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National investigation of semi-volatile organic compounds (PAHs, OCPs, and PCBs) in lake sediments of China: Occurrence, spatial variation and risk assessment

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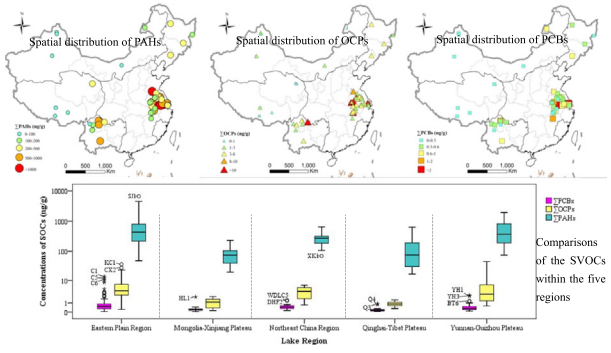
HIGHLIGHTS

- Nationwide analysis of SVOCs are conducted based on 52 lake sediments in China.
- SVOCs contamination in China distributed significant regional variation.
- Human activities are major factors affecting the SVOCs regional variation.
- DDT compound residues exhibit negligible ecological risks in Chinese lakes.

GRAPHICAL ABSTRACT



Comparisons of the SVOCs within the five regions

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ABSTRACT

In this study, a large scale investigation of semi-volatile organic compounds (SVOCs) in sediments from 52 lakes, located in five geographic regions across China, was conducted to assess sediment quality in terms of organic contaminants. Concentrations of polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), and polychlorinated biphenyls (PCBs) in sediment were found to range between 17.00–6,633, 0.12–45.24, and 0.03–13.99 ng g^{-1}, respectively. Lake sediments from different regions exhibited variation in contamination levels, and in the compositions of isomers. The descending order of contamination levels for the three SVOCs groups (i.e., ∑PAHs, ∑OCPs, ∑PCBs) were as follows: the eastern plain region (EPR) > the Yunnan-Guizhou plateau (YGP) > the northeast China region (NCR) > the Qinghai-Tibet plateau (QTP) > the Mongolia-Xinjiang plateau (MXP). High molecular weight PAHs, OCPs and PCBs were found at high concentrations in parts of the EPR and YGP, while lighter isomers were likely to be atmospherically transported, resulting in their nationwide distribution. The sources and type variation (local input or atmospheric deposition) are the primary controls of spatial variation. Additionally, human related socio-economic factors, and geographic conditions also played

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important roles in influencing the spatial differentiation. According to simple sediment quality guidelines (SQG), the risks posed by PCBs in sediments were not as serious as those posed by PAHs and DDT.

1. Introduction

Semi-volatile organic compounds (SVOCs) have attracted great attention due to their potentially adverse health effects on wildlife and humans (Ali et al., 2015). Organochlorine pesticides (OCPs), and polychlorinated biphenyls (PCBs) are common SVOCs, which are notorious for their resistance to degradation, ability to bioaccumulate, and toxicity. Though banned for years, their adverse effects have touched everywhere in the world. Polycyclic aromatic hydrocarbons (PAHs), which are not strictly considered POPs, are also listed on the U.S. Environmental Protection Agency's (EPA) priority pollutant list due to their resistance to degradation, and toxic properties (Guzzella et al., 2016). Due to their strong affinity for particulate matter, these SVOCs are most likely to be associated with sediment in aquatic systems. Thus, sediments are documented as the principal receptors and major reservoirs for SVOCs. Global studies of SVOCs in various aquatic environments have shown that highly urbanized and industrialized areas can suffer severe environmental deterioration (Hilscherova et al., 2007; Rabodonirina et al., 2015), making highly urbanized and/or industrialized areas a focus for research. In China, studies of SVOCs in lakes were typically geographically associated with major industrial districts, including the Yangtze river delta, the Yellow river delta and the Pearl river delta. The occurrences of priority PAHs, DDT (dichlorodiphenyltrichloroethane), HCHs (hexachlorocyclohexane) and some PCBs (mainly tri- to hexa-PCBs) in such lakes, including Lake Tai, Lake Poyang, Lake East, Lake Baiyangdian, and Lake Nansi, have been well documented (Gao et al., 2013; Mai et al., 2004; Li et al., 2016a).

