# Particulate Dry Deposition and Overall Deposition Velocities of Polycyclic Aromatic Hydrocarbons

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**Abstract:** Previous studies have shown that the dry deposition of semivolatile organic compounds to the Great Lakes can account for a significant fraction of their total inputs. However, there is no generally accepted method to directly measure dry deposition. In this study the particulate dry deposition of polycyclic aromatic hydrocarbons (PAHs) was measured using smooth surrogate surfaces during the winter of 1996–1997 in Chicago. Concurrently, ambient air samples were collected. Average particulate $\Sigma_{13}$-PAH fluxes and ambient concentrations were $120 \pm 28$ µg/m² d and $30 \pm 16$ ng/m³, respectively. The measured particulate dry deposition fluxes were similar to those measured in other urban areas. Overall dry deposition velocities of PAHs calculated using the dry deposition fluxes and ambient concentrations averaged $4.5 \pm 3.1$ cm/s. This value is higher than values typically used to estimate PAH particulate deposition, however, it agrees well with values determined using similar techniques. The overall dry deposition velocity for individual PAHs generally decreased with increasing molecular weight. This finding is consistent with the previous experimental studies that have shown that a greater fraction of the higher molecular weight PAHs are associated with fine particles relative to the lower molecular weight compounds.

## Introduction

Previous research indicated that the impact of atmospheric deposition of semivolatile organic compounds (SOCs) to the Great Lakes is large (Hoff et al. 1996). Despite its importance there is no generally accepted method to directly measure or estimate dry deposition. The removal rate of atmospheric particles by dry deposition is a function of the physical (particle size, density, shape) and chemical properties of the aerosol, meteorological conditions (temperature, wind speed, atmospheric stability) and surface characteristics (terrain, vegetation, roughness). The understanding of how these factors influence the dry deposition of particles is far from complete because of the complex interactions between these parameters and deposition (Zhang et al. 2001; Seinfeld and Pandis 1998).

The use of surrogate surfaces is one approach that has been used to directly measure dry deposition (Bidleman 1988). Recently, a greased, smooth surrogate surface was successfully used to measure particulate fluxes of organic and inorganic air pollutants (Tasdemir 1997; Yi et al. 1997; Franz et al. 1998; Cakan 1999; Odabasi et al. 1999; Shahin et al. 1999a; Shahin et al.

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2000; Yi et al. 2001). Since this surrogate surface does not significantly disturb airflow, it gives estimates of the lower limits of dry deposition to rougher, natural surfaces.

Current dry deposition estimation methods often use measured air concentrations and modeled dry deposition velocities. These models assume the dry deposition flux of particles ($F_p$) can be estimated by using an overall particle dry deposition velocity ($V_p$) and particle phase air concentration ($C_p$):

$$F_p = V_p C_p \tag{1}$$

To date there has been no consensus on the appropriate dry deposition velocity to use in these types of models. Estimated (Hoff et al. 1996; Kaupp and McLahlan 1999 and experimental (Holsen et al. 1991; Tasdemir 1997; Franz et al. 1998; Cakan 1999; Odabasi et al. 1999; Yi et al. 2001) dry deposition velocities of SOCs range over an order of magnitude.

One of the reasons for the discrepancy between the estimated and experimental dry deposition fluxes is that deposition velocity is a function of particle size. Gravitational settling has a significant effect on the deposition of coarse particles while Brownian motion dominates the deposition of very fine particles ($<0.1$ µm) (Seinfeld and Pandis 1998). As the particle diameter increases above approximately 1 µm the deposition velocities increase significantly. For this reason a multistep modeling technique, which divides the fine and coarse particle distributions into a number of intervals and assigns an appropriate deposition velocity to each interval, gives a better estimate of dry deposition than the approach shown in Eq. (1) (Holsen and Noll 1992). Using this multistep model, and other techniques, it has been found that coarse particles ($>2.5$ µm) and compounds associated with them are responsible for the majority of dry deposition of polycyclic aromatic hydrocarbons (PAHs), other SOCs, and some elements (Holsen et al. 1991; Holsen and Noll 1992; Lipiatou et al. 1997; Kaupp and McLahlan 1999; Yang et al. 1999; Yi et al. 2001).

