A multimedia fugacity model to estimate the fate and transport of polycyclic aromatic hydrocarbons (PAHs) in a largely urbanized area, Shanghai, China

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ABSTRACT
Increasing PAHs pollution is creating more complex urban pollution system. However, the availability of sufficient monitoring activities for PAHs in multicompartments and corresponding multi-interface migration processes is still not well understood. In this study, a Level III steady state fugacity model was validated to evaluate the detailed local variations, and mass fluxes of PAHs in various environmental compartments (i.e., air, soil, sediment, water, vegetation and organic film). This model was applied to a region of Shanghai in 2012 based on a large number of measured data and brings model predictions in 2020. The model results indicate that most of the simulated concentrations agreed with the observed values within one order of magnitude with a tendency of underestimation for vegetation. Direct emission is the main input pathway of PAHs entering the atmosphere, whereas advection is the main outward flow from Shanghai. Organic film was achieved the highest concentration of PAHs compared to other compartments up to 58.17 g/m³. The soil and sediment served as the greatest sinks of PAHs and have the longest retention time (2421.95–78642.09 h). Importantly, a decreasing trend of PAHs was observed in multimedia from 2012 to 2020 and the transfer flux from the air to vegetation to soil was the dominant pathways of BaP intermediate circulation processes. A sensitivity analysis showed that temperature was the most influential parameter,
1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a diverse class of toxic chemical substances that mainly originate from incomplete combustion, notably vehicle exhaust, domestic heating and industrial activities (Wang et al., 2012; Katsyvannis and Breivik, 2014; Bosch et al., 2015). In recent decades, growing attention has been devoted to discerning the PAHs contaminated dimension in the environment, especially in urban areas (Melymuk et al., 2014). Urban areas act as receptacles that carry high loading of human’s dense activities, which are the significant sources of geographical PAHs distribution (Diamond and Hodge, 2007; Csiszar et al., 2014; Ligaray et al., 2016). Urban areas also vary with relatively higher proportions of impervious surface coverage and this permits elevated PAHs movement during rain events that heterogeneously spread throughout various environmental media (Diamond et al., 2001; Kwamenka et al., 2007; Csiszar et al., 2012). Due to the expanded pollution of PAHs, a diversified and complicated phenomenon in dense urban areas ascribed to population growth and economic development (Melymuk et al., 2012; Ligaray et al., 2016). Hence, addressing the accurate urban-scale patterns of PAHs emissions and fate can be vital for understanding the urban ecosystem (Melymuk et al., 2013).

Different multimedia models have been configured and utilized to quantitatively explore the fate and transport of PAHs in the environment under steady or unsteady assumptions (Wang et al., 2013a,b; Jung et al., 2014; Huang and Stuart, 2014; Liu et al., 2014; Ke et al., 2017; Kim and Lee, 2017). Compared to the other multimedia models, such as Berkeley-Trent (BETR) and Sino Evaluative Simplebox-MAMI Model (SESAMe), which were shown good performance for assessing on a global-, national-, and regional-scale. However, the fugacity model approach is one of the useful tool for compartmentalizing the movement process of PAHs associated with multiple phases and environmental media, which specific to a complex urban system by including films that cover on impervious surfaces which are essential for urban PAHs fate (Tao et al., 2003; Wang et al., 2013a,b; Liu et al., 2014). Notably, Diamond et al. (2001) and Domínguezmorueco et al. (2016) explicitly considered the organic films that become coated onto impervious surfaces in the development of the model and how these films are addressed to trace PAHs in various bulk media in dense urban areas. The different condition of growth and wash off of urban films regulates the movement of PAHs from impervious surfaces to the other media (Csiszar et al., 2012). However, most studies in China have shown only minor attempts to focus on the detailed consideration the role of urban films for mediating contaminant dynamics in a simulation (Tao et al., 2003; Wang et al., 2013a,b; Liu et al., 2014; Song et al., 2016). These predictions of the circulation of PAHs are limited to mass transfers and have not addressed the systematic urban-scale condition of PAHs transport. Optimization and validation of these models have also been impeded due to insufficient monitoring activities (Kim and Lee, 2017). Therefore, there is a research need for a combination of systematic field data and fugacity model simulation to elucidate the variation of PAHs in densely populated urban areas, where the strongly influenced anthropogenic environment is still not well understood (Melymuk et al., 2014; Bosch et al., 2015).

