

# Atmospheric Concentrations and Deposition of Polycyclic Aromatic Hydrocarbons to the Mid-Atlantic East Coast Region

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Atmospheric concentrations of polycyclic aromatic hydrocarbons (PAHs) were measured at urban/industrial, suburban, coastal, and rural areas in New Jersey as part of the New Jersey Atmospheric Deposition Network. Concentrations of 36 PAH compounds were measured in the gas and particle phases in air and in precipitation at nine sites at regular intervals from October 1997 through May 2001. Gas-phase and particle-phase  $\Sigma_{36}$ PAH concentrations ranged from 0.45 to 118 ng m<sup>-3</sup> and from 0.046 to 172 ng m<sup>-3</sup>, respectively, and precipitation concentrations ranged from 11 to 16200 ng L<sup>-1</sup>. PAH concentrations vary spatially across the region, with the highest concentrations occurring at the most heavily urban and industrial locations. Average gas absorption deposition ranged from 0.004 (naphthacene) to 5040 (methylphenanthrenes) ng m<sup>-2</sup> d<sup>-1</sup>, and dry particle deposition PAH fluxes ranged from 0.11 (naphthacene) to 300 (benzo[*b+k*]fluoranthene) ng m<sup>-2</sup> d<sup>-1</sup> at the nine sites. Average atmospheric wet deposition PAH fluxes at the seven sites ranged from 0.40 (cyclopenta[*cd*]pyrene) to 140 (methylphenanthrenes) ng m<sup>-2</sup> d<sup>-1</sup>. These represent the first comprehensive estimates of PAH deposition to New Jersey and the Mid-Atlantic East Coast.

## Introduction

Continuous emissions of polycyclic aromatic hydrocarbons (PAHs) into the atmosphere from combustion-related emission sources such as motor vehicles and residential/commercial fossil fuel use can lead to significant atmospheric deposition to adjacent regions. Semivolatile organic compounds, such as PAHs, are removed from the atmosphere via dry particle deposition, gaseous air deposition, and wet deposition and atmospheric transformation (1, 2). The residence time for these compounds in the atmosphere (and therefore the transport distance) is dependent on the distribution of the compounds between the gas and particle

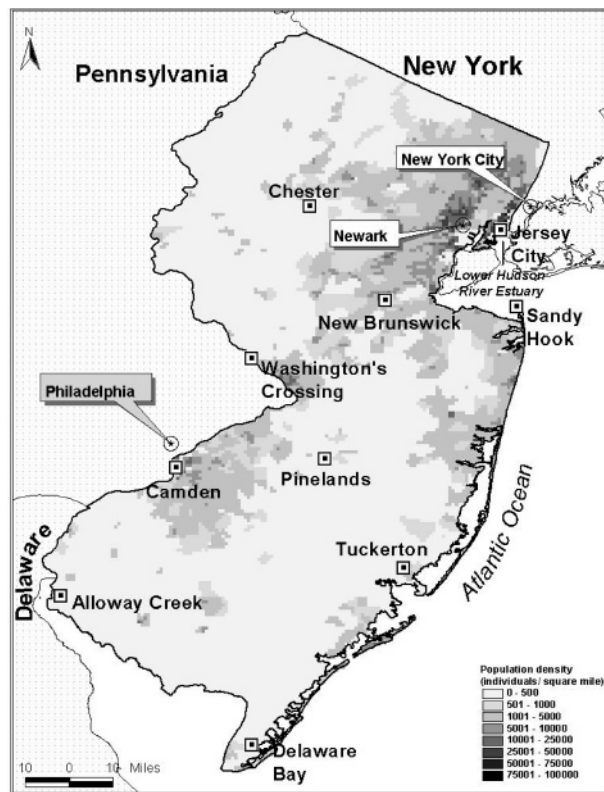


FIGURE 1. Map of New Jersey with population density, showing the NJADN air monitoring stations (squares).

phases in the atmosphere (3). These atmospheric pathways allow for the delivery of SOCs to aquatic and terrestrial ecosystems located both near and remote from emission sources. Thus, atmospheric deposition is likely an important input pathway that must be accounted for in the development of total maximum daily load (TMDL) standards for PAH compounds in the Mid-Atlantic region. One of the primary goals of the New Jersey Atmospheric Deposition Network (NJADN) is to provide estimates of the importance of atmosphere delivery processes to urban water bodies in New Jersey and neighboring states. Because the NJADN included sites on the borders of New Jersey with other states, including Pennsylvania, Delaware, and New York, the resulting data are applicable to much of the Mid-Atlantic East Coast region. This region contains several large urban centers (Boston, MA; New York City, NY; Philadelphia, PA; Washington, DC) with the potential to severely impact their proximate water bodies through atmospheric as well as terrestrial pathways.

NJADN was implemented in October 1997 as a research and monitoring network to assess the magnitude of atmospheric deposition of a suite of SOCs, including polychlorinated biphenyls (PCBs) (4, 5), PAHs (6), and target organochlorine pesticides (7), to water bodies in New Jersey. Trace metals were also measured (8). NJADN was designed to quantify both the local (urban-influenced) and regional (background) concentrations of air toxics by locating monitoring sites in areas with different geographical land-use regimes, including urban, suburban, rural, and coastal environments (Figure 1). The concentrations are then translated into deposition fluxes to aquatic systems in New Jersey. In this paper we focus on the estimates of atmospheric deposition of PAHs from ~4 y of sampling (October 1997 to May 2001) at 10 sites around the state of New Jersey.

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## Experimental Section

**Site Characterization.** To support the objectives set forth for NJADN, air monitoring stations were established at 10 sites around the state of New Jersey (Figure 1) from as far north as Jersey City (latitude/longitude 40.71°N/74.05°W) to as far south as Delaware Bay (39.02°N/74.87°W). At each of the monitoring stations, a high-volume air sampler was deployed to collect gas- and particle-phase SOCs. At 8 of the 10 sites, wet-only integrating precipitation collectors (Meteorological Instrument Centre (MIC) Co., Ontario, Canada) were operated to collect both dissolved and particle-phase SOCs washed out of the atmosphere during precipitation events.

To represent the urban/industrial signal, sampling stations were sited in Jersey City, located across the Hudson River from New York City, and in Camden, located across the Delaware River from Philadelphia, PA. The New Brunswick site was chosen to represent the suburban signal. The Chester and Washington Crossing stations were located to represent lightly suburban to background regions. Four coastal sites were established at Sandy Hook, Tuckerton, Alloway Creek, and Delaware Bay. The Pinelands site is located in a rural, forested pitch pine/oak protected ecosystem remote from the immediate impact of large urban emission sources.

