Dry Deposition of Polycyclic Aromatic Hydrocarbons Associated with Atmospheric Particulate Matters in an Urban Site, Mumbai, India

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Abstract

The rate of dry deposition of polycyclic aromatic hydrocarbons (PAHs) depends on the size of the particulate matter on which they are adsorbed. The distribution of PAHs on size-fractionated aerosol samples has been studied during winter and in the absence of precipitation to estimate the dry deposition flux of individual PAHs. Size-fractionated ambient particulate matter samples were collected during a 72-hour period, using an eight-stage cascade impactor. Filter paper samples were then extracted ultrasonically, and PAH characterization and quantification performed with a high-performance liquid chromatograph (HPLC) system with UV-visible detector. Total PAH concentrations associated with the size-segregated ambient particulate matters collected on different impactor stages were in the range of 18.3 to 66.6 ng/m^3. The size distribution of atmospheric total PAHs was found to be bimodal. The modeled deposition velocity ($V_{d,i}$) at the sampling site for the particle size range of < 0.4 μm, 0.4-1.1 μm, 1.1-3.3 μm and 3.3-9.0 μm were found to be 0.004 cm/sec, 0.037 cm/sec, 0.283 cm/sec and 2.72 cm/sec, respectively. The dry deposition flux for individual PAHs at the sampling site was found to be in the range of 0.17 μg/m^2/day-2.69 μg/m^2/day. The results indicate that the dry deposition of PAHs is mainly due to gravitational settling of coarse particulates .

Keywords: PAHs; Size distribution; HPLC; Cascade impactor; Deposition velocity; Dry deposition flux.

INTRODUCTION

Concentrations of particulate matter in the

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ambient air are typically composed of complex mixtures of chemical species, originating from a wide range of natural sources and human activities. Among the organic compounds of anthropogenic origin, polycyclic aromatic hydrocarbons (PAHs) make up a significant group. Polycyclic aromatic hydrocarbons are a class of aromatic

compounds found in ambient particulate matter and shown by several investigators to be carcinogenic and/or mutagenic (Horikawa *et al.*, 1991; Larsen and Larsen, 1998). PAHs are ubiquitous pollutants in urban atmospheres (Tekasarkul *et al.*, 2008). It is well known that they are formed by pyrolysis of organic material arising mainly from anthropogenic emissions in urban areas, essentially by the combustion of fossil fuels (Benner *et al.*, 1989; Baek *et al.*, 1991). These compounds are usually emitted in the gas phase, but in the free atmosphere they are found, with the exception of naphthalene and tricyclic PAHs, mainly in particulate form. Adsorption of gaseous PAHs on particles has been proposed as the most important mechanism of the gas-particle conversion of PAHs. The more volatile the compound, the less the adsorption. The particle size distribution of all PAHs involves two mechanisms; rapid adsorption of PAH vapor, initially produced, on fine particles, and continuous growth of particles by means of redistribution of PAHs adsorbed on fine particles onto larger particles by condensation (Pistikopoulos *et al.*, 1990). The PAHs are adsorbed on soot particles in a multi-layered system, and hence the subsurface layers are not exposed to sunlight and photochemical decomposition (Von-Dinh, 1989). Therefore, in the absence of precipitation, dry deposition determines the atmospheric residence time of PAHs. The rate of dry deposition depends on the size of the particulate matter on which the PAHs are adsorbed. Characterization and quantification of PAHs in size-segregated aerosol samples

are therefore important to evaluate, particularly their residence time in the atmosphere (Sheu *et al.*, 1996).

Several studies on the concentration of PAHs in urban aerosols have been reported in the literature (Lee *et al.*, 1995; Sahu *et al.*, 2001; Guo *et al.*, 2003). Less attention has been paid to the size distribution of PAHs adsorbed on particulate matter. Also, a great deal of research has been conducted to investigate the dry deposition of atmospheric pollutants using a variety of approaches. But, to date, there is no accepted technique that can be used universally to evaluate the dry deposition of atmospheric pollutants. In the current study, we have collected size-fractionated aerosol samples using a cascade impactor, then analyzed them for PAHs. The dry deposition fluxes were evaluated for individual PAHs in an urban site of Mumbai, India.