Shanghai is the largest industrial and commercial city in China and is the hub of economic activity, and home of millions of people, which suffers from heavy human-induced impacts and drastic increases of built-up areas (Feng et al., 2018). There is a blend of significant loading of PAHs from various sources in Shanghai (Wang et al., 2014). Severe PAHs contamination is expected to continue and show by spatiotemporal differences in strength and duration. This phenomenon has raised questions as to the potential impacts of PAHs on the environment and human health (Wang et al., 2016a, 2016b; Yang et al., 2017a; Bi et al., 2018). Nevertheless, previous studies were only limited to investigations of the behaviors of PAHs in a single media, such as air (Ma et al., 2013; Lv et al., 2015), soil (Wang et al., 2013a,b, 2014, 2015), sediment (Yang et al., 2017b; Bi et al., 2018), water (Liu et al., 2016; Bi et al., 2018), vegetation (Yang et al., 2017a), road dust (Liu et al., 2007; Zheng et al., 2016), and films (Yu et al., 2014). It is crucial to understand and point the hotspot of highly PAHs polluted areas and how the PAHs compounds transported to this wide range of urban environment and impacted by surrounding environment (Ligaray et al., 2016; Domínguezmorueco et al., 2016). Consequently, effective management should be a focus to alleviate PAHs pollutants and reduce the risk of human exposure.

The objective of this study are as follows: (1) to estimate the detailed local variations and mass fluxes of PAHs in various environmental media based on a Level III steady state fugacity model from a highly urbanized and densely populated area in Shanghai, China; (2) to validate the model results with systematic monitoring data on PAHs levels in different media along with a sensitivity and uncertainty analysis of the model parameters; (3) to compare the predicted PAHs concentration between 2012 and 2020 by using local emission data.

2. Materials and methods

2.1. Study area and PAHs monitoring data

Shanghai, which is located in eastern of China (120°52′ E – 122°12′ E, 30°40′ N – 31°53′ N, Fig. 1), has a typical subtropical humid monsoon climate with a mean annual temperature of 17.7 °C, annual rainfall of 1597 mm, and 1668 h annual hours of sunshine in 2016 (SMSB, 2017). With continuous and rapid urbanization (including a population explosion, urban sprawl and expansion of developed areas), this mixed use region, which is thickly settled with a total population of 24.19 million inhabitants in 2016, has endured teeming commercial, industrial activities and intensive land conversion campaigns (many productive areas have been converted to build-up areas) in cities (SMSB, 2017; Feng et al., 2018). A large proportion of PAHs emissions arise from local fossil fuel combustion from heating, transportation, and industry (Wang et al., 2014; Zheng et al., 2016; Bi et al., 2018). For instance, the average daily energy consumptions in Shanghai in 2016 was 32.56% for coal, 12.68% for electric power, 5.45% for gasoline, 5.11% for coke, 4.98% for kerosene, 4.95% for fuel oil, 4.79% for diesel oil, and 0.68% for natural gas; a total of 3.59 million vehicles also leads to traffic exhaust (SMSB, 2017). For this reason, the concentrations of USEPA priority PAHs in multiple matrices (including PM2.5, dry and wet deposition, soil, surface water, suspended particulate matter,
sediment, vegetation, road dust, rainfall runoff and organic film) were conducted by previous monitoring studies covering the entire area of Shanghai from our research group, and these data were used for the model validation (Wang et al., 2016a, 2016b; Yu et al., 2014; Liu et al., 2016; Zheng et al., 2016; Yang et al., 2017a). The detailed descriptions are provided in the supplementary information (Table S1). The PAHs include naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluoranthene (Fluo), phenanthrene (Phe), anthracene (Ant), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (InP), dibenz[a,h]anthracene (DahA), and benzo[ghi]perylene (BghiP).