The monitoring sites were established over a period of about 5 y, and sampling was suspended at the different locations at different times. This paper includes data for the following time periods: New Brunswick (10/1997 to 8/2002), Sandy Hook (2/1998 to 1/2001), Jersey City (7/1998 to 1/2001), Tuckerton (11/1998 to 5/2001), Pinelands (6/1999 to 8/2002), Camden (7/1999 to 8/2002), Delaware Bay (3/2000 to 5/2001), Chester (5/2000 to 5/2001), Alloway Creek (1/2002 to 1/2003). These periods represent the entire active period of each site except for New Brunswick and Camden, where sampling is ongoing.

**Sampling Methodology. Gas and Particle Phase Sampling.** At each of the 10 sites, a high-volume air sampler (Tisch Environmental, Village of Cleves, OH) was operated at a calibrated airflow rate of  $\sim 500 \text{ L min}^{-1}$  to individually collect both particle- and gas-phase PAHs over each 24 h sampling period. The operationally defined particle phase was captured on precombusted quartz fiber filters (QFFs), and the operationally defined gas phase was captured on solvent-cleaned polyurethane foam plugs (PUFs). PUFs were cut from 4 in. thick medium-density polyurethane foam sheets into cylinders 7 cm in diameter, which were cleaned with Alconox, rinsed with Milli-Q water, and extracted with petroleum ether before use. The QFFs were weighed for determination of total suspended particle (TSP) mass.

**Precipitation Sampling.** MIC-B wet-only integrating precipitation collectors (Meteorological Instrument Centre) were deployed at eight of the sampling stations. Each sample was collected in a stainless steel collection funnel that drained through a column filled with XAD-2 resin (Supelco, mesh size 20–60). After sampling, the stainless steel funnel was swabbed with a piece of Milli-Q-water-moistened, solvent-extracted glass wool to collect any remaining particles sorbed to the interior of the funnel. The glass wool was combined with the XAD-2 resin sample during laboratory analysis. The water filtered through the XAD-2 adsorbent was collected in a 40 L carboy for sample volume determination. The volume of precipitation collected over a 12 d sampling period ranged from 0.04 L at Sandy Hook to as much as 50 L at Camden.

**Laboratory Analysis.** All PUF (gas-phase) and QFF (particle-phase) samples were spiked with surrogate standards ( $d_{10}$ -anthracene,  $d_{10}$ -fluoranthene,  $d_{10}$ -benzo[e]pyrene), and each was extracted in a Soxhlet apparatus for 24 h in petroleum ether or dichloromethane, respectively. The XAD-2 samples were extracted in acetone/hexane (1:1 by volume)

in a Soxhlet apparatus for 24 h after the addition of surrogates. The XAD-2 extracts were liquid–liquid extracted with 60 mL of deionized water. The aqueous fractions were back-extracted with  $3 \times 50 \text{ mL}$  of hexane.

The gas, particle, and precipitation sample extracts were concentrated using rotary evaporation, and the solvent was switched to hexane. The extracts were blown-down under a steady stream of purified  $\text{N}_2$  gas to  $\sim 0.5 \text{ mL}$ .

All samples were eluted on a 3% water deactivated alumina column to fractionate the sample on the basis of polarity and to remove any interfering compounds. Fraction 1, containing PCBs and some chlorinated pesticides, was eluted with 13 mL of hexane. Fraction 2, containing PAHs and some chlorinated pesticides, was eluted with 15 mL of 2:1 dichloromethane/hexane. Fraction 2 was reduced in volume to  $\sim 0.5 \text{ mL}$  under  $\text{N}_2$  gas and spiked with  $d_{10}$ -phenanthrene,  $d_{10}$ -pyrene, and  $d_{12}$ -benzo[a]pyrene as internal standards.

The PAHs were analyzed on a Hewlett-Packard 6890 gas chromatograph coupled to a Hewlett-Packard 5973 mass-selective detector (MSD) operating in selective ion monitoring (SIM) mode using a  $30 \text{ m} \times 0.25 \text{ mm i.d.}$ , J&W Scientific 122-5062 DB-5 (5% diphenyldimethylpolysiloxane) capillary column with a film thickness of  $0.25 \mu\text{m}$ . Thirty-six individual PAH compounds were quantified: fluorene, phenanthrene, anthracene, 1-methylfluorene, dibenzothiophene, 4H-cyclopenta[def]phenanthrene (abbreviated 4H-C[def]P), methylphenanthrenes (sum of five compounds), methyl dibenzothiophenes (sum of three compounds, abbreviated MDBTs), fluoranthene, pyrene, 3,6-dimethylphenanthrene (abbreviated 3,6-DMP), 11H-benzo[a]fluorene, 11H-benzo[b]fluorene, retene, benzo[b]naphtho[2,1-d]thiophene (abbreviated BNT), cyclopenta[cd]pyrene, benz[a]anthracene, chrysene + triphenylene (abbreviated CHR + TRI), naphthacene, benz[e]acephenanthrylene + benzo[k]fluoranthene (abbreviated BFLT), benzo[e]pyrene, benzo[a]pyrene, perylene, indeno[1,2,3-cd]pyrene (abbreviated IP), benzo[ghi]perylene, dibenz[a,h]anthracene + benzo[b]triphenylene (abbreviated DBAs), and coronene.

For each PAH, the identity and chromatographic retention time were confirmed by the use of a calibration standard. The calibration standard contained known concentrations of the deuterated surrogate compounds, deuterated internal standard compounds, and all of the compounds of interest in this study. The mass of each PAH was determined by isotopic dilution using the masses of deuterated PAH internal standard added.

**Quality Assurance.** PAH masses in the laboratory blanks were low relative to the masses in the PUF and QFF samples, accounting for  $< 4\%$  of the total PAH mass in both PUF and QFF samples. Therefore, correction for laboratory blanks was not warranted. Precipitation laboratory blanks (XAD-2) contained significant amounts of some PAHs. On average these masses represented  $< 1\%$  to  $> 100\%$  of the mass in the samples. Blank correction was therefore vital. This was particularly problematic for phenanthrene and anthracene. Surrogate recoveries were always better than 50%, averaging better than 78%, in all media, and were used to correct masses of all PAH compounds.

The method detection limit for each PAH is defined as the mean plus three times the standard deviation of the mass in the matrix-specific field blank. PAH method detection limits for gas-phase PUF samples ranged from 0.53 ng (naphthacene) to 29 ng (methylphenanthrenes) ( $n = 52$  field blanks). QFF method detection limits ranged from 0.29 ng (benzo[b]naphtho[2,1-d]thiophene) to 34 ng (methylphenanthrenes) ( $n = 51$ ). Precipitation sample (XAD) method detection limits, calculated using laboratory blank samples, ranged from 0.18 ng (cyclopenta[cd]pyrene) to 130 ng (phenanthrene) ( $n = 25$ ). PUF and QFF field blank masses

accounted for 0.53–3.9% and 0.92–3.7% of the average total sample mass, respectively.

