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PAH distribution and mass fluxes in the Three Gorges Reservoir after impoundment of the Three Gorges Dam

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HIGHLIGHTS

- We calculated PAH mass fluxes for the Three Gorges Reservoir (China) for 2008, 2009 and 2011.
- The PAH mass flux decreased significantly from upstream Chongqing to downstream Maoping.
- High PAH mass input could be linked to industrialized and metropolitan areas with high population density.
- PAHs bind readily to suspended particles and the amount of particle-bound PAH is not negligible.
- Our model predicts the PAH concentration caused by particle-bound PAH to be six times lower than PAHs dissolved in water.

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ABSTRACT

Mass fluxes of polycyclic aromatic hydrocarbons (PAHs) were calculated for the Three Gorges Reservoir (TGR) in China, based on concentration and discharge data from the Yangtze River. Virtual Organisms (VOs) have been applied during four campaigns in 2008, 2009 (twice) and 2011 at sampling sites distributed from Chongqing to Maoping. The total PAH mass fluxes ranged from 110 to 2160 mg s^{-1}. Highest loads were determined at Chongqing with a decreasing trend towards Maoping in all four sampling campaigns. PAH remediation capacity of the TGR was found to be high as the mass flux reduced by more than half from upstream to downstream. Responsible processes are thought to be adsorption of PAH to suspended particles, dilution and degradation. Furthermore, the dependence of PAH concentration upon water depth was investigated at Maoping in front of the Three Gorges Dam. Although considerable differences could be revealed, there was no trend observable. Sampling of water with self-packed filter cartridges confirmed more homogenous PAH depth distribution. Moreover, PAH content of suspended particles was estimated from water concentrations gathered by VOs based on a water–particle separation model and subsequently compared to PAH concentration measured in water and in filter cartridges. It could be shown that the modeled data predicts the concentration caused by particle-bound PAHs to be about 6 times lower than PAHs dissolved in water. Besides, the model estimates the proportions of 5- and 6-ring PAHs being higher than in water phase.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are environmental pollutants that are released mainly by combustion processes in diesel and gasoline engines, coal and wood-burning furnaces as well as from general soot and smoke of municipal, domestic and industrial origin (EPA, 2008). They are known to be toxic and carcinogenic compounds (Mastral et al., 2003). Further transition or metabolism can lead to the formation of even more toxic compounds like nitrated PAHs of which some are suspected to have higher mutagenicity than their precursors (Fu et al., 1994; Topinka et al., 1998).

Detection of persistent organic pollutants (POPs) in the Yangtze River and its Three Gorges Reservoir (TGR) is a demanding task due to high dilution and consequently low concentrations of target compounds in the large water body. At the same time, the pollution of

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the ecosystem is regarded as severe (Müller et al., 2008) and the Yangtze River is listed among the most polluted rivers worldwide (Wong et al., 2007). These at first sight opposing facts can be well understood when the large amount of discharge of the Yangtze is taken into account: the river with its length of 6300 km and catchment area of 1,810,000 km^2 (Yang et al., 2005) has a mean annual water discharge of 29,400 $m^3\ s^{-1}$ (Zhang, 1995). Therefore low concentrations correspond to high mass fluxes in total (Müller et al., 2008). In 2010, the TGR stored 26.43 billion m^3 of flood water and the normal impoundment level of 175 m was reached in October for the first time (CNEMC, 2011). The main sources of pollution within the reservoir are well identified as the huge metropolitan centers Chongqing, Changshou, Fuling, and Wangzhou with 220 million tons of industrial effluent and 486 million tons of urban sewage in 2010 (CNEMC, 2011). With 590 million tons, a large part of the 615 million tons urban sewage has been treated (CNEMC, 2011). Non-point source pollution like scattered domestic garbage (708,000 t in 2010) and oil containing wastewater from ships (481,300 t in 2010) additionally contribute to pollution of the TGR (CNEMC, 2011).

The purpose of this study was to analyze previously collected PAH water concentration data from sampling campaigns in 2008, 2009 and 2011 and put them into perspective using modeled water discharge values from the Yangtze River upstream of the Three Gorges Dam (TGD). Bioavailable PAH water concentrations have been evaluated in a similar manner in all sampling campaigns by deploying virtual organisms (VOs) as passive samplers ensuring compatibility. Sampling locations have been chosen to cover the whole TGR from Chongqing till the TGD. Sample analysis included all 16 PAHs which are on the priority list of the US Environmental Protection Agency. Calculated PAH mass fluxes reveal a better overview of possible pollution sources and processes like degradation, dilution and sorption of the pollutants. In addition, new PAH water concentration data regarding depth distribution are presented. Therefore, sampling was carried out with VOs and self-packed sampling cartridges in 2012 at a single sampling site in front of the TGD in different water depths.