2.2. PAHs inventory

Liu et al. (2015) estimated that emissions of $S_{16}$PAHs in Shanghai were 447.8 tons in 2012, which were recorded from proportion of the following: coking industry (34.66%), domestic coal combustion (21.34%), natural gas (15.62%), the refining industry (10.71%), fuel oil (8.31%), gasoline (5.25%), and diesel (3.63%) in descending order (Fig. S1). In addition, the emissions of $S_{16}$PAHs in 2020 was compiled and was approximately 356.57 tons using the fitting formula calculated from Gross Regional Product (GDP) and energy consumption data. Further details of the inventory calculations can be found in the support information (Tables S2–S4). This mass inventory was the principle model inputs (atmospheric emissions input, $E_A$) for predicting PAHs concentrations in multicompartments.

2.3. Model descriptions

The level III fugacity-based model was applied to depict the dynamics of PAHs in 6 compartments: air, water, sediment, soil, vegetation and film (that accumulates on the impervious surfaces) in Shanghai, China. Each bulk compartment contained sub-compartments, e.g., air included vapor and the PM phase and water consists of the dissolved phase and suspended particles. The details of the model were described in Diamond et al. (2001) and Priemer and Diamond (2002). The framework of the model is shown in Fig. 2. The intercompartmental interactions, e.g., PAHs flux exchange from the air to the film included wet and dry particle deposition, wet gas deposition (rain dissolution), and bidirectional gas absorption/volatilization. The air-soil transfer consists of deposition (dry, wet, and dry gaseous) and volatilization. Briefly, the circulation of PAHs was assumed to be in equilibrium, and PAHs flux exchanges between adjacent compartments were calculated separately using the fugacity approach. The model initialized the compartmental (soil, sediment, vegetation, and film) concentrations to be 0. The initial concentrations in the air and water compartments were adjusted to close to the measured values in Shanghai (Wang et al., 2016a; Liu et al., 2016). In the fugacity formulation, the fugacity for compartment $X$ ($f_X$, Pa), the fugacity capacity constant ($Z_X$, mol/m$^3$/C$^{15}$Pa), and the intercompartment transfer parameter ($D_X$, mol/Pa$^{1/2}$/h) are 3 key variables, which express the transport processes of PAHs among the multicompartments. The mass balance equations used for the PAHs model are shown in Table 1. The concentration of PAHs ($C$, mol/m$^3$) was calculated as follows:

$$C_X = Z_X \times f_X$$

2.4. Model parameterization for Shanghai

Model parameters, including environmental and physicochemical parameters and that were retrieved from the existing literature and databases (Wang et al., 2012; Xu et al., 2013; Liu et al., 2014),
which were used in the model to simulate the fate and transport of PAHs in Shanghai. In this study, a total surface area of 6340 km² was selected. The air compartments were defined in 1000 m according to previous studies (Zhu et al., 2015). The density of the total suspended particulate was generally an empirical value of 1.5E+15 mg/m³ (Paterson and Mackay, 1995). The water area was 6.19E+08 km² with an average depth of 1.67 m. A depth of 5 cm for the sediment underlying to the water was assumed (Bulletin of first water census and second water resource census of Shanghai, 2013). The vegetation was estimated to have an overall thickness of 2.00E-04 m with a leaf area index of 2.75 (Zhang et al., 2007). Other input variables are also summarized in Tables S6–S7.

2.5. Sensitivity analysis

A sensitivity analysis was performed to evaluate how each input parameter affects the model outcomes and to identify the most influential inputs (Zhu et al., 2014; Ligaray et al., 2016). The sensitivity coefficient (S) is calculated by the ratio of the change of outputs compared to that of the test parameters by considering its range as follows:

\[
S = \frac{[(Y_{1.01} - Y_{1.0})/Y_{1.0}]/[(X_{1.01} - X_{1.0})/X_{1.0}]}{
}
\]

where \(X_{1.01}\) indicates that the input parameter is increased by 1%, that is, the value of the parameter is 101% of its mean. \(Y_{1.01}\) represents the output of the model when the test parameter was increased by 1%.

