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Effect of alkyl polyglucoside and nitrilotriacetic acid combined application on lead/pyrene bioavailability and dehydrogenase activity in co-contaminated soils

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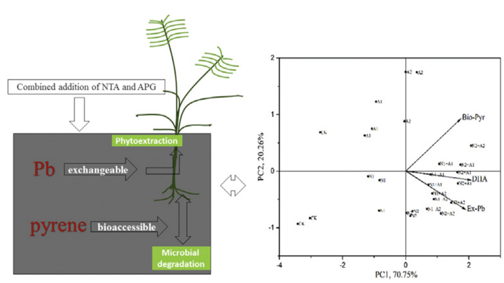
HIGHLIGHTS

- APG played dominant role on increasing bioaccessible pyrene.
- NTA played dominant role on increasing exchangeable Pb.
- NTA and APG can stimulate the bioaccessiblity of pyrene and Pb.
- Combination of NTA and APG significantly enhanced the dehydrogenase activity.
- Variation of bioaccessiblity of pyrene and Pb affected the dehydrogenase activity.

GRAPHICAL ABSTRACT



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ABSTRACT

At present, few research focus on the phytoremediation for organic pollutants and heavy metals enhanced by surfactants and chelate agents in the combined contaminated soils or sediments. In this study, the effect of a novel combined addition of alkyl polyglucoside (APG) and nitrilotriacetic acid (NTA) into pyrene and lead (Pb) co-contaminated soils on bioaccessiblity of pyrene/Pb and dehydrogenase activities (DHA) was studied. Through the comparison of the results with the alone and combined application, synergistic effect on bioaccessiblity of pyrene and Pb was found while APG and NTA was applied together. Results also indicated a significant promotion on the DHA in mixed addition of APG and NTA. In addition, correlation and principal component analysis were performed to better understand the relationship among APG/NTA, bioaccessiblity of pyrene/Pb and the DHA. Results showed that APG and NTA can affect DHA directly by themselves but also can affect DHA indirectly by changing bioaccessible pyrene and exchangeable Pb.

1. Introduction

Co-contaminations of organic and inorganic pollutants are frequently found in the environment (Nadal et al., 2011). As a group of priority control pollutant, polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in the environment (Liao et al., 2015). Some studies have demonstrated that PAHs and heavy metals (HMs) are frequently found together as contaminants in soil. Among heavy metals, lead (Pb) is a poison element, which is known to be a persistent environmental problem and frequently coexist with PAHs (Cachada et al., 2012). Phytoremediation, due to its

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environmentally sound and lower cost, are widely studied for remove PAHs or HMs from soil (Sun et al., 2013; Ehsan et al., 2014). However, PAHs and HMs in soil usually exhibit poor availability, which limits the application of phytoremediation (An et al., 2011; Cheng et al., 2008). The enhanced phytoremediation by chemical method is a promising technology for the removal of PAHs and HMs. Surfactants which are a group of chemicals can assist in increasing the water solubility of organic pollutants. They have been proven to be very effective in promoting phytoremediation for PAHs. For example, studies indicated that the presence of some nonionic surfactants (polyoxyethylene sorbitan monooleate; polyoxyethylene and dodecanol) at relatively low concentrations resulted in significant positive effects on phytoremediation for pyrene-contaminated soil (Gao et al., 2007). Liao et al. (2015) also found that surfactants (Triton X-100, rhamnolipid and saponin) could enhance the removal of pollutants from contaminated soil during phytoremediation. Chelating agents are compounds that can make heavy metals desorbed from soil. Their characteristic of increasing the plant-available fraction and uptake amounts, and transport metals to aboveground parts made them have been widely used in phytoremediation to enhance remediation efficiency (Lee and Sung, 2014). For example, Kanwal et al. (2014) investigated that ethylenediaminetetraacetic acid (EDTA) improves the capability of plants to uptake heavy metals (Pb) from polluted soil.

Alkyl polyglucoside (APG), a nonionic surfactant produced from renewable resources such as fatty alcohols and glucose, was studied to remove PAHs from soil (Liu et al., 2013). As a biodegradable chelating agent, nitrilotriacetic acid (NTA) was found that it can desorb HMs (such as Pb) from soil with no environmental effects (Freitas and Nascimento, 2009). As a consequence, using APG and NTA together may be a promising way to enhance phytoremediation for PAHs and HMs co-contaminated soil without secondary pollution. Besides, the combination of APG and NTA may have synergistic effect on phytoremediation for PAHs or HMs. However, few researches have been concentrated on this combined application.