Four split PUFs were used to quantify breakthrough of gas-phase PAHs. The second half of the split PUF accounted for  $10 \pm 4\%$  ( $n = 4$ ) of the total mass collected on the entire PUF, with the greatest percentages for the lower molecular weight PAHs: fluorene (17%), 1-methylfluorene (20%), phenanthrene (28%), and methylphenanthrenes (29%). The potential breakthrough of these compounds suggests that the gas-phase concentrations reported here may slightly underestimate the actual gas-phase concentration.

**Modeling Framework for Deposition Calculations.** The models used to estimate deposition fluxes from atmospheric concentrations are described in detail by Totten et al. (4) and will be described briefly here.

The gaseous absorption flux represents the magnitude of PAH deposition to freshwater surfaces adjacent to the NJADN monitoring stations. The Whitman two-layer model (9) was used to calculate gas absorption fluxes ( $F_{\text{gas}}$ ):

$$F_{\text{gas}} = v_{\text{aw}}(C_{\text{gas}}/K_{\text{aw}}) \quad (1)$$

where  $v_{\text{aw}}$  is the air–water exchange mass transfer coefficient ( $\text{m d}^{-1}$ ),  $C_{\text{gas}}$  is the gas-phase PAH concentration ( $\text{ng m}^{-3}$ ), and  $K_{\text{aw}}$  is the dimensionless, temperature-specific Henry law constant (10). Only the absorptive flux was calculated, since the dissolved concentration of the PAHs in regional water bodies was not measured. The Henry law constants of Bamford et al. (11) as modified by Gigliotti et al. (12) were used. The error in  $F_{\text{gas}}$  is estimated to be  $\sim 65\%$  (13). This error is described in detail in ref 13 and arises primarily from the uncertainty in the mass transfer coefficient and the Henry law constant.

The dry particle deposition flux,  $F_{\text{dry}}$  ( $\text{ng m}^{-2} \text{d}^{-1}$ ), was calculated by multiplying the concentration of PAHs on atmospheric particles,  $C_{\text{part}}$  ( $\text{ng m}^{-3}$ ), by a deposition velocity,  $v_{\text{d}}$  ( $\text{cm s}^{-1}$  or  $\text{m d}^{-1}$ ):

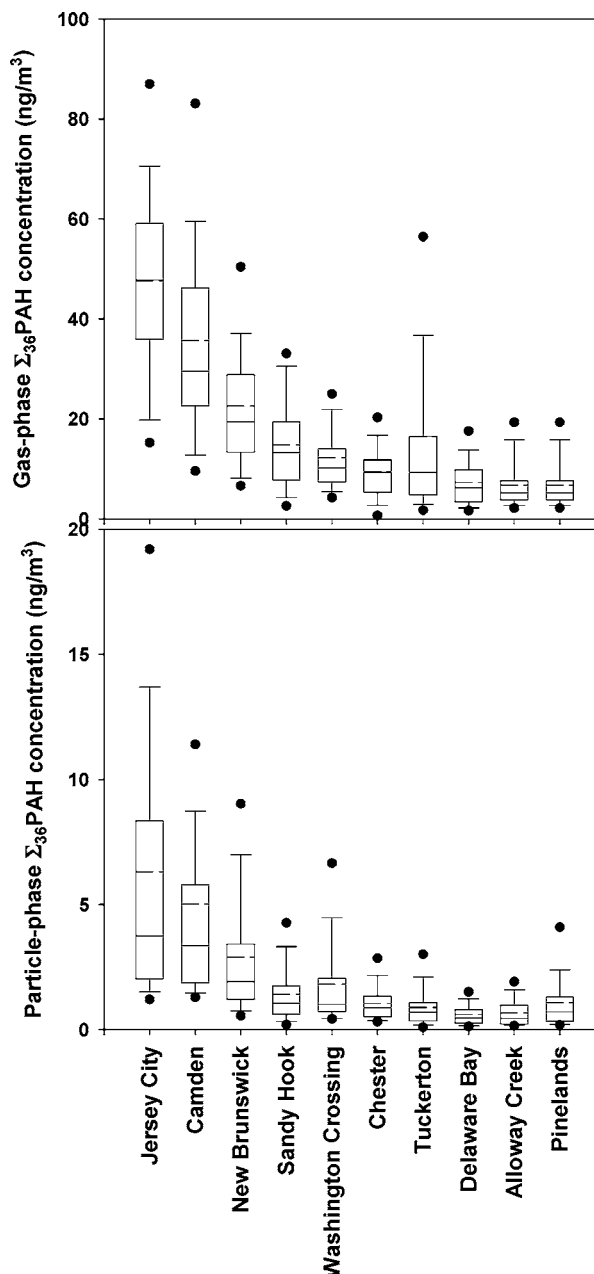
$$F_{\text{dry}} = C_{\text{part}}v_{\text{d}} \quad (2)$$

Franz et al. (14) estimated deposition velocities in the range of  $0.6\text{--}0.9 \text{ cm s}^{-1}$  for PAHs in urban areas where it was hypothesized that large particles dominate the atmospheric particle distribution. In the present study, a deposition settling velocity of  $0.5 \text{ cm s}^{-1}$  was selected to take into account the disproportionate influence that large particles have on the overall settling velocity for areas adjacent to urban and industrial regions. Other researchers (15, 16) have proposed the use of a smaller value of  $v_{\text{d}}$  of  $0.2 \text{ cm s}^{-1}$ . The Chesapeake Bay Atmospheric Deposition Survey (CBADS) used a value of  $0.49 \text{ cm s}^{-1}$  (17). A recent study suggests a large variability in deposition velocities of POPs to marine ecosystems due to the influence of different aerosol size distributions, wind speeds, etc. (18). The difference in the depositional settling velocity ( $\lambda_{v_{\text{d}}}$ ) between the selected value and other reported deposition velocities (range  $0.2\text{--}0.8 \mu\text{m}$ ) yields an error of 250% in estimated dry particle deposition fluxes.

Wet deposition fluxes,  $F_{\text{wet}}$  ( $\text{ng m}^{-2} \text{d}^{-1}$ ), are calculated by multiplying the volume-weighted mean (VWM) concentration of the PAH compound in precipitation,  $C_{\text{precip}}$  ( $\text{ng L}^{-1}$ ), by the total precipitation flux,  $P$  ( $\text{L m}^{-2} \text{d}^{-1}$ )

$$F_{\text{wet}} = C_{\text{precip}}P \quad (3)$$

A 30 y average precipitation flux of  $1.1 \text{ m y}^{-1}$  was used in these calculations, and the error in the flux was assumed to be equal to the standard error of the VWM concentration, usually less than 30% (13). At Tuckerton and Alloway Creek



**FIGURE 2.** Box and whisker plots of gas (top) and particle (bottom) phase  $\Sigma_{36}$ PAH concentrations at the 10 NJADN sites. The upper dot, upper error bar, upper edge of box, lower edge of box, lower error bar, and lower dot represent the 95th, 90th, 75th, 25th, 10th, and 5th percentile concentrations, respectively. Within each box, mean and median concentrations are shown as dashed and solid lines, respectively.

the uncertainties were larger (as high as 74%) due to the smaller number of precipitation samples collected at those sites.