The sensitivity coefficient illustrated the magnitude of the influence of the parameters. Table S6 in the supplementary section shows the list of parameters that were involved in the sensitivity testing for the validity of the PAHs model. According to the physicochemical properties of PAHs (Table S7), the 16 PAHs can be divided into two groups: low molecular weight (LMW) PAHs with 2–3 rings, and high molecular weight (HMW) PAHs with 4–6 rings. In this study, Nap, Phe, Chr, BaP and BghiP were selected as the representative compounds of PAHs for evaluation.

2.6. Uncertainty analysis

A Monte Carlo analysis can effectively reflect the range of the predicted value of the model and verify the stability of the stochastic factors based on the model (Kong et al., 2014). The Monte Carlo simulation was utilized to assess the uncertainty of the
predictions based on prior knowledge of the vital input parameters obtained from the results of the sensitivity analysis (Xu et al., 2013; Huang and Stuart, 2014; Ke et al., 2017). All of the parameters were assumed to follow a logarithmic normal distribution, except for the temperature. The simulation was repeatedly undertaken 3000 times using a built-in function of “normrnd” through the MATLAB 2014a software with new values randomly selected from within the range of the mean ± standard deviation. Furthermore, the maximum, minimum, median, 5th and 95th percentile of the modeled multimedia PAHs concentrations manifested differences compared with the observations for determining uncertainty.

3. Results and discussion

3.1. Model simulation in 2012

The model estimated concentrations are consistent with the measurements within one order of magnitude (less than 0.5 logarithmic units, Fig. 3). Among the 16 PAHs evaluated, the same PAHs component pattern was observed in the most compartments using the steady state version of the multimedia fugacity model, with predominant concentrations by HMW-PAHs (Fig. 4) except for air. LMW-PAHs are abundant in the gas-phase that remain in the air where they are considered lost by advection and photodegradation. The reason is similar to other studies (Choi et al., 2012; Melymuk et al., 2012; Domínguezmorueco et al., 2016). HMW-PAHs mostly tend to stay in particulate phase, which is in accord with previous studies in Shanghai and that are contributed by local controlled emission sources from coal combustion and traffic (Wang et al., 2016c; Li et al., 2016).

The model prediction showed that the highest PAHs concentrations occur in the film compartment (58.17 g/m³), which is followed by sediments (25.72 g/m³, Table S9). The reason for the remaining high concentrations was likely attributed to a huge specific surface area and increasing sorptive capacity associated with black carbon (Unger and Gustafsson, 2008; Csiszar et al., 2012). The high PAHs loadings adhere to urban films, which emphasizes that it might act as a transient receptor for the transmission process of pollutants (Yu et al., 2014). This tendency is in keeping with the results found in Tarragona County, Spain (Domínguezmorueco et al., 2016) and downtown Toronto, Canada (Priemer and Diamond, 2002).

Fig. 3. A comparison of PAHs concentrations between simulated and observed in the multicompartments in 2012.

Fig. 4. Modeled compositional profiles of PAHs in the multicompartments in 2012 (%).
Conversely, soil and sediment were the greatest sinks of PAHs in Shanghai, which accounted for 99.4% (mass of PAHs), and chemical persistence was higher in both compartments (2421.95–78642.09 h, Table S8). This corresponds with the previous reported results (Mackay et al., 1997; Diamond et al., 2001; Domínguezmorueco et al., 2016; Zhu et al., 2018). In this sense and mainly because of the large amounts of atmospheric advection output fluxes and a short half-life in the atmosphere, soil and sediment acted as the main reservoirs. Synchronously, soil and sediment also receive PAHs inputs from water and vegetation. In addition, the average degradation loss rate of \( \Sigma_{16} \)PAHs was estimated to be 1.25 mol/h in soil, 2.84 mol/h in sediment, it reflected that degradation was the dominant loss mechanism in both soil and sediment compartments (Fig. S3 and Fig. 6).

