

# Numerical simulation of PAHs sorption/desorption on soil with the influence of Tween80

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**Abstract:** In this paper, the influences of anionic surfactant Tween80 on polycyclic aromatic hydrocarbons (PAHs) sorption/desorption on artificially contaminated soil were studied, and  $\gamma$  model was applied to simulate the influences. Results showed that, with the use of Tween 80, the sorption behaviors of PAHs on soil altered significantly. Adsorbed Tween 80 increased the sorption amount of PAHs while the dissolved Tween80 increased the apparent solubility of PAHs. These two processes exert influences on the sorption coefficient of PAHs in soil-water system, which can be depicted by apparent sorption coefficient. The partition coefficients (the soil/water partition coefficient of PAHs and surfactants obtained from sorption experiments) and statistical parameters used in the amended  $\gamma$  model were obtained in independent experiments. With these parameters, the  $\gamma$  model could provide a satisfactory independent prediction of PAHs release from soil to aqueous phase at two surfactant concentrations.

**Keywords:** Tween80; PAHs; soil; sorption/desorption;  $\gamma$  model

## Introduction

The emission of polycyclic aromatic hydrocarbons (PAHs) from human production activities poses a serious threat to the quality of soil environment and groundwater. After entering into soils, PAHs undergo physical, chemical and biologic processes such as sorption and degradation. One of the major factors limiting remediation of PAHs-contaminated soil is the slow release of contaminants into the aqueous phase (Yeom and Mriganka, 1998). Owing to low aqueous solubility and high soil-water partition coefficients, the PAH compounds can be strongly sorbed onto the surface of particles and deposit to soils (Doong and Lei, 2003; Kim *et al.*, 2001; Zhu and Feng, 2003). The application of surfactants to soil contaminated with PAHs has become a possible way to enhance the bioavailability of these hydrophobic compounds and to facilitate their biodegradation. Surfactants enhance the release of PAHs from soil, and facilitate PAHs partition into the hydrophobic micelle core and/or monomer of surfactants (Ahn *et al.*, 1996; Brunk *et al.*, 1997; Cuypers *et al.*, 2002; Doong and Lei, 2003; Kim *et al.*, 2001; Tsomides *et al.*, 1995; Yeom *et al.*, 1996; Yeom and Mriganka, 1998; Mulligan *et al.*, 2001; Sobisch *et al.*, 2000). However more need to be known about the mechanism of PAHs desorption from soil.

Based on the experimental research in our laboratory on the PAHs desorption and degradation in soil, Tween80 was selected to carry out a further investigation owing to its low toxicity and high solubility. The purpose of this study is to determine the effects of Tween80 on the desorption of soil-bound PAHs simulated by a kinetic model— $\gamma$  model.

## 1 Theory

As indicated by Liu *et al.* (2001), several alternative models have been developed to describe the rate of desorption of hydrophobic organic contaminants from soil, including radial diffusion models (Wu and Gschwend, 1986; Pignatello *et al.*, 1993), multi-compartment models (van Genuchten and Wagenet, 1989; Gamedainger *et al.*, 1990) and  $\gamma$  model (Connaughton *et al.*, 1993; Liu *et al.*, 2001). The  $\gamma$  model is a kinetic model that incorporated a  $\gamma$  distribution of first-order sorption/desorption rate constants associated with the soil matrix, and was found to better predict the desorption behavior of hydrophobic compounds in batch and column experiments than the commonly used two-site/two-region model that has a comparable number of adjustable parameters (Liu *et al.*, 2001).

With the  $\gamma$  model, the PAHs desorption rate is described by (Liu *et al.*, 2001):

$$\begin{aligned} \frac{\partial M}{\partial t} &= \int_0^{\infty} \frac{\partial C}{\partial t} f(k) dk = - \int_0^{\infty} k(C - K_s^{\text{PAH}} C_{\text{aq}}) f(k) dk \\ &= - \int_0^{\infty} k C f(k) dk + (\alpha/\beta) K_s^{\text{PAH}} C_{\text{aq}} \end{aligned} \quad (1)$$

where  $k$  is the desorption rate constant of PAHs from soil compartment that are  $\gamma$  distributed ( $\text{min}^{-1}$ );  $f(k)$  is the probability density function of  $k$ ;  $C$  [ $=C(k)$ ] is the PAHs concentration in soil compartment with rate constant  $k$  in the  $\gamma$  model ( $\mu\text{g/g}$ );  $t$  is time ( $\text{min}$ );  $K_s^{\text{PAH}}$  is the distribution coefficient for PAHs to soil ( $\text{ml/g}$ );  $C_{\text{aq}}$  is PAHs concentration in the aqueous phase ( $\text{g/ml}$ );  $\alpha$ ,  $\beta$  are adjustable parameters for the  $\gamma$  distribution.  $\beta$  is a “scale parameter” and  $\alpha$  is a “shape parameter” whose effect has been illustrated by Connaughton *et al.*

al. (1993). The values of  $\alpha$  and  $\beta$  can be obtained by fitting experimental data for PAH desorption as a function of time.

