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Enhanced bioremediation of 4-nonylphenol and cadmium co-contaminated sediment by composting with *Phanerochaete chrysosporium* inocula

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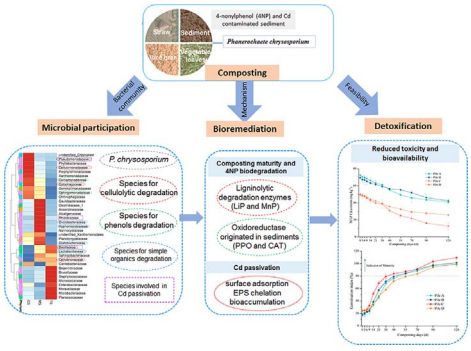
GRAPHICAL ABSTRACT



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ABSTRACT

Composting is identified as an effective approach for solid waste disposal. The bioremediation of 4-nonylphenol (4NP) and cadmium (Cd) co-contaminated sediment was investigated by composting with *Phanerochaete chrysosporium* (*P. chrysosporium*) inocula. *P. chrysosporium* inocula and proper C/N ratios (25.51) accelerated the composting process accompanied with faster total organic carbon loss, 4NP degradation and Cd passivation. Microbiological analysis demonstrated that elevated activities of lignocellulolytic enzymes and sediment enzymes was conducive to organic chemical transformation. Bacterial community diversity results illustrated that *Firmicutes* and *Proteobacteria* were predominant species during the whole composting process. Aerobic cellulolytic bacteria and organic degrading species played significant roles. Toxicity characteristic leaching procedure (TCLP) extraction and germination indices results indicated the efficient detoxification of 4NP and Cd co-contaminated sediment after 120 days of composting. Overall, results demonstrated that *P. chrysosporium* enhanced composting was available for the bioremediation of 4NP and Cd co-contaminated sediment.

1. Introduction

4-Nonylphenol (4NP), a central degradation product and either a raw material of nonylphenol ethoxylates (NPEs) surfactants, exists widely in most environmental samples, including water, soils and sediments (Karahan et al., 2010; Zeng et al., 2013). 4NP is a representative environmental endocrine disruptor (EED) with estrogenic responses, carcinogenicity, teratogenesis and mutagenicity (Gerent and

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Spinelli, 2016; Gong et al., 2009). 4NP is easily adsorbed on particulate matter due to its low solubility in aqueous solution, and thus soils and sediments are crucial destination. Field surveys revealed that 4NP existed at a level of ng L^{-1} to μg L^{-1} in water and mg kg^{-1} concentrations in sediments (Gong et al., 2011). Accordingly, wide range of NP has been detected in surface sediment, a mean value of 73.42 and 115.90 ng g^{-1} dw was reported in East Dongting Lake and Honghu Lake, respectively (Yang et al., 2015). Luo et al. (2017) further reported a maximal bioaccumulation of 3.27 ng g^{-1} in fish samples from lake and rivers within Hunan Province, China. Additionally, significant amounts of heavy metals released to aquatic and terrestrial environments, commonly resulting in the co-contamination of heavy metals and organic pollutants (Xu et al., 2012a; Xu et al., 2017). Numerous studies demonstrated the wide occurrence and great threat of heavy metals in sediment. Recent study reported a range of 2.95–29.15 mg kg^{-1} Cd, especially accompanied with a relative high bioavailable fraction (66.93%), in sediment from Xiangjiang river, Hunan province (Liu et al., 2017a). As such, in consideration of the potential toxicity of 4NP and heavy metals, complete remediation is crucial to the environment.

Microbial biodegradation is a principle mechanism for organic carbon transformation and degradation. Biodegradation, through various biological process, might modify organic molecules or result in microbial destruction of organic pollutants (Alexander, 1999; Yang et al., 2010). Microorganism with available catabolic potential to accessible organic chemicals is essential to the biodegradation. Microorganism is the significant component in sediments, which tends to be major or occasionally sole means in biodegradation in the case of sediment self-purification. However, biodegradation ability of indigenous microorganisms in sediment is limited. Furthermore, as structural and natural diversity, specific degradation is necessary due to the degradative requirement of microbial accessibility to chemicals (Alexander, 1999; Carboneras et al., 2017). Enhanced biodegradation is imperative to complete destruction of organic chemicals.

