

Dry deposition of polycyclic aromatic hydrocarbons and its influence on surface soil contamination in Tianjin, China

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ISCLT3 dispersion model was applied to calculate the flux of dry deposition of polycyclic aromatic hydrocarbons (PAHs) in Tianjin, China based on a high resolution emission inventory. Field observations of dry deposition at 22 sampling sites were used for model validation. The mean deposition flux densities (and standard deviation) of 15 PAHs were $4.62 \pm 2.99 \mu\text{g m}^{-2} \text{d}^{-1}$ and $2.75 \pm 1.78 \mu\text{g m}^{-2} \text{d}^{-1}$ in the heating and the non-heating seasons, respectively, with a time-weighted annual mean of $3.37 \pm 1.81 \mu\text{g m}^{-2} \text{d}^{-1}$ (based on the durations of the heating and non-heating seasons). High fluxes occurred in the urbanized areas as well as near two coke and gasification facilities. It was revealed that PAH concentrations in surface soil were correlated significantly with both dry deposition and soil organic matter content. Accordingly, a linear regression model was developed for predicting surface soil PAH concentration based on deposition flux density and soil organic matter content.

Introduction

Although it has been documented that concentrations of polycyclic aromatic hydrocarbons (PAHs) in the ambient environments of developed countries are declining,¹ the emissions from developing countries are increasing rapidly.² It has been estimated that the annual emission of 16 USEPA priority PAHs was 116 000 tons in 2004 in China, accounting for 27% of the global emissions.³ Located on the North China Plain, Tianjin is one of the most severely PAH contaminated areas in China due to the consumption of large quantities of straw, firewood, and coal for cooking, heating, coke production and other industrial activities.⁴ The high emission rate led to heavy PAH contamination in the ambient air and subsequently imposed severe health impacts on the local population.⁵

According to the results of a multimedia fate modeling, air-to-surface transport is the dominant process for PAH transfer into the soil in Tianjin, while the contribution of wastewater irriga-

tion was much less significant than that of air-to-surface transport, even though irrigation using not-well-treated wastewater was a common and extensive practice in the area.⁶ Of the three air-to-surface transport processes of PAHs, the dry deposition rate in Tianjin was more than one order of magnitude higher than the wet deposition rate and almost two orders of magnitude higher than the molecular diffusion rate.⁶ Therefore, only dry deposition was addressed in this study. In addition to being the major pathway of soil contamination by PAHs, dry deposition is also a key contributor to human exposure *via* dermal contact and accumulation in crops through folia uptake.^{7,8}

In a previous study, a dispersion model was applied to predict the level, geographical distribution and seasonal variation of PAHs in ambient air in Tianjin.⁵ Although air-to-surface transport is a critical process for understanding the fate of PAHs in both atmospheric and terrestrial environments, field data were limited because of the high-cost and time-consuming monitoring procedure. Therefore, deposition modeling is a useful tool for filling the data gap. A number of dispersion models including Industrial Source Complex Long-term (ISCLT3) can be used to quantify the deposition.⁹

The objective of this study was to quantify the dry precipitation of PAHs in Tianjin based on a high resolution emission inventory and to reveal spatial and seasonal variations in the dry deposition. In addition, quantitative relationships between the dry precipitation and PAH concentration in surface soil were addressed. Fifteen parent PAHs were studied: acenaphthylene

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Environmental impact

Although PAHs were not included in the so called “dirty dozen”, they are one of the most concerning organic pollutants in China due to coal combustion, biofuel burning and vehicle emission. Soil is the major sink of PAHs in the environment. The spatial variation in PAH concentrations are often orders of magnitude. This study focuses on the reasons causing such a spatial variation. For the first time, it was revealed quantitatively that the two major driving forces are air-to-surface deposition and soil organic matter content. This finding can provide a better understanding of the fate of PAHs in the environment.

(ACY), acenaphthene (ACE), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYP), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]-fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP) and indeno[1,2,3-cd]pyrene (IcdP). The results are presented either as the concentrations of individual compounds or as the total concentration of the 15 compounds (PAH15).