## Results and Discussion

PAH concentrations are discussed first, followed by an examination of deposition fluxes.

**Concentrations: Spatial Differences.** Box and whisker plots of the gas- and particle-phase  $\Sigma_{36}$ PAH concentrations are shown in Figure 2, with the 10 NJADN sites listed in order of decreasing urban/industrialization. As would be predicted on the basis of increased density of localized, proximate PAH sources in urban and industrial areas, the highest PAH concentrations are measured at the most highly urban and

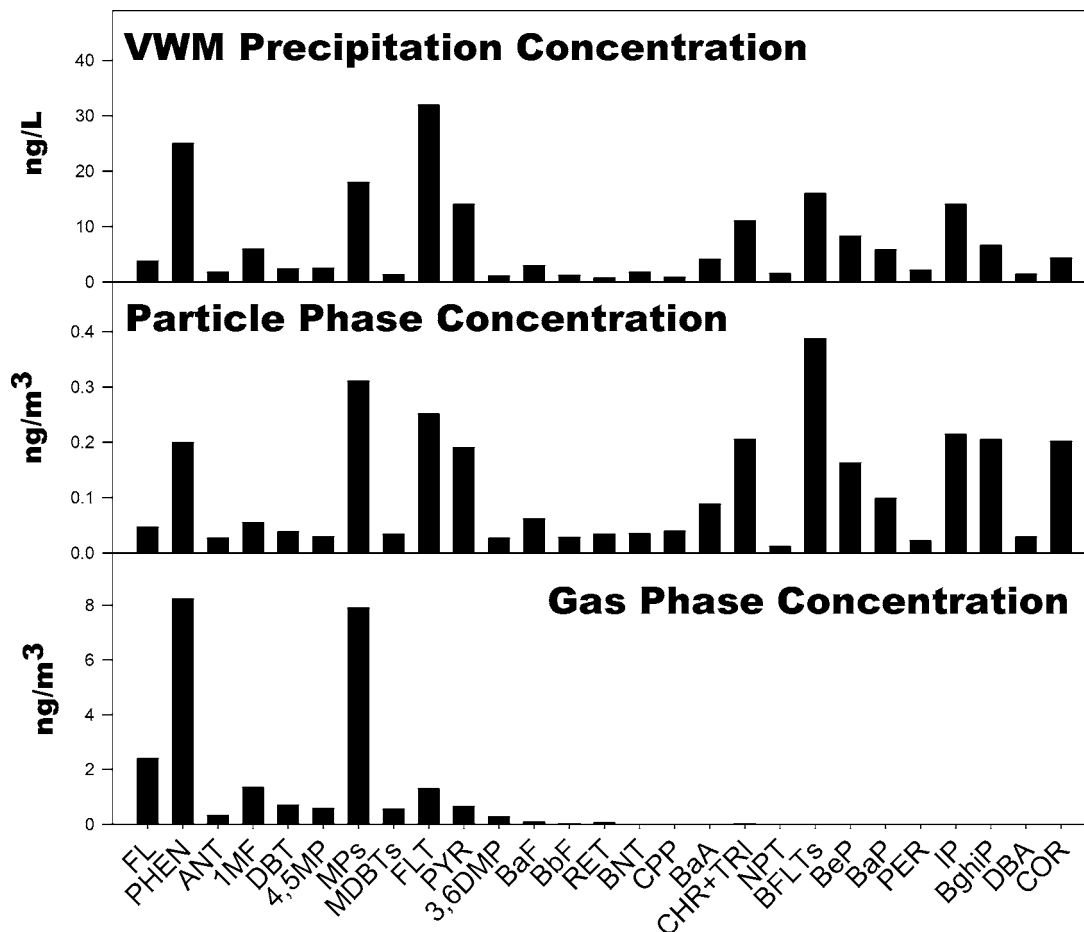


FIGURE 3. Comparison of PAH profiles among precipitation, gas-phase, and particle-phase samples at the New Brunswick station.

industrial sites, Camden and Jersey City. Midrange concentrations are measured in the suburban areas, New Brunswick, Washington Crossing, Chester, and Tuckerton. The lowest concentrations are found in coastal (Delaware Bay, Alloway Creek) and rural (Pinelands) areas, with one notable exception, the coastal Sandy Hook site opposite New York City in the Hudson River Estuary.

The Sandy Hook site, although located in a coastal area away from the immediate impact of heavy traffic arteries, industry, or urbanization, can be classified as “urban/industrial”-impacted, because it is about 15 km south of Brooklyn and 15 km southwest of Staten Island, two of the five boroughs of New York City. This finding is consistent with other studies near Lake Michigan (1) and the Chesapeake Bay (19), which are both impacted due to their proximity to large urban/industrial centers (Chicago, IL, and Baltimore, MD, respectively).

The gas-phase PAH distribution (Figure 3) is dominated by low to medium molecular weight PAHs (fluorene to chrysene) and relatively depleted in higher molecular weight PAHs (benz[*a*]anthracene to coronene). This pattern is determined in part by vapor–particle partitioning, although emission patterns also play a role. Although the average concentrations vary spatially across New Jersey, the average PAH profiles at all sites (except Alloway Creek) are statistically similar ( $r^2 = 0.92–0.99$ ,  $p < 0.05$ ), indicating that while the source strength is different at each site, the mix of sources is similar. Concentrations of gas-phase methylphenanthrenes are significantly lower at Alloway Creek than the other nine sites. If this compound is excluded, the PAH profile at the Alloway Creek site is statistically similar to those at the other nine sites ( $r^2 = 0.91–0.99$ ). Methylphenanthrenes are a component of many emissions sources, including uncom-

busted petroleum, natural gas combustion, oil combustion, and gasoline motor vehicle emissions (20, 21). Statistical analysis of the NJADN data suggests that uncombusted petroleum sources are most closely associated with methylphenanthrenes (20), but it is unclear whether the lower levels of methylphenanthrenes observed at Alloway Creek indicate lower uncombusted petroleum emissions there.

Particle-phase PAH concentrations are often an order of magnitude or more lower than gas-phase PAH concentrations, but they do display a spatial pattern similar to that seen in Figure 2. The PAH profile in the particle phase is shifted toward the high molecular weight compounds (Figure 3). As in the gas phase, the average particle-phase PAH concentrations differ spatially, but the profiles are statistically similar ( $r^2 = 0.70–0.94$ ,  $p < 0.05$ ) at all sites except Alloway Creek. Again, the methylphenanthrenes are lower at Alloway Creek than at any of the other sites. When this compound is removed from the regression, the PAH profile at Alloway Creek is statistically similar to those of all the other sites ( $r^2 = 0.70–0.89$ ).

