



Sedimentary polycyclic aromatic hydrocarbons record recent anthropogenic activities near high-elevation Lake Sayram, northwest China

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ABSTRACT

Mountain lakes are sensitive to natural perturbations and human activities. Profiles of anthropogenic polycyclic aromatic hydrocarbons (PAHs) in lake sediments serve as a natural archive from which human impacts on lakes can be reconstructed. Two parallel sediment cores (34 cm long) were extracted from Lake Sayram in the Tianshan Mountains of arid northwest China and dated using ²¹⁰Pb and ¹³⁷Cs. PAHs in the sediments were used to infer the past ~150 years of anthropogenic impact on the lake. Results showed that PAH concentrations in the lake sediments were generally low, but nevertheless reflect some human activity. Benzo(ghi)perylene (BghiP) and indeno(1, 2, 3-cd)pyrene (IcdP) are typical markers of vehicular traffic, so significant correlations between PAH (especially BghiP and IcdP) and metal concentrations suggest that PAHs in sediments are effective proxies for anthropogenic activities. Furthermore, constrained cluster analysis revealed that the vertical profile of PAHs was characterized by three periods that correspond to changes in human activities. Prior to the 1960s, low PAH concentrations in the sediments generally indicate that the lake was virtually unaffected by human impact. BghiP and IcdP concentrations, however, displayed a minor peak in the 1950s, possibly reflecting transport from a remote polluted region via prevailing westerly winds. From the 1960s through the 1990s, relatively high values of PAHs, especially high molecular weight (HMW) PAHs BghiP and IcdP, and heavy metal enrichment factors (EFs), were related to increasing anthropogenic activities. After the 1990s, maxima of PAHs, HMW PAHs, BghiP, IcdP, EFs, together with highest total phosphorus (TP) and total organic carbon (TOC) values, reflect intensive human-related impact on the lake. The coincidence of limnological changes and local socio-economic development suggest that sedimentary PAHs record recent anthropogenic impact on Lake Sayram.

1. Introduction

Anthropogenic activities during the last 150 years have caused important transformations in aquatic environments, including increased contamination (Halpern et al., 2008), which pose challenges for environmental sustainability. Polycyclic aromatic hydrocarbons (PAHs) are an important class of organic pollutants that have originated mainly from anthropogenic activities in recent years. These compounds, released from incomplete combustion of organic materials, are of great concern because of their widespread occurrence and toxic effects on ecosystems and human health (Choudhary and Routh, 2010). PAH compositions and distributions vary depending on specific processes, which make it possible to use some individual constituents, or a group of PAHs, as tracers in source apportionment models. For example, benzo(ghi)perylene (BghiP) and indeno(1, 2, 3-cd)pyrene (IcdP) are mainly from vehicular emissions (May and Wise, 1984; Harrison et al.,

1996; Larsen and Baker, 2003). High molecular weight (HMW) PAHs are associated with road traffic and certain industrial processes (Vane et al., 2011). Consequently, PAHs in the environment reflect changes in multiple human activities.

Lake sediments constitute an important sink for PAHs, which enter aquatic systems through passive or active long-range transport before finally accumulating on the lake bottom. Sediments, therefore, have become resources for recording and preserving PAHs, and valuable archives of past anthropogenic activities. Historical trends in PAH concentrations in lake sediment cores have been used widely to infer past human activities (Muri and Wakeham, 2009; Mikac et al., 2011; Shen et al., 2016) as well as evaluate changes in PAH sources (Wang et al., 2010).

Mountain lakes are sensitive ecosystems and quickly respond to natural or human-induced changes in their watersheds. Although isolated from direct anthropogenic influence, they are considered ideal

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systems to study atmospherically transported pollutants and track human-induced environmental changes. Lake Sayram is a mountain lake and is sensitive to regional environmental changes. Previously, most studies in the region focused on the climate and lake area changes (Bai et al., 2011; Wu and Ma, 2011; Liu et al., 2014), heavy metal (HM) pollution (Zeng et al., 2014) and aeolian transport (Ma et al., 2015). With rapid industrialization and urbanization in Xinjiang, northwest China, over the last few decades, however, PAHs have become a novel group of environmental contaminants, in addition to metals, that constitute a threat to aquatic environments and now receive serious attention.

In this study, we used the vertical distribution of PAH concentrations in a sediment core from Lake Sayram, combined with a dating model, to reconstruct historical changes in PAH delivery to the lake. We also used the PAH and metal concentration data to investigate anthropogenic impact on the lake over time and assess potential health risks.

2. Materials and methods

2.1. Study area and sampling

Lake Sayram (44°30′–44°42′ N, 81°05′–81°15′ E), located in Bole, western Xinjiang Province, China, is an alpine lake that lies at 2071 m asl in the Tianshan Mountains (Fig. 1). The lake receives water from precipitation, glacial snowmelt and groundwater. There are 39 inflow streams around the lake, among which the Sagakele River is the longest. The lake has a surface area of 462 km², a maximum depth of 99 m, and a watershed area of 1408 km² (Wu et al., 2014). It is free from direct pollution inputs because there are no permanently inhabited areas near the lake, and nomadic grazing remains the most important local activity. Moreover, the low watershed/lake area ratio (~3.1) suggests that pollution inputs are mainly related to atmospheric transport and deposition, and perhaps some surface runoff. The lake is located in the arid hinterland of Eurasia, where Northern Hemisphere westerly winds prevail, and wind-blown dust is prevalent. Westerly winds across the Alatau Pass, a famous gale passage bordering Kazakhstan, can bring gas from vehicle exhaust and dust to Lake Sayram (Zeng et al., 2014). Moreover, with development since the late 1980s, the lake is exposed to more traffic and tourist activities than before.