Methodology

Study area

Located in the coastal zone of northern China and immediately next to Beijing, Tianjin is one of four municipalities directly under the jurisdiction of the central government. The urban area of Tianjin is heavily industrialized and also severely contaminated by many pollutants including PAHs. It was demonstrated that the annual mean BaP equivalent concentration in ambient air in the Tianjin urban area exceeded the national ambient air quality standard which is 10 ng m^{-3} .⁵ The study area is located on an alluvial plain with flat topography. The absolute elevations of 95% of the area vary from 0 to 50 m. The annual mean temperature in Tianjin is $12 \text{ }^\circ\text{C}$ and over 80% of the annual precipitation occurs in the summer from June to August.

Emission sources

The major PAH emission sources identified in Tianjin were classified into three categories: point sources (coal used for power generation, centralized heating and industry; petroleum used in industry; coke production and coal gasification; and natural gas combustion), linear sources (motor vehicle exhaust) and area sources (biomass burning and domestic coal usage). Data on the strengths of these activities in 2003 as well as PAH emission factors were gathered from the literature and have been presented in detail in a previous study.⁵

Deposition modeling and model validation

A total of 3113 sectors ($2 \text{ km} \times 2 \text{ km}$) were defined to provide a receptor grid covering the study area. Dry deposition rates of the 15 individual PAH compounds onto the surface of each sector were modeled under steady-state assumptions. Using a stability array, a Gaussian sector-average plume equation was applied to model the processes of dispersion, dry deposition and degradation. For model validation, observed dry depositions of various PAH compounds at 22 (21 in the heating season) monitoring sites across the study area were adopted. The monitoring was conducted for the non-heating (from March 15 to November 15) and the heating seasons, separately. The detailed information for the sample collection and analysis have been presented elsewhere.¹⁰ In brief, the samples were collected using ceramic buckets ($\Phi 65 \text{ cm} \times 30 \text{ cm}$), extracted using an accelerated solvent extraction (ASE300, Dionex), purified using silica gel columns, and analyzed with GC-MS (Agilent GC6890/5953 MSD).

Parameter identification

The dataset of model parameters used for previous modeling on ambient air concentrations was adopted for this study.⁵ In brief,

meteorological data including temperature, wind direction and wind speed were provided by the Tianjin Meteorological Administration. Based on this data, wind classes, mixing height, average wind speeds, surface roughness heights, the joint frequencies of occurrence of wind direction sectors, stability categories, average temperatures and mixing heights of various stability categories were derived. Default values were adopted for the Monin-Obukhov length, friction velocity, wind profile exponent and vertical potential temperature gradients.⁹ Other model parameters, obtained from the literature, included vapor-particle partition coefficients, degradation rate constants, particle density and particle size distributions from each emission source.^{11,12}

Data analysis

One hundred and eighty eight top soil samples were collected and measured for PAHs and soil organic matter content (SOM, TOC analyzer) in a previous study¹³ and the data was used in the current study for addressing the correlation between PAH concentration in surface soil and the dry deposition rate. SPSS (SPSS Inc.) was used for all statistical analysis and maps were generated using SURFER (Golden Software Inc.).

Results and discussion

Dry deposition of PAHs in Tianjin

Similar to the model predicted PAH concentrations in ambient air,⁸ dry deposition fluxes calculated in this study also followed a typical log-normal distribution pattern. The coefficients of kurtosis and the skewness shifted from 25.5 and 3.75 in the raw dataset to 1.56 and -0.24 , respectively, after log-transformation ($P > 0.05$ in both cases). For the entire Tianjin area, the model predicted dry deposition fluxes of PAH15 ranged from $0.65 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ to $26.0 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ with a mean value (and standard deviation) of $3.37 \pm 1.81 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$, equivalent to approximately 40 tons per year. Among the total flux, a majority (65%) were PAH compounds with 4, 5 or 6 rings which are dominantly associated with particles.¹⁴ To compare our results with those measured in other places in the world, which are often reported in various combinations of individual PAHs, the model calculated deposition fluxes of PAH14 (14 PAHs except FLA), PAH13 (13 PAHs except ACE and ACY), PAH12 (12 PAHs except ACY, IcdP and DahA), PAH6 (6 PAHs including BbF, BkF, BaP, DahA, IcdP, BghiP) and BaP were calculated as $2.69 \pm 1.72 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$, $3.32 \pm 2.07 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$, $3.02 \pm 1.75 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$, $0.95 \pm 0.77 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ and $0.18 \pm 0.18 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$, respectively. The mean dry deposition rates of PAHs in Tianjin including urban and countryside were much higher than those in either rural or urban sites in France and the USA and the only exceptions were the industrial sites in Manchester and Cardiff which were under direct influences of industrial emission sources nearby (Table 1). In fact, the calculated annual mean depositions of PAH12 and BaP in the urban area of Tianjin were $13.6 \pm 4.85 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ and $0.33 \pm 0.12 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ which are significantly higher than those reported values in industrial districts in Manchester and Cardiff one decade ago. The severe contamination in Tianjin is believed to be the result of the consumption of large quantities of straw, firewood, and coal for cooking, heating and