PAH concentrations (Table 1) in urban/industrial Jersey City and Camden are similar to those measured in urban/industrial Baltimore, MD (19), and over the southern basin of Lake Michigan adjacent to Chicago, IL (1). However, concentrations measured within the city of Chicago were as much as 10 times higher than those measured at any of the NJADN sampling sites (1). PAH concentrations measured at Washington Crossing, Chester, Tuckerton, Delaware Bay, Alloway Creek, and the Pinelands are all similar in magnitude to concentrations measured over water in the Chesapeake Bay and at the three rural CBADS sites adjacent to Chesapeake Bay (Wye, Elms, and Haven Beach) (22, 23).

**TABLE 1. Gas-Phase + Particle-Phase PAH Concentrations (ng m<sup>-3</sup>) at Various Locations<sup>a</sup>**

site location	ref	phenanthrene	pyrene	benzo[a]pyrene
<b>NJADN Sites</b>				
Jersey City	this study	15 (0.19–32)	2.1 (0.018–10)	0.19 (0.012–1.4)
Camden	this study	15 (0.66–98)	1.6 (0.062–4.6)	0.14 (0.027–0.90)
New Brunswick	this study	8.4 (0.71–21)	0.85 (0.027–3.0)	0.10 (0–0.37)
Sandy Hook	this study	4.9 (0.078–16)	0.52 (0.008–2.7)	0.038 (0–0.23)
Washington Crossing	this study	3.8 (0.41–10)	0.43 (0.51–1.9)	0.077 (0.006–0.63)
Tuckerton	this study	5.7 (0.029–32)	0.41 (0.034–2.5)	0.021 (0.001–0.16)
Chester	this study	3.7 (0.029–10)	0.32 (0.032–0.94)	0.044 (0.008–0.34)
Delaware Bay	this study	2.7 (0.67–7.5)	0.22 (0.033–0.85)	0.015 (0.001–0.086)
Alloway Creek	this study	3.5 (0.043–10)	0.40 (0.037–1.7)	0.032 (0.0017–0.12)
Pinelands	this study	2.3 (0.62–17)	0.21 (0.035–1.2)	0.031 (0.003–0.26)
<b>Other Locations</b>				
Chicago, IL	1	68 (8.6–250)	14 (1.8–100)	3.1 (1.9–32)
Baltimore, MD	19	21 (8.9–63)	2.1 (1.1–4.6)	0.18 (0.030–0.48)
Lake Michigan	1	10 (0.24–34)	1.8 (0.052–8.0)	0.18 (0.008–0.85)
Chesapeake Bay	19	2.4 (1.1–7.1)	0.38 (0.18–0.88)	0.040 (0–0.090)
Wye, MD	22	2.2 (0–6.2)	0.45 (0–2.5)	0.041 (0–0.30)
Elms, MD	22	1.8 (0–12)	0.4 (0–2.6)	0.045 (0–0.51)
Haven Beach, VA	22	2.6 (0.019–17)	0.76 (0.011–4.4)	0.030 (0.0008–0.37)

<sup>a</sup> All concentration data reported as mean (range).

**Concentrations: Seasonal Differences.** For the gas phase, seasonality was examined quantitatively using a Clausius–Clapeyron-type expression in which the natural log of the partial pressure,  $\ln P$  (Pa), is plotted against inverse temperature,  $1/T$  (K<sup>-1</sup>). Negative slopes indicate that, as temperature decreases, concentrations decrease. Gas-phase concentrations may decrease due to lower emissions during cold periods (less volatilization from surfaces) or due to condensation of gas-phase emissions onto particles or surfaces. The low to medium molecular weight PAHs (those with MW < 235) displayed negative slopes in about 85% of the cases in which the slope was statistically significant ( $p < 0.05$ ), suggesting that air–surface exchange contributes to the movement of these PAHs in New Jersey. This conclusion is supported by statistical analysis of the data set suggesting that 24–27% of the atmospheric PAHs at New Brunswick, Sandy Hook, and Jersey City arise from air–surface exchange (24). The notable exception to this trend is fluorene, which displayed a positive slope at all sites where the regression was significant, indicating higher gas-phase concentrations in winter. Individual PAH compounds were least likely to yield negative slopes at Sandy Hook, Camden, and Jersey City, presumably due to their close proximity to primary sources, and thus lower influence of secondary (recycling) ones. At all other sites, more than 60% of the PAHs with significant slopes displayed negative slopes. Thus, most of the statistically significant regressions result in negative slopes at relatively remote sites such as Alloway Creek, Pinelands, Chester, and Delaware Bay, suggesting (not surprisingly) that air–surface exchange processes are more important in determining gas-phase PAH concentrations at locations further removed from stationary sources. A more complete discussion of the temperature dependence, seasonal variations, and source profiles of PAHs in the NJADN data set is given by Lee et al. (24).

The highest particle-phase PAH concentrations typically occur in the colder winter season. Regressing the natural log of TSP versus  $1/T$  shows statistically significant ( $p < 0.05$ ) negative correlations at all 10 sampling sites, revealing that the TSP mass is higher in the warmer seasons, consistent with increased secondary aerosol formation under warmer temperature conditions and sunlight (25). Particle-phase PAH concentrations expressed as nanograms of PAH per microgram of particles are therefore also highest in winter. Lower temperatures and mixing heights, decreased photolytic oxidative capacity of the atmosphere, and higher emissions can all contribute to increased particle-phase PAH concen-

trations in winter. The winter particulate PAH signal is dominated by higher molecular weight PAH compounds typically associated with atmospheric soot particles of combustion origin (1, 25–27).

**Precipitation.** Absolute PAH concentrations in precipitation vary from below detection limits (naphthacene) to as much as 3770 ng L<sup>-1</sup> (fluorene). Samples with the smallest precipitation volumes typically displayed the highest PAH concentrations. To remove the influence of the volume collected, concentrations are reported as VWMs.

Phenanthrene, the methylated phenanthrenes, fluoranthene, and pyrene dominate the VWM precipitation PAH concentrations. For five of the eight sites (New Brunswick, Sandy Hook, Chester, Tuckerton, and Pinelands), the precipitation profiles are statistically similar to the particle-phase profiles in the air (Figure 3). This similarity was also noted in data from the IADN sites (28). This suggests, as is the case for PCBs in precipitation (29), that atmospheric PAHs associated with atmospheric particles are efficiently scavenged by precipitation.

The similarity of profiles between the atmospheric particle phase and precipitation does not hold true for both the urban/industrial sites, Jersey City and Camden, where the precipitation profile is more similar to that of the gas phase. Other researchers have noted increases in particle washout ratios with distance from emission sources due to an increase in vertical dispersion (30, 31). This phenomenon could account for the differences in PAH profiles between atmospheric particles and precipitation at the urban NJADN sites.

