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Simultaneous enhancement of nonylphenol biodegradation and short-chain fatty acids production in waste activated sludge under acidogenic conditions

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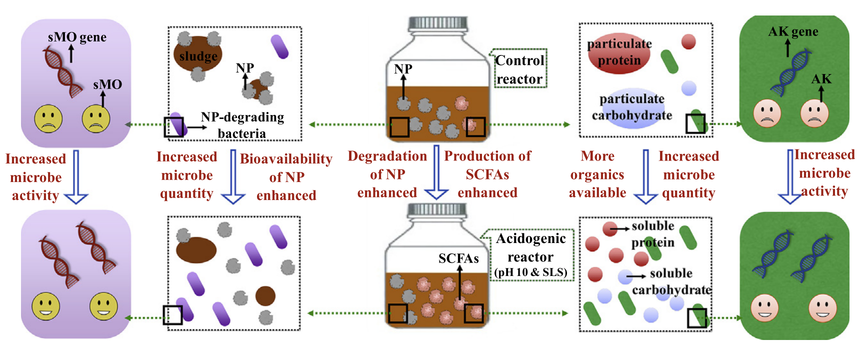
HIGHLIGHTS

- NP degradation and SCFAs yield in WAS were enhanced under acidogenic conditions.
- NP degraders and acidogenic bacteria benefited NP degradation and SCFAs production.
- Enzymatic activity and the quantity of functional genes were promoted.
- Bioavailability of NP and organic substrates used to produce SCFAs was improved.
- NP biodegradation was strengthened by organic substrates involved in acidogenesis.

GRAPHICAL ABSTRACT



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ABSTRACT

Nonylphenol (NP) biodegradation in waste activated sludge (WAS) under anaerobic conditions is usually slow, and no information on NP biodegradation under acidogenic conditions is currently available. In this study, the simultaneous enhancement of NP biodegradation and short-chain fatty acids (SCFAs) accumulation in a WAS fermentation system under acidogenic conditions was accomplished by controlling pH 10 and adding sodium lauryl sulfate (SLS). The biodegradation efficiency of NP was found to be 55.5% within 8 d under acidogenic conditions, much higher than that in the control (24.6%). Meanwhile, the concentration of SCFAs under the same conditions for NP biodegradation was increased from 2234 mg COD/L (control) to 4691 mg COD/L (at pH 10 with SLS). Mechanism study revealed that the abundances of both NP-degrading microorganisms and acidogenic bacteria increased under acidogenic conditions. Altering the enzymatic activity and the quantity of functional genes in the acidogenic fermentation system were beneficial to NP biodegradation and SCFAs accumulation. Furthermore, organic substrates available for uptake by NP-degrading and acidogenic bacteria, *i.e.* NP, protein and carbohydrate, were released from WAS under acidogenic conditions. More importantly, intermediate substrates involved in acidogenic fermentation were advantageous to the cometabolic biodegradation of NP.

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1. Introduction

Nonylphenol (NP) is one of the widely spread xenobiotic compounds used as industrial antioxidants. It is an additive in lubricating oils and a raw material in the production of nonylphenol ethoxylates (NPEOs) surfactants. Since its first synthesis in 1940, the usage of NP has been increasing rapidly (Fan et al., 2018). For example, the annual production of NP in 2015 has reached 48,000 tons in China (Yao et al., 2017). Usually, NP products enter wastewater treatment plants (WWTPs) in substantial amounts and decompose to NP again during wastewater biological treatment. Due to low solubility and high hydrophobicity of NP, it eventually accumulates in waste activated sludge (WAS) in WWTPs. The NP content of WWTPs vary regionally, depending on the use of NPEOs. The NP concentrations range from 64.1 to 200.0 mg/kg dry sludge in China (Hao et al., 2007; Ma et al., 2002; Qiao et al., 2007), 0.5 to 2.2 mg/kg dry sluge in the UK (Paterakis et al., 2012) and 0.3 to 50 mg/kg dry sludge in Germany (Bolz et al., 2001; Verlicchi and Zambello, 2015). Moreover, the concentration of NP could achieve 500 mg/kg dry sludge in some WWTPs (Venkatesan and Halden, 2013). As a typical endocrine disruptor chemicals, NP in WAS may pose a threat to sludge treatment and disposal. Therefore, interest in the removal of NP from sludge has gradually increased.

In China, anaerobic fermentation of sludge has been widely recommended as a preferred method for WAS treatment because of its cost-effectiveness and energy recovery (Chen et al., 2017). Therefore, the effective biodegradation of NP in WAS under anaerobic conditions, especially during anaerobic fermentation is of great significance prior to land application of fermented sludge. NP biodegradation during anaerobic fermentation of sludge has been demonstrated to vary; in the earliest studies no NP removal appeared to occur (RazoFlores et al., 1996), and then anaerobic degradation of NP has been observed and its half-life time was 25 d (Chang et al., 2005). NP degradation during anaerobic fermentation was significantly affected by the anaerobic fermentation system. For example, higher removal of NP was noticed in sludge fermentation reactors under anaerobic thermophilic conditions (Paterakis et al., 2012; Samaras et al., 2014). More recently, efficient anaerobic biodegradation of NP (53%) has been reported in a methanogenic reactor inoculated with sludge from a WWTP (Gonzalez-Gil et al., 2018), which indicated that anaerobic fermentation has been becoming a promising way to eliminate micropollutants with the improvement of fermentation system.