Table 1 Comparison of dry PAH depositions between those calculated and those reported in the literature

PAH	This study/ $\mu\text{g m}^{-2} \text{d}^{-1}$	Location	Reported/ $\mu\text{g m}^{-2} \text{d}^{-1}$	Ref.
PAH15	3.37 ± 1.81	Marne river basin, France	0.219	15
PAH14 ^a	2.69 ± 1.72	Paris, France	0.622~0.641	15, 16
PAH13 ^b	3.32 ± 2.07	Galveston Bay, USA	0.630	17
PAH12 ^c	3.02 ± 1.75	Manchester (industrial districts), UK	5.0 (1.0~23.4)	18
PAH12	3.02 ± 1.75	Cardiff (industrial districts), UK	4.0 (0.8~19.1)	18
PAH6 ^d	0.95 ± 0.77	Paris urban district, France	0.285	15, 16
BaP	0.18 ± 0.18	Manchester (industrial districts), UK	0.300 (0.15~2.22)	18
BaP	0.18 ± 0.18	Cardiff (industrial districts), UK	0.219 (0.036~1.45)	18
BaP	0.18 ± 0.18	Nahant, beside Massachusetts Bay, USA	0.082	18
BaP	0.18 ± 0.18	Truro, beside Massachusetts Bay, USA	0.006	19
BaP	0.18 ± 0.18	Paris urban district, France	0.023	15, 16

^a 14 PAHs except FLA. ^b 13 PAHs except ACE and ACY. ^c 12 PAHs except ACY, IcdP and DahA. ^d 6 PAHs including BbF, BkF, BaP, DahA, IcdP and BghiP.

coke production.³ The high coefficients of variation were primarily due to high spatial variation.

Because of the cold winter in the area, a large amount of fuel is consumed in winter months for indoor heating, resulting in higher emissions in winter than in summer. As a result, the average deposition rate of PAH15 in the heating season ($4.62 \pm 2.99 \mu\text{g m}^{-2} \text{d}^{-1}$) was significantly (t-test, $P < 0.01$) higher than that in the non-heating season ($2.75 \pm 1.78 \mu\text{g m}^{-2} \text{d}^{-1}$). Since population density, and consequentially fuel consumption and emission densities in the urban area were considerably higher than those in the countryside, the seasonal difference in the urban center (2.14 times) was much higher than that in the rural area (1.65 times).

Model validation

A set of field observed data were adopted for model validation.¹⁰ In Fig. 1, the measured deposition rates of PAH15 at 22 sites (21 for the heating season) are plotted against the model predicted ones on a log-scale. For both seasons, more data points appear above the 1 : 1 line, suggesting a trend of moderate underestimation in the model. This was particularly true for the non-heating season. Such an underestimation was likely caused by the following reasons: (1) all measurements were conducted in residential areas either in the city or in towns for technical

reasons and (2) it is expected that the spring dust storm added a significant increment to the observed dry deposition in the non-heating season. In fact, Tianjin is surrounded by a number of large cities including Beijing and Tangshan, which are also heavily polluted by PAHs. Still, the model predictions are acceptable taking into consideration that the emission sources in the area were complicated and the field monitoring data were limited.

Fig. 2 presents the spatial distribution of the differences between the model predicted and the observed deposition fluxes of PAH15 in the heating (left) and the non-heating (right) seasons. The symbols of ‘-’ and ‘+’ represent either under-estimation or over-estimation of the model, respectively and the sizes of the symbols are proportional to the extent of the over- or under-estimations. The distribution patterns of the model error for the two seasons are similar to each other. In general, the model overestimated the dry deposition fluxes in urban districts and underestimated the fluxes in the rural areas. Although it is

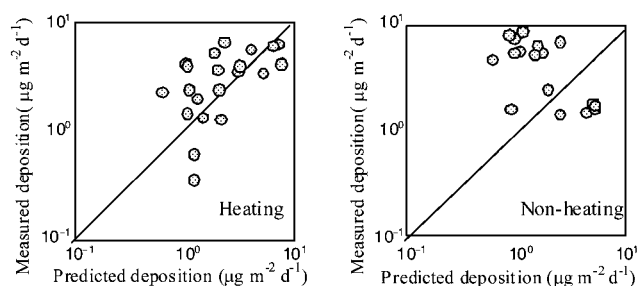


Fig. 1 Comparison between the measured and the model predicted dry depositions of PAH15 in the heating and the non-heating seasons at 22 monitoring stations (21 for the heating season).

