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DOM fractionated from pig waste affecting the solubility of PAHs along with non-ionic surfactant

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ABSTRACT

Studies on the intensification of water solubility for polycyclic aromatic hydrocarbons (PAHs) including phenanthrene and pyrene using individual surfactant and dissolved organic matter (DOM) fractionated from pig wastewater or mixed surfactant–DOM solutions were conducted. Excess surfactant dosages producing critical micelle concentration (CMC) dramatically enhance the solubility of phenanthrene and pyrene in the contaminated soil. The synergetic effect from non-ionic surfactants such as polyoxyethylene sorbitan fatty acid ester, and Tween 80, and DOM on the solubility of PAHs was also evaluated using batch experiments. The current study shows that the solubility of PAHs is respectively increased with an increase in the concentrations of Tween 80 or DOM, especially in the dosage of CMC or high hydrophobic property of DOM. Furthermore, an increase in the solubility of PAHs strengthened by the coexistence of Tween 80 and DOM was strongly affected by the concentration of Tween 80. It proves that the selection of optimal surfactant dosage is the key factor while the way is possibly applied into the remediation technologies in the on-site soil region with pollution.

Keywords: Surfactant; Dissolved organic matter (DOM); Polycyclic aromatic hydrocarbon (PAH); Remediation technologies

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), belonging to non-polar, low solubility, and hydrophobic compounds with low ionization and refractory biodegradability, are one kind of organic pollutants widely existing in the environment [1–4]. The sources, environmental behaviors, ecological effect, and remediation of PAHs in water and sewage sludge have been presented in some studies [5–12]. The solubility of PAHs in water systems is one of the important physicochemical properties to describe their mobilization, transformation, environmental fate,

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ecology effect, and bioavailability [13–15]. The solubility of PAHs represents not only the amount of PAHs in water but also its adsorption coefficient and bioaccumulation factors. Therefore, increasing the solubility of PAHs in water is the key premise for their removal from contaminated soil. Many studies had reported that surfactants can increase the solubility of PAHs in the aqueous phase as well as its mobilization in the environment [16–19].

Further studies proved that sodium dodecyl sulfate (SDS) with Triton X-405 (TX405) [20], and Tween 80, and dissolved organic matter (DOM) [21] have a synergistic effect resulting in the augment of the solubility of PAHs. Owing to the mechanisms of complexation and adsorption, DOM present in environment can significantly affect the fate and transport of hydrophobic organic compounds (HOCs) including PAHs, polychlorinated biphenyls, and chlorinated pesticides [22–24]. Both characteristics of hydrophobic and hydrophilic moieties in DOM can be conceptualized as a separate phase to retain hydrophobic organic contaminants. In addition, aquatic DOM mainly originates from terrestrial region and aquatic biota. The terrestrial organic substances are mainly composed of hydrophobic fractions; however, aquatic biota are attributed to hydrophilic fractions [25]. DOM interacts with PAH pollutants through hydrophobic binding resulting in humid-solute complexes in the aqueous phase, and further increasing the apparent solubility of organic solutes [26]. Moreover, several studies indicated that the evaluation of the mobility of PAHs in wastewater or surface water requires not only in the assessment of the total DOC concentration but also in the contents of the hydrophobic fractions [27,28]. Of course, the decrease of sorption for PAHs on the solid matrix diminishes its transportation in environment. Therefore, the application of DOM on enhancing the movement of PAHs between different phases was worthy of exploring.

As previously stated, both substances, like DOMs and surfactants, can increase the solubility of PAHs in the aqueous phase. Some reports had proved that the specific ability of DOM extracted from soil and from food waste compost could enhance the mobility of HOCs in aquifers and soils [29,30]. The form of hydrophobic sub-phase of microscopic dimension can interact with HOCs like the similar ability for surfactants [31]. According to the previous statements, the effects of DOM or surfactants on PAH solubility were worth for further investigation. Regarding DOM substance in pig waste, this substance was not effectively re-used in the past decades resulting in the pollution of water body in Main China. Actually, DOM in waste and surfactant was simultaneously discharged into aquatic systems with PAHs, possibly changing the PAH solubility. Hence, the objective of this study was aimed at the effect of coexistence between DOM fractionated from pig waste and non-ionic surfactants of Tween 80 on the solubility of PAHs into water to provide the vital information applicable to remedy the contaminated site.