Magee *et al.* (1991) uses a three-component equilibrium model to describe the effect of extracellular polymer on the phenanthrene sorption (Dohse and Lion, 1994; Brunk *et al.*, 1997). In our research, a three-component system (soil-PAHs-surfactant) was studied while Tween80 was added to the system as a surfactant.

In the three component system, interactions among PAHs, surfactant and soil exist. The rate of PAHs desorption from soil is given by:

$$\frac{dC}{dt} = -k(C - K_s^{\text{PAH}} C_w) \quad (2)$$

Where  $k$  is desorption rate content of PAHs from soil ( $\text{h}^{-1}$ );  $C$  is PAHs concentration sorbed to soil (including bound to soil and sorbed surfactant in soil) ( $\text{g/g}$ );  $K_s^{\text{PAH}}$  is the distribution coefficient for PAHs to soil ( $\text{ml/g}$ );  $C_w$  is PAHs concentration in aqueous phase (including dissolved and bound to dissolved surfactant) ( $\text{g/ml}$ ). Combining Equations (1) and (2) gives:

$$\begin{aligned} \frac{\partial M}{\partial t} &= \int_0^{\infty} \frac{\partial C}{\partial t} f(k) dk = - \int_0^{\infty} k(C - K_s^{\text{PAH}} C_w) f(k) dk \\ &= - \int_0^{\infty} k C f(k) dk + (\alpha/\beta) K_s^{\text{PAH}} C_w \end{aligned} \quad (3)$$

Similar to the three-component system of soil-PAHs-extracellular polymer (Liu *et al.*, 2001), we assume that, in the  $\gamma$  model, the distribution coefficients for PAHs to surfactant is unchanged no

matter the surfactant is bounded to soil or dissolved, and the intrinsic binding constant for PAHs with soil is not altered with the presence of sorbed Tween80. We also assume that the distribution of surfactant between the aqueous phase and soil, and the binding of PAHs to Tween80 are at instantaneous equilibrium. The relevant distribution coefficients, e.g. the soil/water partition coefficient of PAHs and surfactants obtained from sorption experiments, and statistical parameters of  $\gamma$  distribution, e.g. the parameters  $\alpha$  and  $\beta$  obtained from the control experiment of PAHs desorption in the absence of surfactant were gained in independent experiments. Using these parameters, the kinetic model was employed to provide an independent prediction on PAHs release from soil to aqueous phase at two test surfactant concentrations.

## 2 Materials and methods

### 2.1 Reagent

99.9% PAHs mixture purchased from Sigma-Aldrich (U.S.A.) was used in this study. Physical and chemical properties of PAHs mixtures are presented in Table 1. 99.7% dichloromethane, 99.7% hexane and 99.8% Tween80 were purchased from Beijing Solvent Company. Selected properties of Tween80 are listed in Table 2. Dichloromethane and hexane were used after distillation purification.

### 2.2 Soil samples

Uncontaminated sub-layer soil sample was collected from Peking University campus. The air-dried soil was homogenized and screened with 10

