



Distribution characteristics of phenanthrene in the water, suspended particles and sediments from Yangtze River under hydrodynamic conditions

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ABSTRACT

The effects of aquatic sediment concentrations, grain size distribution and hydrodynamic conditions on sorption behavior of phenanthrene (PHE) on sediments collected from Yangtze River (Wuhan catchment) were investigated. The results showed that the sorption behavior of PHE was mainly affected by the organic carbon in different phases, i.e. organic carbon contents (f_{oc}) (w/w, organic carbon/dry weight sediment) in the sediments and dissolved organic carbon (DOC) in liquid phase. In this study, sediments were subjected to artificial resuspension under turbulent diffusion coefficients being 24.6, 29.5 and 46.2 cm² s⁻¹ corresponded to 0.4, 0.3 and 0.2 s cycle⁻¹ of the perforated grids, respectively, which were driven by variable speed motor with 150, 180 and 280 rotation per minute (rpm). The suspended particle concentration increased from 1.01 to 6.70 g L⁻¹ as the hydrodynamic strength increased from 150 to 280 rpm, whereas PHE concentration in liquid phase decreased from 0.56 to 0.34 μg mL⁻¹. The amount of DOC was supposed to play an important role in the partition of PHE under hydrodynamic conditions. Moreover, a sorption dynamic model was developed based on the linear isotherm expression and law of conservation of mass. The model was validated by PHE sorption behavior acquired with three different hydrodynamic conditions and the predicted values displayed satisfying accordance with experimental data.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are widespread pollutants with high toxicity and some of them are endocrine disrupting chemicals (EDCs) [1–4]. Once released into the aquatic environment, PAHs tend to bind to particles and subsequently deposit in the sediments due to their high hydrophobicity. Consequently, sediments act as a natural repository for these hydrophobic organic contaminants [5,6]. Unfortunately, under favorable conditions, PAHs will be released from the sediments, which results in a second contamination [7]. As the important source and sink of PAHs, sediment plays an important role in the transport and fate of these chemicals in the aquatic environment. Therefore, there has been considerable interest in the transport and fate of these contaminants in the water [3–5,8].

Investigation on the influence of sediment characteristics, pollutant properties and physicochemical properties of freshwater on

sorption or desorption behavior of organic pollutants has been an issue for several years [9–13]. In the aquatic environment, hydrodynamic condition is a ubiquitous natural process and will induce the sedimentation of suspended particle and the sediment resuspension. A number of laboratory studies indicated that the hydrodynamic conditions could result in the release of organic contaminants into overlying water and could act as a potential pollution source to water column [7,12,13]. For example, Latimer et al. [12] found that variations of dissolved PAHs in the overlying water were not uniform because of different sediment compositions and complicated phase changes. Recently, Feng et al. [7] reported that the distribution of PAHs in the aquatic environment was significantly affected by aquatic particulates or aggregates of numerous complicated organic materials, and the desorption from the entrained particles was the primary source of dissolved PAHs in the liquid phase. Moreover, the composition of sediments is the major factor in determining the amount of released PAHs, more than the level of resuspension energy [9]. So far, the effects of hydrodynamic conditions on the remobilization and fate of PAHs are not fully understood. Especially, little was known about the effects of different hydrodynamic conditions on the distribution characteristics of PAHs in the water, suspended particles and sediments.

The body of literature investigating the absorptive linear partition of hydrophobic organic compounds into natural organic matter is extensive, owing largely to the pioneering work by Chiou,

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Karickhoff, and others [14–16]. These researchers correlated carbon-normalized or organic matter normalized partition coefficients (K_{oc} or K_{om}) with aqueous solubility (S_w) or octanol–water partition coefficient (K_{ow}) on the basis of theory derived from linear-solvation-energy relations (LSERs) [17]. Despite the widespread successes of the organic carbon contents (f_{oc})– K_{oc} paradigm, there are a number of important environmental scenarios (such as the sediment properties and hydrodynamic conditions) in which this paradigm is inadequate [18].