Owing to their physico-chemical properties, SVOCs can be subjected to long range atmospheric transport, which can lead to the contamination of remote, high-altitude, or polar regions, where few people live, and very little industrialization or development occurs (Daly and Wania, 2005; Choi et al., 2008). Recently, scientists have shown increasing interest in quantifying the occurrences of PAHs, and DDTs in remote regions of China, including lakes of Qinghai, Dianchi, and Bosten in western China (Guo et al., 2010; Xu et al., 2014), because understanding transportation and distribution of SVOCs within remote areas could advance researches focused on large-scale atmospheric transport mechanisms generally (Said et al., 2007; Guzzella et al., 2016). Moreover, lakes in remote regions, especially mountainous regions, are often considered pristine environments, and in particular may capture primarily volatile POPs, which are suitable for studying the effects of remote pollution sources.

In China, there are 2693 natural lakes of various types with areas larger than 1 km^2, and they are distributed widely in a vast and complex terrain (Ma et al., 2011). All lakes in China are classified here into five regions, including the Eastern Plain Region (EPR), the Yunnan-Guizhou Plateau (YGP), the Qinghai-Tibet Plateau (QTP), the Mongolia-Xinjiang Plateau (MXP), and the Northeast China Region (NCR). Differences in geographic conditions, and degrees of human impact on these lakes and their watersheds, may lead to different magnitudes and types of pollution. Prior research has focused primarily on POP occurrences either in heavily developed regions, or in a small number of remote lakes, thus it is necessary to conduct a large scale investigation of SVOCs in lakes throughout China. A total of 52 lakes from across five geographic regions were included to study the geochemical and pollution characteristics of PAHs, OCPs and PCBs. The objectives of this study were to (1) investigate the occurrence and spatial distribution of POPs in Chinese lake sediments, (2) fill the data gap on SVOCs contamination in some lesser-known lakes in developing regions, and (3) reveal the differences between the five regions, in terms of anthropogenic activities, sedimentary conditions, and climatic factors.

2. Materials and methods

2.1. Study areas

Lakes in the EPR are found in the eastern coastal region of China, and are associated with the Yangtze, Yellow, Hai, and Huai rivers (Wang and Dou, 1998). The clustered, and abundant lakes here are most often shallow (lakes depth range: 0.6 m–7.6 m, average depth: 2 m), and can easily be influenced by the dense regional population, and local economic developments, resulting in large variations in lake morphology and water quality (Le et al., 2010). The MXP is located in inland, northern China, a region of relatively low precipitation, and frequent dust storms. As such, lakes here often suffer from low discharges to them from runoff or precipitation, and can develop into closed lagoons or salt lakes. The YGP is located in the southwest monsoon region of China, in a predominately karstic area. Lakes in this region are mostly tectonic fault-depressed lakes formed in the early pleistocene, with relatively small surface areas, deep depths, and complicated bottom topography (Xiao et al., 2009). The QTP is the largest and highest elevation plateau in the world, with the greatest density of saline lakes. Lakes in this area experience a relatively harsh climate, and a dramatic elevational temperature gradient, leading to unique precipitation patterns. As a result of the sparse human population and minimal industrial activity, lakes in the QTP are especially sensitive to atmospherically-deposited contaminants (Yang et al., 2011). The NCR is located in relatively cooler northeastern China, with a humid to sub-humid continental monsoon climate. Lakes distributed throughout this topographically complex, mountainous area are often barrier lakes formed by volcanic activity. Many lakes in this region make significant contributions of water for local irrigation.

To comprehensively investigate SVOCs contamination in Chinese lake sediments, 52 lakes were selected, covering five lake regions, and 10 provinces of gradient development (Fig. 1). These lakes differed in geography and limnology, spatial patterns, expected sensitivity to stress, and anticipated contaminant exposure level, the selection standards referred to Stevens study (Stevens, 1994). The geographic and limnological features of the sampled lakes are listed in Table. 1. Descriptions of these lake regions are based on the research of Wang and Dou (1998), and additional information is given in Table S1 (Supplemental materials).

2.2. Sampling and sample preparations

A total of 261 surface sediment sites were selected from 50 lakes and 2 reservoirs in the five regions. The surface sediment samples (0–10 cm) were collected using a stainless steel Peterson grab sampler during the years 2011–2013. Once collected, the sediment samples were wrapped in pre-cleaned aluminum foil, and temporarily stored at 4 °C for transport to the laboratory. All sediment samples were subsequently freeze-dried, then homogenized, then sieved through a 100 mesh sieve, and kept in a freezer at −20 °C until further analysis. Total organic carbon (TOC) was determined for all samples using a TOC analyzer (MultiN/C2100, AnalytikJena AG, Germany) after acidification (Wang et al., 2012).

The sample pretreatment procedures for target SVOCs were performed according to Li et al. (2016b) with some modification. In

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