Status, source and health risk assessment of polycyclic aromatic hydrocarbons (PAHs) in soil from the water-level-fluctuation zone of the Three Gorges Reservoir, China

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ABSTRACT

The paper discussed the distributions, sources and health risk of polycyclic aromatic hydrocarbons (PAHs) in the water-level-fluctuation zone (WLFZ) of the Three Gorges Reservoir (TGR), China, in where the rapid transfer of pollutants between the water and terrene environment cased by the cycle of water fluctuation is of great concern. Twenty-four soil samples were collected in two sampling campaigns, i.e., in September 2012 (the water levels started rising from 145 m) and in June 2013 (the water levels just fell to 145 m), and subsequently they were analyzed for 16 USEPA priority PAHs. Total PAH concentrations ($\Sigma_{16}PAHs$) ranged from 18.40 to 392.29 $ng \cdot g^{-1}$ (mean 87.66 $ng \cdot g^{-1}$) in 2012, and 54.00 to 463.08 $ng \cdot g^{-1}$ (mean 133.84 $ng \cdot g^{-1}$) in 2013. The concentrations of ΣPAHs in soil sample in 2013 were significantly higher than those in 2012. The concentrations of ΣPAHs in the upstream counties (Banan, Changshou, Fuling and Fengdu) were significantly higher than that in the middle and the lower reaches counties (Zhongxian, Wanzhou, Yunyang, Fengjie, Wushan, Badong, Guizhou and Lanlingxi). The compounds of 3-rings and 4-rings were the major components of PAHs in this area. Individual PAH concentrations were not significantly correlated with total organic carbon (TOC) and microbial biomass carbon (MBC), possibly due to the soil microbial mineralized or degraded PAHs and the semi-continuous inputs from anthropogenesis sources. Source identification indicated that PAHs were mainly derived from river transport and combustion. The incremental lifetime cancer risks ranged from 10^{-9} to 10^{-6}, indicated that the carcinogenic risk for the residents was negligible.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of ubiquitous persistent organic pollutants (POPs) (Alebic-Juretic, 2011). They are considered as one of the most serious pollutants due to their persistence in the environment, bioaccumulation and high toxicity (carcinogenic, teratogenic and mutagenic) (Deyerling et al., 2014). Due to their low solubility and high hydrophobic-lipophilic, PAHs in water environment are easily to be combined with suspended solids, then deposited into sediment, and in atmospheric environment final destination also are sediment and soil by atmospheric precipitation and rainfall (Jiang et al., 2014). However, PAHs in sediment and soil can enter water and atmosphere then causes secondary pollution (Sarria-Villa et al., 2016). So the sediment and soil are the sink and source of PAHs. PAHs in the environment from various processes, mainly originate from human activities, such as petroleum producing and leaks, coal and its refined product, and other organic matter combustion (Alebic-Juretic, 2011), many industrial processes (Peng et al., 2011), motor vehicle emissions (Lee and Dong, 2010), and small amounts are produced by forest fires and volcanic eruptions (Ravindra et al., 2008).

The water-level-fluctuation zone (riparian zone) is an effective sink for PAHs due to its integration and deposit of pollutants from both the terrestrial and aquatic environment (Ye et al., 2011). In the non-flood season, PAHs in the riparian zone generally come from natural weathering and anthropogenic activities, such as riverine discharge, industrial wastewater, and domestic sewage. In the flood season, PAHs in the aquatic environment with the inputs from shipping and human activities in the upper streams can accumulate to the riparian zone by flooding. Therefore, PAHs in the riparian zone have been affected by flooding through fluvial transport and frequent material exchanges between the aquatic and terrestrial system. Meanwhile, organic carbon

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fractions play an important role in controlling the adsorption of PAHs in soil, such as total organic carbon (TOC), black carbon (BC) and microbial biomass carbon (MBC) (Luo et al., 2015; Mahmoudi et al., 2013). The TOC and the MBC in soil are quite different between the non-flood season and the flood season for the change of water level. Furthermore, some microbes in soil can degrade certain PAHs (Johnsen et al., 2005; Kanaly and Harayama, 2000), and the various levels of the MBC may change the concentration of PAHs with the different types, quality, and oxidation or reduction conditions of soil. With the completion of the Three Gorges Dam, the Three Gorges Reservoir (TGR) inundates an area of 108,000 ha. The water level of the reservoir fluctuates from 145 m a.s.l. to 172 m a.s.l. in 2008, resulted in formation of the water-level-fluctuation zone (WLFZ) with a total area of 35,000 ha in the reservoir (Ye et al., 2011). There are concerns that PAHs from industrial activities and domestic consumption in the uplands may be carried to and accumulated in the WLFZ by floods (Floehr et al., 2015; Zhu et al., 2015). In this study, the objectives of the present study were to: a) investigate the concentrations and distribution of 16 priority PAHs in order to find out the characters of soil PAHs in a cycle change of water level fluctuation; b) determine the potential sources of the PAHs; and c) evaluate the potential cancer risks of PAHs using the incremental lifetime cancer risk approach. It can offer beneficial reference for the WLFZ ecological restoration and reconstruction.