Recently, comparisons between many of the size-dependent deposition velocity models found that they differ from each other

**Table 1.** Summary of Meteorological Data during Sampling Program

| Sample number | Date | Temperature (°C) | | | Wind direction | Wind speed (m/s) | R.H. (%) ave |
|---|---|---|---|---|---|---|---|
| | | $T_{min}$ | $T_{max}$ | $T_{ave}$ | | | |
| 1 | 10/13/96 | 16.1 | 25.6 | 21.5 | WSW | 3.4 | 66.8 |
| 2 | 10/15/96 | 13.3 | 26.1 | 21.3 | SW | 2.7 | 57.6 |
| 3 | 11/13–14/96 | −5.6 | 1.7 | −1.9 | WNW-NE | 2.6 | 54.3 |
| 4 | 11/26–27/96 | −7.8 | −0.6 | −4.0 | WNW-WSW | 3.9 | 67.5 |
| 5 | 12/4–7/96 | −5.6 | 5.0 | 0.6 | WSW | 3.5 | 77.5 |
| 6 | 1/7–8/97 | −11.1 | −1.7 | −5.8 | WNW-SE | 2.8 | 64.0 |
| | Average | | | 2.1 | | 3.2 | 65.1 |

significantly and the largest uncertainty is for the 0.1–1.0 μm particle size range, for which the deposition velocities can vary by 2–3 orders of magnitude (Zhang et al. 2001). Most of these models suggest that particles in the range of 0.1–1.0 μm diam have deposition velocities smaller than 0.01 cm/s. This value is not comparable to the significantly higher values obtained from field studies investigating some trace species (i.e., sulfate), which are considered to be representative of particles in this size range (Zhang et al. 2001).

The objectives of this work were (1) to measure ambient particulate concentrations and dry deposition of PAHs in Chicago, and (2) to determine overall dry deposition velocities for individual compounds.

## Experiment

### Sample Collection

Dry deposition and ambient air samples were collected during the 1996–1997 winter season at the urban research and monitoring site of the Integrated Atmospheric Deposition Network (IADN) located on the campus of Illinois Institute of Technology (Odabasi et al. 1999).

The particulate phase dry deposition flux was measured using a smooth, greased plate with a sharp leading edge (<10°), mounted on a wind vane. The dimensions of the each greased strip were 5.7×1.8 cm. Five plates and 20 strips with a total collection area of 205.2 cm$^2$ were used.

Particulate PAHs in air were collected on 11-cm-diam glass fiber filters using a modified high-volume sampler model PS-1 (General Metal Works Inc.).

All samples were collected during the daytime when there was no rain. Average sampling time was 24 h over 2 days (2 ×12 h). During each period one dry deposition sample and two air samples were collected (air samples were composited before analysis). The average sampling volume for each 12 h air sample was about 95 m$^3$. A summary of sampling information (sampling dates and meteorological conditions) is provided in Table 1.

### Sample Preparation and Analysis

Glass fiber filters were wrapped loosely with aluminum foil, and baked in a muffle furnace at 450°C overnight. Then they were allowed to cool to room temperature in a desiccator (Odabasi et al. 1999).

Mylar was cut into strips (7.6×2.5 cm) and the area to be greased (5.7×1.8 cm) was marked with a mechanical pen. Then the strips were rinsed with methanol and DI water. Cleaned Mylar strips were coated with ~1.5 mg of Apiezon type L grease. Strips

were mounted on dry deposition plates and ungreased areas were protected with PVC covers to prevent exposure to deposited material during field sampling (Tasdemir 1997; Yi et al. 1997; Cakan 1999; Odabasi et al. 1999).

Cleaned glass fiber filters and dry deposition plates were transported to the field in containers without exposure to ambient air. After sampling PS-1 filters and plates were placed back into their containers, the samples were brought back to the laboratory, and stored in the dark at −20°C until they were analyzed.

