

Chemosphere 65 (2006) 1642-1651

CHEMOSPHERE

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Dissipation of polycyclic aromatic hydrocarbons from soil added with manure or vermicompost

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> Received 26 August 2005; received in revised form 14 February 2006; accepted 17 February 2006 Available online 31 March 2006

Abstract

The dissipation of three PAHs, i.e., 500 mg phenanthrene kg⁻¹ soil, 350 mg anthracene kg⁻¹ soil and 150 mg benzo(*a*)pyrene kg⁻¹ soil, was investigated in soil from Acolman (México) added with cow manure or vermicompost while production of CO₂ and inorganic N was monitored. At day 0, recovery of added phenanthrene was 95%, anthracene 96% and benzo(*a*)pyrene 100% in sterilized soil and concentrations did not change significantly in sterilized soil over time. Application of organic material did not affect the concentration of phenanthrene was detected after 100 days and less than 8.5% of the added anthracene (mean of the two experiments). The decrease in concentration of benzo(*a*)pyrene (BaP) was not fast as that of phenanthrene and anthracene, and 22% was extractable from soil still after 100 days. It was concluded that addition of farm yard manure (FYM) and vermicompost only had an effect on the initial dissipation of phenanthrene, anthracene and benzo(*a*)pyrene in soil of Acolman.

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Keywords: Manure; Polycyclic aromatic hydrocarbons; Dissipation; Vermicompost

1. Introduction

Throughout the 20th century there has been a rapid increase in contamination of soil with oil and its derivatives due to petroleum spills, industrial wastes, and transport and storage accidents (Harrison et al., 2000). According to PROFEPA (2002), an average of 550 environmental emergencies from different sources of contamination occur in Mexico each year and 40% is due to crude oil.

Petroleum hydrocarbons belong to the most widespread contaminants of water and soil. The biodegradation of many components of petroleum hydrocarbons has been reported in a variety of terrestrial and marine systems. Bioremediation of hydrocarbon-contaminated soil by indigenous microflora can be stimulated by adding organic material and nutrients (Sims et al., 1990; Stegman et al., 1991; Wilson and Bouwer, 1997). Readily available organic residues, such as manure, municipal waste water, municipal solid wastes, compost and biosolids have all been added to contaminated soil, but stimulation of microbial degradation activity is still an emerging biotechnology (Wilson and Jones, 1993; Banerje et al., 1997; Wilson and Bouwer, 1997; Namkoong et al., 2002). Kastner and Mahro (1996) reported a degradation of polycyclic aromatic hydrocarbons (PAHs) by microorganisms of compost and this degradation was not caused by sorption to organic matter. The supply of macronutrients, notably N and P, has enhanced PAHs degradation in some cases (Carmichael and Pfaender, 1997; Liebeg and Cutright, 1999; Phillips et al., 2000). Raymond et al. (1976) studied oil biodegradation in soil and found greater degradation in soils receiving fertilizer application and rototilling than in untreated soils. Hence the addition of N and P containing fertilizers can be used to stimulate microbial hydrocarbon degradation (Atlas, 1981).

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^{0045-6535/\$ -} see front matter @ 2006 Elsevier Ltd. All rights reserved. doi:10.1016/j.chemosphere.2006.02.028

Biosolids are known to be effective in stimulating biodegradation of soil contaminants, because they contain large concentrations of inorganic nitrogen, available phosphorous and organic material (Korboulewsky et al., 2002; Namkoong et al., 2002; Rivera-Espinoza and Dendooven, 2004). They could thus be used to remediate hydrocarboncontaminated soil (Eiceman et al., 1989; Gutierrez-Ruíz et al., 1995; Benton and Wester, 1998). Most biosolids in Mexico are low in heavy metal concentrations, but contain large concentrations of pathogens. The biosolid must thus be treated before it could be used to remediate soil (Franco-Hernández et al., 2003). Conventional treatment of biosolid consist in adding lime to pH 12 (Thomas, 1996). However, vermicomposting or composting with earthworms reduces pathogens within a shorter time than in traditional composting and accelerates degradation of organic material while maintaining amounts of nutrients. Vermicompost would be ideal to remediate soils (Pierre et al., 1982; Sinha et al., 2002; Field et al., 2004).

