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Pilot scale aided-phytoremediation of a co-contaminated soil

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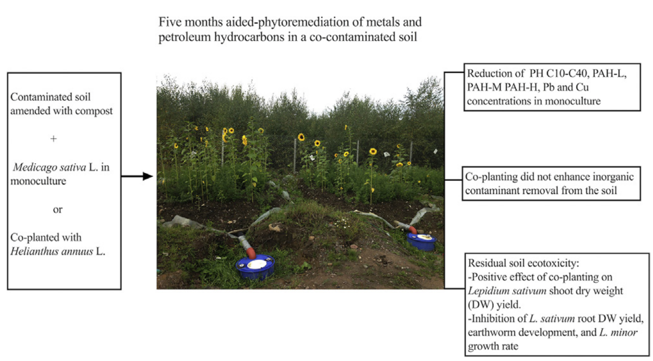
HIGHLIGHTS

- Aided-phytoremediation using alfalfa in monoculture reduced soil PH C10-C40, PAH-L, PAH-M PAH-H, Pb and Cu concentrations.
- Root bioconcentration factors are low but root-to-shoot transfer factor of Mn, Cr, Co and Zn exceeded one.
- Alfalfa co-planting with sunflower did not enhance inorganic contaminant removal from the soil.
- Co-planting was less efficient than alfalfa in monoculture regarding PH C10-C40 and PAH-L degradation.
- After 5 months, co-planting improved the *Lepidium sativum* shoot biomass.

GRAPHICAL ABSTRACT



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ABSTRACT

A pilot scale experiment was conducted to investigate the aided-phytoextraction of metals and the aided-phytodegradation of petroleum hydrocarbons (PHC) in a co-contaminated soil. First, this soil was amended with compost (10% w/w) and assembled into piles (Unp-10%C). Then, a phyto-cap of *Medicago sativa* L. either in monoculture (MS-10%C) or co-cropped with *Helianthus annuus* L. as companion planting (MSHA-10%C) was sown on the topsoil. Physico-chemical parameters and contaminants in the soil and its leachates were measured at the beginning and the end of the first growth season (after five months). In parallel, residual soil ecotoxicity was assessed using the plant species *Lepidium sativum* L. and the earthworm *Eisenia fetida* Savigny, 1826, while the leachate ecotoxicity was assessed using *Lemna minor* L. After 5 months, PH C10-C40, PAH-L, PAH-M PAH-H, Pb and Cu concentrations in the MS-10%C soil were significantly reduced as compared to the Unp-10%C soil. Metal uptake by alfalfa was low but their translocation to shoots was high for Mn, Cr, Co and Zn (transfer factor (TF) > 1), except for Cu and Pb. Alfalfa in monoculture reduced electrical conductivity, total organic C and Cu concentration in the leachate while pH and dissolved oxygen increased. Alfalfa co-planting with sunflower did not affect the extraction of inorganic contaminants from the soil, the PAH (M and H) degradation and was less efficient for PH C10-C40 and PAH-L as compared to alfalfa monoculture. The co-planting reduced shoot and root Pb concentrations. The residual soil ecotoxicity after 5 months showed a positive effect of co-planting on

Abbreviations: BCF, Bioconcentration factor (e.g. root concentration vs. soil concentration); COD, Chemical oxygen demand; CW, Change in weight; DO, Dissolved oxygen; DW, Dry weight; EC, Electrical conductivity; FW, Fresh weight; GRO, Gentle remediation options; ICP-MS, Inductively coupled plasma-mass spectrometry; MS-10%C, Pile planted with alfalfa in monoculture; MSHA-10%C, Pile co-planted with alfalfa and sunflower; MSW, Municipal solid waste; OM, Organic matter; PAHs, Polycyclic aromatic hydrocarbons; PHC, Petroleum hydrocarbons; TF, Translocation factor; TOC, Total organic carbon; Unt, Untreated soil; Unt-10%C, Untreated soil amended with compost at the 10% (*w/w*, air-dried soil) rate; Unp-10%C, Unplanted pile; USEPA, United States Environmental Protection Agency; WC, Water content (%WC); XRF, X-Ray fluorescence.

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L. *sativum* shoot dry weight (DW) yield. However, high contaminant concentrations in soil and leachate still inhibited the L. *sativum* root DW yield, earthworm development, and L. *minor* growth rate.

1. Introduction

Over decades, anthropogenic activities have left about 2.5 million of contaminated sites in European Union (EU) (European Environment Agency, 2014). Most frequent contaminants in soils at these sites are metals (37%), mineral oil (20%) and hydrocarbons (22%) (Evangelou et al., 2012). Metals, e.g. Cu, Co, Pb, and Hg, and petroleum hydrocarbons (PHC), such as the 16 polycyclic aromatic hydrocarbons (PAHs) prioritized by the United States Environmental Protection Agency (USEPA), are of great concern due to their persistence in the environment, and their potentially serious health consequences (Khan et al., 2015; Fu et al., 2012). In Sweden, about 80,000 sites are potentially contaminated, due to more than two hundred years of industrialization, and roughly 60,000 out of these have been risk-assessed (Swedish Environmental Protection Agency, 2016). Open dumpsites and landfills are the most widespread methods for municipal solid waste (MSW) disposal due to relatively low initial investments and operational (Kaczala et al., 2015; Xiaoli et al., 2007). The member countries of the EU have consequently implemented a range of legislation such as the landfill directives (Council Directive 1999/31/EC, 1999) and the waste directives (Directive 2008/98/EC, 2008) to enforce the remediation of contaminated land and to minimize the negative impact on the environment and human health. To provide alternatives to conventional methods of MSW treatment (e.g. disposal to landfill, isolation, soil washing, and pump-and-treat), several methods rely on the use of plants and associated microorganisms and have been alternatively used for remediating polluted soils (Marchand et al., 2016b; Nagendran et al., 2006). Gentle soil remediation options (GRO), including in situ contaminant stabilization ("inactivation" using biological or chemical processes) and plant-based options (i.e. phytoremediation) are gaining social acceptance and have the advantages of being non-destructive, less disruptive to the soil and low-cost (Marchand et al., 2016a; Cundy et al., 2013; Kumpiene et al., 2014; Cundy et al., 2016).