2. Materials and methods

2.1. Preparation and analysis of DOM

The use of DOM in this study was respectively extracted from the pig manure (PM) and from the pig manure compost (PC). PM and PC were extracted with deionized water controlled in a ratio of solid to water of 1:40 (w/v, dry weight basis) in a reciprocal shaker at 200 rpm for 16 h at 25℃. After centrifugation at 12,000 r min^{-1} (4℃) for 10 min by a centrifuge (Eppendorf centrifuge 5417C), supernatant in the upper layer was filtered by 0.45-μm sterile membrane (GN-6 Metrice, Gelman Sciences, and Ann Arbor, MI). The filtrates were analyzed using total organic carbon (TOC) instrument (TOC-5000A, Shimadzu, Kyoto, Japan) with autoanalyzer to measure the amount of TOC [21]. In addition, the non-ionic surfactants of polyoxyethylene sorbitan fatty acid ester type, Tween 80 (15 mg L^{-1} critical micelle concentration (CMC)), was supplied by Sigma Co. (St. Louis, MO).

The properties of DOM were characterized by the molecular-weight separation technique consisting of different cellulose dialysis membrane tubes with a nominal molecular-weight cut-off (MWCO) of 1 k and 25 kDa [32]. All tubes were prewashed with deionized water for 4 h prior to conducting the experiments until the released carbon content became less than 0.5 mg L^{-1}. The dialysate was prepared with deionized water. The operation of dialysis was operated at 4℃ in the dark.

Regarding the procedure of DOM fractionation, 20 mL of sample was fractionated by the tube of 1 k MWCO and stirred in a dialysate with volume of 2,000 mL for 24 h. The 100 mL of dialysate were renewed for the first time and the second time when reaction times were operated at 2 and 18 h, respectively. After equilibrium time of 20 h, 10 mL of the retained solution in the tube was withdrawn (i.e. >1 kDa) for DOC analysis. Sequentially, the remaining 10 mL solution was placed into the tube of 25 k MWCO. The dialysis was repeatedly carried out as previously mentioned, and DOC was measured after 24 h.

2.2. Preparation and analysis of PAHs

In this study, both PAH species including phenanthrene and pyrene (purity of 98%; Aldrich, Milwaukee, WI) were mixed as the targets based on the substantial divergence in water solubility (1.18 mg L^{-1} for phenanthrene and 0.12 mg L^{-1} for pyrene) as well as the obvious difference in hydrophibicity (both values of log K_{ow} of 4.45 for phenanthrene and 4.88 for pyrene) [33]. High purity of phenanthrene and pyrene was directly used in the following experiments without further pretreatment. Mixed solution of phenanthrene (20 mg L^{-1}) and pyrene (10 mg L^{-1}) was prepared in 100 mL of deionized water in 250 mL glass vials.

The concentrations of PAHs in DOM and/or surfactant solution were measured with high performance liquid chromatography (HPLC) (Model no. 515; Waters, St. Milford, MT) equipped with the fluorescence detector (Waters 2,487 dual absorbance detector). Both wavelengths of excitation and emission (EX/EM) were set at 244/360 nm for phenanthrene, and 237/385 nm for pyrene. The operational conditions of under which the chromatography was conducted were constant flow rate of 1.5 mL min^{-1}, mobile phase of 100% acetonitrile (HPLC grade), and 3.6 × 250 mm μ-bondapak C18 reverse phase column (Waters). The results yielded a sensitivity of 1.0 μg L^{-1} of phenanthrene and pyrene. The fluorescent values of different concentrations of DOM or surfactant did not apparently yield fluorescence intensity under these wavelengths. A similar finding had been demonstrated by the study of Chen and Wong [21].

2.3. Experimental procedure

The preparation of Tween 80, DOM, and Tween 80 + DOM were contained of 200 mg L^{-1} $HgCl_2$ for inhibiting microbial activity to biodegrade. Tween 80, DOM and Tween 80 + DOM were added into individual PAH solutions, respectively, the concentrations of DOM with 0, 50, 100, 200, and 400 mg L^{-1}, those of Tween 80 with 0, 7.5, 45, 75, 150, 300, and 600 mg L^{-1}, and same concentrations of Tween 80 prepared plus DOM with 0 and 400 mg L^{-1}, respectively. Meanwhile, the blank control (CK) was prepared using deionized water. After the operation of 72 h at 250 rpm and 25 ± 1℃ in a shaker for whole runs, filtrate was filtered 0.45 μm inorganic membrane filter mounted on a 10 mL syringe to remove undissolved PAHs.