Table 1 Selected properties of PAHs used for sorption and desorption experiments

PAHs	Abbreviation	Ring	Molecular formula	Molecular weight	S'	lgK <sub>ow</sub> **
Naphthalene	Nap	2	C <sub>10</sub> H <sub>8</sub>	128.18	31.5	3.37
Acenaphthylene	Any	3	C <sub>12</sub> H <sub>8</sub>	152.2	3.93	4
Acenaphthene	Ane	3	C <sub>12</sub> H <sub>10</sub>	154.2	3.93	3.92
Fluorene	Fle	3	C <sub>13</sub> H <sub>10</sub>	166.23	1.98	4.18
Phenanthrene	Phe	3	C <sub>14</sub> H <sub>10</sub>	178.24	1.15	4.57
Anthracene	Ant	3	C <sub>14</sub> H <sub>10</sub>	178.24	0.075	4.54
Fluoranthene	Fla	4	C <sub>16</sub> H <sub>10</sub>	202.26	0.206	5.22
Pyrene	Pyr	4	C <sub>16</sub> H <sub>10</sub>	202.26	0.132	5.18
Benz[a]anthracene	Baa	4	C <sub>18</sub> H <sub>12</sub>	228.3	0.009	5.91
Chrysene	Chr	4	C <sub>18</sub> H <sub>12</sub>	228.3	0.002	5.86
Benzo[b]fluoranthene	Bbf	5	C <sub>20</sub> H <sub>12</sub>	252.32	0.002	5.8
Benzo[k]fluoranthene	Bkf	5	C <sub>20</sub> H <sub>12</sub>	252.32	0.0008	6
Benzo[a]pyrene	Bap	5	C <sub>20</sub> H <sub>12</sub>	252.32	0.004	6.04
Indeno[1,2,3-cd]pyrene	Inp	6	C <sub>22</sub> H <sub>12</sub>	276.34	0.0005	6.5
Dibenz[a,h]anthracene	Daa	5	C <sub>22</sub> H <sub>14</sub>	278.36	0.0006	6.75
Benzo[ghi]perylene	Bgp	6	C <sub>22</sub> H <sub>12</sub>	276.34	0.0003	6.5

Notes: \*Water solubility (mg/L, 25°C); \*\* octane-water distribution coefficient; source: Mackay *et al.*, 1992

mesh sieve to remove large particles and debris. Organic carbon content (TOC-5000A analyzer, equipped with SSM-5000 solid injector) is 1.325%, and clay content (FRITSCH A22 granularity analyzer) is 0.5%.

To make artificially contaminated soil, PAHs mixture containing 16 compounds were added to the 100 g soil through a 3-ml hexane carrier. 50 ml deionized water were then added to the soil and agitated at 50 r/min for 24 h to well distribute PAHs and evaporate hexane. After the evaporation, the soil was air-dried and homogenized again and stored in a brown glass (4°C) for one week before use. Concentrations of PAH compounds were listed in Table 3.

Table 2 Selected properties of Tween80

Surfactant	Average molecular weight	HLB <sup>*</sup>	CMC, mg/L <sup>**</sup>
Tween80	1310	15.0	33—45

Notes: <sup>\*</sup> Hydrophilic-lipophile balance; <sup>\*\*</sup> critical micelle concentration (mg/L)

Table 3 Concentrations of PAH compounds in artificially contaminated soil

PAH compound	Concentration, $\mu\text{g/g}$	PAH compound	Concentration, $\mu\text{g/g}$
Nap	1.67	Baa	1.87
Any	1.98	Chr	1.92
Ane	1.73	Bbf	1.95
Fle	1.98	Bkf	1.95
Phe	1.89	Bap	1.87
Ant	1.96	Inp	1.98
Fla	1.98	Daa	1.96
Pyr	1.89	Bgp	1.93

### 2.3 Experiment

The distribution coefficients for PAHs to soil and Tween80 to soil were measured in batch experiments. 25 ml PAHs mixtures solution contains 0.01 mol/ml CaCl<sub>2</sub> and 200  $\mu\text{g/ml}$  HgCl<sub>2</sub> (PAHs concentration ranging from 0 to 0.8  $\mu\text{g/ml}$ ) was added into a 50-ml glass with 10 g uncontaminated air-dried soil inside. The glasses were covered with glass plug and rapped with aluminum foil to keep the PAHs from evaporation and light-degradation. Then these glasses were put in an air batch vibration implement (20 r/min) (HZQ-C). The temperature was kept on (25  $\pm$  1)°C. After 24 h equilibration, the samples were separated by centrifugation at 3000 r/min for 5 min. The deposits were collected for further extraction and analyses. The sorption experiment condition for Tween80 was kept unchanged as PAHs sorption experiment, except that 50 ml Tween80 solution was used instead of PAHs mixtures (Tween80 concentration ranging from 0 to 30 mg/ml). After 24 h equilibration, the aqueous phase was separated by

centrifugation at 3000 r/min for 5 min, the supernatant was filtered using 0.45  $\mu\text{m}$  glass fibred film and leachate was withdrawn. The dissolved Tween80 was determined by UV-visible light spectrophotometer at 752 nm (HACH DR/4000U Spectrophotometer, Japan).

The effect of Tween80 on PAHs sorption in soil was investigated under the same experimental condition, while Tween80 (with concentration ranging from 0 to 75 mg/L) was added into the PAHs mixture solutions. After equilibration, aqueous and deposit phases were analyzed respectively.

The effect of Tween80 on PAHs desorption from artificially contaminated soil was studied under the same experimental condition, while 10 g artificially contaminated soil was used, instead of the uncontaminated air-dried soil. Two Tween80 concentrations (27 and 65 mg/L) were selected.