The modeled PAHs concentration in vegetation (89.62 ng/g) is lower than the measured values (290.6 ng/g, Yang et al., 2017a), and the underestimation is possible because the current study did not account for the process of the vegetation absorption of PAHs from soil. The estimates of PAHs burdens were much higher than those found in Tarragona County, Catalonia, Spain (Domínguezmorueco et al., 2016), which may be caused by variations in vegetation types and meteorological conditions from different geographical situations (Diamond et al., 2001). This ultimately affected the deposition budget of 16 priority PAHs pollutants listed by the USEPA (Zhang et al., 2015).

The modeled \( \Sigma_{16} \)PAHs water concentrations ranged between 1488.2 and 379,862.9 ng/m\(^3\) for PAHs compounds. The most PAHs loading inputs into water were from the impervious surfaces (0.53–12.27 mol/h) rather than atmospherically deposition (0.06–0.70 mol/h), except for NaP, Ace and Acy (Fig. S3). Shanghai has a subtropical monsoon climate characterized by abundant rainfall that dislodges PAHs to storm water from the surface area (SMWAB, 2016). The persistence of PAHs in water is only 180.91–538.01 h (Table S8) due to the densely covered and tidal Huangpu River network (Liu et al., 2016; Bi et al., 2018).

The model results showed that air flow is the main route of \( \Sigma_{16} \)PAHs loadings entering Shanghai, whereas the greatest loss was due to advection from the air (Fig. S3 and Fig. 6). The transfer fluxes of LMW-PAHs across the air-vegetation exchange made the highest contribution, followed by air-film interfaces, and the process of rain wash-off from film to water. In contrast, the diffusive transportation of HMW-PAHs consistently achieved more circulations across the film-water and vegetation-soil interaction.

**Fig. 5.** Comparison of the mass distribution of PAHs accumulated in various media between 2012 and 2020.

**Fig. 6.** Estimated rates of BaP movement and transformation around the multicompartments (left is 2012 and right shows 2020).
3.2. Predicted transfer fluxes of PAHs in 2020

The model was used to predict the $\Sigma_{16}$ PAHs emissions for 2020 by projecting the emissions and parameters of Shanghai in various compartments (Tables S6–S7). Modeled multicompartments concentrations were 1.99E-07 g/m$^3$ in air, 1.16E-03 g/m$^3$ for water, 0.45 g/m$^3$ for soil, 18.26 g/m$^3$ for sediment, 0.08 g/m$^3$ for vegetation, and 56.06 g/m$^3$ for film, and these values were lower than those simulated in 2012 (Table S9). Nevertheless, similar composition profiles of PAHs were assessed for 2020 and were dominated by HMW-PAHs (Fig. S4). The predicted accumulation of PAHs in sediment, and soil were 342.06 tons and 134.96 tons respectively, which were still the greatest sinks for PAHs. The estimated mass of PAHs showed a decreasing trend in multicompartments compared to those in 2012, especially in organic film (86.28%) followed by sediment (14.03%) (Fig. 5). This is related to a series of policies surrounding the displacement of traditional fuels with new cleaner energy resources, such as liquefied petroleum gas and natural gas, which were put forward by the government (SMDRC, 2006; 2011, 2017).

BaP is one of the most toxic PAHs due to its comparatively high carcinogenicity, which is an example used to address the detailed transfer fluxes among multimedia (Shrivastava et al., 2017). The rate of BaP emission inputs decreased and the mass accumulated in various compartments were diminished (Fig. 6). However, the change of the degradation rate of BaP was not obvious. The difference of the transfer fluxes between air and water was reduced with the increase of water area from 2012 to 2020, which showed that BaP was apt to being detained in water and then remained. The interaction of air-film and film-water were weakened with decreasing impervious surface coverage simultaneously. To a certain extent, this reduces the mobility of BaP in the system (Csizsar et al., 2012). The corresponding exchange of BaP from the air to vegetation to soil (inclusive of gas diffusion, wet gas deposition, wet and dry particle-phase deposition, canopy drip, wax erosion, and litterfall) was more prominent, which acted as the

![Fig. 7. The sensitivity coefficient for the parameters.](image-url)
Fig. 8. The Monte Carlo simulation results of Σ_{16}PAHs compared with measured values.
main intermedia transfer pathway. This is consistent with previous studies in the Bohai coastal region, China (Liu et al., 2014; Song et al., 2016).

