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Enhanced micropollutant biodegradation and assessment of nitrous oxide concentration reduction in wastewater treated by acclimatized sludge bioaugmentation

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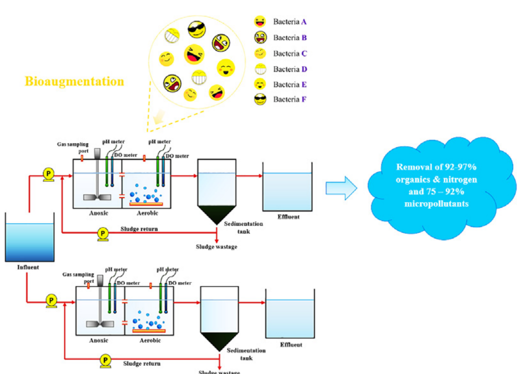
HIGHLIGHTS

- Acclimatized sludge promotes bacterial diversity and enhances micropollutant biodegradation of the AS system.
- Increased *nosZ* genes in the elevated biodiversity environment reduce N_2O concentration.
- Acclimatized sludge bioaugmentation is highly effective for the influent with low compounds concentration.

GRAPHICAL ABSTRACT



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ABSTRACT

This research investigated the micropollutant biodegradation and nitrous oxide (N_2O) concentration reduction in high strength wastewater treated by two-stage activated sludge (AS) systems with (bioaugmented) and without (non-bioaugmented) acclimatized sludge bioaugmentation. The bioaugmented and non-bioaugmented systems were operated in parallel for 228 days, with three levels of concentrations of organics, nitrogen, and micropollutants in the influent: conditions 1 (low), 2 (moderate), and 3 (high). The results showed that, under condition 1, both systems efficiently removed the organic and nitrogen compounds. However, the bioaugmented system was more effective in the micropollutant biodegradation and N_2O concentration reduction than the non-bioaugmented one. Under condition 2, the nitrogen and micropollutant biodegradation efficiency of the non-bioaugmented system slightly decreased, while the N_2O concentration declined in the bioaugmented system. Under condition 3, the treatment performance and N_2O concentration abatement were substantially lowered as the compounds concentration increased. Further analysis also showed that the acclimatized sludge bioaugmentation increased the bacterial diversity in the system. In essence, the acclimatized sludge bioaugmentation strategy was highly effective for the influent with low compounds concentration, achieving the organics and

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nitrogen removal efficiencies of 92–97%, relative to 71–97% of the non-bioaugmented system. The micropollutant treatment efficiency of the bioaugmented system under condition 1 was 75–92%, indicating significant improvement in the treatment performance ($p < 0.05$), compared with 60–79% of the non-bioaugmented system.

1. Introduction

High strength wastewater is wastewater with high concentrations of organics, inorganics, nitrogen, micropollutants, and toxic compounds. The micropollutants normally present in high strength wastewater are the phenol and phthalate plasticizer groups (Boonyaroj et al., 2012). Phenol and phthalate are classified by the World Health Organization and the U.S. Environmental Protection Agency as the endocrine disrupting chemicals because of the harmful effects on the reproductive system, neural development, and immune system. The micropollutants and toxic compounds normally found in the agriculture wastewater and municipal leachate are *N,N*-diethyl-*m*-toluamide (DEET) and pharmaceutical compounds. These micropollutants are biodegradation-resistant and pose serious risks to the ecosystem, aquatic environment, and human health (Muter et al., 2017; Park et al., 2017).

Existing biological wastewater treatment technologies are effective in removing organic and inorganic compounds, but less effective in removing nitrogen, micropollutants, and toxic compounds. Thus, advanced treatment technologies have been adopted to improve the biodegradation efficiency of micropollutants in wastewater, including the membrane-distillation-enzymatic bioreactor (Asif et al., 2018), the ferrate (VI) technology for the degradation of emerging micropollutants (Jiang et al., 2016), the graphene adsorption and simultaneous electrocoagulation/electrofiltration process (Yang et al., 2017), and the short-chain organic acids (SCOAs) to accelerate the reactivity of zerovalent iron nanoparticles with polychlorinated aromatic pollutants (Ou et al., 2016). Moreover, the micropollutant biodegradation was improved by optimizing the operating conditions, e.g., hydraulic retention time (HRT), solid retention time (SRT), and C/N ratios (Boonnorat et al., 2017; Jiang et al., 2018); and the nitrification, nitratation, and heterotrophic conditions (Fernandez-Fontaina et al., 2016).

