



Formation of oxygenated polycyclic aromatic hydrocarbons from polycyclic aromatic hydrocarbons during aerobic activated sludge treatment and their removal process



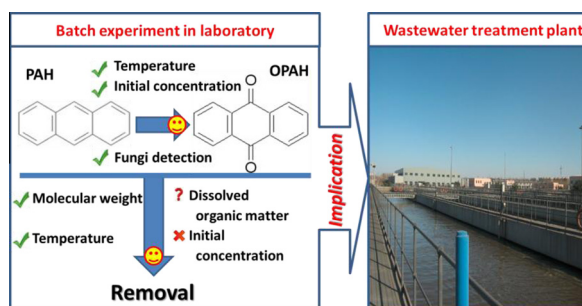
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HIGHLIGHTS

- Transformation from anthracene to anthraquinone was validated in biological treatment.
- Functional fungi for PAH transformation to OPAH were detected in activated sludge.
- The low molecular weight compounds were easier removed than the high ones.
- The middle temperature was fit for compounds removal than the low and high ones.
- The dissolved organic matter probably significantly influenced the compounds removal.

GRAPHICAL ABSTRACT



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ABSTRACT

Though previous studies have focused on the removal of polycyclic aromatic hydrocarbons (PAHs) and their oxygenated derivatives (OPAHs) in wastewater treatment plants, there is a lack of information on the transformation from PAHs to OPAHs during the activated sludge treatment. In this study, a batch experiment was carried out to simulate the aerobic biological treatment process, and the fungi species was detected by high-throughput sequencing. In the result, the transformation from PAHs (anthracene) to OPAHs (anthraquinone) was confirmed. A higher temperature (25 °C/35 °C) and higher initial concentration (5.0 µg/L) of anthracene promoted the formation of anthraquinone, compared with a lower temperature (10 °C) and lower initial concentration (0.5 µg/L). The relevant functional fungi for the transformation were detectable in the activated sludge (*Candida parapsilosis*, *Candida tropicalis* and *Bjerkandera*-sp), confirming the biotransformation. After 168 h, anthraquinone was totally biodegraded, indicating that activated sludge treatment was satisfactory for PAHs removal from the aspect of the biodegradation of the intermediate OPAHs. Furthermore, the possible factors influencing the biodegradation of both PAHs and OPAHs were also determined. The lower molecular weight compounds were more easily biodegraded than the higher ones. The temperature around 25 °C was more appropriate for compounds removal than 35 °C or 10 °C, because the microbial species were more plentiful in activated sludge at 25 °C. Different initial concentrations (0.5 µg/L, 5.0 µg/L) did not significantly influence the removal efficiency. However, the compounds we added to the samples could be more easily removed than those inherent in the samples, probably resulting from the stronger combination of the inherent compounds with the dissolved organic matters. The results from the batch experiment could be valid indicators for the effect of real wastewater treatment process.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) and their derivatives, such as oxygenated PAHs (OPAHs), methyl PAHs (MPAHs) and nitrated PAHs (NPAHs) are emitted from incomplete combustions and diesel engines [1,2]. The derivatives could also be created by transformation from their corresponding PAHs by chemical and/or biological processes [3,4]. Some of the derivatives are as toxic as or more toxic than their corresponding parent compounds [5,6]. Previous studies reported the existence of MPAHs and OPAHs in wastewater treatment plants (WWTPs) as well as in rivers [7,8]. Although these compounds can be partially removed in biological WWTPs, the effluent from the corresponding WWTPs is a major source for PAHs and their derivatives in the receiving river systems, especially in Beijing, China [8,9].

It has been reported that PAHs can be transformed to OPAHs by the white rot fungi, which produce the laccase, manganese peroxidase and lignin enzymes. In order to determine the function of a particular white rot fungus, a number of studies investigated the biotransformation from PAHs to OPAHs using a single white rot fungus. Fluorene could be transformed to 9-hydroxyfluorene and 9-fluorenone (the major product) by manganese peroxidase and lignin enzyme [10]. Under the same conditions, anthracene and phenanthrene could transform to 9,10-anthraquinone and then to CO₂, and 9,10-phenanthraquinone then to 2,2'-diformylbiphenyl and finally to CO₂, respectively [11]. Vyas et al. investigated various functions of the white rot fungi, and concluded that all these fungi could transform anthracene to anthraquinone [12]. *Phanerochaete chrysosporium* and strain Px could accumulate anthraquinone, and *Pleurotus ostreatus*, *Coriopsis polyzona* and *Trametes versicolor* could further degrade anthraquinone, while *P. ostreatus* and *C. polyzona* could not degrade anthraquinone in the presence of anthracene [13]. There are still some studies reported the formation of anthraquinone from anthracene in real soil environment by the function of white rot fungi [4]. However, there has been few study focused on the performance of activated sludge in transforming PAHs to OPAHs. The formation of OPAHs in the wastewater treatment process has never been reported before, except a speculative conclusion in our previous study [7]. Meanwhile, whether the white rot fungi exist in WWTPs is also unknown.