The objective of this study was to investigate the effect of hydrodynamic conditions on the sorption behavior of phenanthrene (PHE) in the sediments from the Yangtze River (Wuhan catchment) using a designed syntonetic turbulence-simulation device. The influences of sediment concentrations and grain size on PHE distribution in static experiments were also studied. Moreover, based on the linear isotherm expression and law of conservation of mass, a sorption dynamic model was developed from the view of the hydrodynamic conditions.

2. Materials and methods

2.1. Chemicals

Standard PHE was purchased from the National Research Center for Certified Reference Materials of China. The PHE standard solutions (1.25 g L^{-1}) were prepared by diluting appropriate volumes of the standard PHE with methanol solution (HPLC grade, Merck, Germany). Other solvents and reagents included acetone (HPLC grade, Tedia, USA), dichloromethane, n-hexane (HPLC grade, Dikma, USA), and anhydrous sodium sulfate (analytical grade, Beijing Chemical Reagent Co., China). Anhydrous sodium sulfate was baked in a furnace oven at 650°C for 6 h, kept in a sealed desiccator prior to use. Additionally, the background solution was prepared by CaCl_2 electrolyte, which was composed of 1 mM CaCl_2 , and some of 100 mg L^{-1} NaN_3 as bacterial inhibitor, in order to make the aquatic environment condition close to the natural freshwater. All solutions were prepared using distilled water that was passed through a MilliQ UV-plus water purification system (Millipore, Bedford, MA, USA) with a specific resistivity of $18.2 \text{ M}\Omega \text{ cm}$.

2.2. Sample collection

Sediment samples were collected using a Van Veen stainless steel grab sampler (Eijkelamp, Netherlands) from Yangtze River (Wuhan catchment) in July 2005. The top 10 cm sections from five cores (8 cm in diameter) were combined and passed through a $500\text{-}\mu\text{m}$ mesh sieve to remove macrofauna. The sediment, which was black, fine-grained, muddy sand, was allowed to settle for 7 days at 4°C prior to analysis and use in the microcosms.

2.3. Sorption experiments

Kinetics of PHE sorption onto the Yangtze River sediments was carried out in a series of conical flasks to determine the time required to reach the fast adsorption equilibrium. PHE ($20 \mu\text{L}$) dissolved in ethanol was added to each empty flasks for a total mass of $2.5 \mu\text{g}$ PHE per flask. After the ethanol evaporated, the equivalent sediment was added to each flask (2.0 g dry weight) along with 25 mL of CaCl_2 electrolyte. Flasks were sealed, and shaken at 150 rotation per minute (rpm) in darkness at $20 \pm 1^\circ\text{C}$ for various periods of time ranging from 0.6 to 12 h. At the end of each agitation period, aliquots of 10.0 mL from duplicate flasks were collected, and centrifuged at 4000 rpm for 20 min. The supernatant was moved to a clean centrifuge tube, and the PHE concentration in

the supernatant was analyzed with high-performance liquid chromatography (HPLC). In order to select an isotherm with the best validity and stability, the relative errors of partition coefficient (RE) and the coefficient of variation (CV) were used to test the stability of isotherms.

A series of mass of sediments with different grain size were added to conical flasks containing 0.8 mL L^{-1} of PHE CaCl_2 electrolyte. After the initial sorption phase was obtained, the PHE concentration was determined to exhibit the effects of sediment concentration and grain size on the sorption behavior of PHE.

2.4. Analytical methods

Prior to testing, the sediments were size fractionated using sequential sieve sizes to obtain the following fractions: $>200 \mu\text{m}$, $125\text{--}200 \mu\text{m}$, $63\text{--}125 \mu\text{m}$, $25\text{--}63 \mu\text{m}$, and $<25 \mu\text{m}$. The grain size analysis of the sediment samples used for classification test was performed by X-ray sedimentography using a SALD-3001 Particle Analyzer ($0.269\text{--}2000 \mu\text{m}$, RSD $<3\%$) (Shimadzu, Japan). The total organic carbon (TOC) contents of sediment samples were determined using a Liqui TOC analyzer (Elementar, Germany) after treatment with 2N HCl to remove carbonate-containing minerals. The values of dissolved organic carbon (DOC) in liquid phase were also measured by Liqui TOC analyzer (Elementar, Germany) by detecting the organic carbon contents in solution samples.

