



Distribution of phthalate acid esters in lakes of Beijing and its relationship with anthropogenic activities



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HIGHLIGHTS

- Effective analysis methods for 15 phthalate acid esters (PAEs) were developed.
- PAEs were widespread occurrence in different matrices of research lakes.
- The PAE concentrations in solid samples were higher than those in water ones.
- The distribution of PAEs in sediments increased linearly with human activities.
- Di-(2-ethylhexyl) phthalate and di-butyl phthalate were predominated PAEs.

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ABSTRACT

The distribution of 15 phthalate acid esters (PAEs) in water, sediment and suspended particle samples from representative lakes of Beijing was investigated and its relationship with anthropogenic activities was also assessed. The sample preparation and analysis methods for PAEs, using solid-phase extraction or microwave assisted extraction coupled with gas chromatography–mass spectrometry, were developed to achieve the satisfactory detection limits and recoveries. The analysis results indicated occurrences of most target PAEs were widespread in the researched lakes with the total PAE concentrations ranged from 0.386 to 3.184 $\mu\text{g/L}$ in water, from 52.6 to 8216.4 ng/g dry weight in sediments and from 138.7 to 2052.4 $\mu\text{g/g}$ dry weight in suspended particles. Di-(2-ethylhexyl) phthalate and di-butyl phthalate were the predominant PAEs, frequently being detected in all matrices, followed by di-iso-butyl phthalate and butyl benzyl phthalate. It was also found that anthropogenic activities were associated with the spatial distribution of phthalates in sediments considering its relationship with population distribution characteristics of Beijing.

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1. Introduction

Endocrine disruptor chemicals (EDCs) are any externally originating chemical compounds, either natural or synthetic, that interfere with normal endocrine function (Ponzo and Silvia, 2013). Phthalate acid esters (PAEs) are one of the top offenders in the growing list of suspected EDCs with several million tons being produced worldwide annually (LaFleur and Schug, 2011; Guo et al., 2012). Since these compounds have been found to bio-accumulate in fats (Keresztes et al., 2013; Staples et al., 1997) and shown to be carcinogenic and estrogenic (Wu et al., 2013; He et al., 2013), the potential risk to human and animals' health generated from PAEs has drawn great attention all over the world. Several phthalates have been listed as priority pollutants by many national and international regulatory organizations (Ventrice et al., 2013; Yang et al., 2013).

Low-molecular-weight PAEs are used in cosmetics and personal care products, such as perfumes and nail polishes (Gómez-Hens and Aguilar-Caballo, 2003; Koniecki et al., 2011). Most of the high- to mid-molecular-weight PAEs are widely used as plasticizers to improve the flexibility and workability of polymeric materials, and about 80% of all PAEs produced are used for this purpose (Cao, 2010). Since PAEs are physically rather than chemically incorporated in the polymeric matrices, PAEs can easily migrate from the products into the hydrosphere, atmosphere and biosphere during processes of production, usage and ultimate disposal (Okamoto et al., 2011). Like the ubiquitous PCB's and DDT's, the phthalate plasticizers are refractory to the environmental microorganisms (Wu et al., 2013) and were found in almost all environment matrices, such as soil (Chen et al., 2012), water (Shi et al., 2012), sediment (Liu et al., 2010), biota (Blair et al., 2009), air (Kang et al., 2012) and sewage (Dargnat et al., 2009). Di-(2-ethylhexyl) phthalate and di-butyl phthalate were also found in marine water in the North Atlantic (Giam et al., 1978) and remote atmosphere at Enewetak Atoll in the North Pacific Ocean (Atlas and Giam, 1981).

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Aquatic ecosystems are susceptible to pollutants in various media of water bodies, such as water, sediments and suspended particles. According to the literature survey, the integrated researches about spatial distribution of PAEs in all of these water body matrices are limited, although there are many references about PAE spatial distribution or PAEs in solo water body matrix. Furthermore, since PAEs are anthropogenic pollutants, the relationships between PAE spatial distribution and human activities should also be taken into consideration (Vitali et al., 1997; Sun et al., 2013). Based on these factors, the relative researches and investigations were conducted in this article.

Six PAEs listed as priority pollutants by the U.S. Environmental Protection Agency (EPA) and nine other prevalent PAE congeners, which were demonstrated to have negative effects on humans, were selected as target compounds in this work. Guanting Reservoir, the Lakes Shichahai and the Lakes in Summer Palace, which are important water bodies of Beijing, were selected as representative research objects. The preparation and analysis methods for the PAEs in water, sediment, and suspended particle samples were developed to research PAE distribution in these lakes. It should be mentioned that this work indicated anthropogenic activities were associated with the spatial distribution of phthalates in sediments, which would provide useful information for future management and remediation efforts for PAEs. This work might also improve our understanding of the status and fate of these compounds in these water environments.

