



Assessing the environmental impact of phenanthrene in different types of land use based on the binding characteristics with dissolved organic matter

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ABSTRACT

The binding characteristics of phenanthrene with dissolved organic matter (DOM) were studied by the excitation emission matrix fluorescence spectroscopy with parallel factor analysis in four types of land use which derived from forest (F), meadow (M), cropland (C), and greenhouse (G). The results showed that the humification degree and binding characteristics of phenanthrene with DOM were distinct differences in the four soils. The binding capacities of humic-like components with phenanthrene were stronger than those of protein-like components. The log *K* derived from the Stern-Volmer equation significantly correlated with the humification degree of DOM ($p < 0.05$) in different types of land use. Besides, correlation analysis demonstrated that the potential binding index (Fk) obtained from the modified Stern-Volmer model was a more accurate parameter to describe the combination degree of DOM with phenanthrene than log *K*, which presented a decrease order of C > F > M > G. Therefore, the environmental impact of phenanthrene in different types of land use could be assessed deeply based on the Fk and DOM concentration.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), with stable ring structures, poor water solubility and chemical stability, have been general concern because of their toxicity and carcinogenicity in recent years. PAHs are mainly produced by combusting fossil fuels incompletely, and pose a serious threat to the environment and human health. The three benzene-ring containing compound, phenanthrene, is one of the common PAHs in soils, water and sediments. Generally, phenanthrene occurs in low concentrations (dozens of nanogram per liter) in the marine environment (Zhang et al., 2014). However, exceptionally high levels of phenanthrene ranging from 14.6 µg/L (Vrana et al., 2001) to 1460 µg/L (Anyakora et al., 2005) have been reported in the marine environment. These levels are high enough to impair the growth and physiological functions of fish (Cypher et al., 2017; Hano et al., 2017). In addition, the vapor pressure of phenanthrene is low and volatile. It has a strong migration activity in the environment due to its strong affinity with organic matter. Therefore, soil is one of the most important sinks for phenanthrene in the environment. For example, in agricultural soils of China, the concentrations of phenanthrene range from 11.1 ng/g to 13,067 ng/g (soil dry weight) (Ma et al., 2015; Wang

et al., 2010; Xing et al., 2011). Phenanthrene can be accumulated in crops and subsequently transferred to humans through the food chain, which poses a serious hazard to human health (Deng and Zeng, 2017). Moreover, phenanthrene in the atmosphere may enter the human body through respiratory tract, causing harm to human cardiovascular health (Brette et al., 2017). Thus, these can be seen that phenanthrene causes serious toxic effects. However, the adverse effects can be reduced when they bind to dissolved organic matter (DOM).

DOM plays a significant role in ecosystem by influencing carbon and nitrogen cycles. Various biotic and abiotic processes, such as degradation, mineralization, partitioning, dissolution, and aggregation, are associated with the dynamics of DOM (Inamdar et al., 2012; Leenheer and Croué, 2003; Schindler et al., 1997). Moreover, DOM can control toxicity and bioavailability of pollutants in the environment (Fu et al., 2016; Yuan et al., 2015). Many previous studies reported that the fate, transport and bioavailability of metals and PAHs are largely affected by DOM (He et al., 2015, 2014; Matar et al., 2013). Although many literatures have reported that microbial degradation was the way to remove PAHs from soils (Cai et al., 2016; Han et al., 2015; Hur et al., 2011), this way is limited in natural environments of northern China because of the low temperature. Therefore, the combination between

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DOM and pollutants may be an important way to reduce the bioavailability and toxicity of pollutants.

Different fractions of DOM have distinct effects on adsorption, migration, and accumulation of PAHs in the environment (Hu et al., 2016; Ishii and Boyer, 2012). These components may differ in aspects such as functional groups, molecular weights, and redox sites (Jia et al., 2013; Tan et al., 2017). These discrepancies can result in various quenching effects that may significantly affect the degradation of pollutants. Thus, the binding capacities between DOM fractions and phenanthrene are diverse. Moreover, the concentration and composition of DOM can be changed by land use. Land use can influence soil properties, such as pH, moisture, temperature and nutrient availability (Jiang et al., 2014). There are great differences in structure and composition of DOM derived from different sources, e.g. compounds with high aromaticity and molecular weights are often dominated in DOM leachates from humified organic soils (Derrien et al., 2017; Hu et al., 2016). The humification degree of DOM is correlated with binding capacity for PAHs (Mei et al., 2009). Therefore, understanding the function of individual fractions and effect of land use on DOM are essential to better elucidate the binding characteristic of DOM with phenanthrene.

We analyzed and compared DOM composition from four different patterns of land use through excitation emission matrix (EEM) fluorescence spectroscopy coupled with parallel factor analysis (PARAFAC). The method of fluorescence quenching and mathematical models, including the Stern-Volmer and modified Stern-Volmer models, were used to figure out the binding characteristics. The objectives of the present study were to: (1) compare the differences of DOM composition in different land uses; (2) compare the differences of binding characteristics of each DOM component with phenanthrene in different land uses; (3) discuss the fate of the complex DOM-phenanthrene (DOM-phe) in the environment.