The extraction procedure was a modification of the method described elsewhere [7]. Briefly, the sediments were extracted three times with 30 mL hexane/acetone mixture (1:1, v/v) for 1.0 h in an ultrasonic bath (KQ-502B, Kunshan Ultrasonic Instruments, China). The extract was partitioned against solvent-extracted, deionized water and reduced in volume to about 1.0 mL in a rotary evaporator (RV 05 basic, IKA, Germany). Cleanup of the extracts was performed using an open glass column containing 2 g of 5% deactivated silica gel ($100\text{--}200$ mesh, Qingdao Ocean Chemical Plant, China). Water samples were extracted with dichloromethane and the aqueous extracts were then reduced to a volume of about 2 mL , re-extracted with methanol, and again concentrated to 1.0 mL for analysis. Following the extraction from sediments or water samples, quantification analysis of PHE was performed on a Waters 1525 HPLC system (Waters, USA) equipped with fluorescence detector (excitation wavelengths were 292 nm , emission wavelength were 366 nm for PHE). The injector was a Rheodyne 7725 high-pressure manual injector valve with a $20\text{-}\mu\text{L}$ injection loop. Separation was carried out using a ChromSep guard column ($10 \text{ mm} \times 4.6 \text{ mm}$, grain size $5 \mu\text{m}$, Varian, USA) followed by a ChromSep C_{18} Column ($250 \text{ mm} \times 4.6 \text{ mm}$, grain size $5 \mu\text{m}$, Varian, USA). The elution was conducted with 95% acetonitrile/water (v/v). PHE quantification was based on an external standard (NIST SRM1647d). Extraction solvents were regularly checked for blank levels.

2.5. Turbulence-simulation device

In the natural aquatic environment, the adsorption of organic matter on sediments or suspended particles is carried out in the turbulent water rather than the still water. The improved syntonetic turbulence-simulation device (ISTSD) was used to simulate the natural aquatic environment, and perform hydrodynamic experiments to estimate the effect of hydrodynamic condition on the adsorption of PHE on resuspended particles. As shown in Fig. 1, the improved ISTSD consists of a vertical acrylic cylinder ($\Phi = 25 \text{ cm}$, $h = 60 \text{ cm}$) and a set of oscillating vibratory grids driven by a variable speed motor. The positions of sample ports on the cylinder are 5 cm intervals from bottom to top.

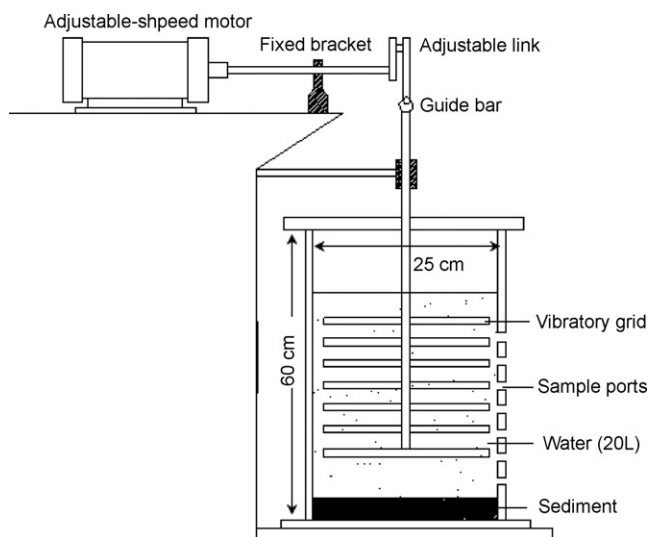


Fig. 1. Schematic diagram of improved turbulence-simulation device.