Two parallel sediment cores, each 34 cm long, were obtained within 5 m of one another from a site in eastern Lake Sayram (Fig. 1). Cores were retrieved with a gravity corer in January 2013, and the coring location was identified using GPS. The core used for ²¹⁰Pb and ¹³⁷Cs dating was sectioned at 0.5-cm resolution, whereas the core for analysis of other variables was sectioned at 1-cm intervals. Prior to analysis, all sediment samples were freeze-dried, powdered and homogenized.

2.2. Sediment dating

Samples were analyzed for ²¹⁰Pb and ¹³⁷Cs activities by direct gamma spectrometry using Ortec HPGe GWL series well-type coaxial low-background intrinsic germanium detectors. The unsupported ²¹⁰Pb (²¹⁰Pb_{ex}) activity was calculated by subtracting ²²⁶Ra activity from the total ²¹⁰Pb activity. The sediment chronology was determined using the constant rate of supply (CRS) model (Appleby and Oldfield, 1978).

2.3. Sample extraction and analysis

The standard solution contains 16 United States Environmental Protection Agency (USEPA) priority PAHs, including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[ah]anthracene (DahA), benzo[ghi]perylene (BghiP), and indeno[1,2,3-cd]

pyrene (IcdP). The deuterated recovery surrogates standard (consisting of naphthalene-D8, acenaphthene-D10, phenanthrene-D10, chrysene-D12, and perylene-D12), and internal standard (hexamethylbenzene) were purchased from Supelco (Bellefonte, Pennsylvania). All organic solvents for PAHs analysis were chromatographic grade and also purchased from Supelco.

Detailed description of the method for PAH analysis is described in Shen et al. (2016). It is briefly summarized here. Sediments were extracted using dichloromethane by accelerated solvent extraction (ASE-100, Dionex, Sunnyvale, CA, USA), and cleaned up with 70-mL n-hexane/dichloromethane (5:2 v/v) using a silica gel chromatography column, followed by volume reduction and instrumental analysis. Before extraction, a suite of deuterated PAHs was added to the sediment as a surrogate standard. Known quantities of an internal standard were added to the sample prior to instrumental analysis.

PAHs were quantified with a high performance liquid chromatograph (Agilent 1200 HPLC) equipped with a diode array detection (DAD) and fluorescence detector (FLD), using a WATERS PAHs C18 (4.6 mm × 250 μm, particle size: 5 μm) column. The gradient elution program consisted of 40% water and 60% acetonitrile at first, after 20 min, a linear gradient to 100% acetonitrile, held 10 min. PAH concentrations were quantified by peak identification and retention times of corresponding standard components. Blanks, duplicate samples, and internal standard additions were employed for analytical quality assurance. Detection limits for individual PAHs were in the range of 0.03–3.57 ng g⁻¹, whereas spiked recoveries varied from 76% to 103%. All analyses are reported on a dry weight basis.

2.4. Total organic carbon (TOC) analysis

Samples for TOC analyses were treated with HCl (~3 N) to remove carbonates and subsequently rinsed with deionized water to remove remaining chloride. A CE-440 elemental analyzer (EAI Company) was used for TOC analysis.

2.5. Statistical analysis

Pearson correlation tests (P < 0.01) were carried out in SPSS Version 18 to assess linear relations among variables in the data matrix. The constrained incremental sum of squares cluster analysis (CONISS), a multivariate method for quantitative definition of stratigraphic zones in a sediment core, was performed using the program PAST version 3.01 (Hammer et al., 2001).

3. Results

3.1. Chronology of the sediment core

The ²¹⁰Pb_{ex} and ¹³⁷Cs profiles for the sediment core are shown in Fig. 2a. The ²¹⁰Pb_{ex} activity displays a generally exponential decrease with depth from the surface of the sediment core (~709 Bq kg⁻¹) to negligible concentrations at 28.5 cm. Ages and sediment accumulation rates were computed for each sample interval using the CRS model. The ¹³⁷Cs activity profile appeared at 16 cm and there is a well-defined peak at 11.5 cm depth. These two markers were attributed to 1954 (first atmospheric fallout attributable to atmospheric nuclear tests) and 1963 (maximum fallout) (Appleby et al., 2001; García-Orellana et al., 2006). The ¹³⁷Cs dates were in general agreement with the ²¹⁰Pb dates (Fig. 2b).

3.2. PAHs in the sediment core

Total concentrations of PAHs ranged from 129.2 to 642.9 ng g⁻¹. The low molecular weight (LMW) PAHs (2 and 3 ring PAHs; Nap, Acy, Ace, Fl, Phe and Ant) were in the range of 126.3–536.6 ng g⁻¹, whereas high molecular weight (HMW) PAHs (4, 5 and 6 ring PAHs; Chr, BaA,

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