All samples were spiked with PAH surrogate standards prior to extraction in order to determine analytical recovery efficiencies. Air filters and greased strips were Soxhlet extracted with a mixture of dichloromethane (DCM): petroleum ether (PE) (20:80) for 24 h.
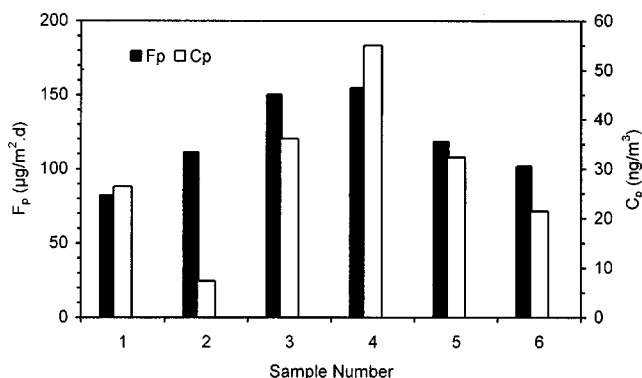
All sample extracts were concentrated and transferred into hexane using a rotary evaporator and a high-purity stream of nitrogen. After volume reduction to 2 mL and transfer into hexane, samples were cleaned on an alumina–silicic acid column containing 3 g silicic acid (3% water) and 2 g alumina (6% water). The column was prewashed with 20 mL DCM followed by 20 mL PE. The sample in 2 mL hexane was added to the top of the column and PAHs were eluted with 20 mL DCM. The solvent was exchanged into hexane, and the final sample volume was adjusted to 1 mL by nitrogen blow-down (Odabasi 1998).

Samples were analyzed for 13 PAHs (acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,1-cd]pyrene, and benzo[g,h,i]perylene) using a HP model 5890 Series II gas chromatograph and a HP model 5971 A mass selective detector (MSD). An HP-5 column (60 m×0.32 mm id., Hewlett Packard Corp.) was used. The MSD was operated in selected ion monitoring (SIM) mode. Instrument operating conditions and the ions used for PAH identification and quantification have been detailed elsewhere (Odabasi 1998).

### Quality Control

All samples were spiked with PAH internal standards prior to extraction to determine analytical recovery efficiencies. Recoveries of PAH internal standards ranged from 64 to 79%. The recoveries of the following internal standards were used to correct the amounts of specific PAHs found in the samples: Acenaphthene-$d_{10}$ for acenaphthene and fluorene, phenanthrene-$d_{10}$ for phenanthrene, anthracene, and fluoranthene, chrysene-$d_{10}$ for pyrene, benz[a]anthracene and chrysene, and perylene-$d_{12}$ for benzofluoranthenes, benzo[a]pyrene, indeno[1,2-cd]pyrene and benzo[g,h,i]perylene.

Quantifiable PAH amounts were determined from sequential injections of diluted standard solutions ranged from 0.015 ng (for acenaphthene) to 0.24 ng (for benzo[g,h,i]perylene). Blank filters
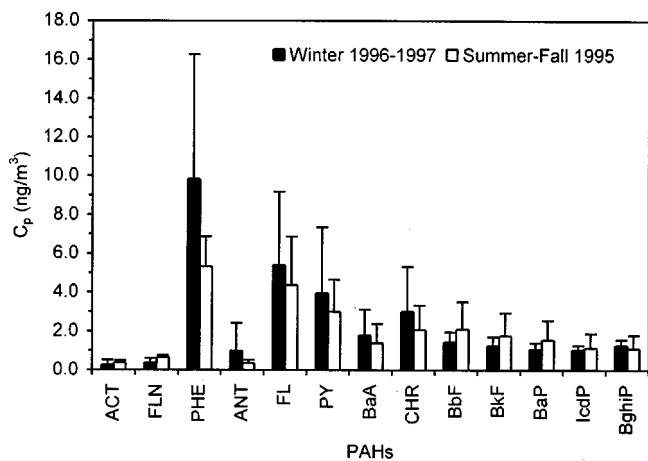
**Fig. 1.** Particulate $\Sigma_{13}$-PAH air concentrations and dry deposition fluxes