Prior to testing, the calibration was conducted empirically in order to produce the similar resuspension concentrations for the same sediment under identical environmental conditions. In this study, sediments were subjected to artificial resuspension under turbulent diffusion coefficients being 24.6 , 29.5 and $46.2 \text{ cm}^2 \text{ s}^{-1}$ corresponded to 0.4 , 0.3 and 0.2 s cycle^{-1} of the perforated grids, respectively, which were driven by variable speed motor with 150 , 180 and 280 rpm . The resuspended particle concentrations were under those conditions were 0.42 , 1.14 and 6.25 g L^{-1} , respectively, in which 0.42 and 1.14 g L^{-1} were typical and 6.25 g L^{-1} was occasional of Yangtze River sediment concentrations [19].

For dynamic testing, about 20 L deionized water and 200 g sediments, passed through a $200\text{-}\mu\text{m}$ mesh sieve, were added to the cylinder and kept static for 48 h . The cylinder was positioned to the ISTSD unit and the perforated grid was lowered into the water. During resuspension, the grid was oscillated with vertical amplitude of 3.2 cm and operated between 5.0 and 8.2 cm from the top of the sediment surface. After a 10 h process of resonant turbulence, duplicate samples were collected from given sample ports on the device. After being centrifuged at 4000 rpm for 20 min , the sediment was air-dried, and then the mass of sediments, distributions of grain size and the content of total organic carbon were determined.

In order to evaluate the distribution of PHE under different hydrodynamic conditions, the same experiment was performed except a given quantity of PHE solution was added into the equipment after static phase of 48 h . In addition, the PHE concentration in superior-clear liquid was analyzed. To avoid the volatilization of the PHE in sediments, the analysis of PHE in sediments was conducted after the sediments were freeze-dried.

All collected resuspended sediment samples were evaluated for the PAHs, organic carbon and particle size. Composite filtrate samples were evaluated for dissolved phase organic contaminants. Organic contaminants, organic carbon and particle size were also measured for bulk sediments.

2.6. Quality control

All analytical operations were conducted under strict quality control guidelines. Before the extraction and analysis program, the recovery study of PHE was undertaken to demonstrate the availability of this method. The sediment samples and water samples were spiked with the PHE standard solution, then extracted, purified, and analyzed in the same way as the samples. The six parallel

experiments indicated that recoveries for PHE were $89\text{--}101\%$ for the sediment samples and $85\text{--}98\%$ for water samples. PHE content in original sediment samples used in our experiments was about $12.2 \mu\text{g kg}^{-1}$, which was so less to be omitted in the adsorption experiments. The relative standard deviations (RSDs) of the mass balance determination experiments in water, suspended, and sediment phase were less than 10% . The RSDs of PHE were all below 4.81% for the sediment samples and below 4.77% for water samples. Each experiment was accompanied by two blank experiments. Under analytical conditions, detection limits of PHE were 0.14 and $0.96 \mu\text{g kg}^{-1}$ (dry weight) for water and sediment samples, respectively. The retentive ratio of $98.7 \pm 1.5\%$ showed that the PHE loss in the sorption experiments was well controlled and the experimental designs performed as expected.

3. Results and discussion

3.1. Sorption isotherms

The adsorption experiments for sediments including original sediments and different size sediments demonstrated that a quasi-steady state (initial sorption phase) was achieved in 6 h (shown in Fig. 2), indicating that initial sorption takes place rapidly. Previous studies showed that sorption and desorption included two phase: the fast (initial) phase and the slow (final) equilibrium. Usually, the initial phase could reach a quasi-steady state in a short time and final equilibrium of all sorption sites could require months to years to fill the slow and very slowly desorbing sites [20–22]. Unfortunately, the geochemical equilibrium for suspended particles, sediments and water in dynamic systems was not achieved in our experimental conditions. In this study, the sorption experiments were conducted for 8 h ($>6 \text{ h}$) to further ensure the quasi-steady state. To investigate the sorption behavior of PHE on sediments, three isotherms (Linear, Freundlich and Langmuir sorption isotherms) were tested to describe the sorption model, i.e. Linear: $q = KC_e$; Freundlich: $q = K_F C_e^n$; Langmuir: $q = a C_e / (K_L + C_e)$; in which K_d , K_F , and K_L are the partition coefficients, C_e stands for the equilibrium concentration in liquid phase, n ($n \neq 1$) and a are constants. In addition, the dissolved organic carbon was also measured as the DOC commonly plays an important role in sorption procedures.

