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Profiles and removal efficiency of polycyclic aromatic hydrocarbons by two different types of sewage treatment plants in Hong Kong

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ABSTRACT

Sewage discharge could be a major source of polycyclic aromatic hydrocarbons (PAHs) in the coastal waters. Stonecutters Island and Shatin Sewage Treatment Works (SCISTW and STSTW) in Hong Kong, adopted chemically enhanced primary treatment and biological treatment, respectively. This study aimed at (1) determining the removal efficiencies of PAHs, (2) comparing the capabilities in removing PAHs, and (3) characterizing the profile of each individual PAHs, in the two sewage treatment plants (STPs). Quantification of 16 PAHs was conducted by a Gas Chromatography. The concentrations of total PAHs decreased gradually along the treatment processes (from 301 ± 255 and 307 ± 217 ng/L to 14.9 ± 12.1 and 63.3 ± 54.1 ng/L in STSTW and SCISTW, respectively). It was noted that STSTW was more capable in removing total PAHs than SCISTW with average total removal efficiency 94.4% ± 4.12% *vs.* 79.2% ± 7.48% ($p < 0.05$). The removal of PAHs was probably due to sorption in particular matter, confirmed by the higher distribution coefficient of individual and total PAHs in solid samples (dewatered sludge contained 92.5% and 74.7% of total PAHs in SCISTW and STSTW, respectively) than liquid samples (final effluent-total contained 7.53% and 25.3% of total PAHs in STSTW and SCISTW, respectively). Despite the impressive capability of STSTW and SCISTW in removing PAHs, there was still a considerable amount

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of total PAHs (1.85 and 39.3 kg/year, respectively for the two STPs) being discharged into Hong Kong coastal waters, which would be an environmental concern.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous organic pollutants consisting of two or more aromatic rings fused together in a molecule. They are produced from incomplete combustion of organic substances through natural as well as anthropogenic sources (Kim et al., 2005). The anthropogenic PAHs are mainly released into the environment through incomplete combustion of fossil fuels, burning coke and discharge of petroleum-related materials (Pham and Proulx, 1997). They are toxic, carcinogenic, and mutagenic (ATSDR, 1995). The US Environmental Protection Agency (EPA) has classified 16 PAHs as priority pollutants and their levels in effluents need extensive control and monitoring (Keith and Telliard, 1979; Roger, 1996; Blanchard et al., 2004). Among all PAHs, the US EPA identified seven PAH compounds as probable human carcinogens (Group B2), including benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-cd]-pyrene (ATSDR, 1995). The International Agency for Research on Cancer (IARC, 2010) has further indicated that benzo[a]pyrene as Group 1 carcinogen, based on sufficient evidence in humans and sufficient evidence in animals and strong mechanistic data in humans.

The PAHs have acute effects on both terrestrial and aquatic environment (Douben, 2003). Our early study reported that PAHs could be bioaccumulated within fish muscle (*Nile tilapia*) (186 ± 33.9 ng/g dw, body weight: 181 ± 35.9 g and $N = 17$) in Mai Po Mashes, Hong Kong (the most important site for biological conservation in South China), which may be biomagnified in food chains, imposing adverse effects on local and migratory birds and even human beings (Liang et al., 2007).

Effluents of municipal sewage treatment plants (STPs) could be a significant source of PAHs contamination (Peng et al., 2009; Song et al., 2006). In fact, PAHs have been included in the list of organic contaminants (Directive 2006/11/CE), for monitoring sewage discharge from STPs (European Parliament and The Council, 2006). The levels commonly found PAHs in wastewater could be up to ppb level (μg/L) (Céspedes et al., 2005; Porte et al., 2006; Fatone et al., 2011), which could impose toxic effects, neurotoxicity and endocrine disruption in marine organisms at the ecosystem level (Chapman, 2004; Tiffany-Castiglioni et al., 2006).

Most PAHs are generally hydrophobic with high boiling and melting points, and possess low water solubility (Haritash and Kaushik, 2009). The solubility of PAHs decreases when more aromatic rings fused in a molecule (Fatone et al., 2011). Therefore, smaller PAH molecules tend to have smaller log octanol–water partition coefficient K_{ow} and log partitioning coefficient K_p between solid and liquid phases (ATSDR, 1995). Because of the low solubility, high hydrophobicity and biodegradation resistant properties of PAHs, they are adsorbed onto the solid particle and could be removed from wastewaters by activated sludge treatment (Roger, 1996). Therefore, considerably high concentrations of PAHs were found in sludge from STPs ranged from 0.0610 to 6.60 mg/kg in Guangzhou, China (Cai et al., 2007).

It has been noted that the removal efficiency of PAHs in STPs decreased with molecular sizes (Wang et al., 2013). Although part of the highly hydrophobic PAHs could be removed by sedimentation of suspended solids, the remaining less hydrophobic compounds might not be completely removed and thus released to receiving bodies (Peng et al., 2009), and subsequently affect the drinking water quality (Rayne and Ikonomou, 2005). Therefore, it is necessary to determine the source, occurrence and fate of PAHs input into STPs, in order to minimize the release of this toxic pollutant to aquatic environment.

The Pearl River Delta (PRD), located in southern China, has become a key developing region with high levels of urbanization and industrialization, releasing different toxic organic pollutants (including PAHs) into the environment. Concentrations of PAHs found in dry sludge collected from Guangdong Province near PRD, China varied from 2534 to 6927 μg/kg (Zeng et al., 2010). Hong Kong, located near to the estuary of PRD, is also exposing to serious water pollution through different human activities. In Hong Kong, the majority of sewage goes through either chemically enhanced primary treatment (CEPT) or secondary treatment (plain primary settling and biological treatment) (DSD, 2013). The removal of PAHs in STPs could be largely related to the sorption onto particulate matter (Fatone et al., 2011). Two STPs in Hong Kong were chosen for this study: Stonecutters Island Sewage Treatment Works (SCISTW) uses CEPT process and Shatin Sewage Treatment Works (STSTW) adopts biological treatment in the secondary stage (DSD, 2009a, 2009b).

A number of previous research studies have reported the occurrence and source of PAHs in municipal wastewater in different countries including China (Song et al., 2006; Wang et al., 2013). However, there is still insufficient information regarding the removal efficiency and release of PAHs in the STPs in Hong Kong. Moreover, several studies only focus on the concentrations of PAHs in particular phase of STPs such as sludge (Sánchez-Avila et al., 2009; Zeng et al., 2010; Zhang et al., 2012). There are limited studies that have comprehensive evaluation on the fate of PAHs in different stages of STPs.

Therefore, we hypothesized that the two STPs have different removal efficiencies of PAHs and different sewage samples: influent including liquid portion and particulate matter, effluent including liquid, particulate matter and sludge may have different profiles of PAHs. More specifically, the objectives of this study were to (1) determine the removal efficiencies of PAHs in the two major STPs, (2) compare the capabilities of the two STPs in removing PAHs, and (3) characterize the profile of each individual PAHs by evaluating their percentages in the total PAHs in the two STPs.