PAHs in leachate were extracted with liquid-liquid extraction method. Leachate was filtered through 0.45  $\mu\text{m}$  glass fiber filter, and then 10 ml H<sub>2</sub>SO<sub>4</sub> was added to destroy emulsion, and a few drops of ethanol were added to eliminate foam. Dichloromethane and hexane were used for extraction. The organic phase was collected and determined using GC/MS. PAHs in soil was extracted with accelerated solvent extraction (ASE) method. The extracts were analyzed for PAHs using Agilent 6890 GC coupled with Agilent 5973 mass spectrometer and 7683 auto-sampler (Agilent Technology). The extraction and analysis were carried out using a procedure (including quality control) developed by Tao and Cui (2002).

## 3 Results and discussion

### 3.1 PAHs sorption on soil

The sorption isotherm equation for low concentration PAHs could be fitted by linear equation. The sorption coefficient  $K_s^{\text{PAH}}$  which reflects PAHs sorption ability on soil can be presented as:

$$K_s^{\text{PAH}} = Q/C_e \quad (4)$$

where  $K_s^{\text{PAH}}$  is the PAHs-soil distribution coefficient (ml/g);  $Q$  is the sorptive quantity ( $\mu\text{g/g}$  soil);  $C_e$  is the concentration of PAHs in aqueous phase ( $\mu\text{g/ml}$ ).  $K_s^{\text{PAH}}$  calculated from sorption experiment is 47.41 ml/g.  $K_{\text{OC}}^{\text{PAH}}$  (the partition coefficient of PAHs in the organic fraction of the soil) is 3578.11 ml/g.

### 3.2 Tween80 sorption on soil

Experimental data for Tween80 sorption on soil is shown in Fig.1. Tween80 sorption isotherm is linear at the low concentration section, while the sorptive quantity decreased slightly with high Tween80 concentrations. The partition coefficient for Tween80 to soil can be given by:

$$q = K_s^{surf} C \quad (5)$$

where  $q$  is the sorptive quantity ( $\mu\text{g/g}$  soil);  $C$  is the concentration of Tween80 in aqueous phase ( $\mu\text{g/ml}$ );  $K_s^{PAH}$  is the partition coefficient for Tween80 to soil ( $\text{ml/g}$ ).  $K_s^{PAH}$  calculated from sorption experiment was  $9.43 \text{ ml/g}$ .  $K_{oc}^{surf}$  is  $711.70 \text{ ml/g}$ .

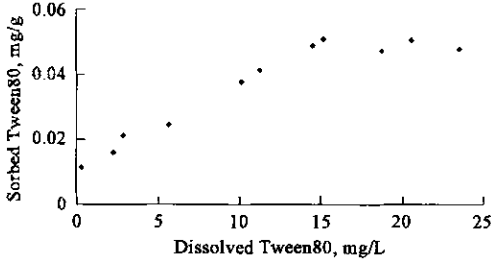


Fig.1 Experimental data for Tween80 sorption on soil

### 3.3 PAHs sorption on soil in the presence of Tween80

Sorption isotherm curves of PAHs on soil in the presence of Tween80 are showed in Fig.2. Low concentration Tween80 (below  $39 \text{ mg/L}$ ) enhances the sorption of PAHs on soil, while higher concentration Tween80 (higher than  $39 \text{ mg/L}$ ) hinders the sorption of PAHs on soil. PAHs sorption in soil-water-surfactant system can be illustrated by apparent sorption coefficient  $K_{surf}^{PAH}$ :

$$K_{surf}^{PAH} = K_s^{PAH} \frac{1 + f_{st} K_{st} / K_d}{1 + X_{mn} K_{mn} + X_{mc} K_{mc}} \quad (6)$$

where  $K_{surf}^{PAH}$  is the apparent sorption coefficient of PAHs;  $K_s^{PAH}$  is the sorption coefficient for PAHs to soil;  $K_{st}$  is the partition coefficient for PAHs to Tween80;  $f_{st}$  is the concentration of sorbed Tween80 ( $\text{mg/g}$ );  $X_{mn}$  and  $X_{mc}$  are concentrations of Tween80 in monomer and micelle in aqueous phase ( $\text{mg/ml}$ ), respectively (if  $X \leq \text{CMC}$ ,  $X_{mn} = X$ ,  $X_{mc} = 0$ ; if  $X > \text{CMC}$ ,  $X_{mn} = \text{CMC}$ ,  $X_{mc} = X - \text{CMC}$ );  $K_{mn}$  is the partition coefficient of PAHs between monomer and water, and  $K_{mc}$  is the partition coefficient of PAHs between micelle and water. The partition coefficient  $K_{surf}^{PAH}$  calculated from the experiment is  $681 \text{ ml/g}$ .

