



Evaluation of pyrene sorption–desorption on tropical soils



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ABSTRACT

Sorption–desorption processes control soil–pollutant interactions. These processes determine the extent of pyrene transport in soils. Understanding sorption characteristics of pyrene is necessary in ascertaining its fate in soil. Laboratory batch experiments were conducted to study the sorptions–desorption of pyrene on eight soils from varying tropical agro-ecological zones (AEZs). The results showed that pyrene sorptions equilibria were attained within 720 min. Solution pH had a reciprocal effect on pyrene sorptions. Sorption was exothermic and increased with pyrene concentration in solution. The quantities of pyrene sorbed by each soil as well as the hysteresis were proportional to the percentage organic matter, and to some degree, the clay mineralogy. Sorption isotherms showed distributed reactivity involving several linear and non-linear isotherms. The present investigation showed that pyrene is likely to be more available to biota and reach the aquifer faster in low organic matter soils than those with relatively higher organic matter and more so in warmer climates.

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1. Introduction

Natural polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in the environment; however, their amounts (especially in soils) have been enormously supplemented by human activities resulting in unprecedented soil pollution levels (USEPA, 2007). Soil, the ultimate sink for most pollutants, has aroused widespread concern by scientists and environmentalists because of its importance in global development issues such as food security and global burden of disease (ATSDR, 1995).

The toxicities of PAHs (pyrene, fluorene, naphthalene, Phenanthrene, etc.) are well documented in several biota species (USEPA, 2007), and in some instances it can be amplified by bio-accumulation in the food chain. They have been shown to induce some cancers and genetic defects in humans, and thus PAHs have been classified as priority environmental pollutants by USEPA (USEPA, 2007; An et al., 2010). Hence, understanding the fate of these pollutants in soil is of paramount importance. Although PAHs are among the most intensively studied pollutants (An et al., 2010; Hwang and Cutright, 2002; Teixeira et al., 2011; Sun and Yan, 2007; Ramirez et al., 2001; Haftka et al., 2010), there is dearth of information concerning their fate on tropical sub-Saharan soils especially in Nigeria.

Sorption is a critical process in the control of environmental pollution because the fate of most organic pollutants depends both on the interactions between the soil components and organic pollutant as well as the prevailing conditions of the soil/organic pollutant environment (Hwang et al., 2003). Thus, PAHs will exhibit different fates in different soils formed from different parent materials under different environmental conditions, and this has led to studies concerning their bioavailability and remediation (Hwang and Cutright, 2002; Teixeira et al., 2011; Ramirez et al., 2001). Hence, understanding the sorption–desorption behavior of these contaminants in soils is vital for predicting their fate and transport, and ultimately their bioavailability.

Soils in Nigeria vary extensively in physical and chemical properties in the various agro-ecological zone (AEZs) due to different soil forming parent materials and the prevailing environmental conditions (temperature, moisture, and biota). Considering this diversity of soils in the country, pyrene sorption study using these soils is important in order to obtain technical and environmental safety recommendations for the different soils and climate conditions. Thus, in this study, soils from different AEZs in Nigeria were used.

Pyrene was chosen as a model PAH in this study due to its wide prevalence in soils and the dearth of information on pyrene sorption on Nigerian soils. When in soil, it is subjected to numerous physical, chemical and biological processes that determine its fate. Sorption and desorption are predominant of these, and they determine, to a large extent, the quantity of the organic pollutant

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retained by the soil as well as that leached to the aquifer (Teixeira et al., 2011). Hence, this work was specifically designed to study the sorption and desorption of pyrene on eight soils having varying physicochemical properties in order to understand the effects these properties on pyrene sorption and desorption on soils of African origin. The kinetics and adsorption isotherm models as well as thermodynamics parameters were used to describe the sorption data.

2. Materials and methods

2.1. Soil sampling, pretreatment and characterization

Eight soil types were obtained from surface soils (0–30 cm) from eight AEZs in Nigeria – Table 1a and b and Fig. 1. Prior to sorption experiments, the soil samples were air-dried, crushed gently and sieved through a 230-mesh size sieve and the fine fractions retained for the study.

The pH of the soils was determined in milipore ultra-pure water and 1.0 M KCl (ratio 1:1) (Benton, 2001). Particle size determination was done using the laser particle analyzer (Mastersizer-2000). Organic matter was determined by the Walkley-Black (1934) wet digestion method. Metals (oxides, exchangeable and trace) in the soils were determined using inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 5300DV), except for Cd which was done by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500).