A major challenge of GRO for co-contaminated soils is the simultaneous removal or/and control of multiple contaminants. Therefore, a combination of different set of technologies is often required to achieve effective performance on soil remediation. Among the five main subsets of phytoremediation, phytoextraction, i.e. the use of plants to remove inorganic or organic compounds from soil by accumulating them in the biomass of plants and phytodegradation, i.e. the use of plants to uptake, store and degrade organic compounds have received great attention. The selection of appropriate plant species is critical to optimize the phytoremediation and co-planting are often used for a simultaneous removal of multiple contaminants (Wang et al., 2013). *Medicago sativa* (alfalfa) is widely used for phytoremediation of organic (Wei and Pan, 2010; Hechmi et al., 2014) and inorganic pollutants (Zaefarian et al., 2013; Vamerali et al., 2011; Bonfranceschi et al., 2009). This plant species has a fibrous root system suitable for the PHC rhizodegradation (Wang et al., 2012) and can contribute to TE phytostabilisation (Zribi et al., 2015). Phytoremediation can be applied on contaminated soil in combination with soil conditioners (so-called aided-phytoremediation) to promote the biomass production (Hattab-Hambli et al., 2016). Alfalfa in combination with compost was selected on the basis of previous results at mesoscale level (Marchand et al., 2016a). Compost originated from the organic fraction of MSW is increasingly used as soil conditioner as well as a fertilizer for meeting both nitrogen and organic matter addition (Cesaro et al., 2015). Compost amendment has been also effectively used for the phytostabilisation of metal-contaminated soil (Ruttens et al., 2006; Park et al., 2011; Ogundiran et al., 2015), the rhizodegradation of PHC in contaminated soils (Zhang et al., 2012; Ghanem et al., 2013; Wang et al., 2012) and remediation of metal and PHC co-contaminated soil (Marchand et al., 2016a; Chirakkara et al., 2016). *Helianthus annuus* (sunflower) is also used to remediate metal(loid)-contaminated soils (Kolbas et al., 2011; Kidd et al., 2015a) and facilitation/intercropping is claimed to promote the efficiency of several phytotechnologies for remediating contaminated soils (Brooker et al., 2008; Wang et al., 2014; Kidd et al., 2015b). Co-planting of alfalfa with sunflower may be a promising option to optimize the remediation of metal and PHC co-contaminated soils.

This study aimed at investigating: (1) the efficiency of alfalfa either in monoculture or co-planting with sunflower to remediate a PHC and metal co-contaminated soil in a pilot scale plant, (2) the residual soil ecotoxicity using *Lepidium sativum* L. and earthworm (*Eisenia fetida* Savigny, 1826) and (3) the residual soil leachate ecotoxicity using *Lemna minor* L. The study also aimed to verify the hypothesis that alfalfa co-planting with sunflower would increase PHC degradation and metal removal as compared to alfalfa monoculture, thus decreasing soil and soil leachate ecotoxicity.

2. Material and methods

2.1. Site and pile construction

The studied area is a 40-year old MSW landfill at Moskogen, in southern Sweden (56°41′26″ N; 16°10′49″ E). This landfill receives approximately 65,000 tons of wastes per year from three communities with a total population of 90,000 inhabitants. Among these wastes, 3.5% are hazardous wastes including the oil-contaminated soil used in this study. The climate is typically inland but seasonally affected by the Baltic sea, with an annual rainfall of 650–700 mm year^{-1} (30-year average 470 mm). The landfill area has a facility for treating about 150,000 m^3 year^{-1} of leachate. This facility, described by Thorneby et al. (2006), consists of three consecutive ponds and a constructed wetland used to collect and treat the water. This water was used in this study as well as the compost obtained from the food waste of the whole municipality. Contaminated soil was amended with compost at 10% (w/w, fresh soil FW).

Nine piles of 9 m^2 and 0.7 m in height were constructed on May 2015 at the Moskogen facility (Fig. 1: pile design). The pile was lined with a polyethylene waterproof membrane to prevent the leachate from draining into the ground and ensure the correct inflow-outflow water balance. A 10 cm drainage layer made of gravels and sand was laid manually on the bottom. The ecotextile was placed over a 50 mm perforated drainage pipe and then pile was raised with compost-amended soil (10%C). The piles were constructed with 15° of inclination to allow adequate flow/drainage of leachate that was further collected in 1000 L underground tanks.

2.2. Experimental set up

The experimental set-up was a full randomized design of three treatments with three replicates: unplanted pile (Unp-10%C), pile planted with *M. sativa* alone (MS-10%C) and pile co-planted with *M. sativa* and *H. annuus* (MSHA-10%C) (Fig. 2). Seeds of alfalfa and sunflower were obtained from Weibulls Seed Company, Sweden. Growth was allowed for 5 months (week 23 to week 44). During dry days, piles were automatically irrigated every two days with water from the landfill leachate treatment.

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