MATERIALS & METHODS

Study area

In the present study, we have chosen a typical urban site where all the sources are mixed, and a period when the atmospheric stability is high and the photochemistry reduced. Measurements were carried out in Mumbai (latitude $18^o53'$ to $19^o04'$N; longitude $72^o48'$ to $72^o53'$E), India, during the winter and in the absence of precipitation. As a consequence, the effects of certain parameters, such as the elimination of particles by wet deposition, the disappearance of PAHs related to atmospheric radical and oxidants, as

well as the formation of artifacts, are markedly reduced.

Sample collection

Samples were collected over 72 hrs. average sampling period using an eight stage cascade impactor (Andersen Instruments Inc., USA) at 2 m height from ground level. The impactor was connected to a continuous duty, carbon-vane pump and a constant airflow rate of 28.8 L/min was maintained with an in-line rotameter. The samples were collected on glass-fiber filter paper (Whatman EPM 2000) cut to the size of impactor stages.

Sample extraction

The filter paper samples were then extracted ultrasonically with 30 mL of dichloromethane (E. Merck, HPLC grade). The extract was filtered using a sintered glass disc to eliminate other particulate impurities. The extract was then subjected to cleanup procedure. The cleanup procedure has been given elsewhere (Pandit *et al.*, 1996). The extract was then dried by a flow of dry nitrogen. The residue obtained was re-dissolved in 1 mL of acetonitrile for analysis by HPLC. The recovery efficiencies were determined by spiking filter paper samples with a PAH standard mixture. The mean recovery varied from 80-97%.

Sample analysis

The characterization and quantification of PAHs were done by a high performance liquid chromatograph (HPLC) system (Shimadzu, LC-10 AD) with UV-visible detector. The analytical column was of 250 mm length and 4.6 mm i.d., packed with totally porous spherical RP-18 material (particle size 5 μm). A guard column packed with totally porous spherical RP-18 material (10 mm long and 4.6-mm i.d.) preceded the analytical column. Acetonitrile-water mixture (75:25) was used as mobile phase at a flow rate of 1.0 mL per minute. Samples of 100 μL were injected into the column through the sample loop. A UV detector set at 254 nm for absorption was used for detection of the compounds. The data was processed with a CR7A Chromatopac data processor. Several dilutions corresponding to 0.2 to 20 ng absolute of synthetic standard mixture of individual component of PAHs, supplied by Polyscience, USA, dissolved in HPLC grade acetonitrile was used for determining the retention data and for studying the linearity of the detector. The response was linear for a wide range of concentrations mentioned above. The detection limit for the HPLC system ranged from 1.21 to 6.53 pg/μL depending on the compound (Table 1).

QUALITY CONTROL AND ASSURANCE

A strict regime of quality control and assurance was operated during every stage. Field blanks, which accompanied samples to the sampling sites, were used to determine any background contamination. Method blanks (solvent) and spiked blank (standards spiked into solvent) were analyzed. Blanks were run with each sample batch. PAHs were not

Table 1. Detection limits for the PAH compounds analyzed with HPLC system and analysis results of the NIST SRM-1649 urban dust sample.

PAHs	Detection Limit (pg/μL)	SRM Certified Value (μg/Kg)	SRM Measured Value (μg/Kg)
PHE	1.21	4.14 ± 0.37	4.57 ± 0.49
ANT	1.92	0.43 ± 0.08	0.45 ± 0.06
FLT	2.75	6.45 ± 0.18	5.75 ± 0.18
PYR	4.84	5.29 ± 0.25	4.67 ± 0.22
BaA	1.73	2.21 ± 0.07	1.94 ± 0.59
PER	3.24	0.64 ± 0.07	0.76 ± 0.10
BbF	2.48	6.45 ± 0.64	5.83 ± 0.40
BkF	1.72	1.91 ± 0.03	2.07 ± 0.29
BaP	1.34	2.51 ± 0.09	2.82 ± 0.72
BghiP	6.53	4.01 ± 0.91	3.78 ± 0.69

detected in any of these blanks. Efficacy of the method was verified using the standard reference material (SRM) 1649 Urban Reference Dust (supplied by the U.S. National Institute for Standards and Technology). The resulting values were in good agreement with the certified values given for this material (Table 1).