According to the Linear, Freundlich, and Langmuir sorption models described above, the q values versus C_e values were regressed to ascertain the relationship between the adsorption mass of PHE and equilibrium concentration. The results were statistically analyzed with the linear and nonlinear regression model procedure, respectively, using SPSS software (SPSS 9.0). The PHE-fitted sorption parameters (i.e., K , K_F , K_L , a and n) and their standard deviations (S.D.) were determined using SPSS software. The results showed

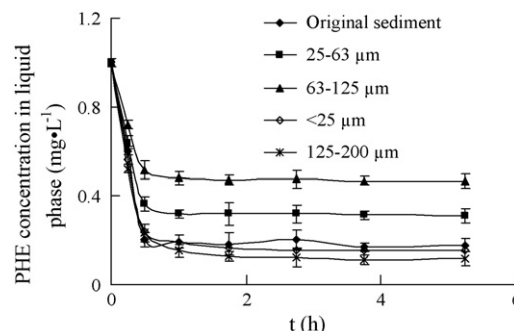


Fig. 2. The adsorption kinetic curves of PHE on different size sediments.

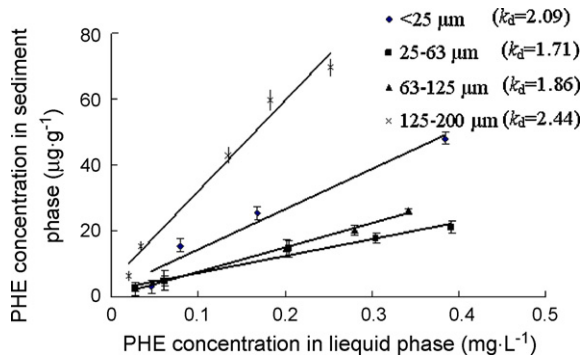


Fig. 3. The isotherm of PHE on the sediments with different grain sizes.

that the correlation was significant ($R^2 = 0.946\text{--}0.964$) for all the three isotherms. However, the largest RE (%) and CV (%) values of Linear adsorption isotherm (3.28 and 2.56) were lower than those of Freundlich isotherm (83.3 and 33.32) and Langmuir isotherm (104.30 and 41.72). This suggested that Linear adsorption isotherm was the optimal equation to interpret the sorption behavior of PHE in the sediments, which was consistent with the previous studies and it is indeed applicable to numerous important environmental scenarios [23]. Moreover, considering the DOC value of $115.1 \mu\text{g mL}^{-1}$ in the sorption experiment, the partitioning data was 6.76 times larger than the apparent partition coefficient, which was consistent with the result coming from Gauthier et al. [24]. Thus, the DOC is supposed to affect the partition data of PHE in the sediment–water system.

3.2. Effects of sediment concentrations and grain sizes

To investigate the isotherm parameters of PHE in aquatic environment, static experiments including effects of sediment concentration and grain size on isotherm were carried out. The partitioning of PHE between solid and liquid phases is commonly portrayed using a distribution coefficient (K_d) as the following equation

$$K_d = \frac{C_p}{C_d} \quad (1)$$

where C_p is the concentration of PHE in the particle phase, $\mu\text{g g}^{-1}$; C_d stands for the concentration in dissolved phase, $\mu\text{g mL}^{-1}$.

