



Impact of upgrading wastewater treatment plant on the removal of typical methyl, oxygenated, chlorinated and parent polycyclic aromatic hydrocarbons

Meng Qiao, Wei Cao, Bochuan Liu, Yaohui Bai, Weixiao Qi, Xu Zhao*, Jiuhui Qu

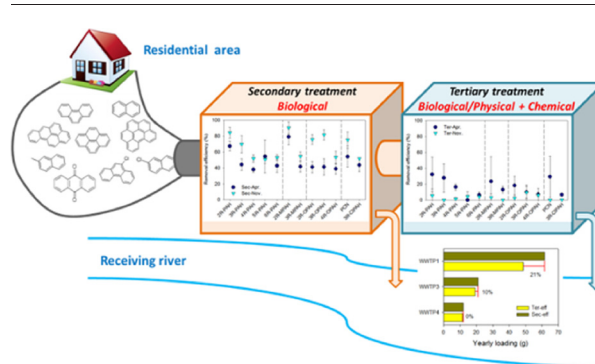
Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China



HIGHLIGHTS

- Chlorinated PAHs exist in WWTPs in Beijing with low concentration.
- SPAHs should not be ignored due to the similar total concentration to PAHs.
- PAHs and SPAHs mainly removed by secondary treatment but not tertiary treatment.
- Dissolved organic matter might prevent the removal of PAHs and SPAHs in WWTPs.
- Upgrading of WWTPs may not efficiently decrease the discharge of PAHs and SPAHs.

GRAPHICAL ABSTRACT



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ABSTRACT

Wastewater treatment plant (WWTP) secondary effluent is a main source for polycyclic aromatic hydrocarbons (PAHs) and their derivatives (SPAHs) to wastewater receiving rivers in Beijing. The treatment technologies are being upgraded in the WWTPs as the tertiary treatment. To assess the improvement of the removal efficiencies of PAHs and SPAHs after the treatment upgrading, we investigated 16 PAHs and 4 types of SPAHs in the secondary and tertiary treatment process in 5 major WWTPs. Most of the parent PAHs, methyl PAHs, oxygenated PAHs and chlorinated PAHs were detected in the influent, secondary and tertiary effluent. The concentrations of Σ SPAHs (61 ng/L–529 ng/L) were similar to Σ PAHs (89 ng/L–474 ng/L), indicating that SPAHs should not be ignored when studying the PAH contamination. Σ PAHs and Σ SPAHs were largely removed by the secondary treatment (45%–82%) and less by the tertiary treatment (0%–24%). The removal efficiencies were lower in the secondary and tertiary treatment in WWTPs than in the lab-scale experiment conducted previously, probably a result of the association of PAHs and SPAHs with dissolved organic matters (DOMs) in wastewater. DOMs might be a limiting factor for the removal of PAHs and SPAHs in WWTPs. The estimated yearly loadings of the total PAHs and SPAHs decreased only 21% in the tertiary effluent compared with the secondary effluent in WWTP1 and 9% in WWTP3. Therefore, the upgrading of WWTPs did not efficiently improve the removal of PAHs and SPAHs. DOMs should be further considered for improving the removal of PAHs, SPAHs and similar contaminants in WWTPs.

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* Corresponding author.

E-mail address: zhaoxu@rcees.ac.cn (X. Zhao).

1. Introduction

Wastewater treatment plant (WWTP) effluent is a main source for polycyclic aromatic hydrocarbons (PAHs) and some of their derivatives (SPAHS) in wastewater receiving rivers in Beijing (Qi et al., 2013; Qiao et al., 2014b). Until now, the secondary treatment (biological treatment) effluent directly discharges to the receiving rivers. During the biological treatment process in WWTPs, the occurrence and removal of PAHs have been well studied previously (Bergqvist et al., 2006; Fatone et al., 2011; Manoli and Samara, 1999; Mezzanotte et al., 2016; Ozaki et al., 2015). However, to the best of our knowledge, SPAHS, such as methyl PAHs (MPAHs), oxygenated PAHs (OPAHs), polychlorinated naphthalenes (PCNs) and chlorinated PAHs other than PCNs (CIPAHs) have been seldom reported in WWTPs. The toxicities of some SPAHS, especially some CIPAHs, are higher than their corresponding PAHs (Bhatia et al., 1987; Iino et al., 1999; Ohura, 2007). CIPAHs and PCNs, emitted from traffic and incineration facilities, are widely spread in the atmosphere environment (Ohura, 2007). Similar to OPAHS, CIPAHs can be formed by PAHs during chemical reactions, such as PAHs incineration at 400–600 °C in the existence of O₂ or Cl (Iino et al., 1999; Yoshino and Urano, 1997). CIPAHs have also been detected in tap water, formed during water chlorination (Johnsen et al., 1989; Shiraishi et al., 1985). PCNs have been detected in activated sludge from WWTPs (Stevens et al., 2003; Zhang et al., 2014). So, we suppose that CIPAHs and PCNs exist in the dissolved phase of the wastewater from WWTPs. MPAHS and OPAHS were detected during the secondary treatment in WWTPs in one study from our group (Qiao et al., 2014a). In addition, OPAHS were proved to be formed from PAHs during activated sludge treatment process (Qiao et al., 2016b). Therefore, besides PAHs, the occurrence and removal of SPAHS should be taken into consideration in wastewater treatment process.