Average VWM precipitation concentrations do not follow the same spatial pattern as the air concentrations. Although the highest concentrations occur at one of the urban/industrial sites (Jersey City), urban/industrial Camden and suburban New Brunswick demonstrate statistically similar concentrations ( $p < 0.05$ ). In contrast to air PAH concentrations, precipitation PAH concentrations were lower at the “impacted” Sandy Hook site than at the lightly suburban Tuckerton, Chester, and Alloway Creek stations.

As Table 2 shows, PAH concentrations in Chicago, IL (32), precipitation are often more than an order of magnitude larger than those measured in urban/industrial New Jersey (Jersey City and Camden). Offenber (33) measured even higher concentrations of PAHs in event-based sampling of precipitation in Chicago. Precipitation concentrations in Jersey City and Camden are similar in magnitude to those measured over the southern basin of Lake Michigan and on

**TABLE 2. Site Comparisons for Select VWM Precipitation PAH Concentration Data (ng L<sup>-1</sup>) (±Standard Error)**

site location	land-use classification	ref	phenanthrene	pyrene	benzo[a]pyrene
Jersey City, NJ	urban/industrial	this study	39 ± 6.0	23 ± 4.3	8.2 ± 1.6
Camden, NJ	urban/industrial	this study	27 ± 4.4	15 ± 3.0	5.8 ± 1.2
New Brunswick, NJ	suburban	this study	18 ± 2.5	10 ± 1.2	3.8 ± 0.47
Sandy Hook, NJ	coastal-impacted	this study	9.0 ± 1.6	5.0 ± 0.72	2.0 ± 0.33
Chester, NJ	light suburban	this study	12 ± 3.0	8.5 ± 1.7	2.9 ± 0.68
Tuckerton, NJ	coastal/light residential	this study	17 ± 4.5	14 ± 4.7	2.2 ± 0.71
Alloway Creek, NJ	rural/coastal	this study	10 ± 7.7	7.0 ± 4.1	2.3 ± 1.6
Pinelands, NJ	rural	this study	4.0 ± 0.55	3.3 ± 0.41	1.5 ± 0.20
Chicago, IL	urban/industrial	32	360	340	200
Lake Michigan	over water	33	31	26	15
South Haven, MI	coastal-impacted	33	39	28	21
Wye, MD	rural	22	3.0	1.9	0.63
Elms, MD	rural	22	4.7	9.0	1.8
Haven Beach, VA	rural	22	4.3	2.1	0.70

**TABLE 3. Average Annual Gas Absorption Fluxes of PAHs at NJADN Sites (ng m<sup>-2</sup> d<sup>-1</sup>)<sup>a</sup>**

PAH	JC	CC	NB	SH	CH	WC	TK	DB	AC	PL
fluorene	1587	812	214	582	99	372	580	46	96	128
phenanthrene	4923	3860	1015	1764	310	1038	1977	136	579	351
anthracene	241	149	31	47	4.3	26	63	3.0	12	5.2
1-methylfluorene	685	386	120	332	25	175	153	12	67	44
dibenzothiophene	517	298	65	169	24	103	189	12	52	32
4H-C[de]P	477	320	66	134	16	76	168	8.2	33	21
methylphenanthrenes	5039	3683	1214	2160	201	1070	1875	113	55	298
MDBTs	429	350	58	185	23	174	169	17	12	39
fluoranthene	1147	755	204	373	65	158	395	26	115	44
pyrene	706	477	113	199	29	101	151	14	69	31
3,6-DMP	275	169	31	84	6.1	39	57	3.1	26	9.5
benzo[a]fluorene	106	60	9.1	19	2.0	11	14	1.4	6.4	2.8
benzo[b]fluorene	26	15	3.2	5.0	0.72	2.7	4.4	0.48	3.1	0.55
retene	31	28	8.7	17	3.2	18	16	10	17	7.2
BNT	7.6	11	0.57	4.6	0.32	2.6	3.3	0.47	2.3	1.4
cyclopenta[cd]pyrene	3.5	1.3	1.4	0.23	0.12	0.33	0.43	0.10	0.12	0.34
benz[a]anthracene	4.5	3.3	0.61	0.68	0.17	0.84	0.60	0.18	0.30	0.22
CHR + TRI	21	20	4.5	5.5	1.4	5.0	6.2	1.5	7.6	1.7
naphthalene	1.5	0.41	0.46	0.087	0.037	0.017	0.15	0.0038	0.19	0.10
BFLTs	4.4	2.9	1.1	0.83	0.13	0.78	0.99	0.14	0.55	0.14
benzo[e]pyrene	1.9	2.0	0.56	0.64	0.10	0.38	0.80	0.14	0.63	0.079
benzo[a]pyrene	1.8	1.4	0.40	0.44	0.057	0.22	0.49	0.043	0.81	0.055
perylene	0.40	0.28	0.091	0.097	0.0081	0.038	0.094	0.0041	0.46	0.0085
IP	1.3	1.3	0.34	0.37	0.077	0.12	0.31	0.040	0.17	0.042
benzo[ghi]perylene	0.99	0.66	0.51	0.29	0.024	0.062	0.14	0.013	0.26	0.021
DBAs	0.18	0.10	0.038	0.054	0.044	0.053	0.089	0.022	0.017	0.023
coronene	0.77	0.63	0.29	0.38	0.23	0.30	0.51	0.13	0.12	0.16
Σ <sub>36</sub> PAHs	16239	11407	3165	6084	810	3374	5824	405	1155	1017

<sup>a</sup> The error in these fluxes is about 65%. PAHs are listed in order of decreasing vapor pressure. JC = Jersey City, CC = Camden, NB = New Brunswick, SH = Sandy Hook, CH = Chester, WC = Washington Crossing, TK = Tuckerton, DB = Delaware Bay, AC = Alloway Creek, and PL = Pinelands

its eastern shore at South Haven (33). Precipitation concentrations at the rural Pinelands were similar to those measured at the three rural CBADS sites adjacent to Chesapeake Bay (Wye, Elms, and Haven Beach) (22).

**Deposition.** The distribution of target PAHs between the gas and particle phases in the atmosphere is important in determining the dominant deposition process and the magnitude of total atmospheric deposition. For example, phenanthrene occurs predominantly in the gas phase and thus can be loaded by gas absorption to surfaces of aquatic or terrestrial environments. In contrast, benzo[a]pyrene is primarily particle-bound in the atmosphere and thus is removed by wet and dry particle deposition with only a minor contribution from gas-phase absorption. This demonstrates the importance of measuring individual compounds in the atmospheric gaseous and particle phases for the estimation of atmospheric deposition. The data presented herein represent the first comprehensive estimates of PAH deposition to New Jersey.