The removal process of PAHs in WWTPs has been well investigated. In the biological combined with chemical precipitation treatment process, the removal efficiencies of $\Sigma 16$ PAHs (94–100%) were higher than those in the mechanical and the chemical combined mechanical process (25–78%) [14]. The influence of the molecular weight and temperature on the removal of PAHs in WWTPs has also been considered. Generally speaking, compared to the higher molecular weight (HMW) PAHs, the removal efficiencies of the lower molecular weight (LMW) PAHs were much higher, because the LMW PAHs could be more easily biodegraded/biotransformed than the HMW PAHs [15]. Combined with different treatment process, the HMW PAHs were easier to remove by chemical precipitation, while the LMW PAHs were necessary to remove by biological treatment [14]. The removal efficiencies of PAHs varied in different seasons with different temperatures. In summer, the removal efficiencies of PAHs were higher than those in winter, due to a more appropriate temperature in summer for microorganisms [16]. Regarding to SPAHs, only a few studies have reported the removal of MPAHs and OPAHs in WWTPs. Pham and Proulx investigated the concentrations of MPAHs both in the influent and effluent from a WWTP using physical–chemical treatment [17]. The average concentrations of 5 individual MPAHs in the influent ranged from 53 to 94 ng/L, and in the effluent from 15 to 35 ng/L; the removal efficiencies for individual MPAHs were

between 53% and 75%. Berqvist et al. examined the removal efficiencies of 5 most abundant MPAHs [15]. The concentrations of the individual naphthalene derivatives were 21–130 ng/L in the influent and 1–27 ng/L in the effluent. For other PAH derivatives, to the best of our knowledge, only one study from our group reported the removal of OPAHs in a biological WWTP [7]. Thus, there is a lack of study focused on the removal process and influencing factors of SPAHs. Although a number of study reported the behavior of PAHs in WWTPs, the process in real WWTPs would be influenced by varies environmental factors. Accordingly, the simulation of a batch experiment is indicated to be necessary, and it will be studied.

Therefore, the aims of this study were to: (1) validate the transformation from PAHs to OPAHs; (2) figure out the definite influencing factor for the biodegradation/biotransformation of PAHs and OPAHs; (3) compare the behavior of the compounds in a real WWTP and the batch experiment, to make the results from the batch experiment as indicators for the treatment effect of the real WWTPs.

2. Materials and methods

2.1. Chemicals

Six d-PAHs, including d-fluorene (d-Fluo, 10 $\mu\text{g/mL}$), d-phenanthrene (d-Phe, 200 $\mu\text{g/mL}$), d-anthracene (d-Ant, 2000 $\mu\text{g/mL}$), d-pyrene (d-Pyr, 500 $\mu\text{g/mL}$), d-benz[a]anthracene (d-BaA, 2000 $\mu\text{g/mL}$) and d-benzo[a]pyrene (d-BaP, 10 $\mu\text{g/mL}$); one d-OPAH, d-anthraquinone (d-AQ, 100 $\mu\text{g/mL}$); three OPAHs, encompassing 9-fluorenone (9-FL, in solid 100%), anthraquinone (AQ, 100 $\mu\text{g/mL}$), benz[a]anthracene-7,12-dione (BA-7,12-D, 50 $\mu\text{g/mL}$); and 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 $\mu\text{g/mL}$), were purchased from AccuStandard, Inc., New Haven, USA. Internal standards for 2-fluorobiphenyl (2-FB, in solid >96%) and decachlorobiphenyl (PCB209) were obtained from Aldrich Chemical Co., Inc. (Gillingham, Dorset, UK).

2.2. Biodegradation and transformation test system

The influent and activated sludge were collected from Qinghe WWTP in Beijing, China, in April, 2014. After transport to the laboratory, the influent and activated sludge were mixed in a 1:1 ratio, as in the WWTP. The average suspended solid volume (SSV) in each incubator was 4069 ± 368 mg/L.

The experiment simulated three temperature conditions, low temperature (10 °C, LT), middle temperature (25 °C, MT) and high temperature (35 °C, HT). Every sample was acclimated for 3 days, in aerobic conditions. The acclimation experiment was conducted in biochemical incubators, at constant temperature and in darkness. After the pretreatment, 0.5 $\mu\text{g/L}$ d-PAHs (d-Fluo, d-Phe, d-Ant, d-Pyr, d-BaA, d-BaP) were spiked into each sample.

To determine the influence of the initial PAH concentrations, 5.0 $\mu\text{g/L}$ of 4 types of d-PAH (d-Phe, d-Ant, d-Pyr, d-BaA) were added at 25 °C. In addition, for investigating the removal process of the 16 PAHs and 3 OPAHs, at the same temperature and the same initial d-PAH concentrations, certain amounts of the 3 OPAHs (5.0 $\mu\text{g/L}$) and 16 PAHs (5.0 $\mu\text{g/L}$) were added. Thus, in the same

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