The treatment technologies are being upgraded in some of the WWTPs in Beijing (tertiary treatment). The tertiary treatment effluent will discharge into the receiving rivers instead of the secondary effluent in the future years. Therefore, it is necessary to investigate the removal of the toxic organic pollutants during the tertiary treatment process to forecast the improvement of water quality by the upgraded treatment. The removal of pharmaceutical and personal care products has been well studied during the tertiary treatment process, as well as in the WWTPs in Beijing (Hollender et al., 2009; Li et al., 2013; Sui et al., 2010). Ozonation is a commonly used technique in the tertiary treatment process, especially in the WWTPs in Beijing. PAHs ozonation has been studied in lab-scale experiment. PAHs were found to be easily eliminated by ozone (Trapido et al., 1995). However, in WWTPs, the removal of PAHs and SPAHS has rarely been investigated in the tertiary treatment process including the ozonation process.

The objective of this study was to: 1) identify the existence of typical PCNs and CIPAHs in WWTPs; 2) investigate the removal efficiency and deduce the removal mechanism of PAHs and typical SPAHS both in the secondary and tertiary treatment processes; 3) estimate the discharge reduction of PAHs and SPAHS to the wastewater receiving rivers after the treatment technology upgraded. The result will provide bases for forecasting the improvement for the water quality of the WWTP effluent receiving river on the aspect of PAHs and typical SPAHS after the techniques upgraded in the WWTPs.

2. Materials and methods

2.1. Wastewater treatment process

The removal of PAHs and typical SPAHS were investigated in 5 major WWTPs in Beijing by the secondary and tertiary treatment process. All the secondary treatment processes are biological treatment in the 5 WWTPs. The tertiary treatment processes are chemical treatment combined with biological or physical treatment in 3 WWTPs. The 5 WWTPs

treat 70% of the domestic wastewater in Beijing. The details of the WWTPs are listed in Table 1.

2.2. Sample collection

The influent, secondary effluent and tertiary effluent of the WWTPs were collected in April and November 2015, representing the non-heating season and the heating season respectively. The concentrations of PAHs and SPAHS were relatively stable on different days during one week based on our previous study (Qiao et al., 2014a). Additionally, during the sampling period in this study, there was no large precipitation which might influence the inflow velocity and quality. Therefore, the samples were collected once in each season. The sampling sites are shown in Table 1. Some samples could not be collected, including the secondary influent in WWTP3 in April, the tertiary effluent in WWTP4 and the secondary influent in WWTP5 in November. 4 L water was collected for each grab sample. The sampling was limited by the lack of auto samplers and also the sampling time limitation. Grab samples could be still deemed acceptable, because the concentrations of PAHs and SPAHS were relatively stable at 3 sampling times during one day period based on our previous study (Qiao et al., 2014a). Furthermore, as indicated above, during this sampling period, the inflow velocity and water quality were relatively stable.

2.3. Chemicals and materials

Seventeen individual SPAH standards included 4 CIPAH standards, 9-chlorophenanthrene (9-ClPhe, 50 µg/mL in isooctane), 2-chloroanthracene (2-ClAnt, 50 µg/mL in isooctane), 1-chloroanthraquinone (1-ClAQ, solid, 98%), 9,10-dichloroanthracene (9,10-DCIAnt, solid, 96%) purchased from Chiron (Norway); 5 PCN standards, 1-chloronaphthalene (1-CN, solid, 95.5%), 2-chloronaphthalene (2-CN, 100 µg/mL in methanol), 1,4-dichloronaphthalene (DCN, solid, 99.4%), 1,2,3,4-tetrachloronaphthalene (TeCN, solid, 100%) and octachloronaphthalene (OCN, 100 µg/mL in methanol); 4 MPAH standards, 2-methylnaphthalene (2-MN, solid, 100%), 1-methylfluoranthene (1-MF, 10 µg/mL), 2,6-dimethylnaphthalene (2,6-DMN, solid, 100%), 3,6-dimethylphenanthrene (3,6-DMP, solid, 100%); 4 OPAH standards, 9-fluorenone (9-FL, solid, 100%), anthraquinone (AQ, 100 µg/mL), 2-methylanthraquinone (2-MAQ, solid, 99.0%), benz[*a*]anthracene-7,12-dione (BA-7,12-D, 50 µg/mL) purchased from AccuStandard, Inc. (USA). The sixteen USEPA priority PAHs, including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fluo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flua), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Chry), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene (IcdP), dibenz[*a,h*]anthracene (DBA), and benzo[*g,h,i*]perylene (BghiP), in a mixture (200 µg/mL) were purchased from AccuStandard, Inc. (USA). The physical and chemical properties are listed in Table S1. Surrogate standards including 4 deuterated PAHs (d-PAHs) acenaphthene-d10 (d-Ace), phenanthrene-d10 (d-Phe), chrysene-d12 (d-Chry) and perylene-d12 (d-Pery) in a mixture (2000 µg/mL) were also purchased from AccuStandard, Inc. Internal standards including 2-fluorobiphenyl (2-FB, in solid > 96%) and decachlorobiphenyl (PCB209) were obtained from Aldrich Chemical Co., Inc. (Gillingham, Dorset, UK).

Hexane (HEX, Fisher Scientific, USA), dichloromethane (DCM, J. T. Baker, USA) and methanol (MeOH, Fisher Scientific, USA) were HPLC grade solvents. Silica gel (0.06–0.2 mm) and alumina (100–200 mesh) for chromatography were purchased from Acros Organics, Inc. (USA). Before use, silica gel and alumina were baked at 180 °C and 250 °C respectively for 12 h, and then deactivated with 3% ultrapure water, and kept in HEX until use. Analytical grade anhydrous sodium sulfate (Tianjin, China) was baked at 450 °C for 5 h before use.

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