3. Results and discussion

3.1. Characterization for PM and PC

The percentages of each fractions of MWCO for PM and PC, including <1 kDa, 1 k-25 kDa, and >25 kDa, were calculated by individual TOC for each fractions divided by the sum of TOC from whole DOM. The property of DOM with respect to hydrophilic and hydrophobic components was characterized by the separation technique using non-ionic macroporous resins of XAD-8 [34]. The variations of both molecular weight and hydrophobicity for DOM are listed in Table 1. Table 1 reveals that the opposite organic differences existed between PM and PC, indicating that less than 1 kDa of 71% in PM was higher than 26.7% in PC, and larger than 25 kDa of 22.4% in PM was less than 57.2% in PC. Small organic fraction in PM had the higher hydrophilic fraction than large molecular weight in PC.

3.2. Effects of surfactants and DOM on the solubility of PAH solution

Fig. 1 reveals the effects of different concentrations of Tween 80 or DOM on the apparent solubility of PAHs including phenanthrene and pyrene. The solubilities of both phenanthrene and pyrene in deionized water were respectively 0.644 and 0.058 mg L^{-1}. Fig. 1(A) demonstrates that the apparent solubility of PAHs increased with the ascending concentrations of Tween 80. Tween 80 concentration of 7.5 mg L^{-1} existing in phenanthrene and pyrene, resulted in the increase of the apparent solubilities 1.71 and 2.31 times higher than both corresponding PAHs in deionized water. The apparent solubilities of phenanthrene and pyrene were respectively increased to 2.62 and 13.25 times relative to PAHs in deionized water while the concentration of Tween 80 was 75 mg L^{-1}.

Surfactant molecules can form micellar aggregates in the water phase when a specific threshold, known as critical micellar concentration (CMC), is exceeded [35]. Incorporation of hydrophobic compounds in the micelles is termed as solubilization. In equilibrium situation, the amount of solubilized PAHs linearly depends on the surfactant concentration above the CMC [36–38]. The current result shown in Fig. 1(A) may be related with surfactants consisting of a hydrophilic and a hydrophobic moiety, resulting in the aggregation of PAHs at surfaces and interfaces, thereby decreasing the levels of surface tension and interfacial tension between DOM and surfactant.

Regarding to the effect of DOM on apparent solubility of PAHs, Fig. 1(B) reveals that the presence of

Table 1
Organic characteristics of PM and PC

	Proportion of molecules weight (%)			Hydrophobic or hydrophilic distribution (%)	
	<1 kDa	1 k-25 kDa	>25 kDa	Hydrophilic	Hydrophobic
PM	71.04	6.59	22.37	58.34	41.66
PC	26.67	16.18	57.15	44.62	55.38