In the current study, distribution coefficients between solid and liquid phase for PHE were calculated. The results are plotted in Fig. 3. It can be calculated from this study that $\log K_d$ values were 2.44, 2.09, 1.86 and 1.71 for 125–200 μm , <25 μm , 63–125 μm and 25–63 μm particle size sediments, respectively. Moreover, f_{oc} (w/w, organic carbon/dry weight sediment) values in the sediments with different grain sizes were 2.64% ($\Phi = 125\text{--}200 \mu\text{m}$), 1.33% ($\Phi < 25 \mu\text{m}$), 0.81% ($\Phi = 63\text{--}125 \mu\text{m}$), and 0.24% ($\Phi = 25\text{--}63 \mu\text{m}$), respectively. The results showed that the K_d values decreased with the decreasing f_{oc} values, implying that the sorption of PHE was affected by f_{oc} values of the sediments. In order to sort out differences in binding mechanism for each grain size sediment, the partition coefficient data was normalized to the organic carbon content ($K_d = K_{oc}f_{oc}$). K_{oc} values for different grain-size sediments were found to be 4.02 ($\Phi = 125\text{--}200 \mu\text{m}$), 3.97 ($\Phi < 25 \mu\text{m}$), 3.95 ($\Phi = 63\text{--}125 \mu\text{m}$), and 4.33 ($\Phi = 25\text{--}63 \mu\text{m}$), respectively. It can be seen that most of the variation in partitioning among particle sizes disappears except for the 63–125 μm particle size sediment, implying that there could be another binding mechanism for very fine sediment particles. The similar K_{oc} values for different grain-size sediments ascertain that the sorption of PHE on sediments is mainly

Table 1

The sediment concentrations at various heights under different dynamic conditions (g L^{-1}).

Height (cm)	150 rpm	180 rpm	280 rpm
30	0.90 ± 0.05	2.57 ± 0.53	6.11 ± 0.95
25	0.90 ± 0.08	2.70 ± 0.92	6.65 ± 1.28
20	1.10 ± 0.12	2.94 ± 0.72	6.70 ± 1.10
15	1.02 ± 0.05	3.05 ± 1.32	6.72 ± 0.51
10	1.08 ± 0.09	3.20 ± 1.51	7.41 ± 0.23
5	1.12 ± 0.11	3.41 ± 1.03	7.19 ± 1.2

due to partition, and the effect of grain size on isotherm parameters is caused by f_{oc} values. The conclusions drawn from this study is consistent with previous reports by other researchers [25,26], who found that the solute sorption was mainly affected by partition into the sediment organic carbon, and the sorption coefficients normalized to the sediment organic carbon content.

The experiment exhibiting the effect of sediment concentration on PHE adsorption behavior indicated that K_d values varied from 129.7 to 117.3 with the sediment concentration ranging from 1 to 40 g L^{-1} . The result demonstrates that the sediment concentration exhibits a certain effect on K_d value, and the sorption coefficient decreases slightly with the increasing sediment concentration. The f_{oc} in the samples with different sediment concentration were consistent, however, the DOC varied among them. The correlation between the sorption coefficient and DOC for different sediment concentrations was then calculated. The result of the correlation is $K_d = -0.153\text{DOC} + 141.76$ ($R^2 = 0.972$), suggesting that the sorption coefficient was affected by the sediment concentration due to the amount of DOC in liquid phase in the sediment–water system.

3.3. Dynamic experiments

Generally, influence of hydrodynamic conditions on the sorption behavior of organic pollutants is mainly caused by two factors: the change of sediment concentrations and grain sizes resuspended by the change of aquatic turbulent conditions. In order to simulate the physical condition of the Yangtze River, the hydrodynamic condition was designed according to the sediment concentrations and median diameters of sediments in the key hydrology stations of Yangtze River. Data collected from the Yangtze River Water Resources Committee [19] in different hydrology stations along Yangtze River showed that the average sediment contents per annum were between 0.49 and 1.76 g L^{-1} , the maximal sediment contents per annum ranged from 0.70 to 2.9 g L^{-1} , and the minimal sediment contents per annum ranged from 0.26 to 1.06 g L^{-1} . The average median diameters per annum were between 17 and 31 μm , the maximal median diameters per annum varied from 39 to 63 μm , and the minimal median diameters per annum varied from 6 to 13 μm . For the simulation device, the sediment concentrations and grain size distributions at various heights under experimental conditions are presented in Tables 1 and 2, respectively. It can be seen that the sediment concentrations and median diameters of sediments under designed dynamic conditions produced by the improved syntonc turbulence–simulation device can simulate the field dynamic conditions of Yangtze River to some extent. It can be

Table 2

Median diameters (μm) at various heights under different hydrodynamic conditions.