Theory of dry deposition

The deposition velocities for atmospheric particles can be obtained by the following equations (Allen *et al.*, 1996).

$$V_d = V_{st} + 1.12 \times U^* \times \exp(-30.36/D_p) \quad (1)$$

Where V_{st} is the particle settling velocity (cm/sec), U^* is the friction velocity (cm/sec) and D_p is the particle diameter (μm).

When the aerodynamic diameter of the particle (D_p) ≥ 5 μm, the particle settling velocity can be given by the following relationship:

$$V_{st} = V_{T,Stoke} \quad (2)$$

where $V_{T,Stoke}$ is the terminal settling velocity (cm/sec) and can be expressed as:

$$V_{T,Stoke} = (\rho_p \times D_p^2 \times g)/(18 \times \mu) \quad (3)$$

where ρ_p is the density of particle (1.0 g/cm³); g is the gravitational constant (980 cm/sec); and μ is the absolute viscosity of air (1.81 × 10⁻⁴ g/cm. sec).

When the aerodynamic diameter of the particle (D_p) < 5 μm, the particle settling velocity can be given by the following relationship:

$$V_{st} = V_{T,Stoke} \times K_c \quad (4)$$

where Cunningham correction factor and can be expressed as:

$$K_c = 1 + 2 \times \lambda \, [1.257 + 0.4 \exp(-0.55 \times D_p/\lambda)]/D_p \quad (5)$$

where λ is the mean free path (cm).

Friction velocity (U^*) and surface roughness (Z_0) are two meteorological parameters that influence atmospheric turbulence. The relationship between these two parameters for near neutral stability condition is:

$$U = (U^*/k) \times \ln[(Z-d)/Z_0] \qquad (6)$$

Where U (cm/sec) is the mean wind speed at height Z. Z is the measured height above ground (m); k is the Von Karman's constant (0.4); d is the datum height (80% of the average structure height); and Z_0 is the surface roughness height (m). The roughness coefficient was estimated by the general relationship that Z_0 is approximately 1/30 of the average roughness height. The average height of the structures surrounding the sampling site was taken as 8.0 m.

The total dry deposition flux (F_t) for individual PAHs (µg/m²/day) can be estimated by using the following relationship:

$$F_t = \sum_{i=1}^{n} C_i \times V_{d,i} \qquad (7)$$

where C_i is the concentration of individual PAHs in each size-fractionated aerosol samples. $V_{d,i}$ is the dry deposition velocity calculated for each fraction and n is the number of size fractions considered.

RESULTS AND DISCUSSION

The distribution of PAHs on size-fractionated aerosol samples has been studied in Mumbai, India. The range and mean concentrations of particle-bound individual PAHs in size-fractionated aerosol samples are presented in Table 1. Total PAH concentrations (13 compounds) associated with the size-segregated aerosols collected on different impactor stages were in the range of 18.3 to 66.6 ng/m³. It has been observed that in case of phenantherene, anthracene, fluoranthene and pyrene there is an increase in PAH concentration with increased particle size. This may be attributed to the evaporation and recondensation of these semivolatile, low molecular weight PAHs onto bigger sized aerosols, as most of them exist mainly or partly in the gas phase (Van Vaeck and Van Cauwenberghe, 1985). Benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene and benzo(a)pyrene show a decrease in concentration with increased particle size. This could be due to the fact that these PAHs were attached to fine particulates (soot) from combustion sources. In addition, smaller particles have a higher specific surface area and a higher attachment rate for organic pollutants, and hence, contain a greater amount of organic carbon, which allows more PAH adsorption. The concentrations of perylene and benzo(ghi)perylene have not shown any dependence on particle size. These PAHs were mostly associated with fine particles (aerodynamic diameter < 4.7 µm), but the lower molecular weight PAH compounds (2-4 rings compounds) predominated in the aerosol fraction > 1.1 µm,

whereas the higher molecular weight PAH compounds were predominant in the aerosol fractions < 1.1 μm. The predominating nature of lower molecular weight PAH compounds (2-4 rings compounds) in the aerosol fraction > 1.1 μm could be due to the cyclic volatilization and adsorption of these semivolatile species on accumulation-mode urban aerosols.