In the biological treatment, greenhouse gases (GHG), including carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), are produced and emitted into the atmosphere, contributing to global warming. By comparison, N_2O contributes more to global warming than CO_2 over a 100-year cycle. Therefore, N_2O from the biological wastewater treatment received more interest in recent years (Mannina et al., 2016). During the biological wastewater treatment, N_2O is released during the nitrogen removal process, by which ammonia (NH_3) is transformed into nitrogen gas (N_2) by various bacterial groups (Toor et al., 2015). Thus, efforts have been made to reduce N_2O during the biological wastewater treatment, including optimizing the treatment conditions, e.g., the optimized dissolved oxygen (DO) concentration for the N_2O-producing ammonia oxidizing bacteria (AOB); and the advanced N-removal schemes, e.g., the anammox process (Massara et al., 2017).

According to Herrero and Stuckey (2015), bioaugmentation is a remedial strategy to enhance the wastewater treatment efficiency by introducing microorganisms into the wastewater reactor to catalyze the catabolism of refractory organic compounds. According to Boonnorat et al. (2014a, 2017), in the biological treatment system bioaugmented with acclimatized sludge (older than one year), the predominant bacterial communities were effective bacterial consortia, including heterotrophic bacteria (HB), heterotrophic denitrifying bacteria (HDB), heterotrophic nitrifying bacteria (HNB), and nitrifying bacteria (ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB)).

There exist research works on bioaugmentation to enhance the wastewater treatment efficiency. Qu et al. (2009) investigated the removal of bromoamine acid (dye) in a synthetic wastewater using *Sphingomonas xenophaga* QYY; and achieved the removal efficiency of 80–90%. Yu et al. (2010) examined the removal of *o*-Nitrobenzaldehyde (ONBA) in the synthetic wastewater augmented with the single strain *Pseudomonas putida* ONBA-17gfp, using a lab-scale sequencing batch reactor (SBR); and reported the complete removal of the compound. Hamjinda et al. (2017) experimented with *Pseudomonas putida*-entrapped PVA/SA gel beads in a bioaugmented MBR to remove nitrogen and pharmaceutical compounds. They found that *P. putida* enhanced the treatment efficiencies, despite short HRT (3, 6, and 12 h). *P. putida* is a heterotrophic-nitrifying-aerobic-denitrifying bacterial strain (Su et al., 2015).

For the abatement of N_2O, Ikeda-Ohtsubo et al. (2013) bioaugmented a wastewater treatment system with nitrous oxide-reducing denitrifier *Pseudomonas stutzeri* strain TR2. They reported that *P. stutzeri* strain TR2 survived and could effectively reduce the N_2O production in the biological treatment system.

Song et al. (2014) documented that high N_2O reductase-producing genes (*nosZ* genes) contributed to lower N_2O concentrations in the treatment system. They found that the bacterial genera *Rhodobacter*, *Oligotropha*, *Shinella*, and *Mesorhizobium* were the nitrous oxide-reducing bacteria that produce nitrous oxide reductase, which converts N_2O to N_2. Toor et al. (2015) noted that oxygen and N_2O in the treatment system were positively correlated. Yoon et al. (2016) found that the kinetic constants of nitrous oxide reductase greatly influenced the ability to suppress N_2O concentrations of microbes. There are two clades of nitrous oxide reductases characterized by different affinity constants.

There exists no publication on the use of acclimatized sludge to enhance the micropollutant biodegradation and reduce N_2O concentration in the biological AS system. Thus, this research investigates the micropollutant biodegradation and N_2O concentration reduction in high strength wastewater treated by two-stage activated sludge systems with (bioaugmented) and without acclimatized sludge bioaugmentation (non-bioaugmented). The high strength wastewater was a mixture of leachate and agriculture wastewater (1:1, v/v), diluted with RO water for lower influent concentrations. The target micropollutants were bisphenol A (BPA), 2,6-di-*tert*-butyl-phenol (2,6-DTBP), di-butyl-phthalate (DBP), di-(ethylhexyl)-phthalate (DEHP), carbamazepine (CBZ), diclofenac (DCF), and *N,N*-diethyl-*m*-toluamide (DEET).

2. Materials and methods

2.1. Two-stage activated sludge systems and operation

Two 20-L acrylic tanks, one containing the anoxic mixed liquor (DO = 0.5 mg/L) and the other the aerobic mixed liquor (4 mg/L), were used in the bioaugmented and non-bioaugmented systems (i.e., two tanks each for the bioaugmented and non-bioaugmented systems). Each tank had a working volume of 10 L. The HRT of both systems was 24 h (Boonnorat et al., 2014b). The sludge in the sedimentation tank was recirculated to the anoxic tank on a daily basis (50% of influent flow) to maintain the anoxic condition and the mixed liquor suspended solids (MLSS) (10 mg/L). The solid retention time (SRT) of the seed sludge of the bioaugmented and non-bioaugmented systems was 30 d, which is a normal length of time adopted by local AS treatment plants.

The seed sludge was from a local AS wastewater treatment plant in Thailand's capital Bangkok (10 g/L MLSS). The acclimatized sludge