



# Remediation of phthalates in river sediment by integrated enhanced bioremediation and electrokinetic process



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## HIGHLIGHTS

- ORC injection was found to improve the EK-assisted biodegradation of PAEs in river sediment.
- Applying polarity reversal and extending reaction time were beneficial to biodegradation of PAEs.
- In this work four strains were found to be responsible for degradation of PAEs in river sediment.

## ARTICLE INFO

### Article history:

Received 19 October 2015

Received in revised form

11 December 2015

Accepted 13 December 2015

Available online 28 December 2015

Handling Editor: Shane Snyder

### Keywords:

Sediment

Phthalate esters

Enhanced bioremediation

Electrokinetic process

Oxygen releasing compound

Molecular biotechnology

## ABSTRACT

The objective of this study was to evaluate the feasibility of enhanced bioremediation coupling with electrokinetic process for promoting the growth of intrinsic microorganisms and removing phthalate esters (PAEs) from river sediment by adding an oxygen releasing compound (ORC). Test results are given as follows: Enhanced removal of PAEs was obtained by electrokinetics, through which the electroosmotic flow would render desorption of organic pollutants from sediment particles yielding an increased bioavailability. It was also found that the ORC injected into the sediment compartment not only would alleviate the pH value variation due to acid front and base front, but would be directly utilized as the carbon source and oxygen source for microbial growth resulting in an enhanced degradation of organic pollutants. However, injection of the ORC into the anode compartment could yield a lower degree of microbial growth due to the loss of ORC during the transport by EK. Through the analysis of molecular biotechnology it was found that both addition of an ORC and application of an external electric field can be beneficial to the growth of intrinsic microbial and abundance of microflora. In addition, the sequencing result showed that PAEs could be degraded by the following four strains: *Flavobacterium* sp., *Bacillus* sp., *Pseudomonas* sp., and *Rhodococcus* sp. The above findings confirm that coupling of enhanced bioremediation and electrokinetic process could be a viable remediation technology to treat PAEs-contaminated river sediment.

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## 1. Introduction

PAEs are widely used in many industrial and household applications such as adhesives, paints, inks, rubbers, and surface treatment. In addition, they are massively used in the composition of polyvinyl chloride (PVC) and other resins for polymers because of the stability, fluidity and low volatility of heavier congeners (Fountoulakis et al., 2006). It is known that PAEs are only physically bound to the polymer chains. Because of the widespread usage of

phthalate-containing products, the residues of PAEs have been routinely detected in various matrices such as soil (Xia et al., 2011), water (Sha et al., 2007; Liu et al., 2014), air (Wang et al., 2008, 2012), and sediments (Yuan et al., 2002; Sha et al., 2007; Sun et al., 2013; Yang et al., 2015), and even drinking water (Liou et al., 2014; Yang et al., 2014a,b).

Recently, more and more concerns have been raised on harmful effects of PAEs on human health, even in ultra trace to trace levels. Humans get exposed to phthalates via multiple exposure pathways including inhalation, dietary intake and dermal absorption (Clark et al., 2011). After PAEs reach natural environment, these compounds may have health risks to human due to their xenoestrogenic effects (Nielson and Larsen, 1996), and carcinogenic

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and mutagenic effects (Nielsen and Larsen, 1996). Several PAEs are considered as hazardous pollutants due to their suspected mutagenicity and carcinogenicity. For instance, di (2-ethylhexyl) phthalate (DEHP) has been classified by the International Agency for Research on Cancer (IARC) as Group 2B carcinogen that is possibly carcinogenic to humans. Some PAEs have been found to bioaccumulate in fats (Keresztes et al., 2013) and shown their carcinogenicity and estrogenicity (Wu et al., 2013). Thus, the potential risk to human and animals' health from PAEs has drawn much attention all over the world. Several PAEs have been listed as priority pollutants by many national and international regulatory organizations (Ventrice et al., 2013; Yang et al., 2013).

PAEs have been detected in sediments around the world. Table SM-1 shows that the concentrations of PAEs are in the range of ND–881.18 mg/kg dw for sediments over the world.

Bioremediation is defined as the process whereby organic wastes are biologically degraded under controlled conditions to an innocuous state, or to levels below concentration limits established by regulatory authorities (Mueller et al., 1996). Most bioremediation systems are run under aerobic conditions. Generally, the oxygen supply includes injection of air (or oxygen), injection of hydrogen peroxide, and injection of oxygen releasing compounds (ORC) (Anid et al., 1993). However, hydrogen peroxide can be toxic to some subsurface microorganisms at concentrations as low as 0.003% (Pardieck et al., 1992). Thus, it is necessary to control the concentration of hydrogen peroxide when it is used.

Biodegradation has been proposed as an effective way to reduce PAEs in the environment (Navacharoen and Vangnai, 2011; de Moura Carrara et al., 2011; Fang et al., 2014; Pranaw et al., 2014; Sompornpailin et al., 2014; Chi and Gao, 2015). However, it has some restrictions in application including: (1) mass transfer between electron acceptor and specific nutrients (Simoni et al., 2001); (2) bioavailability of contaminants (Lohner and Tiehm., 2009); and (3) acclimatization and biodegradability of aboriginal microorganisms to contaminants (Mrozik and Piotrowska-Seget, 2010).