The size distribution of ambient particulate matter and that of atmospheric total PAHs are illustrated in Fig. 1 and Fig. 2 The maximum of the particle size distribution of the PAHs coincides with the maximum of the ambient aerosols, which suggests that the gaseous PAHs are adsorbed, after production, on preexisting particles during cooling. This observation is in good agreement with the earlier observation (Venkataraman *et al.*, 1999). The size distribution of atmospheric total PAHs was found to be bimodal and in agreement with the earlier studies reported in the literature (Noll and Fang, 1989). The size distribution of PAHs in the ambient aerosols was influenced by the vapor pressure of the compounds and on the chemical compositions. The two peaks in the total PAH size distributions were located at the particle size range 1.1 – 2.1 μm and at 3.3 – 4.7 μm, which mostly belongs to the fine particle mode. This result indicates that the PAH mass in the ambient air of the sampling locations originated mainly from the combustion of fossil fuels. The peak of the size-fractionated total PAHs localized at the particle size range 1.1 – 2.1 μm essentially reflects the gas to particle condensation process in the young

aerosols, whereas the peak located at the size range 3.3 – 4.7 μm shows the occurrence of accumulation processes in these aerosols.

We have estimated different concentration ratios for PAHs, differing either in reactivity or in the particle size range to which they belong in order to find out the dominant removal mechanism of atmospheric PAHs in the study area. Removal of atmospheric PAHs by wet deposition is not taken into account in this discussion as no wet episodes were observed during the sampling period. The benzo(a)anthracene-to-pyrene ratio decreased markedly (by a factor of 10) as we move from the smaller particle size range to larger. This could be due to the slower deposition of benzo(a)anthracene, since it was concentrated on the fine particles. Since benzo(a)anthracene is much more reactive than pyrene, the observed ratios indicate a less-dominant photochemical depletion of atmospheric PAHs compared to dry deposition. This result is in agreement with the sampling period, as all the samples were collected in winter season when the disappearance of PAHs related to atmospheric radical and oxidant are markedly reduced. There was not much variation observed in the ratio of anthracene to pyrene as we move from smaller to bigger particle size ranges. This may be due to the fact that both anthracene and pyrene were associated with larger particles and hence similar deposition pattern. On the other hand, anthracene is much more reactive than pyrene, which again implies that the dry deposition is the dominant disappearance mechanism for PAHs during
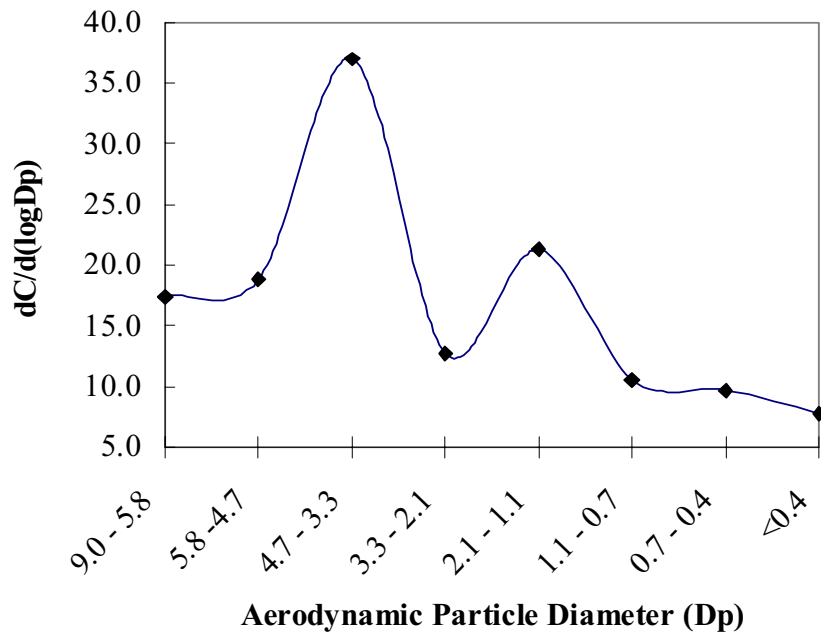
Fig. 1. Distribution of atmospheric total PAHs in size segregated aerosol samples.
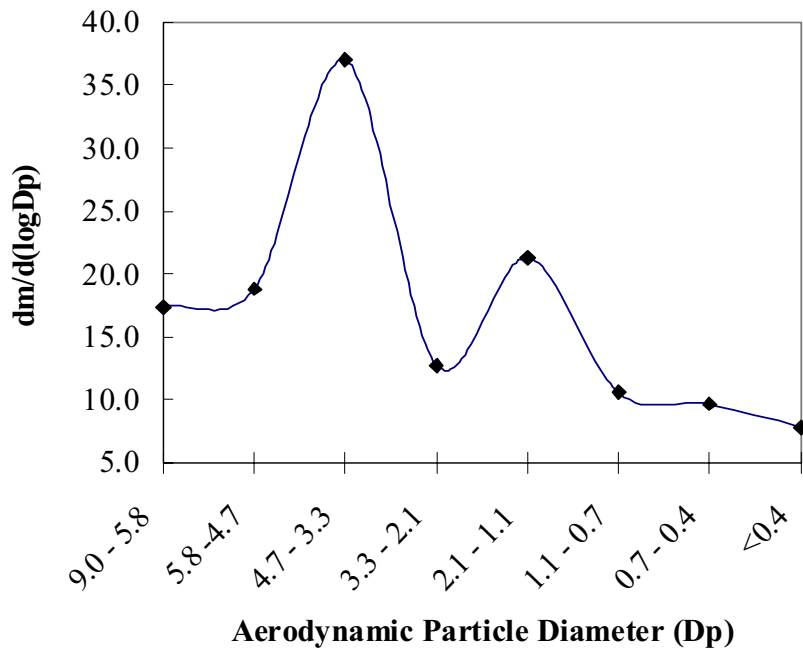