3.3. Sensitivity analysis

The sensitivity of the parameters of PAHs were identified (Fig. 7). The first 22 sensitive parameters of PAHs species based on absolute value showed that both physicochemical parameters and environmental parameters made a difference in the degree of influence on the variation of PAHs. Temperature plays a pivotal role and was the most influential, especially for Phe. Modeled PAHs' concentrations were positively correlated with the atmospheric advection input rate (Q0) and showed a negative correlation with output rate (Q10). The results agree with those reported in the Bohai Rim, China (Liu et al., 2016; Song et al., 2016). However, most of the input parameters were sensitive to PAHs compounds. The input of atmospheric emissions (Ea) remarkably affected the concentrations of Nap, BaP and BghiP but did not affect Phe and Chr. In addition, the sensitivity analysis resulted in octanol-air partition coefficient, particle density and organic carbon fraction that have closely resembled the effects on the model outputs. As shown in Fig. 7 there is a more profound impact on the distribution of LMW-PAHs by these parameters than HMW-PAHs.

3.4. Uncertainty analysis

The Monte Carlo simulation was conducted to determine the uncertainty of the model (Fig. 8). The modeled concentrations of total dissolved PAHs in water mostly fell into the 5% and 95% percentile range, which showed that the model has in good stability. Nevertheless, in the suspended particulate matter and soil granular phase, the modeled LMW-PAHs concentrations had higher uncertainty than HMW-PAHs, which is an underestimate because of the ignorance of the potential sources and the variation of half-life in the soil (Liu et al., 2014, 2016). For the sediment particle phase, the model results tend to overestimate the distribution of PAHs especially for HMW-PAHs, which could be caused by regular silting of the river in Shanghai (Bi et al., 2018). In comparison with other compartments, modeled PAHs concentrations in vegetation still underestimated LMW-PAHs and overestimated HMW-PAHs, which did not consider the activities of volatile compounds and the complex metabolic processes of vegetation (Yang et al., 2017a). The modeled PAHs concentrations in the organic film considered a higher organic content and stronger absorbability so that the model outputs were at a higher level except for NaP. However the cumulative characteristics of pollutants in organic films that coats in imperious surface are not very significant regarding heavy precipitation in Shanghai (Yu et al., 2014).

4. Conclusions

A steady state Level III fugacity model was successfully performed to estimate the fate and transfer of PAHs in Shanghai, China, which is an area with multiple industrial and heavy human-induced impacts. The model generally performed well for 16 EPA priority PAHs as shown by the comparison between predictions and measurements. The results suggest that similar patterns of PAHs accumulated in multicompartments and were dominated by the HMW-PAHs. The organic film was the compartment that achieved the hotspots of PAHs (58.17 g/m² and 56.06 g/m², respectively) in both 2012 and 2020. Soils and sediments act as the greatest sinks for PAHs as a consequence of high inputs three transmission pathways include air, water and vegetation. The persistence time of PAHs in soils and sediments was estimated to range between 2421.95 and 78,642.09 h. In addition, the simulated PAHs concentrations in 2020 showed a decreasing trend by jointly accounting for energy consumption compared to 2012, which is attributed to clean energy alternatives. The sensitivity analysis highlighted that the temperature was the most profound parameter that affected critical processes in the model. The results of the Monte Carlo analyses suggested that modeled HMW-PAHs concentrations were overestimated in organic film and sediment, but LMW-PAHs were generally underpredicted in air and vegetation, which is helpful in capturing the approximate and possible ranges of predictions. This study provides a good scientific understanding of the environmental pollutants dynamics in urban areas for the land conservation and sustainable development.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2018.10.172.

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