2. Materials and methods

2.1. Sampling

The WLFZ was exposed to the air after submergence in May when the reservoir's water level was 145 m. From September 2012 (the water level started rising from 145 m) to June 2013 (the water level just fell to 145 m), We selected 12 sampling sites based on the geographical characteristics of the WLFZ from upstream to downstream in the TGR (Fig. 1), and totally 24 soil samples were collected from the WLFZ of the TGR in September 2012 and in June 2013, to find out the changes of soil PAHs in a cycle of water level fluctuation. In each site, five sampling plots (1 m × 1 m) were randomly selected in a 100 m² field of the elevation from 165 m to 175 m. At each plot, we collected five topsoil samples (0–10 cm) and the samples were well mixed to form a composited sample. All the samples were wrapped in aluminum foil and stored in sealed polythene bags and freeze dried at −4 °C before analysis.

2.2. Analysis of PAHs

The procedures of preparing for the samples were similar to those described previously (Zhang et al., 2015). The soil samples were dried naturally, and then a 10 g soil sample was spiked with 1000 ng mixed-recovery surrogates (napthalence-d_8, acenapthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12} and perylene-d_{12}), and Soxhlet-extracted with 125 mL dichloromethane (DCM) for 24 h. Elemental sulphur was removed by adding activated copper sheets to the collection flasks. The sample extract was concentrated and the solvent exchanged with hexane and further reduced to 2–3 mL by a rotary evaporator (Heidolph-4000, Germany). A 1:2 (v/v) alumina/silica gel column (48 h extraction with DCM, then baked muffle for 12 h at 180 °C and 240 °C, both deactivated with 3% of its weight water before use) was used to cleanup the extract; then the PAHs were eluted with 30 mL of DCM/n-Hexane (v:v = 2:3). The eluate was concentrated to 0.2 mL under a gentle nitrogen stream (purity ≥ 99.999%). Prior to analysis, a known quantity of hexamethylbenzene (1000 ng) was added to the eluate as an internal standard.

All the soil samples were analyzed for 16 PAHs listed by the U.S. Environmental Protection Agency as priority PAHs: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluorantene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1, 2, 3-cd)pyrene (IcdP), dibenzo(a, h)anthracene (DBA) and benzo(g, h, i)perylene (BghiP).

The determination of PAHs was performed by a GC–MS (Agilent 7890 A-5975C) equipped with a DB-5 capillary column (30 m × 0.25 mm i.d × 0.25 μm film thickness). The carrier gas was high-purity helium at a constant flow rate of 1.0 mL/min. Mass selective detector operated in the electron impact mode at 70 eV. The chromatographic conditions were as follows: injector temperature and detector temperature were kept at 270 °C and 280 °C. The oven temperature program
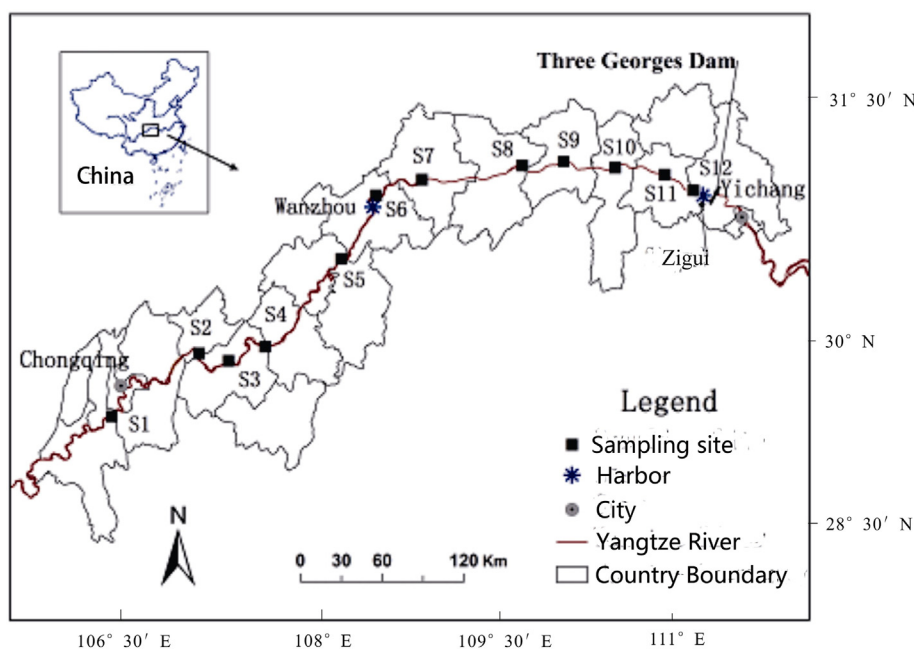
Fig. 1. Location of the sampling sites. S1. Banan, S2. Changshou, S3. Fuling, S4. Fengdu, S5. Zhongxian, S6. Wanzhou, S7. Yunyang, S8. Fengjie, S9. Wushan, S10. Badong, S11. Guizhou, S12. Lanlingxi.