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Factors affecting annual occurrence, bioaccumulation, and biomagnification of polycyclic aromatic hydrocarbons in plankton food webs of subtropical eutrophic lakes

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ABSTRACT

The biological pump plays a critical role in the occurrence and fate of hydrophobic organic contaminants (HOCs) mostly in temperate and frigid oligotrophic waters. However, the factors for the long-term occurrence and fate of HOCs in subtropical eutrophic waters remain largely unknown. This study provides novel insights into biogeochemical and physical factors on the annual occurrence, bioaccumulation, and biomagnification of 16 polycyclic aromatic hydrocarbons (PAHs) in the plankton food webs of four Chinese subtropical eutrophic lakes by one-year simultaneous field observations for five compartments. The annual mean ΣPAH_{16} in the water columns ranged from 359.69 ± 31.52 ng L^{-1} to 682.69 ± 65.41 ng L^{-1}, and increased with the annual mean trophic state index, and phytoplankton biomass of these lakes, but was independent on the proximity of the lakes to urban areas. Biodilution effect played an important role in the occurrence of the PAHs in both phytoplankton and zooplankton. In contrast to previous studies in oligotrophic waters, not only the biological pump but also the equilibrium partitioning and the indirect influence of eutrophication (high pH induced by phytoplankton, and phytoplankton life cycling) modulated the annual occurrence of the PAHs in the water columns of these eutrophic lakes. Biphasic correlations were found between the bioaccumulation factors of the PAHs by plankton and the temperature ($n = 97–136$, $R^2 = 0.06–0.24$, $p \leq .008$), and were related to plankton phenology. Bioaccumulation factors by plankton were dependent on the hydrophobicity of the PAHs ($n = 16$, $R^2 = 0.27–0.31$, $p \leq .023$), and decreased with plankton biomass ($n = 94–103$, $R^2 = 0.09–0.27$, $p \leq .010$). Trophic transfer of the PAHs from phytoplankton to zooplankton increased with phytoplankton biomass ($n = 26$, $R^2 = 0.27$, $p = .004$), and the temperature ($n = 102–135$, $R^2 = 0.06–0.13$, $p \leq .004$), but decreased with lake trophic state index. Biomagnification only occurred during phytoplankton bloom periods.

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1. Introduction

Hydrophobic organic contaminants (HOCs) such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and organochlorine pesticides (OCPs) are ubiquitous in aquatic environments, posing great risks to organisms and humans due to their toxicity, persistence, and bioaccumulation potential (Berglund et al., 2000; Dachs et al., 2002; Jeremiason et al., 1999; Tao et al., 2017a). Phytoplankton, and zooplankton are the primary producer, and primary predator in aquatic environments. Bioaccumulation and trophic transfer in plankton food webs are the first two steps for HOCs to enter food webs. Elucidating and quantifying the factors for the occurrence and fate of HOCs in plankton food webs will be of great help to understand and eliminate their risks in waters (Nizzetto et al., 2012).

One of the underlying mechanisms for the occurrence and fate of HOCs in aquatic environments is uptake by phytoplankton and subsequently transferred to higher trophic level organisms or to deeper water and sediment by phytoplankton – derived organic matter. When phytoplankton bloom, phytoplankton deplete the dissolved HOCs in the water column, reduce their concentrations in

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aquatic organisms, and enhance their air − water exchange fluxes, sinking fluxes, and sequestration in sediments (Dachs et al., 2002; Everaert et al., 2015; Jeremiason et al., 1999; Nizzetto et al., 2012). The role of this biogeochemical process, known as the biological pump, in the sequestration of HOCs has been intensively examined on the spatial scale in oceans in one growth season along the navigational route of a research vessel (Berrojalbiz et al., 2011a, 2011b; Casal et al., 2017; Dachs et al., 2002; Everaert et al., 2015; Morales et al., 2015). In contrast, empirical evidence of the working of the biological pump has been less studied on the temporal scale. Nizzetto et al. (2012) investigated the coupling between phytoplankton biomass dynamics and the distribution and fate of PCBs in Italian Lake Maggiore in the time frame of a few weeks. Berglund et al. (2001) investigated the influence of trophic status on PCB distribution in sediments and biota in Swedish lakes in spring and summer. Jeremiason et al. (1999) compared biogeochemical cycling of PCBs in two experimental lakes of variable trophic status in Ontario, Canada in the time frame of a few months. Everaert et al. (2015) applied a model to explore the links between phytoplankton dynamics and PCB concentrations in marine sediments and biota in the North Sea on a three-decadal time scale. All these studies provided very valuable information on the spatial and temporal occurrence and fate of HOCs mostly in temperate and frigid regions and a few tropical and subtropical regions.

