured according to GB 7857–87 (Chinese national standard method) (Zhang and Xu, 1984). The sediment particle size (SPS) was determined using a laser particle sizer (Malvern Mastersizer 2000). The results of the water quality parameters, TOC content and SPS of sediment samples are listed in Table S2, Supplementary.

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