2. Materials and methods

2.1. Sample collection and preparation

The soil samples were randomly collected from the surface layer (0–20 cm) of soils located at Suihua, China, including forest soil (F), meadow soil (M), cropland soil (C), greenhouse soil (G). Four types of land use soils were collected to compare the characteristics of DOM binding to phenanthrene. Each soil type contained four sampling sites. Detailed information about sample location can be found in Supplementary information Table S1. The content of soil organic matter was about 11.4%, however, the content of forest soil organic matter was about 24.8%. The average content of total nitrogen and total phosphorus were 0.4% and 0.12%, respectively. The content of Pb and Hg were 28.36 mg/kg and 0.55 mg/kg, respectively. Cropland, forest and meadow have been maintained for at least ten years, and at least three years in greenhouse. Corn and soybeans were grown mainly in cropland, while greenhouse was mainly for commercial production of vegetables.

DOM samples were extracted by mixing one part of dried soil sample with five parts of ultrapure water and continuously shaking them for 24 h. The liquid was centrifuged at 10,000 rpm for 20 min at 4 °C and filtered through a pre-washed 0.45- μ m membrane. Then the filtered liquid was used for the fluorescence analysis. The concentration of DOM was measured by the total organic carbon analyzer (TOC-VcpH). DOM concentration are summarized in Table 1.

In this study, SUVA represents the aromatic character of DOM from different soils. The UV absorbance at 254 nm measured divided by the organic carbon concentration measured is defined as the Specific UV absorbance at 254 nm (SUVA₂₅₄) index. This index is used as an indicator for the aromaticity of the DOM, however, SUVA₂₈₀ is positively correlated to DOM molecular weight (Peng et al., 2015). A number of optical properties of DOM were calculated (Table 1). SUVA₂₅₄ and SUVA₂₈₀ can provide important insights on aromaticity degree,

Table 1

TOC values, specific UV absorbance and fluorescence characteristics of four types soil DOM. C1-C3 and C1' represent component 1–3 and 1' based on the PARAFAC analysis - derived components, respectively.

	DOM concentration (mg/L)	SUVA ₂₅₄ (L/mg·m)	SUVA ₂₈₀ (L/mg·m)	C1 (%)	C2 (%)	C3 (%)	C1' (%)
F	127.18	0.012064	0.008995	32.61%	47.34%	20.15%	–
M	53.40	0.011139	0.008415	–	49.30%	20.05%	30.64%
C	51.13	0.015361	0.013805	31.02%	48.34%	20.65%	–
G	47.12	0.012759	0.009550	31.38%	48.34%	20.29%	–

(C cropland, G greenhouse, F forest, M meadow).

humification degree, and molecular weight of DOM.

2.2. Quenching titration experiment

Phenanthrene dissolved in methanol was used as a stock solution with varying concentrations. Quenching titration was carried out by adding phenanthrene solutions into DOM samples. The concentration of all DOM samples were diluted to 12 mg(C)/L. The pH of the sample solution was adjusted to 7.0 by addition of 0.1 M HCl or 0.1 M NaOH. An 8 ml of each sample was pipetted into a cuvette and then added the standard solution of phenanthrene. The final phenanthrene concentrations of the total solutions were 0, 20, 40, 60, 80, 100, 120, 140, 160, 180, 200 μ g/L for the eleven soil DOM samples. The volumetric content of methanol was lower than 0.01% (v/v), at which point no apparent influence on fluorescence quenching was found. All solutions after titration were continuously shaken at 25 °C for 5 min to ensure an equilibrium binding between phenanthrene and DOM. Then the solutions were used for the determination of EEM fluorescence spectra.

The EEM fluorescence spectra were recorded by F-7000 fluorescence spectrometer (Hitachi High Technologies, Japan) at room temperature (20 \pm 2 °C). The measured excitation and emission wavelengths were set from 220 to 490 nm and from 250 to 550 nm with the scan steps of 10 nm and 2 nm, respectively.

2.3. PARAFAC analysis

The approach of PARAFAC analysis of EEMs has been well documented elsewhere (Ohno et al., 2008; Stedmon and Bro, 2008; Wu et al., 2011), therefore, only a brief description is provided here. The PARAFAC modeling was carried out in MATLAB 2010b using the free-download DOM Fluor toolbox (www.models.life.ku.dk). During PARAFAC analysis, a nonnegative constraint was applied to the parameters and two to eight components were computed for the EEMs. And split half analysis and residual analysis were used to identify the number of component. In the study, 176 EEMs were analyzed by the model, and four components were obtained from four land uses of soil samples.

The relative concentration of each component is reflected by fluorescence intensity which is represented by F_{max} . The contribution of each component to the total fluorescence (i.e., %C1, %C2 and %C3) is considered as DOM quality indices (Table 1).

2.4. Mathematical models

The Stern-Volmer and modified Stern-Volmer models are widely used for characterizing the hydrophobic organic contaminants or metals binding behavior of DOM although the models still have the limitation that they neglect the potential existence of multiple complexes.

The Stern-Volmer equation can explain the static quenching of a fluorophore derived by Gauthier et al. (Gauthier et al., 1987). Previous investigations showed fluorescence quenching of PAHs by DOM were attributed to static quenching mechanisms in light of temperature,

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