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Phenanthrene degradation in soil by ozonation: Effect of morphological and physicochemical properties



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ABSTRACT

The aim of this study was to characterize the ozone reaction with phenanthrene adsorbed in two types of soils (sand and agricultural). The effect of soil physicochemical properties (texture, bulk density, particle density, porosity, elemental composition, permeability, surface area and pore volume) on the phenanthrene decomposition was evaluated. Commercial sand has a uniform morphology (spherical) with a particle size range between 0.178 and 0.150 mm in diameter, regular elemental composition SiO₂, specific density of 1701.38 kg/m³, a true density of 2492.50 kg/m³, with an effective porosity of 31%. On the other hand, the agricultural soil had heterogeneous morphology, particle size between 0.1779 and 0.05 mm in diameter, elemental composition was montmorrillonite silicon oxide, apparent density of 999.52 kg/m³, a true density of 2673.55 kg/m³, surface area of 34.92 m²/g and porosity of 57%. The percentage of phenanthrene decomposition in the sand was 79% after 2 h of treatment. On the other hand, the phenanthrene degradation in the agricultural soil was 95% during the same reaction time. The pore volume of soil limited the crystal size of phenanthrene and increased the contact surface with ozone confirming the direct impact of physicochemical properties of soils on the decomposition kinetics of phenanthrene. In the case of agricultural soil, the effect of organic matter on phenanthrene decomposition efficiency was also investigated. A faster decomposition of initial contaminant and byproducts formed in ozonation was obtained in natural agricultural soil compared to the sand. The partial identification of intermediates and final accumulated products produced by phenanthrene decomposition in ozonation was developed. Among others, phenanthroquinone, hydroquinone, phenanthrol, catechol as well as phthalic, diphenic, maleic and oxalic acids were identified.

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1. Introduction

In 2010, around 161 sites were identified as potentially

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contaminated in Mexico. A third of all these sites (55) was mainly polluted by compounds derived from petroleum. With respect to oil spills and leaks, the local petroleum company (PEMEX) reported that 8031 tons of hydrocarbons (oil, diesel and gasoline) were spilled, mostly in soil of four pipeline sectors placed in the country (PEMEX, 2001a; 2001b; Salvador Vega et al., 2012).

Polycyclic aromatic hydrocarbons (PAHs) are by-products formed in the pyrolysis of petroleum, in acetylene synthesis from natural gas, refinery operations, etc. These compounds, considered as priority contaminants, have been detected in air, water and soil (Fouillet et al., 1991). In the case of soils, PAHs concentration was above 10 g/kg, which is considered an extremely high value (Lundstedt, 2003). PAHs are characterized by their hydrophobic nature and therefore, they tend to be adsorbed on solid particles. This characteristic converts them into a relevant source of soil contamination. Due to their low solubility in water, PAHs can be adsorbed on the surface of the soil that decreases their bioavailability. In consequence, the biodegradation of these compounds is usually difficult (Cerniglia, 1992; Luthy et al., 1994; Mulder et al., 2001; Hansen et al., 2004).

Degradation of PAHs in soils has been performed by two types of treatments: physical treatment (extraction with subcritical and supercritical fluids, surfactants, vegetable oil, solvents, etc.) and chemical treatment (oxidative treatment with ozone and/or hydrogen peroxide with or without iron ions) (Ferranese et al., 2008; Iossari et al., 2007; Gómez -Alvarez et al., 2012; Helena et al., 2012; Wu et al., 2015; Li, et al., 2015). Some other conventional methods such as incineration and thermal desorption (Javorská et al., 2009; Osgerby, 2006), vapor extraction and bioventing have shown poor results when PAHs are the target pollutants to be eliminated from soil (Choi et al., 2001; Goi et al., 2006). Hence, mixed treatment strategies based on the sequence of chemical oxidation with ozone or hydrogen peroxide coupled to biodegradation seem to be a plausible option to remove PAHs adsorbed in soil (Choi et al., 2001; Srehr et al., 2001; Haapea and Tuhkanen, 2006; Derudi et al., 2005; Kulik et al., 2006; Poznyak et al., 2007; Gómez -Alvarez et al., 2012; Ahn et al., 2005; Alcantara et al., 2008).