Rotate speed (rpm)	30 cm	25 cm	20 cm	15 cm	10 cm	5 cm
150	5.10	5.13	5.10	5.11	5.12	5.25
180	8.99	9.03	9.43	9.42	9.52	9.52
280	16.99	17.15	17.20	17.35	17.28	17.64

Table 3

Distributions of phenanthrene in three phases and organic carbon in liquid and suspending phase under different hydrodynamic conditions.

Rotate speed (rpm)	Liquid phase ($\mu\text{g mL}^{-1}$)	DOC ($\mu\text{g mL}^{-1}$)	Suspending phase ($\mu\text{g g}^{-1}$)	f_{oc} (%)	Sediment phase ($\mu\text{g g}^{-1}$)
150	3.22	365.7	0.41	1.38	1.37
180	2.95	317.9	1.06	1.18	0.99
280	2.20	293.5	1.71	1.19	1.10

seen from Tables 1 and 2 that the relationship between the heights and concentration/median diameters is not significant under the same hydrodynamic conditions, indicating a thoroughly turbulence in the apparatus. Thus, the sediment distribution was found to be relatively homogenous under the scale of 30 cm height from the top of the sediment surface. However, with the increase of turbulent diffusion coefficient (namely, rotate speed), the sediment concentrations and the median diameters increased dramatically. This phenomenon indicates that the hydrodynamic condition plays an important role in the distribution of sediment concentration and sediment grain size in the aquatic environment. The distribution of PHE in three phases (liquid phase, suspended phase and sediment phase) was investigated (see Table 3). The results showed that the sediment concentration increased from 1.01 to 6.70 g L^{-1} with the hydrodynamic strength from 150 to 280 rpm, whereas the PHE concentration in liquid phase decreased from 0.56 to 0.34 $\mu\text{g mL}^{-1}$. Moreover, the results of the hydrodynamic experiments also indicated that the content of PHE in liquid phase decreased with the increase of turbulent diffusion coefficient. Contrarily, the content of PHE in suspended phase increased with the increase of turbulent diffusion coefficient and the PHE content in sediment phase was consistent basically. As shown in the static experiments, organic carbon (OC) is apparently the dominant factor affecting the distribution of PHE in the sediment–water system. Thus, the amount of OC in the hydrodynamic experiments was also investigated (see Table 3). It can be seen from Table 3 that as the turbulent conditions improved, the amount of DOC in the liquid phase decreased obviously whereas the values of f_{oc} in suspended particles was relatively consistent. PHE concentrations in three phases had no relationship with f_{oc} , but were relevant with the DOC values. The result showed that in the hydrodynamic experiments, the DOC played an important role in the partition procedure of PHE. The amount of DOC had a sufficiently negative relationship with PHE in suspending phase ($R^2 = 0.966$). It was supposed that the distribution of PHE among the three phases provides insight into processes that control the transport and fates of PHE under hydrodynamic conditions, and the amount of DOC acted as an important factor.

3.4. Dynamic model

The current results showed that the sorption behavior of PHE on sediments from Yangtze River was in accordance to linear sorption. Thus, the Linear isotherm could be used to describe the sorption behavior of PHE on sediments. Meanwhile, the blank experimental results displayed that the effect of both vessel wall sorption and PHE volatilization was less than 2.8%, which could be negligible. Combining the Linear isotherm expression and the law of conservation of mass, a sorption dynamic model was developed and expressed as follows:

$$K_i = \sum_{j=1}^4 K_{ij} p_{ij} = K_{OC} \sum_{j=1}^4 f_{OC,ij} p_{ij} = -0.153 \text{ DOC} + 141.76 \quad (2)$$