It is well known that methanogenic and acidogenic fermentation are two common methods of sludge anaerobic treatment (Perimenis et al., 2018; Zhang et al., 2018), which can be influenced by operational parameters such as pH and SRT, as well as organic substances present in the fermentation systems, such as surfactant, anti-inflammatory analgesic and cationic polyacrylamide (Hu et al., 2018; Hussain et al., 2017; Leano and Babel, 2012; Luo et al., 2013; Wang et al., 2018; Ye et al., 2018). The biodegradation of NP under methanogenic conditions has been documented more extensively (Chang et al., 2005; Gonzalez-Gil et al., 2018; Paterakis et al., 2012; Samaras et al., 2014) since methane is the final product of anaerobic fermentation. Nevertheless, few data concerning the biodegradation of NP during acidogenic fermentation of sludge are available. During acidogenic fermentation in WWTPs, complex organic substances are converted to short-chain fatty acids (SCFAs), which has drawn more attentions because of its potential end-uses, such as the energy and carbon source for microorganisms carrying out other biological processes (Wang et al., 2017), and the monomers for polyhydroxyalkanoates (PHA) production (Zhang et al., 2018). In addtion, acidogenic fermentation system enriched with acidogenic bacteria is highly suitable for organic pollutants biodegradation (Lin et al., 2018; Perimenis et al., 2018; Zhang et al., 2018). Large-molecular-weight refractory organic pollutants are biodegraded to low-molecular-weight soluble compounds during acidogenesis (Elefsiniotis and Oldham, 1994; Lin et al., 2018). Thus, acidogenic fermentation is a potential way of producing valuable bioproducts and degrading organic pollutants simultaneously.

The main objective of this study was therefore to investigate anaerobic biodegradation of NP during acidogenic fermentation of sludge, which has been considered as a sustainable and practical solution for sludge treatment. The performance of NP biodegradation and SCFAs production under acidogenic conditions at pH 10 in the presence of sodium lauryl sulfate (SLS) was investigated. The relative abundance of NP-degrading microorganisms and acidogenic bacteria under acidogenic conditions was first analyzed. The activity of key enzymes and the quantity of functional genes related with NP biodegradation and SCFAs production under acidogenic conditions were then examined. Finally, the bioavailable organic substrates released from WAS and the cometabolic biodegradation of NP under acidogenic conditions were explored to determine the mechanisms of enhancement of pollutants biodegradation and SCFAs production.

2. Materials and methods

2.1. NP-containing WAS, NP and SLS

WAS without NP used in this study was collected from the secondary sedimentation tank of a WWTP in Shanghai, China. The WAS was first filtered with 1 mm × 1 mm screen and then concentrated by settling at 4 °C for 1 d. Table 1 shows the the main characteristics of the sludge.

4-*n*-NP with a purity >98% was purchased from Alfa Aesar (Shanghai, China). NP was dissolved in methanol (high-performance liquid chromatography (HPLC) grade, Sigma-Aldrich) to a concentration of 1000 mg/L to form the stock solution. NP content in WAS has been reported to be about 200 mg/kg dry sludge in China (Hao et al., 2007; Ma et al., 2002; Qiao et al., 2007). Thus, during investigation of NP degradation in sludge, NP stock solution was added into the anaerobic fermentation reactor and the final concentration of NP was 200 mg/kg dry sludge. SLS with a purity ≥99% was purchased from Sigma-Aldrich.

2.2. Anaerobic degradation of NP and production of SCFAs under acidogenic conditions

Batch anaerobic experiments on NP degradation and SCFAs production in the WAS fermentation systems were conducted in a series of batch reactors containing 240 mL NP-existing sludge. Anaerobic fermentation of WAS is significantly affected by operating conditions and the characteristics of WAS, such as pH, SRT and surfactants present in sludge (Hu et al., 2018; Hussain et al., 2017; Leano and Babel, 2012; Luo et al., 2013; Wang et al., 2018; Ye et al., 2018). In this study, alkaline pH and surfactant addition were combined to create acidogenic conditions. The optimal acidogenic condtions was investigated by adjusting the fermentation pHs within the range of 5 to 11 using 4 M sodium hydroxide (NaOH) or hydrochloric acid (HCl) and adding surfactants (SLS, Octaethylene glycol monododecyl ether (C12E8) and rhamnolipid (RL)). In addition, a reactor without pH adjustment and SLS addition was used as the control. After the reactors were flushed with nitrogen to eliminate oxygen, they were sealed with rubber stoppers and placed

Table 1
Characteristics of the sludge used in the study.

Parameters	Mean value[a]
pH	6.9 ± 0.1
TSS (Total suspended solid, mg/L)	15,723 ± 460
VSS (Volatile suspended solids, mg/L)	10,680 ± 315
TCOD (Total chemical oxygen demand, mg/L)	13,710 ± 185
SCOD (soluble chemical oxygen demand, mg/L)	140 ± 10
Total protein (mg COD/L)	8532 ± 445
Total carbohydrate (mg COD/L)	1240 ± 56
Lipid and oil (mg COD/L)	165 ± 15

[a] The data are the averages and their standard deviations in triplicate tests.