2.2. Pyrene sorption studies

Pyrene (100 µg/L) was prepared in 20% v/v acetonitrile/milipore ultra-pure water using 0.005 M CaCl₂ and 0.01 M NaN₃ as background electrolyte and biocide to eliminate microbial degradation, respectively. Sorption experiments were carried by adding 10 mL of 100 µg/L (except where otherwise stated) pyrene solution into brown vials containing 0.50 g of the soil sample and then tightly sealed with Teflon lined screw caps. Aluminum foil was used to wrap the vials to minimize possible losses by photochemical decomposition. The vials were equilibrated in the dark by shaking at 100 rpm in a temperature (25 °C) controlled shaker. For the effect of time, the vials were incubated from 10 to 1440 min. Equilibrium studies and effect of temperature were investigated in the concentration range of 20–100 µg/L at 25 °C and 40 °C. After the equilibrium experiments pyrene desorptions were carried out immediately by thoroughly decanting the incubated solution and replacing with a fresh 20% v/v acetonitrile/milipore ultra-pure water solution containing the same concentration of CaCl₂ and NaN₃ but no pyrene. These were incubated in similar conditions as above, centrifuged after 1440 min, and pyrene concentration determined. All experiments were done at the

ambient soil pH. Control experiments without soil samples were done to account for possible losses due to volatilization and sorption onto the vials walls (and these were found to be negligible). After incubation, the vials were centrifuged at 4000 rpm for 20 min, and the concentration of pyrene in solution determined by Perkin Elmer fluorescence spectroscopy (model LS 55) using the synchronized scan at a wavelength difference (Δλ) of 36 nm and scanning from 300 to 400 nm. The synchronous peak at 335 nm was used to monitor the sorption.

2.3. Data treatment

The amounts of pyrene sorbed were calculated from the initial (C₀) and final (C_e) concentrations in solutions after the incubation using the Eq. (1):

$$q_e = \frac{(C_0 - C_e)V}{M} \quad (1)$$

where q_e , V and M are the amount of pyrene sorbed (µg/g), volume of the solution (mL) and mass (g) of sample, respectively.

Three adsorption models – Langmuir (1916) (Eq. (2)), Freundlich (1906) (Eq. (3)), and Distributed reactivity model (DRM) (Weber et al., 1992) (Eq. (7)) in their linear forms were employed in describing the adsorption process.

$$\frac{C_e}{q_e} = \frac{1}{Q_0 b} + \frac{C_e}{Q_0} \quad (2)$$

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (3)$$

where Q_0 (µg/g) is the soil maximum adsorption capacity; b the binding energy constant; K_f and n are Freundlich isotherm constants; and other parameters are as stated above.

The DRM describes sorption comprised of both linear and non-linear isotherms. Here, the linear isotherm was represented by Eqs. (4) and (5), and the non-linear isotherm by Eq. (6). A combination of Eqs. (4) and (6) gives the DRM in Eq. (7).

$$q_e = \left(\sum_{i=1}^m x_i K_{D_i} \right) C_e = \sum_{i=1}^m x_i K_{D_i} C_e \quad (4)$$

where q_e and C_e are same as above; K_{D_i} and K_D are the partition coefficient for reaction i expressed on a per mass of component i basis and the mass-averaged partition coefficient for the summed linear components, respectively. For true partitioning processes, Eq. (4) is rearranged to accommodate the mass fraction of the organic matrices

Table 1a
Physico-chemical properties of the soil samples.

| Soil | AEZ ^a | Sampling coordinate | pH water | pH 1 N KCl | EA ^c | OM ^d (%) |
|--------------------|---------------------------------|----------------------------|----------|------------|-----------------|---------------------|
| Lokoja (LF) | Southern Guinea SV ^b | 6° 45'00" E; 7° 49'00" N | 6.73 | 5.86 | 0.14 | 6.25 |
| Agbor (AG) | Fresh water swamp | 6° 11'36" E; 6° 15'06" N | 6.29 | 5.39 | 0.10 | 5.26 |
| Gembu (GSF) | Montane | 11° 15'00" E; 6° 43'00" N | 5.14 | 4.25 | 0.14 | 2.80 |
| Jos (JB) | Northern Guinea SV ^b | 8° 52'00" E; 9° 48'00" N | 5.17 | 4.08 | 0.44 | 3.57 |
| Port Harcourt (PH) | Mangrove swamp | 6° 59'55" E; 4° 47'21" N | 7.66 | 7.24 | 0.06 | 5.29 |
| Ibadan (IB) | Moist-lowland/Rainforest | 3° 54'00" E; 7° 26'30" N | 6.57 | 5.88 | 0.08 | 4.68 |
| Maiduguri (MA) | Sudan SV ^b | 13° 09'42" E; 11° 50'36" N | 6.67 | 5.21 | 0.02 | 1.67 |
| Monguno (MG) | Sahel SV ^b | 13° 36'51" E; 12° 40'14" N | 8.40 | 7.59 | 0.12 | 0.18 |

^a AEZ – Agroecological zone.

^b SV – Savanna.

^c EA – Exchange acidity.

^d OM – Organic matter.

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