2. Material and methods

2.1. Data sources

For this work, concentration data gathered during previous sampling campaigns in the TGR have been used which are already published to a large extent (Wang et al., 2009, 2013). The VO sampling campaigns took place in May 2008, April–May 2009, May–June 2009 and April–May 2011 and covered the whole TGR. The single sampling sites (from upstream to downstream) were located at Chongqing, Changshou, Wanzhou, Badong, Guojiaba and Maoping (see Fig. 1). In 2011, five additional sites in Yunyang, Fengije and Wushan were investigated. At each sampling site, seven VOs were deployed whereas usually a single VO was sufficient for analysis. Details regarding deployment of the samplers can be found in Section 2.2.

Moreover, VOs were deployed at a single site in the middle of the Yangtze River near the TGD in August/September 2012 in 1, 11, 21, 31, 41, 51 and 61 m water depth to investigate the PAH distribution between different water layers. In a similar sampling campaign which took place at the same place and time, PAH concentrations in water and suspended particles have been evaluated by active sampling on self-packed XAD-cartridges (see Section 2.3 for details) in 11, 31 and 61 m water depth.

Daily data at the three hydrological stations Cuntan, Wanzhou and Yichang were used to estimate the discharges at different locations in the Three Gorges Reservoir (TGR). According to different sampling periods, averaged daily discharges were calculated for Cuntan and Yichang. These discharges represent upstream and downstream flow boundaries for the TGR. Since discharge data at Wanzhou were only available from February 2004 to May 2007 (afterwards only water level data was reported), the discharge data at Wanzhou were used as reference for the discharge estimation. Average annual discharge data at the confluences or major control hydrological stations of major tributaries in the TGR were considered to estimate the mainstream discharge increase of the Yangtze River from upstream to downstream. This

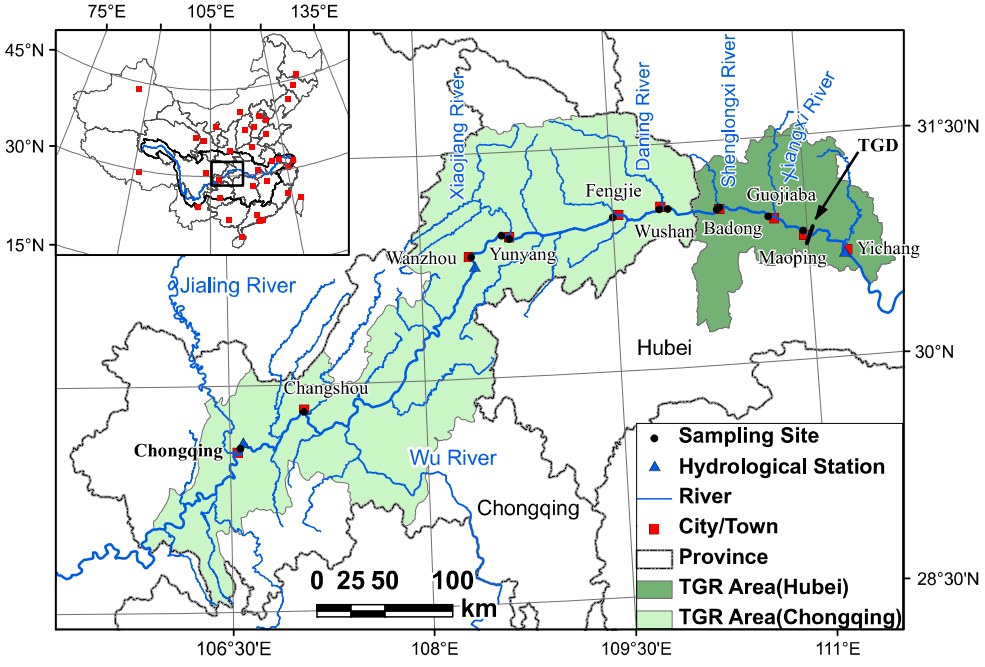

Fig. 1. Map of the Three Gorges Reservoir; the sampling sites and hydrological stations (used in discharge calculation) are marked.