We are not aware of any studies that used vermicompost to stimulate remediation of hydrocarbon-contaminated soil. The aim of this work was to investigate the addition of vermicompost obtained from biosolid and the earthworm *Eisenia fetida* and how this affected the degradation of PAHs. Soil was contaminated with phenanthrene, anthracene and BaP. Production of CO₂, dynamics of inorganic N (NH₄⁺, NO₂⁻ and NO₃⁻) and concentrations of the PAHs were monitored in an aerobic incubation at $22 \pm$ 2 °C for 100 days. FYM, partly used to generate the vermicompost, was also added to the hydrocarbon-contaminated soil and its effect on remediation was studied.

2. Material and methods

2.1. Chemical products

Hydrocarbons were obtained from Sigma (USA) with purity of >96% for phenanthrene, 99% anthracene and 97% benzo(*a*)pyrene. Acetone was obtained from J.T. Baker (USA) with purity 99.7%. It would have been interesting to use ¹⁴C labelled PAHs to study mineralization, but they cannot be imported into Mexico for security reasons.

2.2. Characteristics of the manure and vermicompost

Manure was collected from a cowshed and had an organic C content of 432 g C kg⁻¹, total phosphorous 12 g P kg⁻¹, available phosphorous 0.87 g P kg⁻¹ and total nitrogen content 21.9 g N kg⁻¹ on a dry matter base. *E. fetida* was cultivated in biosolid obtained from a wastewater treatment plant at Lerma, Edo. de México. Details of the biosolid used and the vermicompost obtained can be found in Contreras-Ramos et al. (2005). Briefly, the vermicompost was obtained from a mixture of 1800 g biosolid and 800 g manure at 70% water content added with 40 *E. fetida* and conditioned for three months. The vermicom-

post with the best stability and maturity and a weight loss of 18% was obtained with 1800 g biosolid, no straw and 800 g manure at 70% water content. This vermicompost had the following properties: pH 7.9; organic C content of 117 g kg⁻¹; an electrolytic conductivity of 11 mS cm⁻¹: a humic-to-fulvic acid ratio of 0.5 (HA/FA); total N content of 9 g N kg⁻¹; water soluble C (C_w) less than 0.5%; cation exchange capacity of 41 cmol_c kg⁻¹; a respiration rate of 188 mg CO_2 -C kg⁻¹ compost-C day⁻¹; a NO_3^-/CO_2 ratio greater than 8; and a NH_4^+/NO_3^- ratio lower than 0.16. The vermicompost gave a germination index for cress (Lepidium sativum) of 80% after two months while the earthworm production increased 1.2-fold and volatile solids decreased five-times. In addition, the vermicompost contained less than three CFU g^{-1} Salmonella spp., no faecal coliforms and Shigella spp. and no eggs of helminthes. Concentration of sodium was $152 \text{ mg kg}^{-1} \text{ dry}$ compost, while concentrations of chromium, copper, zinc and lead were below the limits established by USEPA (1995).

2.3. Experimental site and soil sampling

The experimental site is located near the ex-convent of Acolman in the State of Mexico (Northern Latitude 19° 38' Western Longitude 98° 55'). Its average altitude is 2250 m above sea level and characterized by a sub-humid temperate climate with a mean annual temperature of 14.9 °C and average annual precipitation of 624 mm mainly from June through August (http://www.inegi.gob.mx). The soil in this area is mainly cultivated with maize and oat and that for >20 years, receiving a minimum amount of inorganic fertilizer without being irrigated (http://www.inegi. gob.mx). Soil for the first experiment was sampled in spring and for the second one in summer. The sandy loam soil (Gee and Bauder, 1986, USDA modified soil texture triangle) with water holding capacity (WHC) 68.25%, pH in water 6.58 (Thomas, 1996), organic C content 18 g kg⁻¹ soil (Amato, 1983), inorganic C 0.7 g kg^{-1} soil (Nelson and Sommers, 1996), and total N 0.84 g kg^{-1} soil (Bremner, 1996) contained 220 g clay kg⁻¹, 140 g silt kg⁻¹ and 640 g sand kg⁻¹ (Gee and Bauder, 1986). Three plots of ca. 400 m² were sampled at random from an agricultural field of 1 ha. Thirty samples were collected by augering the top 0–15 mm layer of each plot.

2.4. Addition of PAHs

In experiment one in which FYM was added to soil, 1 ml acetone was used to add 500 mg phenanthrene kg⁻¹, 350 mg anthracene kg⁻¹ and 150 mg benzo(*a*)pyrene kg⁻¹ together. The soil was left to stand for 30 min to allow evaporation of the acetone and thoroughly mixed. However, it appeared that not all acetone evaporated as the production of CO₂ was larger in soil to which PAHs were added as compared to untreated soil. The CO₂ can be generated by the degradation of residual acetone, PAHs or/and killed biomass.

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