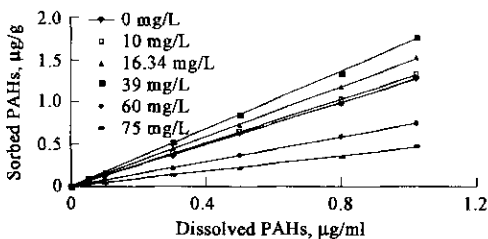


Fig.2 Sorption isotherm curve of PAHs on soil in the presence of Tween80 at different concentrations

### 3.4 Model simulation

The  $\gamma$  model has been applied to simulate the desorption of phenanthrene from soil in the presence of bacterial extracellular polymer (Liu *et al.*, 2001). In this paper, this model was used to illustrate the influence of Tween80 on PAHs desorption from soil and predict the release of PAHs from soil to aqueous phase at two surfactant concentrations.

The partition coefficients for PAHs to soil ( $K_s^{PAH}$ ,  $47.41 \text{ ml/g}$ ), PAHs to Tween80 ( $K_{surf}^{PAH}$ ,  $681 \text{ ml/g}$ ) and Tween80 to soil ( $K_s^{surf}$ ,  $9.43 \text{ ml/g}$ ) were obtained from batch experiments. With Equation (4),  $\alpha$  and  $\beta$  ( $\alpha = 0.023$ ,  $\beta = 14\text{h}$ ) were calculated through a simulation of PAHs desorption from soil in the absence of Tween80. These parameters obtained from batch experiments and model simulation were used in the  $\gamma$  model (Equation (4)) to simulate the desorption dynamics of PAHs (Fig.3).

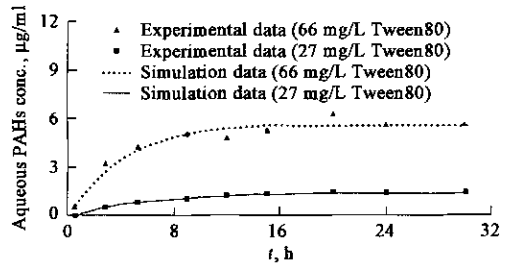


Fig.3 PAHs desorption dynamics and simulation with  $\gamma$  model in the presence of Tween80

Good fitness is observed from Fig.3 between the experimental data and model simulation at the Tween80 concentrations of  $27$  and  $66 \text{ mg/L}$ .  $F$  statistic was used to evaluate the fitness of the model. Results show that the model is significant at the level of  $\alpha = 0.001$  and  $f(2, 9)$  since the two  $F$  values obtained ( $32.1$  and  $29.4$  at the Tween80 concentrations of  $27$  and  $66 \text{ mg/L}$ , respectively) are higher significantly than  $F_{0.001, [2, 9]}(16.4)$ .

From Fig.3, we can find that the simulation for Tween80 concentration of  $27 \text{ mg/L}$  is better than Tween80 concentration of  $66 \text{ mg/L}$ . This could be explained by the unstable sorption of high concentration Tween80 on soil. Previous research showed that surfactant may increase the diffusivity of PAHs in the soil matrix by changing the physical properties of the medium. Surfactant molecules may penetrate into the organic matrix of the soil, swell the medium, and create more space for PAHs diffusion (Yeom *et al.*, 1996).

In summary, PAHs sorption/desorption on/from soil in the presence of Tween80 include at least several steps: (1) competition of PAHs and Tween80 for active sorption sites in soils; (2) PAHs sorbed to

active sorption sites of soil and Tween80 micelles; (3) the swell of soil organic matrix profits to PAHs transport; (4) sorbed Tween80 enhances PAHs sorption; (5) the increase of Tween80 concentration in aqueous phase lowers the water solubility of PAHs after Tween80 reach saturated sorption. (1), (2) and (3) promote PAHs desorption from soil, while (4) and (5) enhance PAHs sorption on soil.

#### 4 Conclusions

The presence of Tween80 could change the sorption/desorption behaviors of PAHs on soil. Adsorbed surfactant enhances the sorption amount of PAHs while the dissolved surfactant increases PAHs apparent solubility. These two processes influence the PAHs sorption coefficient in soil-water system, which can be depicted by apparent sorption coefficient. The  $\gamma$  model provides a satisfactory independent prediction on PAHs release from soil to aqueous phase at two test surfactant concentrations.

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