**Gas Absorption.** Gross gas absorption fluxes to the surfaces of water bodies (Table 3) are driven by a combination of atmospheric concentration and wind speed. In general, the gas-phase concentration is the most important parameter, such that urban/industrial sites experience higher gas absorption fluxes. However, annual mean wind speeds were significantly lower at New Brunswick (averaging 1.8 m s<sup>-1</sup>) than at Sandy Hook (5.4 m s<sup>-1</sup>) and Tuckerton (5.0 m s<sup>-1</sup>). As a result, gross gas absorption fluxes are lower at New Brunswick despite higher gas-phase concentrations there. If the fluxes at all sites are calculated using an average wind speed of 5.0 m s<sup>-1</sup>, fluxes and gas-phase concentrations are perfectly correlated. There is no obvious seasonal trend in gas absorption fluxes.

Fluxes measured at sites adjacent to Lake Superior, Lake Michigan, and Lake Ontario (16) are on the same order as those measured at the Chester and Pinelands NJADN sites, which represent two of the sites with the lowest measured

**TABLE 4. Atmospheric Deposition Fluxes ( $\text{ng m}^{-2} \text{d}^{-1}$ ) of Select PAHs at IADN Sites (16), CBADS Sites (22), and Selected NJADN Sites<sup>a</sup>**

	phenanthrene	pyrene	benzo[ <i>b+k</i> ]fluoranthene	benzo[ <i>a</i> ]pyrene
<b>Gross Gas Absorption</b>				
IADN sites				
Eagle Harbor	260	67	NM	3.3
Sleeping Bear Dunes	308	41	NM	4.4
Sturgeon Point	1173	192	NM	5.4
Point Petre	246	52	NM	1.1
NJADN sites				
Jersey City	4923	706	4.4	1.8
New Brunswick	1015	113	1.1	0.40
Sandy Hook	1764	199	0.83	0.44
Pinelands	351	31	0.14	0.055
<b>Dry Particle Deposition</b>				
IADN sites				
Eagle Harbor	3.3	4.0	NM	1.9
Sleeping Bear Dunes	5.2	6.6	NM	3.6
Sturgeon Point	9.2	12	NM	6.7
Point Petre	6.8	10	NM	8.7
CBADS sites				
Wye	9.9	7.7	13	3.3
Elms	25	26	43	9.0
Haven Beach	20	2.4	35	7.9
NJADN sites				
Jersey City	132	184	302	83
New Brunswick	90	86	179	45
Sandy Hook	43	39	71	18
Pinelands	31	33	52	13
<b>Wet Deposition</b>				
IADN sites				
Eagle Harbor	8.7	5.3	NM	4.7
Sleeping Bear Dunes	17	10	NM	8.1
Sturgeon Point	53	35	NM	19
Point Petre	10	9.5	NM	8.1
CBADS sites				
Wye	10	6.0	7.1	2.7
Elms	11	6.3	8.5	1.9
Haven Beach	8.2	3.3	5.2	1.1
NJADN sites				
Jersey City	118	69	50	25
New Brunswick	54	31	38	11
Sandy Hook	27	15	20	6.0
Pinelands	12	10	11	4.4

<sup>a</sup> NM = not measured.

fluxes in New Jersey (Table 4). The annual average flux at Lake Erie resembles most closely the Washington Crossing and New Brunswick fluxes, which are midrange fluxes relative to that of the rest of New Jersey.

**Wet Deposition.** Precipitation intensity is fairly constant across New Jersey at about  $1.1 \text{ m y}^{-1}$ , so that VWM concentrations of contaminants in precipitation give a relatively accurate picture of the spatial variations in PAH wet deposition fluxes at the NJADN sites. As with the gas absorption fluxes, the wet deposition fluxes are not well correlated with the degree of urbanization/industrialization (Table 5). If they were, Camden would have the second-highest  $\Sigma_{36}\text{PAH}$  wet deposition flux. Instead, New Brunswick claims that distinction. The Sandy Hook, Chester, Tuckerton, Alloway Creek, and Pinelands sites all have similar annual average fluxes.

Wet deposition fluxes at all of the rural CBADS (22) and background IADN (16) sites are, on average, lower than those calculated at the NJADN sampling stations (Table 4). Fluxes at the Sturgeon Point site, about 30 km from Buffalo, NY, are most similar in magnitude to those at the light residential/coastal Tuckerton site.

**Dry Deposition.** By the rubric used to calculate dry deposition fluxes, they must necessarily follow the same trend as the average particle-phase PAH concentrations (Table 6).

Dry particle deposition fluxes are therefore correlated with the degree of urbanization such that urban/industrial sites > suburban sites > light suburban sites > coastal sites.

In Table 4, all flux data from the IADN (16) and CBADS (22) studies have been recalculated using a deposition settling velocity of  $0.5 \text{ cm s}^{-1}$  to perform a more valid comparison with NJADN dry deposition fluxes. Dry particle deposition fluxes at the remote IADN sites and the CBADS coastal Chesapeake Bay sites are similar in magnitude to those of the nonurban/industrial NJADN stations. The ratio of phenanthrene to benzo[*b+k*]fluoranthene is nearly unity at all three coastal CBADS sites, but is significantly lower at the majority of NJADN sites. However, both this ratio and the PAH concentrations are similar to those of the CBADS sites at Tuckerton and Delaware Bay. This similarity suggests that the coastal meteorology and PAH sources may be similar among these CBADS and NJADN sites.

**Annual Deposition across the State of New Jersey.** Examination of the various deposition fluxes reveals some trends. For primarily gas-phase PAHs with molecular weights of less than 200, gas absorption represents >90% of their total (wet + dry + gas) atmospheric deposition to water bodies. For PAHs with molecular weights between 200 and 235, all three modes of deposition to water surfaces are important. Particle-bound PAHs with molecular weights greater than 235 undergo

**TABLE 5. Annual Average Wet Deposition Fluxes (ng m<sup>-2</sup> d<sup>-1</sup>) at NJADN Sites Assuming 1.1 m y<sup>-1</sup> Precipitation<sup>a</sup>**