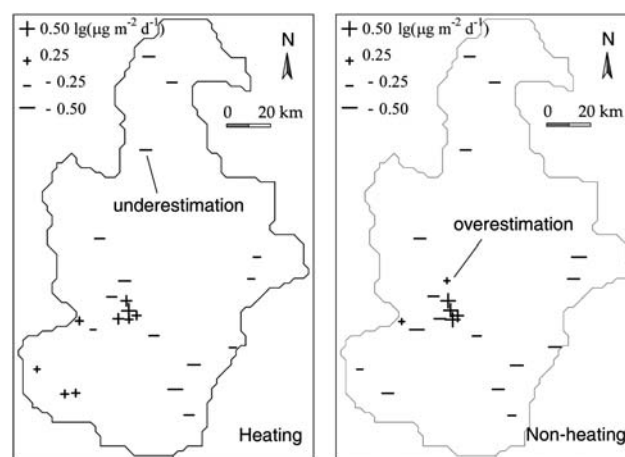


Fig. 2 The differences between the predicted and the observed dry depositions of PAH15 in Tianjin in the heating and non-heating seasons. The overestimation and the underestimation are represented using ‘+’ or ‘-’, respectively. The symbol sizes are proportional to the differences.

rather difficult at this stage to identify all the reasons causing such differences, the underestimation in non-urban areas can be at least partially attributed to the field measurements. As suggested previously, the dust cans in rural areas are all deployed in small towns or villages, rather than in an open field of 'true' rural sites. As a result, the observed dry depositions in rural areas were systematically higher than the real situation. This is also one of the reasons causing the overall underestimation of the model (Fig. 1).

Spatial variation in dry deposition

The model calculated PAH dry depositions of the two seasons are mapped in Fig. 3. Although the deposition rates in the heating season were significantly higher than those in the non-heating season, the distribution patterns were similar to each other. In fact, the patterns were also similar to those of ambient air concentrations at a 2 m height derived from the same model.⁸ The annual mean deposition rate in the urban area during the heating season was as high as $13.6 \pm 4.85 \mu\text{g m}^{-2} \text{d}^{-1}$ and

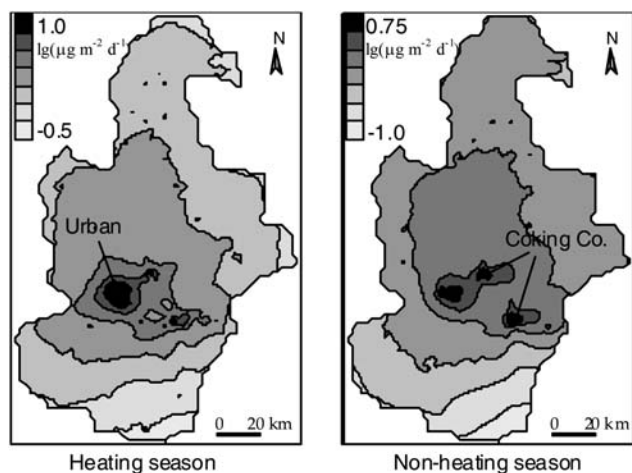


Fig. 3 Geographical distributions of dry deposition rates of PAH15 in Tianjin in the heating (left) and the non-heating (right) seasons.