Composting is an appropriate enhanced biodegradation approach for organic contaminant removal and heavy metal passivation (Jonkers et al., 2001; Rawoteea et al., 2017). How to reduce active metals and organic contaminants to alleviate the toxicity of the composting products needs to broaden concerns. Our previous studies suggested that composting with white rot fungi inocula is one of the most prospective approaches for soil and solid waste treatment (Huang et al., 2008). *Phanerochaete chrysosporium* (*P. chrysosporium*) are extensively studied in environmental remediation (Xu et al., 2012b). Degradation of NPs or 4NP by *P. chrysosporium* has been reported previously (Cajthaml et al., 2009; Subramanian and Yadav, 2009). Cajthaml et al. (2009) reported that *P. chrysosporium* was efficient to degrade 4NP in aqueous solution within 14 d with the acceptable activity of lignin peroxidase (LiP) and manganese peroxidase (MnP). However, scarce study focused on the *P. chrysosporium* enhanced bioremediation of 4NP in sediment, especially in the metal co-contaminated sediment. Hence, the aim of this study was to investigate the bioremediation of 4NP and Cd co-contaminated sediment via composting with *P. chrysosporium* inocula. An attempt was made to determine the potential roles of oxidoreductase in organic chemical biodegradation and the possibility in composting maturity assessment accompanied with physico-chemical parameters. Importantly, bacterial community analysis was conducted to investigate the bacterial species participated in composting and bioremediation.

2. Materials and methods

2.1. Materials preparation and fungal strain

Sediment samples were collected from the 5–15 cm layer from Xiangjiang River in Changsha, China. After air dried, sediment samples were grinded and sieved to 2 mm prior to the experiments. The sediment had a neutral pH (6.76) and an organic carbon content of 24.9 g kg^{-1}. The 4NP concentration was detected at 31.5 μg kg^{-1} in the prepared sediments. 500 mL of 4NP methanol solution (200 mg L^{-1}) was added to 1 kg of dry sediment to prepare the 4NP contaminated sediment. The slurry was then stirred for 12 h in dark and left to stewing in fume cupboard until dried. Thereafter, the contaminated sediments were grinded and sieved to 2 mm for further use. The straw was air-dried and cut into about 20 mm length. Rice bran and vegetable leaves were also air dried as composting substrate.

The *P. chrysosporium* (BKMF-1767) obtained from China Center for type Culture Collection (Wuhan, China). Spore suspensions were prepared by scraping and blending in the sterile distilled water. All reagents were analytical grade and used without further purification. Distilled water was used for the preparation of all the solutions throughout this study.

2.2. Composting setup

Four piles with each about 3 kg of composting materials (dry weight) were set up indoors in 70 L polystyrene boxes with the internal dimensions of 0.58 × 0.42 × 0.38 m (length × width × height). In detail, sediment, straw, vegetable leaves and rice bran were mixed in the designed ratios to obtained four piles with C/N ratios at the value of 9.53 (Pile A), 18.77 (Pile B), 25.51 (Pile C) and 25.46 (Pile D), respectively. Initial 4NP concentration was 45.86 mg kg^{-1} (Pile A), 37.01 mg kg^{-1} (Pile B), 30.59 mg kg^{-1} (Pile C) and 29.87 mg kg^{-1} (Pile D). Initial total Cd concentration was 79.28 mg kg^{-1} (Pile A), 73.51 mg kg^{-1} (Pile B), 52.34 mg kg^{-1} (Pile C) and 51.91 mg kg^{-1} (Pile D). The moisture content was adjusted to about 70% with the distilled water. Pile A, B and C was inoculated with 1% of *P. chrysosporium* spore suspensions, pile D was used as the control without the inoculants. The moisture was controlled by addition of distilled ultrapure water, and the piles were turned over twice a week for eventual oxygen supply and temperature distribution.

2.3. 4NP analysis and Cd sequential extraction

For chemical and microbiological analysis, triplicate samples were collected and mixed from four symmetrical locations in each pile at designated time intervals. 4NP was extracted by ultrasonic extraction method modified from Yang et al. (2014). 2.0 g composting samples were dried via vacuum freeze-drying equipment for 24 h, and then 15 mL of acetone and n-hexane (1:1, v/v) extracting solution and ultrasonic extracted for 30 min. The supernatant was collected by centrifugation at 4000 rpm for 20 min at 4 °C. The ultrasonic extraction process was repeated for three times, and the supernatant was mixed. The supernatant was removed by rotary evaporation and then 1 mL of methyl alcohol was added to dilute the extracted 4NP. All the extracts were filtrated through 0.45 μm membrane. 4NP concentration was detected by HPLC as described by Gabriel et al. (2005). Detection was carried out at 277 nm, and acetonitrile/water (85:15, v/v) was used as the mobile phase at a flow rate of 1.0 mL min^{-1}. The limit of detection (LOD) for NP was 0.01 mg kg^{-1} sediment in this study.

Metal speciation was investigated by improved BCR procedure according to the previous study (Rauret et al., 1999). Metal fraction was classified as exchangeable fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). The TCLP tests were conducted with the TCLP leachant (0.1 M glacial acetic acid solution, pH 2.88 ± 0.05). The TCLP leachant was mixed with the samples with a leachant-to-sample ratio of 20:1, vibrated at 30 rpm for 16 h. The supernatant was centrifuged and filtrated by 0.45 μm membrane for the Cd testing. TCLP levels were normalized to mg kg^{-1} by multiplying the TCLP values by the mass/volume ratios.

2.4. Chemical and microbiological analysis

The temperature was recorded at a depth of 20 cm in each pile. For