Bioavailability of PAHs and HMs in soil is the key factor in determining efficiency of phytoremediation (Megharaj et al., 2011; Ayanka et al., 2015). Chelating agents can increase the plant-available fraction and uptake amounts, and transport HMs by desorbing HMs from soil (Shen et al., 2002; Lee and Sung, 2014). Surfactants have been proven to be very effective in promoting the mobilization of organic compounds of relatively low water solubility (Sun et al., 2013). So, to measure the possibility of this approach for enhancing phytoremediation, it is of great importance to evaluate bioaccessiblity of PAHs and HMs in soil by application of APG and NTA. In addition, when the mixed compounds of APG and NTA are applied their respective effect on bioavailability of PAHs or HMs will be influenced each other. Exchangeable fraction of metal extracted by $MgCl_2$ and bioaccessible PAH extracted by butanol, the most easily available for plant uptake, was used to discuss the bioavailability of metal and PAH in soil (Kim et al., 2010; Wei et al., 2014). However, there is no report showed that whether and how APG and NTA affect each other on the effects of increasing bioavailability of PAHs and Pb.

Soil enzymes, derived primarily from soil microorganisms, plant roots, plant and animal residues, can be the indicator to measure the effectiveness of soil to support biochemical process involving the decomposition of PAHs (Zhou et al., 2011). Enzymes involved in the degradation of PAHs are oxygenase, dehydrogenase and lignolytic enzymes (Haritash and Kaushik, 2009). Dehydrogenase, an intracellular enzyme, is the catalyst for important metabolic process which includes the decomposition for organic inputs and detoxification of xenobiotics (Zhang et al., 2014). So, this study uses the dehydrogenase activity (DHA) as a model to reveal the effects of application of APG and NTA on soil enzymes.

In a short, the aim of this study was to discuss the possibility of the combined application of NTA and APG by investigating the effects of alone application of APG or NTA and combined application of APG and NTA on pyrene and Pb bioaccessiblity, as well as on DHA.

2. Materials and methods

2.1. Chemicals

Pyrene (purity: 98%) was purchased from Aladdin Reagent. APG used in the test was C12/14-APG (APG1214) obtained from the China Research Institute of Daily Chemical Industry (Shanxi, China). The other chemicals, analytical grade or better, were bought from Sinopharm.

2.2. Soil preparation

The soil (air-dried, 2 mm sieved) in this study was collected from the top of the soil profile of a field without previous exposure to pyrene and Pb contamination in Shanghai University, China. Its properties are as follows: pH 8.3; organic matter 19.6 g kg^{-1}; total nitrogen 0.52 g kg^{-1}; clay 7.4%; silk 60.4% and sand 32.2%. Co-contaminated soil with pyrene and Pb were prepared by a step-by-step process: prepared soils were firstly mixed with Pb [$(CH_3COO)_2Pb$] solution. After adding for few days later, pyrene dissolved in acetone was added into the Pb contaminated soil. After the solvent evaporated, the mixed contaminated soil was transferred to a box and aged for about two months before the experiments. The final concentrations of pyrene and Pb in the soil were measured as 184.5 and 454.3 mg kg^{-1}, respectively.

2.3. Experimental design

2.3.1. Bioavailability of pyrene and Pb by application NTA and APG

The co-contaminated soil with pyrene and Pb (1.00 g) was placed in centrifuge tubes followed by pre-incubating 7d. Then, APG and NTA with different dosage with the form of solution was added into soil. The final concentrations of NTA and APG in soil were as follows: CK (none of NTA and APG); A1 (1 g APG kg^{-1} soil); A2 (2 g APG kg^{-1} soil); N1 (1 g NTA kg^{-1} soil); N2 (2 g NTA kg^{-1} soil); N1+A1 (1 g NTA+1 g APG kg^{-1} soil); N1+A2 (1 g NTA+2 g APG kg^{-1} soil); N2+A1 (2 g NTA+1 g APG kg^{-1} soil); N2+A2 (2 g NTA+2 g APG kg^{-1} soil). The solution of NTA and APG were added NaOH to adjust the pH to 7.3 in order to limit soil property modification and 0.01% w/w NaN_3 to inhibit microbial activity. Soil water content of each tube was controlled at 60% of WHC. These samples were then replaced to incubator for 48 h at 25 ± 1 °C in dark environment for analysis of bioaccessible pyrene and Pb.

2.3.2. Variations of the DHA with NTA and APG

Co-contaminated soil with pyrene and Pb (4.00 g) was placed in conical flask. After 7 d' pre-incubation, APG and NTA with different dosage in the form of solution were added by following the same steps mentioned in 2.3.1 without adding NaN_3.

2.3.3. Influence of NTA on the effect of APG on the solubility of pyrene

Sufficient pyrene (0.100 g) was respectively added into glass centrifuge tubes. Then, solution of APG and NTA with different concentration was added into the tubes. The concentrations of NTA and APG in each tube were shown in Table 1. Solutions without pyrene were the black control. All tubes were replaced into