**Table 2.** Dry Deposition of Polycyclic Aromatic Hydrocarbons (PAHs) ($\mu g/m^2$ day) Measured with Greased Dry Deposition Plates

| PAH[a] | Chicago[b] | Chicago[c] | Chicago[d] | Taiwan[e] | Taiwan[f] |
|---|---|---|---|---|---|
| ACT | 1.6±1.1 | 3.3 | 0.12 | 1.6 | 6.4 |
| FLN | 2.6±1.2 | 4.2 | 0.11 | 2.1 | 4.1 |
| PHE | 54.6±30.9 | 47.1 | 1.56 | 2.8 | 10.1 |
| ANT | 1.4±0.8 | 1.6 | 0.19 | 3.3 | 2.8 |
| FL | 17.0±3.9 | 25.5 | 2.71 | 2.0 | 14.5 |
| PY | 17.1±6.4 | 22.9 | 2.24 | 3.9 | 24.3 |
| BaA | 3.5±1.6 | 7.0 | 1.09 | 4.1 | 27.7 |
| CHR | 4.9±2.1 | 9.1 | 1.88 | 2.1 | 13.0 |
| BbF | 5.5±2.1 | 9.6 | 2.91[g] | 9.4 | 22.3 |
| BkF | 5.0±2.1 | 8.6 | | 6.0 | 140.0 |
| BaP | 3.8±1.5 | 7.7 | 1.10 | 7.2 | 311.0 |
| IcdP | 1.6±0.7 | 6.3 | 0.77 | 8.9 | 234.0 |
| BghiP | 1.2±0.5 | 5.4 | 0.99 | 7.5 | 206.0 |

[a]Acenaphthene (ACT), fluorene (FLN), phenanthrene (PHE), anthracene (ANT), fluoranthene (FL), pyrene (PY), benz[*a*]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[*1,2,3-cd*]pyrene (IcdP), benzo[*g,h,i*]perylene (BghiP).
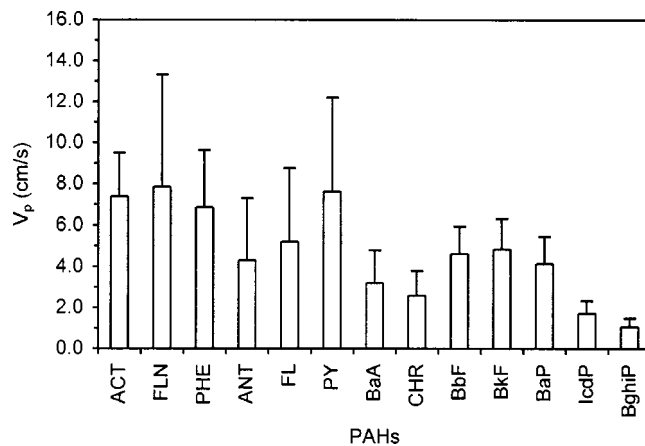
[b]This study.

[c]Odabasi et al. (1999).

[d]Franz et al. (1998) (geometric means).

[e]Shen et al. (1996) (traffic intersection).

[f]Sheu et al. 1996 (residential area).

[g]Bbf + BkF.

and dry deposition plates were routinely taken to the field to determine if there was any contamination during sample handling and preparation for analysis. The limit of detection (LOD, ng) was defined as the mean blank mass plus three standard deviations (Halsall et al. 1994; Cotham and Bidleman 1995; Falconer et al. 1995). Benz[*a*]anthracene through benzo[*g,h,i*]perylene were not detected (nd) in blanks. LODs for PAHs ranged from nd to 1016 ng for PS-1 filters and nd to 1997 ng for dry deposition plates. The largest amounts found in the blanks were for phenanthrene. In general, PAH amounts in the samples were substantially higher than LODs. Average sample amount (ng) to LOD (ng) ratios were 1.8–5.5 and 2.4–4.8 for filters and plates, respectively. Sample quantities exceeding the LOD were quantified and blank corrected by subtracting the mean blank amount from the sample amount.