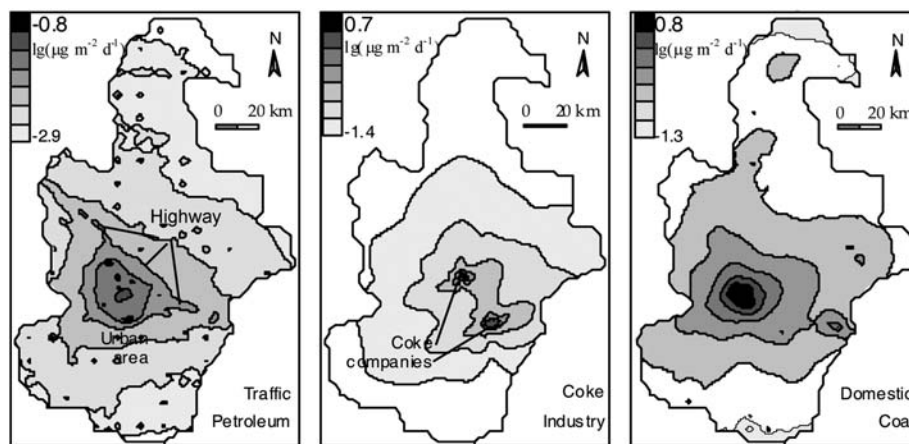


Fig. 4 The model predicted spatial variations in PAH15 dry depositions from three typical emission sources in Tianjin. From left to right are: traffic petroleum consumption, coke production and domestic coal usage.

the highest deposition rates (up to $26.0 \mu\text{g m}^{-2} \text{d}^{-1}$) occurred in the urban center of Tianjin during the heating season, followed by the places where two coking and coal gasification companies are located. Along the southern border, the deposition rates in both seasons were much lower than those of other places.

The contributions of various sources to the dry deposition were derived by running the model using individual emission sources separately. The spatial variations in the contributions from three typical sources including traffic petroleum usage as a linear source, coke companies as point sources and domestic coal consumption as an area source are presented in Fig. 4. The deposition originating from motor vehicle emissions was concentrated in the urban area as well as along the Beijing-Tianjin-Tanggu expressway running across the area. The influence of the two major coke and coal-gasification companies were localized and heavy deposition occurred primarily in the areas close to the two factories. Since domestic coal burning stoves and boilers were spread all over the area, the emissions were treated as an aerial source in the model.

In a previous study, it was found that the contributions of various sources to PAHs in ambient air were remarkably different from those emission sources in Tianjin. For example, although domestic coal combustion contributed to 43.3% of the total emissions, only 22.9% of PAHs in ambient air were from that source. The reasons causing such differences were the differences in emission height, source composition profile and particle size distribution, which led to differentiation in decay after emission.⁸ For the same reason, similar changes are expected between the emission and dry deposition. Fig. 5 presents the relative contributions of major sources to total emissions, ambient air concentration and dry deposition of PAH15. Two minor sources, centralized heating and natural gas combustion which contributed to less than 0.5% of the total emissions, are not included. Although contribution of domestic coal consumption dropped from 43.3% at emission sources to 22.9% in ambient air, it accounted for 56.0% of dry precipitation. Obviously, both low emission heights and relative abundances in high molecular weight PAHs associated with particles from domestic coal combustion led to a quick deposition from the air to the surface and resulted in a relatively larger fraction of total

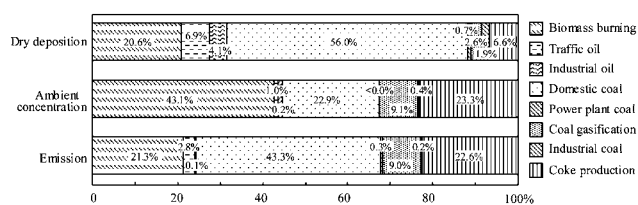


Fig. 5 Relative contributions of various sources to emission (bottom), ambient air concentration (middle) and dry deposition (top) of PAH15 (gaseous + particulate PAHs). Of the ten emission sources modeled, centralized heating and natural gas combustion contributed to less than 0.5% of the total and are not shown.

deposition and a smaller fraction in the total air concentration. Emissions from motor vehicles fall into a similar pattern as domestic coal burning with a reduced fraction in ambient air but an increased fraction in dry deposition. Again, very low emission heights were the primary reason for such shifting. For the two emission sources at relatively high heights, namely coal gasification and coke production, the contributions to the total dry deposition were much smaller than those of the total emissions, as well as ambient air concentration. Emissions from straw and firewood appeared to be more persistent in air, likely because the fraction of high molecular weight PAH compounds in the straw and firewood emissions were higher than those from other sources.