In the case of treating contaminated soil by ozone, the majority of previous research studies controlled the treatment efficiency by the decomposition only of the initial compound. Some results focused on the decomposition of anthracene by ozone have reported the 9,10-anthraquinone, the anthrone and the phthalic acid as relevant by-products. These studies usually consider sand as model soil to evaluate the treatment efficiency (Gómez -Alvarez et al., 2012; Zhang et al., 2005; Perraudin et al., 2007).

Some studies have shown that ozonation is an effective pretreatment step before considering the application of any biodegradation treatment by reducing the toxicity of the PAHs. This methodology assumes that byproducts formed after ozonation are less toxic and more biodegradable than initial compounds (Goi and Trapido, 2004; Liang et al., 2009; Nam and Kukor, 2000a; 2000b; Sutton et al., 2011). Then, sequential treatments based on the combination of oxidative process (as pre-treatment) and biodegradation may seem to be an efficient option to remove PAHs from complex solid phases (Goi et al., 2006; Kulik et al., 2006; Ahn et al., 2005; Nam and Kukor, 2000a, 2000b; Zeng and Hong, 2002).

These processes are strongly influenced by some specific operation variables such as dose of oxidant, soil particle size, content and nature of organic matter etc. (Volke and Velasco, 2002; Wang, et al., 2012). Additionally, efficiency of the PAHs decomposition for either single or combined treatment must consider not only the chemical structure of the contaminant, but also the physicochemical properties of soils such as texture, bulk density, particle density, porosity, elemental composition, permeability, surface area and

pore volume (Van Deuren et al., 2001; Masten and Davies, 1994).

The aim of this work was to study the phenanthrene decomposition in different soils (model and real) by ozonation. In particular, this work focused on study the effect of morphological and physicochemical properties of soils and the presence of organic matter on the phenanthrene decomposition dynamics, as well as on the distribution of by-products. The partial identification of these compounds was carried out by the HPLC analysis to characterize the reaction pathway.

2. Materials and methods

2.1. Soil characterization

The sand used as model soil was commercial and has been donated by the Mexican Petroleum Institute. Agricultural land was obtained from a farming site located in the municipality of Allende, Cuernavaca, Morelos, Mexico. This site was exclusively used for organic (without pesticides or some other chemicals) cultivation of agave. Therefore, it is considered free of contaminants.

The soil characterization was based on determining the following physicochemical properties: granulometry, apparent and real density, moisture, permeability, morphology, texture, porosity, heavy metals, organic matter concentration, pH, conductivity and BET area. Size, morphology and distribution of soil aggregates were made by SEM technique. A scanning electron microscope JSM-6300 JEOL was used to obtain all the aforementioned morphological properties. Also, pore volume was evaluated by filling empty space of soil contained in a vessel. This procedure was executed with distilled water as filling substance. The coating of the porous media was prepared with a carbon film or metal (gold or platinum) to provide sufficient electrical conductivity in the system.

To determine the mineralogical composition and elemental composition of soil samples, the XRD technique was employed. Two equipment were used to complete these morphological analyses: X-ray Spectroscopy and Energy Dispersive Spectroscopy Brand NORAN 2000. To determine the heavy metals content in the sample of agricultural land the technique reported in (Cresser and Hargitt, 1976) was used. This methodology is similarly to the reported in (Yanzheng et al., 2006). All analyses were performed by Atomic Absorption; model Perkin-Elmer, Series 2380. Table 1 summarizes the physicochemical properties of soils used in this study.

Table 1
Summary of the physicochemical properties of porous media.

Parameter	Sand	Agricultural soil	
		Before calcination	After calcination
^a Specific gravity, kg/m ³	1701.38	999.52	1105.67
Real density, kg/m ³	2492.50	2673.55	2581.88
Permeability (k, cm/s)	0.075	0.0049	0.0059
Porosity, %	31.71	57.14	57.14
Moisture, %	0.016	0.42	_
Texture	sandy	sandy	sandy
^b Composition, %			
Sand	99.33	94.77	
Clay	0.063	5.012	
Slime	0.004	0.011	
pН	10.19	7.88	6.97
Conductivity, μS	525.51	307.69	289.16
BET surface area (m ² /g)	7.87	34.92	13.027
Pore volume (cm ³ /g)	0.0036	0.0160	0.0059

^a Standard method ASTM D 854.

^b Standard method ASTM D 422a.

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