PAH	JC	CC	NB	SH	CH	TK	AC	PL
fluorene	23	14	10	7.3	7.1	11	6.4	3.9
phenanthrene	118	83	54	27	36	52	31	12
anthracene	11	6.5	4.1	3.2	2.8	4.1	3.7	0.71
1-methylfluorene	18	11	16	9.1	2.7	4.5	4.1	3.2
dibenzothiophene	12	8.2	6.7	9.8	3.9	4.4	2.9	1.6
4H-C[def]P	15	9.1	6.3	3.6	3.6	5.1	2.8	1.5
methylphenanthrenes	112	65	48	31	23	30	4.0	10
MDBTs	8.4	4.2	3.5	3.2	1.9	1.9	0.71	0.64
fluoranthene	98	71	51	23	44	71	41	15
pyrene	69	47	31	15	26	41	21	10
3,6-DMP	7.6	4.0	3.0	3.2	1.5	1.1	3.1	0.71
benzo[a]fluorene	15	9.5	6.9	5.1	4.7	6.5	3.4	2.1
benzo[b]fluorene	6.2	8.4	2.7	1.7	1.9	2.5	3.6	0.88
retene	3.9	5.6	2.1	1.3	1.5	0.71	3.3	1.7
BNT	8.1	6.5	3.4	2.5	3.9	5.0	3.0	1.5
cyclopenta[cd]pyrene	2.3	6.1	2.4	1.5	1.0	1.9	1.3	0.57
benz[a]anthracene	23	15	8.7	4.8	5.6	9.1	3.3	2.5
CHR + TRI	41	32	23	11	18	26	19	7.0
naphthacene	46	9.4	4.3	2.3	3.0	4.9	1.3	1.5
BFLTs	50	42	38	20	25	19	22	11
benzo[e]pyrene	31	24	17	9.1	15	15	16	7.5
benzo[a]pyrene	25	17	11	6.0	8.7	6.6	7.1	4.4
perylene	8.2	5.9	4.3	2.6	2.7	1.3	4.0	1.9
IP	42	32	22	13	19	12	9.6	8.8
benzo[ghi]perylene	24	18	13	7.3	8.4	5.6	10	5.5
DBAs	6.4	4.3	2.5	1.3	1.5	1.1	1.6	0.77
coronene	17	9.6	8.1	5.3	4.4	1.6	6.4	2.7
Σ <sub>36</sub> PAHs	816	533	402	224	231	342	64	117

<sup>a</sup> Abbreviations are the same as in Table 3.

**TABLE 6. Annual Average Dry Particle Deposition Fluxes (ng m<sup>-2</sup> d<sup>-1</sup>)<sup>a</sup>**

PAH	JC	CC	NB	SH	CH	WC	TK	DB	AC	PL
fluorene	21	7.9	18	9.6	2.8	4.6	3.2	9.4	1.9	3.2
phenanthrene	132	141	90	43	28	44	16	30	19	31
anthracene	38	16	12	7.2	3.8	3.9	2.4	3.6	2.5	2.6
1-methylfluorene	23	8.5	19	5.1	3.8	7.4	3.0	3.8	3.2	4.8
dibenzothiophene	39	15	17	8.3	2.8	4.7	2.5	2.7	2.7	2.5
4H-C[def]P	30	18	13	6.5	4.5	6.5	2.4	2.9	2.5	4.2
methylphenanthrenes	251	160	142	77	39	59	19	25	3.7	42
MDBTs	32	17	15	8.8	3.6	6.3	2.3	2.6	0.89	3.7
fluoranthene	196	130	113	49	41	57	21	23	26	38
pyrene	184	106	86	39	33	55	20	19	21	33
3,6-DMP	28	14	12	5.0	2.5	4.5	1.3	1.0	2.8	2.5
benzo[a]fluorene	79	32	29	11	8.7	13	4.0	3.6	5.7	7.1
benzo[b]fluorene	37	12	13	4.7	4.9	5.6	1.7	1.9	2.8	2.8
retene	34	28	15	7.9	14	21	13	20	12	15
BNT	41	19	16	6.3	4.9	6.0	2.5	3.0	3.0	3.3
cyclopenta[cd]pyrene	33	18	17	4.5	5.8	11	2.2	1.9	3.3	3.9
benz[a]anthracene	106	53	42	14	14	23	6.8	5.7	5.9	10
CHR + TRI	192	107	93	38	34	51	15	18	18	25
naphthacene	24	10	4.9	1.4	7.3	2.5	0.11	0.79	1.2	0.82
BFLTs	302	203	179	71	62	94	22	31	30	52
benzo[e]pyrene	179	90	74	33	26	46	13	16	30	23
benzo[a]pyrene	83	53	45	18	19	29	4.8	7.2	12	13
perylene	25	15	10	4.3	6.8	6.8	2.5	9.0	2.9	6.6
IP	236	152	100	50	31	74	13	20	23	37
benzo[ghi]perylene	185	133	96	38	25	55	13	9.5	23	28
DBAs	23	13	14	5.6	3.4	6.0	1.6	1.7	2.8	2.4
coronene	229	143	93	40	25	44	15	13	19	27
Σ <sub>36</sub> PAHs	2781	1713	1379	608	459	742	224	284	283	427

<sup>a</sup> Abbreviations are the same as in Table 3.

atmospheric deposition to water surfaces primarily (>95%) through dry particle and wet deposition. These conclusions are true regardless of whether a dry deposition velocity of 0.5 or 0.2 cm s<sup>-1</sup> is used. Differences in gas-particle distribution are therefore primarily responsible for the relative importance of the three atmospheric deposition processes discussed above. The importance of dry particle and wet deposition

for PAHs is due to the strong association of these compounds to aerosol soot carbon (27). Conversely, for other compounds without this strong tendency to partition to soot, such as PCBs, absorption dominates as a deposition process (18).

For the Σ<sub>36</sub>PAHs, gas absorption represents the single largest component of total atmospheric deposition to water surfaces for each of the sites (55–92%), followed by dry

particle deposition (4–31%). Wet deposition constitutes the smallest fraction of the total atmospheric flux (3–16%).

Annual average  $\Sigma_{36}$ PAH total atmospheric deposition fluxes to water surfaces in New Jersey and the region range from 540 to 7300  $\mu\text{g m}^{-2} \text{y}^{-1}$ . The order of highest to lowest fluxes follows the trend Jersey City, Camden, Sandy Hook, Tuckerton, New Brunswick, Alloway Creek, Pinelands, and Chester. Delaware Bay and Washington Crossing are not included in the trend evaluation due to a lack of precipitation data. Overall, the fluxes increase with degree of urbanization.

It is useful to consider whether atmospheric deposition (that is, inputs from the atmosphere) exceeds volatilization (outputs to the atmosphere) for urban systems. In particular, the New York/New Jersey Harbor Estuary is of interest due to the economic and ecological importance of this system. Gigliotti et al. (12) indicated that while most PAHs exhibit positive (volatative) fluxes during a sampling cruise conducted in 1998, a few compounds, particularly phenanthrene and methylphenanthrenes, typically exhibited negative (absorptive) net instantaneous air–water exchange fluxes. It is difficult to extrapolate these instantaneous fluxes to an assessment of the behavior of PAHs over longer time scales. Using NJADN data, we estimate that atmospheric deposition (wet + dry + gas) contributes  $\sim 500$ – $1500 \text{ kg y}^{-1}$  of phenanthrene,  $\sim 75$ – $300 \text{ kg y}^{-1}$  of pyrene, and  $\sim 7$ – $30 \text{ kg y}^{-1}$  of benzo[*a*]pyrene to the New York/New Jersey Harbor. Data from the Contaminant Assessment and Reduction Project (CARP) (34) indicate that most PAHs in the harbor are at concentrations that put them in a near-steady-state condition relative to their atmospheric inputs (wet + dry + gas).

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