$$q_i = K_i C_e \quad (3)$$

$$Q = \sum_{i=1}^6 \frac{1}{6} v q_i s_i \quad (4)$$

$$H = C_e v \quad (5)$$

$$S = N_0 - Q - H \quad (6)$$

where K_{ij} is the sorption coefficient of sediment with j -given grain size in i layer; P_{ij} is the percent of j grain size in i layer; K_i stands for the sorption coefficient of sediment in i layer; q_i stands for the amount of PHE sorbed per unit mass of sediment ($\mu\text{g g}^{-1}$) in i layer ($\mu\text{g g}^{-1}$); C_e stands for equilibrium aqueous-phase PHE concentration ($\mu\text{g mL}^{-1}$); Q is the mass of PHE in suspended phase (μg); H is the mass of PHE in liquid phase (μg); S is the mass of PHE in sediment phase (μg); v stands for the water volume in turbulent device (mL); s stands for the sediment concentration (g L^{-1}); N_0 is the total mass of PHE in turbulent device (μg).

Both the correlation between linear sorption coefficient and sediment concentration in different heights and the distribution ratio of four kinds of grain sizes were predicted based on Eqs. (2)–(6). As the f_{oc} in suspended particles was relatively consistent in the hydrodynamic experiments, the relationship of DOC and K_i was used to estimate the sorption coefficient under hydrodynamic conditions. The distribution of PHE in liquid phase, suspended particles and sediment phase at every height was obtained and discussed

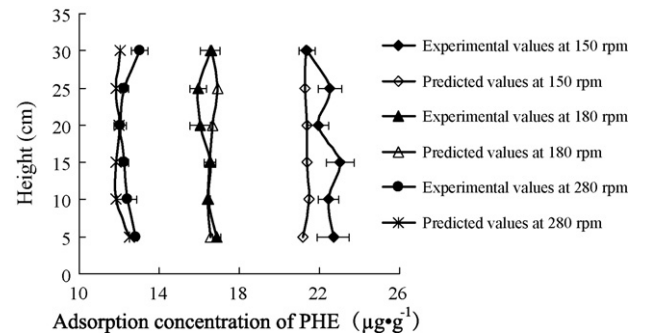


Fig. 4. The vertical distribution of PHE concentration on suspended particles under different hydrodynamic conditions.

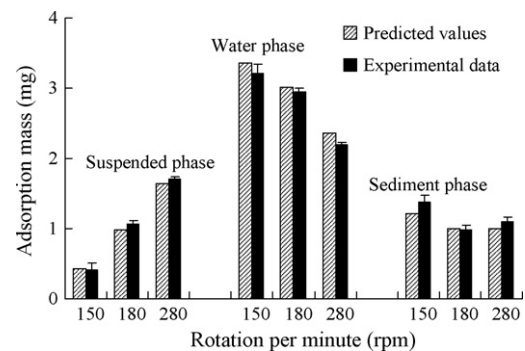


Fig. 5. The distribution of PHE in three phases under different hydrodynamic conditions.

based on the concentration of PHE in aqueous phase. Similarly, the distributions of PHE in three phases at different heights were calculated based on the sorption dynamic model under three different dynamic conditions, and the results were compared with the experimental data in Figs. 4 and 5. The referred data are available on request through the corresponding author. The results showed that the predicted values were in good agreement with the experimental data in three phases under different hydrodynamic conditions. This approved again that the increase of turbulent coefficient could lead to the decrease of the PHE concentration of PHE in the aqueous phase.

4. Conclusions

This research showed that linear isotherm could describe sorption behavior of PHE in sediments from Yangtze River. Both sediment concentrations and grain sizes have influences on sorption characteristics due to the effect of organic carbon. The findings from our research showed that the hydrodynamic conditions exhibited significant influence on the distribution of PHE among water, suspended particles and sediments. The hydrodynamic experiments indicated that the sediment concentration increased from 1.01 to 6.70 gL⁻¹ as the hydrodynamic strength increased from 150 to 280 rpm, whereas the PHE concentration in liquid phase decreased from 0.56 to 0.34 μg mL⁻¹. The results in this study can provide some useful information to better understand the transport and fate of PHE in the aquatic environment.

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