To overcome the above limitation of bioremediation, the electrokinetic (EK) process has been used to enhance the efficiency of biodegradation of organic contaminants. In so doing, an improved transport of nutrients and terminal electron acceptors and increased bioavailability of contaminants were found (Wick, 2009). It has also been reported that EK can aid the release of organic contaminants bound to clay particles and organic matter in soils and sediments yielding an improved treatment performance (Maini et al., 2000; Luo et al., 2005a). Positive effects associated with EK also include the generation of oxidizing and reducing zones favorable for biodegradation of contaminants close to the electrodes (Kim et al., 2010; Lohner et al., 2011). Shi et al. (2008b) reported that at the single cell level, low intensity electric field can stimulate adenosine triphosphate (ATP) production and have minimal effect on the cell surface charge. However, at a high intensity electric field (e.g., >40 mA) the cell surface becomes more hydrophobic and can cause the cell to change shape (Luo et al., 2005b).

This study aimed to evaluate the feasibility of coupling enhanced bioremediation and electrokinetic process for remediation of PAEs in river sediment. To meet this end, batch experiments were first conducted to evaluate the influence of H<sub>2</sub>O<sub>2</sub> and nutrients concentration to aboriginal microorganisms in degradation of PAEs in river sediment and further influence integrated by the oxygen releasing compound (ORC) in EK performance. Additionally, bacterial counts and molecular biotechnology were used to monitor the change of microflora in this integrated remediation process.

## 2. Materials and methods

### 2.1. Chemicals and standards

HPLC-grade methanol was purchased from Mallinckrodt Baker, USA. American Chemical Society (ACS)-grade formic acid, ammonium formate, di-n-butyl phthalate (DnBP), di-iso-nonyl phthalate (DiNP) and di-(2-ethylhexyl) phthalate (DEHP) were purchased from Sigma–Aldrich, USA. ACS grade di-iso-decyl phthalate (DiDP), di-(2-ethylhexyl) phthalate-d4 (DEHP-d4), di-n-octyl phthalate-d4 (DnOP-d4), and di-n-butyl phthalate-d4 (DnBP-d4) were purchased from AccuStandard, USA. ACS grade butyl benzyl phthalate (BBP) and di-n-octyl phthalate (DnOP) were purchased from Supelco, USA. ACS grade di-iso-decyl phthalate-d4 (DiDP-d4) and di-iso-nonyl phthalate-d4 (DiNP-d4) were purchased from Toronto Research Chemical, Canada. ACS grade butyl benzyl phthalate-d4 (BBP-d4) was purchased from Chem Service, USA. Silica gel (70–230 mesh) was purchased from Macherey–Nagel, Germany. Mixed standard working solutions (10 mg L<sup>-1</sup>) were prepared by the dilution of the standard stock solutions in methanol. Japanese Industrial Standards (JIS)-grade sodium phosphate dibasic 12 hydrate (Na<sub>2</sub>HPO<sub>4</sub> · 12H<sub>2</sub>O; 99.5% in purity), potassium phosphate monobasic (KH<sub>2</sub>PO<sub>4</sub>; 99% in purity), ammonium chloride (NH<sub>4</sub>Cl; 98.5% in purity), magnesium sulphate-7-hydrate (MgSO<sub>4</sub> · 7H<sub>2</sub>O; 99.8% in purity), calcium chloride anhydrous (CaCl<sub>2</sub>; 95% in purity), potassium phosphate dibasic (K<sub>2</sub>HPO<sub>4</sub>; 98% in purity) and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>; 35%) were supplied by Nihon Shiyaku Reagent, Japan.

### 2.2. Sampling of sediment and its characterization

The sampling site for the sediment is located under Hongmaokang-Road Bridge of the Cianjhen River, Kaohsiung, Taiwan (see Fig. SM-1). The top layer of sediment (surface sediment; ca. 0–15 cm) was collected following the standard method NIEA S104.31B (Taiwan EAL, 2012a) using an Ekman dredge. The collected sample was sealed with Teflon-lined caps in 120-mL clear straight sided rounds. Prior to the analysis, the sediment sample was stored at 4 °C. To ensure homogeneity, the collected sediment sample was thoroughly mixed before testing. All characterization methods followed the standard methods: (1) the pH determination by NIEA S410.62C (Taiwan EAL, 2009); (2) the moisture content determination by NIEA S280.62C (Taiwan EAL, 2012b); (3) the specific gravity determination by ASTM D854 (ASTM, 2014); and (4) the heterotrophic plate count determination by NIEA E204.55B (Taiwan EAL, 2013). Table SM-2 shows the characterization results for the original sediment. The concentrations of DnBP, DiDP, and DnOP, however, were all lower than 50 µg kg<sup>-1</sup> in the sediment. To evaluate the performance of the batch degradation tests and coupled remediation technology against various PAEs, spiking of PAEs of concern to the original sediment became necessary in this work. Following the method reported by Minten et al. (2011), the river sediment was spiked with 250 mg L<sup>-1</sup> stock solutions of DnBP, DiDP, and DnOP to yield a respective concentration of about 500 µg kg<sup>-1</sup> for each compound. Then, the spiked sediment sample was air dried at ambient temperature.

### 2.3. Sample extraction and analysis

The spike-and-recovery experiments for PAEs in sediment were performed by spiking 1000 mg kg<sup>-1</sup> of standards in blank samples collected from the Cianjhen River. The recovery yields for all spiked concentrations were 94–99% for PAEs in sediment samples. For sediment samples, the sonication assisted extraction method using ultrasonic probe (Sonic, VCX 750, USA) was employed following

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