The analytical method used was tested by analyzing three aliquots of NIST Standard Urban Dust Reference Material (SRM-1649). Concentrations of PAHs found in the SRM-1649, as the percent of NIST certified values, were: benz[*a*]anthracene 96%, benzo[*a*]pyrene 88%, benzo[*g,h,i*]perylene 96%, fluoranthene 106%, and indeno[*1,2-cd*]pyrene 117%.

Possible sampling artifacts associated with the dry deposition plates have been discussed in detail elsewhere and will only be briefly discussed here (Odabasi et al. 1999). Quantities of gas-

phase PAHs absorbed by grease were estimated previously using temperature adjusted octanol-air partitioning coefficients, the average amount of grease used, and the measured gas-phase ambient concentrations during each sampling period. Estimated quantities absorbed by the grease were not significant (Odabasi et al. 1999). Shahin et al. (1999b) also reported that the amount of deposited PAH due to gas-phase deposition was not significant compared to particle deposition. In this study, it was assumed that reactive losses of deposited PAHs from dry deposition plates due to exposure to solar radiation and atmospheric oxidants during sampling were also insignificant.



**Fig. 2.** Average particulate air concentrations of individual polyclic aromatic hydrocarbons (PAHs) in Chicago, measured in Winter 1996–1997 (this study, $n=6$) and Summer–Fall 1995 (Odabasi et al. 1999, $n=12$). Error bars represent one standard deviation.



**Fig. 3.** Overall dry deposition velocities of polycyclic aromatic hydrocarbons (PAHs). Error bars represent one standard deviation.
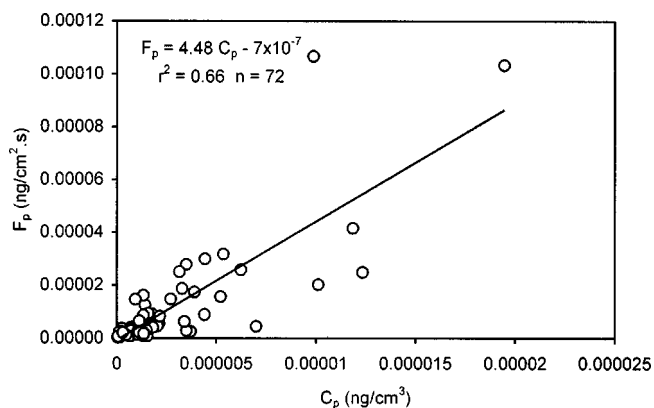
**Fig. 4.** Relationship between ambient particle phase polycyclic aromatic hydrocarbon (PAH) concentrations and particle PAH fluxes measured with the dry deposition plate.
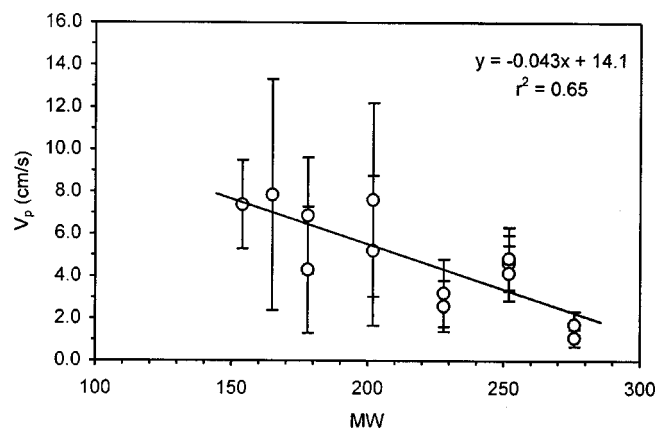


**Fig. 5.** Relationship between the molecular weights and average overall dry deposition velocities. Error bars represent one standard deviation.