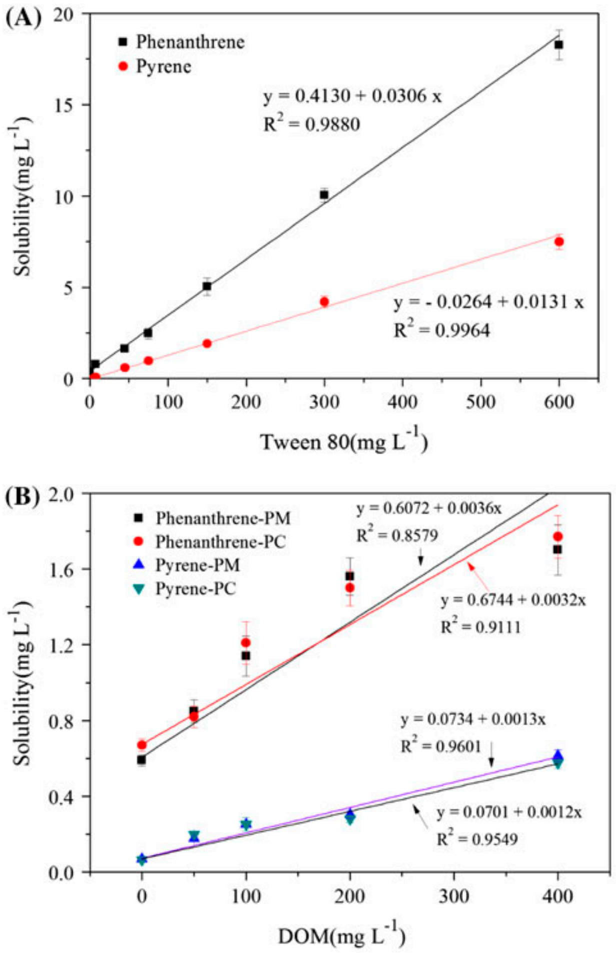

Fig. 1. Effects of (A) surfactant Tween 80 and (B) DOM derived from PM and PC on the apparent solubility of phenanthrene and pyrene.

PM or PC enhanced the solubility of phenanthrene and pyrene. As DOM concentration reached 100 mg L^{-1}, the apparent solubility of phenanthrene was respectively higher 1.58 times for PM and 1.94 times for PC compared with that in deionized water. For the apparent solubility of pyrene, 3.29 times for PM and 3.48 times for PC was higher than that in deionized water. The solubility of PAHs was affected by both DOM substances, those in PC were superior to those in PM which may be explained by the differences between molecular weight distributions and hydrophobic/hydrophilic property. This inference was in accordance with the result shown in Table 1. High hydrophobic property or large molecular weight could promote more solubility of HOCs than opposite organic property.

The apparent solubility of PAHs increased with the ascending concentrations of individual presence of Tween 80 and DOM. Water solubility enhancement of PAHs resulting from surfactant was directly related with concentration of single molecule and micelle of surfactant and homologous partition coefficient in the solution [39]. Owing to the amphiphilic structure existing in DOM and anionic surfactant, this phenomenon of the synergistic effect could further enhance the solubility of PAHs. A similar report was found in the research of Wu and Yang [40].

3.3. *Solubility of PAHs coexisted with DOM and surfactant*

Individual test for DOM or anionic surfactant on the effect of solubility of PAHs was completed and discussed in earlier paragraph. The coexistence of a similar structure like DOM and anionic surfactant on the effect of the solubility of PAHs was becoming an attractive issue. Compared with Figs. 1 and 2, it indicates that while the concentration of Tween 80 is less than 150 mg L^{-1} with DOM, the apparent solubility of phenanthrene was higher than that of phenanthrene with DOM or surfactant solution alone. Researchers demonstrated that the extent of desorption of PAHs in soil-water systems containing both Tween 80 and DOM was higher than the sum of the amount of PAHs desorbed from soils in the systems containing only either Tween 80 or DOM [21]. However, that value was still less than the summation from individual solubility of phenanthrene in DOM and Tween 80.