China has the largest subtropical lake density in the Northern Hemisphere (Downing et al., 2006), and most of them are shallow lakes (Yang et al., 2010). Due to the rapid development of China, currently 85.9% of these lakes are eutrophic or hyper-eutrophic (Yang et al., 2010), and suffer from the contamination of HOCs (Fang et al., 2014; Li et al., 2017; Qin et al., 2013; Qiu et al., 2010; Ren et al., 2017a; Shi et al., 2017), especially the PAHs (Li et al. 2016, 2017; Tao et al., 2010, Tao et al., 2017b, 2017c). In contrast to previous studied oligotrophic waters, water chemistry and cycling of carbon, nitrogen, phosphorus, and HOCs in Chinese subtropical lakes change faster due to higher annual mean water temperatures and plankton biomass, and higher background levels of HOCs in waters (Chuai et al., 2011; Tao et al., 2017b, 2017c; Wu et al., 2012; Ye et al., 2011), and air (Qin et al., 2013; Qiu et al., 2008; Ren et al., 2017b; Wang et al., 2012). The occurrence and fate of HOCs in Chinese subtropical lakes, and the relevant factors may consequently be different from previous studies in oligotrophic waters. To date the knowledge on the long-term occurrence and fate of HOCs in plankton food webs in Chinese subtropical lakes has received little attention due to the lack of field observation.

The main objectives of this work are: (i) to report a one-year data set of the concentrations of 16 priority PAHs in five compartments (water, dissolved organic carbon, phytoplankton, zooplankton, and total suspended particles (TSP)) of four Chinese subtropical lakes with different trophic status on a biweekly basis, (ii) to investigate the annual biogeochemical and physical factors for the occurrence, bioaccumulation, and biomagnification of the PAHs in the plankton food webs of these lakes, and (iii) to provide novel knowledge on the annual occurrence, bioaccumulation, and biomagnification of HOCs in plankton food webs of subtropical eutrophic waters.

2. Materials and methods

2.1. Chemicals and materials

Sixteen priority PAHs including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo[*a*]anthracene (BaA), benzo[*b*]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*c*,*d*]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), and benzo[g,h,i]perylene (BghiP) were studied. Standard solutions of 16 priority PAHs with purity >99% bought from Accustandard (New Haven, USA) were used for quantification. All solvents used were of HPLC grade (Merk, Germany).

2.2. Site description and sample collection

Four Chinese subtropical lakes with different trophic state index were chosen for this study (Fig. 1 and Table S1). Trophic state index was calculated according to the studies of Carlson (1977) and Hu et al. (2014). Lake Xuanwu and Lake Baijia are shallow urban lakes located in Nanjing, with areas of 3.68 and 1.60 km^2, and mean depths of 1.14 and 1.60 m. Lake Jinniu is a rural lake 103 km far from Nanjing, with an area of 17 km^2 and a mean depth of 5.74 m. Lake Taihu is the third largest freshwater lake of China, 230 km far from Nanjing, with an area of 2338 km^2 and a mean depth of 1.89 m. The annual mean temperature of this area is 15–16 °C. Sampling campaigns were performed simultaneously at one site in each lake every ten to fifteen days from October 2015 to September 2016. All together 26 sampling campaigns were conducted for each lake. Water, phytoplankton, zooplankton, and TSP were collected simultaneously from each lake during each sampling campaign. Twenty liters of water sample were collected separately from the depths of 30, 60, and 90 cm, and combined together as one sample. Zooplankton (>112 μm, composed of copepods and cladocerans) were collected and concentrated from 50 L of combined water sample with a 112-μm mesh net, and then transferred onto a pre-combusted and pre-weighed GF/F glass fiber filter. Two liters of water sample after filtered with 112-μm mesh net was used to collect phytoplankton and TSP by sinking (Chen et al., 2006; Tao et al., 2017b, 2017c). After sinking for 30 min, an aliquot of 500 mL supernatant water was filtered through a GF/F glass fiber filter to collect phytoplankton. The TSP was collected by filtering the bottom water with a GF/F glass fiber filter. The result of Chl *a* showed that more than 90% of the phytoplankton and TSP were separated by this method. The samples of phytoplankton, zooplankton, TSP, and surface sediments were freeze-dried, weighed, and stored at −18 °C until chemical analysis. An aliquot of 500 mL of the filtered water was concentrated with a preconditioned C_{18} solid phase extraction column to collect the dissolved PAHs. An aliquot of 50 mL of the filtered water was analyzed for dissolved organic carbon (DOC). The mean temperature at the beginning (the first ten days), the middle (the second ten days), and the end (the third ten days) of each month was collected from local meteorology, and used as the corresponding mean water temperature of each lake for each sampling campaign.

2.3. Chemical analysis

The freeze dried filters with TSP, phytoplankton, and zooplankton were ground separately in a mortar to obtain homogeneous samples. Each of them was sonic extracted three times with 20 mL of *n*-hexane and dichloromethane (1: 1, v/v) each time. The extract was reduced to approximately 5 mL with a rotary evaporator, purged to 1–2 mL under a gentle nitrogen stream, and then cleaned with a Waters gel permeation chromatography (GPC) composed of 1515 isocratic pump, 2707 auto sampler, 2489 UV/Vis detector, Fraction Collector III, and an Envirogel GPC Cleanup column (19 mm × 150 mm). The mobile phase was consisted of *n*-hexane/dichloromethane (1: 1 v/v), and pumped at a rate of 1.5 mL/min. The eluate was evaporated, solvent-exchanged to *n*-hexane (100 μL), and analyzed immediately. The C_{18} solid phase extraction column was eluted with 70 mL of *n*-hexane/dichloromethane (7: 3 v/v). The eluate was dried with anhydrous sodium sulfate, then

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