## Results and Discussion

### *Ambient Particle Phase Concentrations*

$\Sigma_{13}$-PAHs refers to sum of the particle concentrations of the 13 measured compounds. In this study, particle phase $\Sigma_{13}$-PAH concentrations ranged from 7 to 55 ng/m$^3$ (average $30\pm16$ ng/m$^3$)

(Fig. 1). These concentrations were similar to those measured at the same site in Chicago in 1995 (10–48 ng/m$^3$) (Odabasi et al. 1999) (Fig. 2).

Phenanthrene was the most abundant compound measured in this study (Fig. 2). Phenanthrene, fluoranthene, and pyrene accounted for 46, 14, and, 14 of $\Sigma$PAHs, respectively. The concen-

**Table 3.** Dry Deposition Velocities for Semivolatile Organic Compounds (SOCs) Associated With the Particle Phase

| Compound[a] | $V_p$ (cm/s) | Method | Reference |
|---|---|---|---|
| PCB | 0.50 | Estimated for submicron particles at 14 m/s wind speed using the model by Sehmel and Sutter | Doskey and Andren (1981) |
| PAH | 0.99 | Calculated by a mass balance model | McVeety and Hites (1988) |
| SOC | 0.20 | Estimated using the model by Slinn and Slinn for small (0.5 μm) and large (5 μm) particles assuming a small to large ratio of 1.5:1 | Hoff et al. (1996) |
| PAH and PCDD/F | 0.05 | Weighted average calculated using selected deposition velocities for each particle size interval and the fraction of the substance in the corresponding interval | Kaupp and McLahlan (1999) |
| OC | $5.0\pm2.0$ | Dry deposition plates | Cakan (1999) |
| PCN | $3.0\pm2.3$ | Dry deposition plates | Cakan (1999) |
| PCB | 5.0 | Dry deposition plates | Holsen et al. (1991) |
| PCB | $6.5\pm5.0$ | Dry deposition plates | Tasdemir (1997) |
| PCB | $4.4-7.2$ | Dry deposition plates | Franz et al. (1998) |
| PAH | $0.67-1.8$[b] | Dry deposition plates | Sheu et al. (1996) |
| PAH | $0.4-3.7$ | Dry deposition plates | Franz et al. (1998) |
| PAH | $6.7\pm2.8$ | Dry deposition plates | Odabasi et al. (1999) |
| PAH | $4.5\pm3.1$ | Dry deposition plates | This study |

[a]Polychlorinated biphenyls (PCB), organochlorine pesticides (OC), polychlorinated naphthalenes (PCN), polychlorinated dibenzo-*p*-dioxins and dibenzo-furans (PCDD/F).

[b]$V_p$ was calculated by dividing the flux measured with dry deposition plate by total (particulate+gas) air concentration.

tration profile was very similar to the one observed at the same site in Chicago in 1995 (Odabasi et al. 1999) (Fig. 2). Concentrations of fluoranthene through benzo[*g,h,i*]perylene were very similar. However, concentrations of acenaphthene and fluorene were approximately two times lower in winter while concentrations of phenanthrene and anthracene were approximately two times higher than concentrations measured during Summer–Fall 1995. Higher concentrations in winter may be due to the less blow off from the filter as a result of substantially lower temperatures. However, this was not observed for all relatively more volatile compounds. Therefore, this difference was probably due to varying source strengths (i.e., domestic heating) of PAHs with season.

### Particulate Fluxes

The range of particulate $\Sigma_{13}$-PAH fluxes measured with the dry deposition plates was 82–155 $\mu g/m^2$ d (average 120 $\pm 28 \mu g/m^2$ d) and was related to the measured particle concentrations (this relationship will be discussed in detail below) (Fig. 1). These fluxes were similar to the ones measured in other urban areas. For example, the particulate $\Sigma_{14}$-PAH flux for a residential area in Taiwan was reported as 60.9 $\mu g/m^2$ d by Sheu et al. (1996). Ranges of 3.4–140 $\mu g/m^2$ d (Franz et al. 1998) and 27–229 $\mu g/m^2$ d (average $144 \pm 60 \mu g/m^2$ d) (Odabasi et al. 1999) for particulate $\Sigma$PAH flux was measured in Chicago, recently.