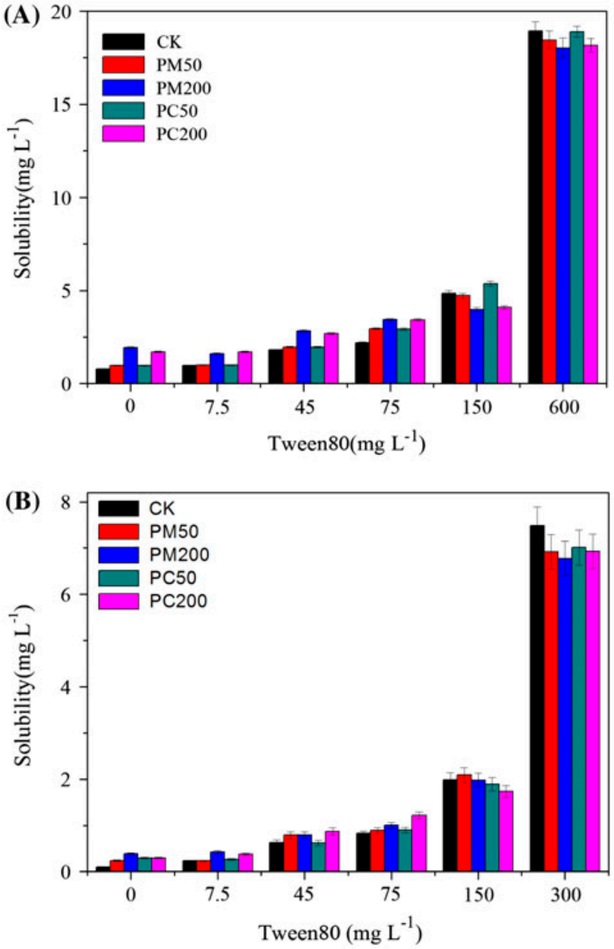

Fig. 2. Combined effect of surfactant Tween 80 and DOM derived from PM and PC on the apparent solubility of (A) phenanthrene and (B) pyrene (CK = solubility of PAHs in deionized water containing 200 mg L^{-1} $HgCl_2$).

This result was in contrast to the report of Hyun et al. [41]. Hyun proved that the combined effects of DOM with surfactant had an additive effect on solubilities of phenanthrene and pyrene. The divergence may be explained from different test conditions and the types of DOM and surfactant. Hydrophile-Lipophile Balance number of anionic surfactant in the mixed solution and octanol-water partition coefficient (K_{ow}) of PAHs may be directly related with synergistic solubilization of mixed solution [42]. Compared with Fig. 2(A) and (B), the enhancement of phenanthrene solubility in the presence of Tween 80 and DOM was much higher than that of pyrene. The octanol–water partition coefficient (K_{ow}) of PAHs, named as K_{ow}, is regarded as the key factor for explaining the previous result [43].

As shown in Fig. 2, Tween 80 and DOM may have the combined effect on the solubility of phenanthrene and pyrene in deionized water, especially in DOM property. In order to ensure the above results, available information was analyzed using linear regression equations in different DOM concentrations at three Tween 80 dosages, as shown in Table 2. Table 2 reveals the slope values of phenanthrene and pyrene were positive while the dosages of Tween 80 ranged from 0 to 75 mg L^{-1}, however, the drop of slope in phenanthrene was opposite to the increase of slop in pyrene. For the variations of solubility of phenanthrene by DOM substances, the higher effect by PM than PC was found. A similar result was found for pyrene while PM was compared with PC. It proves that surfactant of Tween 80 could weaken the soluble effect of DOM on phenanthrene and pyrene, but positive correlation between DOM and the solubility of phenanthrene and pyrene was kept. Regarding the high Tween 80 dosage of 600 mg L^{-1}, the slope value of linear regression equation was negative, implying that the solubility of PAHs decreased with the increasing concentrations of surfactant. The interaction of PAH fraction in DOM substance may be a possible mechanism [44,45].

Table 2
Correlation between phenanthrene or pyrene solubility and Tween 80 at both DOM substances

		Phenanthrene		Pyrene	
Tween 80 (mg L^{-1})	DOM[a]	Linear Regression Equation	Correlation Coefficient (R^2)	Linear Regression Equation	Correlation Coefficient (R^2)
0	PM	$Y = 0.0028X + 0.7658$	0.8317	$Y = 0.0013X + 0.0654$	0.9719
	PC	$Y = 0.0028X + 0.8018$	0.89	$Y = 0.0011X + 0.0805$	0.9453
75	PM	$Y = 0.0025X + 2.4443$	0.9581	$Y = 0.0016X + 0.8275$	0.939
	PC	$Y = 0.0017X + 2.3712$	0.8906	$Y = 0.0023X + 0.8085$	0.9912
600	PM	$Y = -0.0085X + 19.506$	0.9305	$Y = -0.0009X + 7.6687$	0.4166
	PC	$Y = -0.0054X + 19.474$	0.9332	$Y = -0.0017X + 7.7686$	0.8577

[a] Concentrations of DOM are 0, 50, 100, 200, and 400 mg L^{-1}.

4. Conclusions

Several findings were summarized below. High concentrations of nonionic surfactant of Tween 80, especially for the condition over the CMC, could promote the solubility of both PAHs including phenanthrene and pyrene. The ascending solubility of PAHs increased linearly with increasing DOM concentration. The DOM with high hydrophobic property resulted in more solubility of PAHs than that with the low hydrophobic property. For coexistence of Tween 80 and DOM, apparently, the solubility of PAHs was mainly affected by Tween 80 concentration.

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