2. Materials and methods

2.1. Chemicals and materials

Standards of individual phthalate esters, including di-methyl phthalate (DMP), di-ethyl phthalate (DEP), di-butyl phthalate (DBP), di-amyl phthalate (DAP), di-hexyl phthalate (DHXP), butyl benzyl phthalate (BBP), di-*n*-octyl phthalate (DNOP) and di-(2-ethylhexyl) phthalate

(DEHP) were supplied by AccuStandard (New Haven, USA). Di-propyl phthalate (DPrP), di-*iso*-butyl phthalate (DIBP), bis-(2-methoxyethyl) phthalate (BMEP), bis-(4-methyl-2-pentyl) phthalate (BMPP), bis-(2-ethoxyethyl) phthalate (DEEP), bis-2-*n*-butoxyethyl phthalate (DBEP), and di-cyclohexyl phthalate (DCHP) were bought from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Ultra resi-analysis grade organic solvents, including acetone, ethyl acetate, methylene dichloride, *n*-hexane and HPLC grade acetonitrile and methanol were acquired from J.T. Baker (Phillipsburg, USA). 0.45 μm millipore membrane was obtained from Jiuding High-tech Filtration Equipment Ltd. Co. (Beijing, China). Single PAE standard stock solutions (1000 mg/L) and mixed standard stock solutions (50 mg/L) were prepared in methanol. All solutions were kept at 4 °C in the dark.

In order to avoid overestimating concentrations, the laboratory equipments made of glass, stainless steel and polytetrafluoroethylene were used during the sampling and execution of experiments. All glass apparatuses were soaked in nitric acid solution for at least 24 h, rinsed with purified water at least 10 times, and finally baked at 320 °C for 4 h.

2.2. Sample collection

Water samples (sites shown in Fig. 1) from Guanting Reservoir (n = 10, n, number of sample sites), the Lakes Shichahai (n = 4) and the Lakes in Summer Palace (n = 5), were collected (as shown in Fig. 1) using a stainless steel bucket from April to May, 2012. Physico-chemical parameters including temperature, pH, conductivity, turbidity, dissolved oxygen and total dissolved solids, were recorded using a multi water quality checker (Horiba U-50, Kyoto, Japan) at the same time. The sensor was placed at 0.25 m below the water surface to make the measurements. Water samples were sealed in 2.5 L wide mouthed amber bottles with foil-lined caps. Sediment samples were collected at the corresponding positions, using a column sediment sampler (Eijkkelkamp

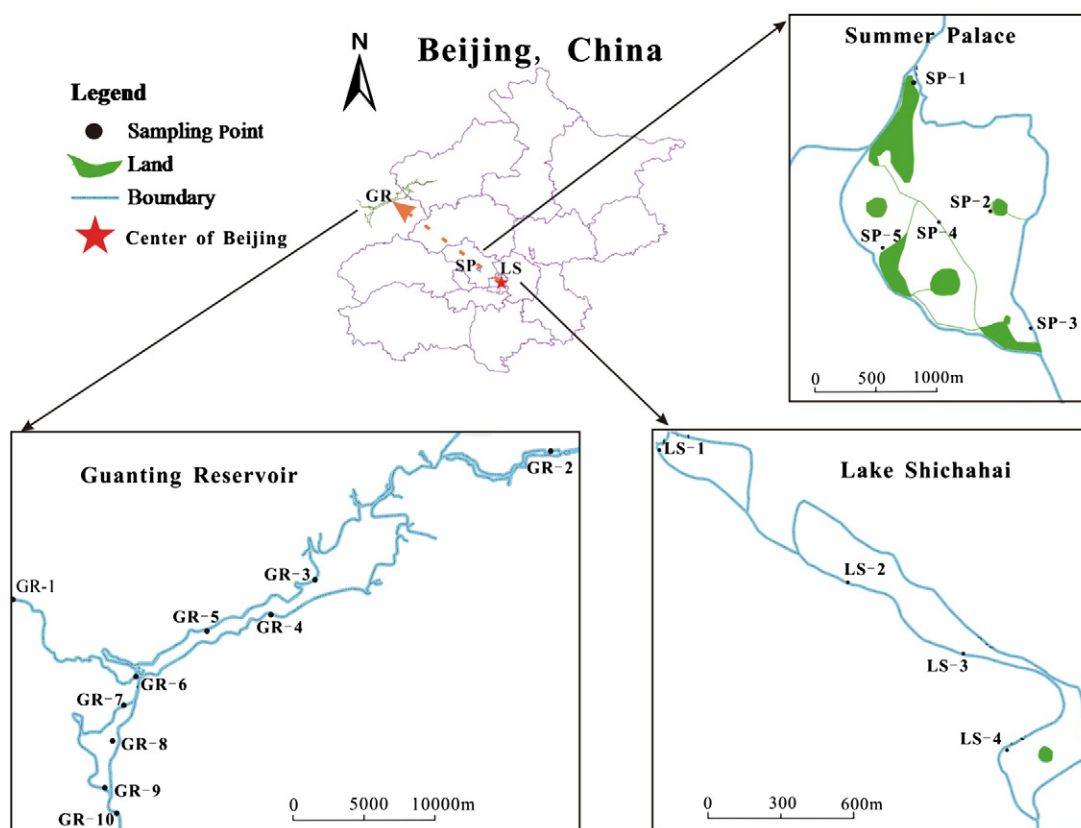


Fig. 1. Maps of sampling sites in Guanting Reservoir (GR), Lakes Shichahai (LS) and Summer Palace (SP) and their geographical locations in Beijing, China.

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