Similar to the air concentrations, particulate $\Sigma_{13}$-PAH fluxes were dominated by phenanthrene, fluoranthene, and pyrene (Table 2). These compounds accounted for 33, 18, and 13% of particulate $\Sigma_{13}$-PAH fluxes, respectively. Generally, fluxes of individual compounds were comparable to the ones measured previously (Table 2) at the same site in Chicago in 1995 (Odabasi et al. 1999). Fluxes of individual compounds benz[*a*]anthracene through benzo[*g,h,i*])perylene measured in this study were 2–4 times lower than the ones measured in Summer–Fall 1995, probably due to lower particulate associated concentrations and deposition velocities.

### Overall Dry Deposition Velocities of PAHs

The overall dry deposition velocities for PAHs calculated by dividing the particulate fluxes measured with the surrogate surfaces by ambient particulate associated concentrations [Eq. (1)] ranged from 1.1 to 7.8 cm/s with an overall average of $4.5 \pm 3.1$ cm/s (Fig. 3). The particulate phase PAH fluxes were correlated with the ambient particulate phase concentrations (Fig. 4). This correlation was statistically significant (at the 95% confidence level) ($r^2 = 0.66$). The slope of the linear regression line (4.5 cm/s) is the apparent best-fit overall dry deposition velocity, which is the same as the average value reported above.

Reported values for the particle phase dry deposition velocities of semivolatile organic compounds are summarized in Table 3. The ratio between the dry deposition velocities calculated in this study and previously reported values varied between 0.7 and 90. However, some of these values are not directly comparable to the results of this study because of differences in experimental procedures, estimation techniques used and physical properties of the compounds.

The agreement between the calculated dry deposition velocities in this study and the reported values using similar techniques is good (Holsen et al. 1991; Tasdemir 1997; Franz et al. 1998; Cakan 1999; Odabasi et al. 1999). The values reported by Sheu et al. (1996) were lower than the dry deposition velocities calcu-

lated in this study although the sampling procedures were similar. This difference is because Sheu et al. (1996) calculated the dry deposition velocities using the measured dry deposition fluxes and total (particulate+gas) ambient concentrations assuming that the dry deposition plate collects both gas and particulate dry deposition. Based on the analysis of gas-phase deposition to the greased surfaces discussed above (which indicated little sorption), this would result in an underestimation of particle associated PAH dry deposition velocities.

The discrepancy between the experimental and estimated dry deposition velocities may be due to large particles ($>10$ $\mu m$) that were not taken into consideration in the dry deposition velocity estimates by Kaupp and McLahlan (1999), Hoff et al. (1996), and Doskey and Andren (1981). However, results of the study by Holsen et al. (1991) indicated that the contribution of coarse particles to the dry deposition fluxes of PCBs was important. Recent studies also reported that the dry deposition fluxes of PAHs were dominated by the coarse particles (Lipiatou et al. 1997; Kaupp and McLahlan 1999; Yang et al. 1999).

The overall dry deposition velocity for individual PAHs generally decreased with increasing molecular weight. The average overall dry deposition velocity for PAHs with molecular weights between 154 and 202 was $6.3 \pm 3.2$ cm/s, and for PAHs with molecular weights between 228 and 276 it was $3.2 \pm 1.8$ cm/s. The overall dry deposition velocity for individual PAHs were well correlated with molecular weight ($r^2 = 0.65$) (Fig. 5). Correlation was statistically significant at the 95% confidence level. This decrease in deposition velocity with increasing molecular weight is supported by other experimental studies which have shown that a greater fraction of the higher molecular weight PAHs are associated with fine particles relative to the lower molecular weight compounds (Pistikopoulos et al. 1990; Aceves and Grimalt 1993; Poster et al. 1995; Allen et al. 1996; Kiss et al. 1998; Kaupp and McLahlan 1999; Kaupp and McLahlan 2000). Previously, a decrease in deposition velocity with increasing molecular weight was reported for PAHs (Odabasi et al. 1999) and organochlorine pesticides (Cakan 1999).

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