Contribution of dry deposition to surface soil contamination

The air-to-surface deposition is an important pathway for PAHs entering surface soil. According to the results of a multimedia fate model in a wastewater irrigated area in Tianjin, the majority of PAHs in surface soil originated from air-to-surface deposition.²⁰ Using the model to calculate dry deposition in Tianjin, the dependence of soil PAH concentration on the deposition rate was examined. It was revealed that the correlation ($r = 0.337$) between the measured soil PAH15 concentrations (ln-transformed) at 188 sampling locations¹³ and the model calculated deposition rates (also ln-transformed) at the same sites was significant ($P < 0.01$). However, in spite of the significant correlation, a regression analysis using the deposition rate (D) as

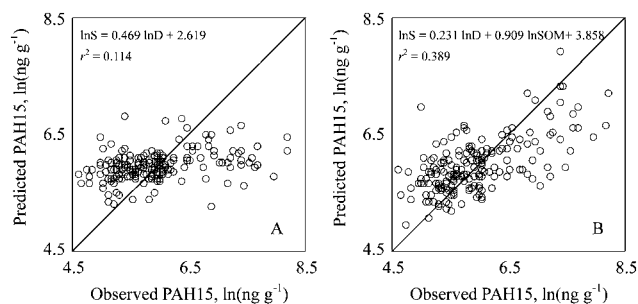


Fig. 6 Relationship between the observed and the model predicted PAH15 (ln-transformed) in surface soil in Tianjin ($\ln S$, $n = 188$). The two models applied were (A) simple regression with deposition flux ($\ln D$) as the dependent variable and (B) bivariate regression model with deposition flux and soil organic matter content ($\ln \text{SOM}$) as the dependent variables.

the sole dependent variable failed to provide an acceptable model for predicting soil PAH concentration (S) and the coefficient of determination was only 0.11 ($\ln S = 0.469 \ln D + 2.619$). As shown in Fig. 6A, the simple model overestimated or underestimated the soil concentrations at the sites with low or high deposition rates, respectively. In addition to the air-to-soil input, soil organic matter content (SOM) is often found to be positively correlated to concentrations of persistent organic pollutants in soil.²¹ In investigations on PAHs in background soils in western Europe and the world, it was found that soil PAH concentrations depended on both source- and sink-related factors and the latter included SOM and black carbon.^{22,23} In a multimedia modeling study on the fate of PHE, evidence collected suggested that both deposition rate and soil organic matter content (SOM) were important factors governing the spatial variation in PHE concentrations in top soil.⁶ To test the influence of SOM, the residuals of the above mentioned simple linear regression model were plotted against SOM at the 188 sampling sites and a significant negative correlation was demonstrated, suggesting that SOM is another critical and independent factor controlling PAH concentrations in soil. Accordingly, a bivariate linear model with both deposition flux and SOM as independent variables was developed ($\ln S = 0.231 \ln D + 0.909 \ln \text{SOM} + 3.858$, $r^2 = 0.39$) and 39% of the variation in the soil PAH15 can be explained (Fig. 6B). In addition to the two independent variables used in the model, there are obviously other factors which may affect soil PAH concentrations. For example, composition of SOM as well as soil moisture content could also be important to the accumulation of PAHs in soil. Unfortunately, spatially resolved data were not available in the study area.

A limitation of the statistical model was that only dry deposition was included in this study. Both wet deposition and air-to-surface exchange of gaseous phase PAHs could affect PAH abundances in soil. More studies are necessary for a better understanding of the source-receptor relationship. In addition to the important role of dry deposition on surface PAH concentrations, dry deposition rates are also expected to be related to PAHs in vegetables and grains, because vegetation accumulates PAHs chiefly from the atmosphere through deposition on leaves.²⁴ Finally, human exposure to PAHs through dermal contact is also related to dry deposition. Further studies are required to address the effects of deposition on vegetation accumulation and human exposure.

Conclusions

The results of this study showed that not only the input *via* deposition but also SOM are governing factors for the accumulation of PAHs in soil. Although the effect of SOM is expected to occur also in other areas for PAHs as well as for other hydrophobic organic contaminants, the regression equation derived in this study should be further tested and the regression coefficients are likely site specific. More studies are necessary to reveal the mechanism of the influence of SOM on the PAH level in soil. Although the high affinity of SOM for PAHs is often believed to be the main reason leading to the correlation between SOM and PAHs, sequestration at a steady-state was also proposed as an explanation.⁶

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