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Historical records of polycyclic aromatic hydrocarbon deposition in a shallow eutrophic lake: Impacts of sources and sedimentological conditions

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ABSTRACT

Sediment core samples collected from Lake Chaohu were analyzed for 15 priority polycyclic aromatic hydrocarbons (PAHs) to assess the spatial and temporal distributions of the PAHs during lacustrine sedimentary processes and regional economic development. Assessing the PAH sedimentary records over an approximately 100-year time span, we identified two stages in the PAH inputs and sources (before the 1970s and after the 1970s) in the eastern lake region near a village, whereas three stages (before the 1950s, 1950s–1990s and after the 1990s) were identified in the western lake region near urban and industrial areas. Rapid increases in the PAH depositional fluxes occurred during the second stage due to increased human activities in the Lake Chaohu basin. The composition and isomeric ratios of the PAHs revealed that pyrolysis is the main source of PAHs in this lake. Strong positive relationships between PAH concentration and the total organic carbon concentration, sediment grain size (<4 μm), as well as the local population and Gross Domestic Product indicated that the sedimentary conditions impact the depositional characteristics of the PAHs; simultaneously, socioeconomic activities, such as energy consumption and the levels of urban industrialization and civilization, affect both the composition and abundance of the PAHs.

Introduction

Polycyclic aromatic hydrocarbons (PAHs), a group of ubiquitous organic pollutants listed as priority pollutants by the international environmental protection agencies, have raised significant environmental concern due to their toxic, carcinogenic, and mutagenic characteristics, on regional and global scales (Xu et al., 2005; Wilcke, 2007; Okuda et al., 2010; Li et al., 2012a; Nie et al., 2014). Although they can arise from both natural and anthropogenic sources, the PAHs released into the environment stem mainly from anthropogenic sources, including the incomplete combustion of fossil fuels and biomass, waste incineration, and traffic emissions (Yunker et al., 1996; Leite et al., 2011; Chen et al., 2012; Azoury et al.,

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2013). Previous studies have shown that PAHs correlate well with anthropogenic activities (Liu et al., 2012b); thus, PAHs are treated as a useful geochemical marker to trace the impact of anthropogenic activities, such as socioeconomic changes and energy use (Schneider et al., 2001; Lima et al., 2002; Liu et al., 2012b;). Because they are highly hydrophobic and weakly soluble, PAH compounds are typically attached to fine particles in the environment and are eventually incorporated into lake or sea sediment *via* runoff and atmospheric fallout (Mouhri et al., 2008; Hui et al., 2009; Guo et al., 2011b). However, the PAH pollutants trapped in lake or sea sediments can sometimes return to the water column, causing secondary pollution (Machado et al., 2014). Using dated, segmented sediment core samples is a well-established method for reconstructing the historical records of anthropogenic PAH input into the environment and for assessing temporal environmental changes, such as human population growth and land settlement (Kannan et al., 2005; Itoh et al., 2010; Arp et al., 2011; Li et al., 2012b). In addition, lake sediments are more efficient in revealing historical contamination than marine sediments due to the more limited post-depositional mixing processes in lacustrine systems (Guo et al., 2010).

Lake Chaohu is one of the five largest freshwater lakes in China and is located in the lower reach of the Yangtze River (Huo et al., 2013). Lake Chaohu plays an important role in supporting the more than 9.1 million people living in Anhui Province, Eastern China (Wang et al., 2013). Several studies have investigated the historical contamination characteristics of different pollutants in relation to human activities in Lake Chaohu, such as eutrophication and pollution by heavy metals (Zan et al., 2011a, 2011b; Wang et al., 2013; Wang et al., 2014). Nevertheless, reports on the sedimentary records of PAHs are rare for Lake Chaohu. During recent decades, China has experienced rapid economic development, especially after the implementation of the Reform and Opening policy in 1978 (Liu et al., 2012b). This social trend resulted in an increasing consumption of energy, particularly the massive combustion of coals, biomass and foil oils, which consequently caused vastly increased emissions of PAHs (Yunker et al., 1996). However, most studies of PAHs have focused on near-shore estuarine areas (Liu et al., 2000, 2005; Zhang et al., 2009), and few studies have examined the influence of socioeconomic development on PAH contamination in inland shallow water lakes in China (Liu et al., 2012b). Because sediment records in the semi-enclosed Lake Chaohu have been well preserved by stratification, using the depositional fluxes and concentrations of PAHs in the sediment can reveal both the historical impact of socioeconomic development and the sedimentological changes in the inland Lake Chaohu.

In the present study, four dated sediment cores from different regions of Lake Chaohu were used to (1) reconstruct the PAH contamination history to provide a comprehensive picture of the temporal trends in PAH emissions in a typical inland, shallow lake in Eastern China, *i.e.*, Lake Chaohu; (2) identify the possible sources of PAHs; and (3) evaluate the relationship between PAH pollution and socioeconomic development together with sedimentological changes.

1. Materials and methods

1.1. Study area and sampling

Lake Chaohu, located in Anhui Province, Eastern China (117°16′–117°51′N, 31°25′–31°43′E), has a mean depth of approximately 3.0 m and a surface area of 780 km^2. Six main rivers, providing approximately 80% of the runoff volume from the catchment area, feed into Lake Chaohu, and the outlet, the Yuxi River, is the only channel linking the lake to the Yangtze River (Fig. 1). The Nanfei River and the Shiwuli River are the principal sources of pollutant inflows into the western lake region (Zan et al., 2012).

The sampling sites were chosen away from lake banks, stream inlets and the lake outlet to diminish any disturbance of the sediment by scouring or re-suspension. Eight cores were obtained in July, 2009 at four sites, C4, C5, C6 (western lake region) and C10 (eastern lake region), with two cores taken at each site. A coring sampler was used to insert an 8 cm inner diameter 50 cm long PMMA tube into the lake sediment. After siphoning overlying water on the sediment–water interface, the sediment in each core (ca. 30 cm) was immediately sectioned at 2-cm intervals. The labeled slices were placed in aluminum foil, which had been prewashed by methylene chloride, and temporarily kept in iceboxes at −4°C. Then, the samples were transferred to the laboratory and stored frozen below −20°C. All the sediment samples were freeze-dried at −50°C, homogenized *via* grinding with an agate mortar and pestle, and passed through a 100 mesh sieve before analysis.

1.2. Sample preparation and analysis

A detailed description of the analytical methods has been provided in a previous study (Zeng et al., 2012). Briefly, the freeze-dried and homogenized samples (10 g) were Soxhlet extracted with a mixed solvent (200 mL) of hexane and dichloromethane (1:1 by volume) for 48 hr. In addition, a mixture of deuterated PAHs (naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12}) was spiked as the recovery surrogate. About 2 g of activated copper was added to the extract solvent for desulphurization. The obtained extract was filtered, solvent-exchanged to hexane and then concentrated to less than 1 mL. The purification and fractionation process was performed on a two-layer 3% deactivated alumina/silica gel (1:2 by volume) column. The first fraction, eluted with 15 mL hexane, was collected for another purpose, which is not further discussed here. The second fraction, containing the PAH components, was eluted with a 70 mL mixture of hexane:dichloromethane (7:3, V/V), then solvent-exchanged and concentrated to 0.2 mL under a gentle nitrogen stream. The internal standard hexamethylbenzene (HMB) was added to each sample extract before instrumental analysis.

The identification and quantification of 15 priority PAHs (acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (IcdP),