Fig. 2. Size distribution of ambient particulate matter.

Table 2. Range and mean concentration of the individual PAH in the size segregated samples in ng/m^3.

PAHs	BF		0.4 –1.1 µm		1.1 – 3.3 µm		3.3 –9.0 µm	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
PHE	0.07-0.31	0.12 ± 0.05	0.06-0.51	0.23 ± 0.10	0.02-0.79	0.36 ± 0.14	0.02-0.97	0.42 ± 0.20
ANT	0.02-0.26	0.10 ± 0.08	0.02-0.38	0.15 ± 0.08	0.03-0.58	0.21 ± 0.13	ND-0.76	0.28 ± 0.13
FLT	0.49-1.00	0.61 ± 0.21	0.20-1.26	0.75 ± 0.37	0.06-1.49	0.79 ± 0.48	0.12-2.21	0.92 ± 0.58
PYR	ND-0.82	0.43 ± 0.12	0.06-0.92	0.51 ± 0.23	0.56-1.23	0.73 ± 0.36	0.22-2.95	1.06 ± 0.67
BaA	0.12-0.58	0.29 ± 0.10	0.09-0.26	0.17 ± 0.09	0.04-0.12	0.09 ± 0.03	0.04-0.09	0.06 ± 0.02
PER	0.91-1.71	1.15 ± 0.67	0.07-0.62	0.38 ± 0.21	0.05-0.93	0.47 ± 0.21	ND-0.71	0.34 ± 0.18
BbF	0.82-1.65	1.16 ± 0.54	0.05-1.02	0.84 ± 0.57	0.12-0.87	0.53 ± 0.29	ND-0.32	0.19 ± 0.08
BkF	0.10-1.15	0.4 ± 0.18	0.10-0.98	0.35 ± 0.19	0.15-0.43	0.29 ± 0.11	ND-0.26	0.14 ± 0.06
BaP	0.44-1.02	0.76 ± 0.34	0.03-0.91	0.57 ± 0.27	0.03-0.57	0.28 ± 0.09	ND-0.10	0.07 ± 0.02
BghiP	0.63-1.11	0.81 ± 0.49	0.07-0.72	0.54 ± 0.32	0.05-0.85	0.68 ± 0.36	ND-0.08	0.05 ± 0.02

Table 3. Modeled dry deposition flux (µg/m^2/day) for individual PAH.

PAHs	0.0 – 0.4 µm	0.4 –1.1 µm	1.1 – 3.3 µm	3.3 –9.0 µm	Total
PHE	0.42×10^{-3}	7.37×10^{-3}	8.81×10^{-2}	9.84×10^{-1}	1.07
ANT	0.35×10^{-3}	0.48×10^{-3}	5.13×10^{-2}	6.58×10^{-1}	0.66
FLT	2.16×10^{-3}	2.42×10^{-3}	1.93×10^{-1}	21.6×10^{-1}	2.35
PYR	1.51×10^{-3}	1.64×10^{-2}	1.81×10^{-1}	24.9×10^{-1}	2.69
BaA	1.04×10^{-3}	5.44×10^{-3}	2.20×10^{-2}	1.40×10^{-1}	0.17
PER	4.06×10^{-3}	1.22×10^{-2}	1.15×10^{-1}	7.95×10^{-1}	0.91
BbF	4.06×10^{-3}	2.69×10^{-2}	1.30×10^{-1}	4.43×10^{-1}	0.58
BkF	1.73×10^{-3}	1.12×10^{-2}	7.08×10^{-2}	3.26×10^{-1}	0.41
BaP	2.68×10^{-3}	1.81×10^{-2}	6.84×10^{-2}	1.63×10^{-1}	0.25
BghiP	2.85×10^{-3}	1.73×10^{-2}	1.66×10^{-1}	1.17×10^{-1}	0.30

the sampling period. The fluoranthene-to-pyrene ratios allowed us to compare two species associated mostly with the bigger particle size fractions. The fluoranthene-to-pyrene ratios were found to decrease as we moved from fine to bigger particle size ranges. This result indicates that dry deposition is clearly dominant and that perhaps fluoranthene is more reactive than pyrene in the particulate phase.

Table 2 shows the estimated dry deposition flux for individual PAH. The procedure for calculating dry deposition flux divided the particle distribution into four intervals (0 ~ 0.4 µm, 0.4 ~ 1.1 µm, 1.1 ~ 3.3 µm and 3.3 ~ 9.0 µm) and assigned a modeled deposition velocity to each interval. The calculated flux for each interval was then summed to calculate the total dry deposition flux. The modeled deposition velocity ($V_{d,i}$) at the sampling site for the particle size range of < 0.4 µm (mean

Dp = 0.2 μm), 0.4 ~ 1.1 μm (mean Dp = 0.75 μm), 1.1 ~ 3.3 μm (mean Dp = 2.2 μm) and 3.3 ~ 9.0 μm (mean Dp = 6.20 μm) were found to be 0.004 cm/sec, 0.037 cm/sec, 0.283 cm/sec and 2.72 cm/sec, respectively. The dry deposition flux for the individual PAHs at the sampling site was found to be in the range of 0.17 μg/m^2/day (benzo(a)anthracene) to 2.69 μg/m^2/day (pyrene). The higher deposition flux at the sampling site for fluoranthene and pyrene may be attributed to their higher concentrations in the particles having aerodynamic diameters > 1.1 μm. The results indicate that even though PAHs existed mainly in fine particulate (< 2.5 μm), their dry deposition was mainly due to gravitational settling of coarse particulates, particularly those larger than 3.3 μm. The atmospheric dry deposition due to the fine particulates by initial impaction was very minor.

CONCLUSIONS

In this study, PAHs in different sizes of aerosols were characterized and their dry deposition fluxes were estimated. It was found that most of the PAHs were associated with fine particles. The size distribution of atmospheric total PAHs was found to be bimodal. The maximum of the particle size distribution of the PAHs coincides with the maximum of the ambient aerosols, which suggested that the gaseous PAHs are adsorbed, after the production, on preexisting particles during cooling. We have used different concentration ratios for PAHs to find out the dominant removal mechanism of atmospheric

PAHs in the study area. The results indicate that dry deposition is the dominant disappearance mechanism for PAHs during the sampling period for the current study location.

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