Atmospheric Deposition of Polycyclic Aromatic Hydrocarbons at Casco Bay, Maine

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Summary - A novel collector has been designed, constructed, deployed and used for measuring wet and dry deposition of Polycyclic Aromatic Hydrocarbons (PAHs) at the CBEP monitoring site, Wolfe's Neck, on the shore of Casco Bay, about 30 km northeast from Portland, Maine. The wet/dry collector is a modified Aerochem-Metrics acid deposition collector. Wet deposition (rain and snow) is collected in a funnel which drains into a shielded and temperature controlled receiving bottle. Dry deposition of gaseous and particulate PAHs is also collected in a funnel which is filled with water almost to the rim. The water is constantly pumped into the funnel, and the overflow is drained into a receiving bottle. In this fashion dry deposition occurs on a water surface, thus simulating the uptake of PAHs onto the ocean surface. The receiving bottles are brought to the laboratory where PAHs are extracted from the aqueous solution/suspension by Solid Phase Extraction (SPE) for analysis by Gas Chromatography-Mass Spectrometry (GC-MS). Measurements started in March 1998 and ended in May 2000.Weekly samples were taken, one or more samples per month. Wet samples were obtained for 32 weeks, and dry samples for 41 weeks. For 6 weeks two simultaneous wet samples and for 7 weeks two simultaneous dry samples were obtained using the "old" and "new" configuration of the wet/dry collector, making the total count 38 wet and 48 dry samples. The old and new collectors gave comparable results. Sixteen PAH species were analyzed, ranging from acenaphthylene to coronene. The 17th specie, naphthalene, gave spurious results, and is not included in the data.

The average wet deposition of the 38 samples is 831 ng m^{-2} per cm precipitation. The normal annual precipitation at Portland, ME is 109 cm. Thus, the estimated annual wet deposition of the sum of 16 PAH species in the 1998-2000 period at Wolfe's Neck is 90.5 µg m^{-2} y^{-1}. The average wet deposition for the heating season (15 Oct-15 Apr) is 872 ng m^{-2} cm^{-1}; for the non-heating season (15 Apr-15 Oct), 790 ng m^{-2} cm^{-1}. Evidently, during the heating season more PAHs are emitted due to increased use of fossil fuels for space heating.

The average dry deposition of the 48 samples is 9.3 ng m^{-2} hr^{-1}. Multiplying by 8760 hours, the estimated annual dry deposition of 16 PAH species in the 1998-2000 period at Wolfe's Neck is 81.5 µg m^{-2} y^{-1}. The average dry deposition for the heating season is 10.4 ng m^{-2} hr^{-1}; for the non-heating season 8.2 ng m^{-2} hr^{-1}. This demonstrates again that PAH emissions are greater

during the winter months than in the summer months.

The average composition of the wet samples is: phenanthrene 27%, fluoranthene 13%, pyrene 10.5%, fluorene 9%, benzo(b and k)fluoranthene 9.5%, chrysene 4%, indeno(1,2,3,-c,d)pyrene 4%, benzo(g,h,i)perylene 4% and others 19%. The average composition of the dry samples is: fluoranthene 22%, pyrene 18%, acenaphthylene 12%, benzo(b and k)fluoranthene 11.5%, benzo(a)pyrene 6%, chrysene 6%, anthracene 4%, benzo(g,h,i)perylene 4%, indeno(1,2,3,-c,d)pyrene 4% and others 12.5%. The different composition of wet and dry samples indicates different sources, solubility in cloud droplets, and different pathways for PAHs in wet and dry deposition, respectively. Most likely, wet deposition originates from regional sources, whereas dry deposition is of local origin.

1. Introduction

Sediments in Casco Bay, Maine, show high concentrations of toxic metals, polycyclic aromatic hydrocarbons (PAHs) and other toxic pollutants [1]. The sources of these pollutants are unknown, but it is suspected that atmospheric deposition is a major source [2]. Especially, PAHs are thought to originate mainly from atmospheric deposition, because PAHs are found in the emissions of fossil fuel (coal, oil and natural gas) and biomass (e.g. wood) combustion. Fossil fuels and their products are combusted in boilers, kilns, furnaces, open fires and internal combustion engines, such as automobiles, busses, trucks, tractors, lawn mowers, off-road vehicles, ships, boats, locomotives and aircraft. Other sources of PAHs in the sediments could be oil spills and accidental or deliberate dumping into the sea of coal tar, asphalt, crank case oil and creosote leaching from wood piles. Some of the PAHs are known or suspected carcinogens to humans and animals. Especially, bottom feeding fish and crustaceans are likely to ingest PAHs, causing tumors and lesions to the animals, and possibly passing them on to humans in the food chain.

The Casco Bay Estuary Project (CBEP) established a major monitoring effort to determine the likely sources of toxic contamination of Casco Bay [3]. A monitoring site has been set up at Wolfe’s Neck, near Freeport, Maine, a peninsula jutting out into Casco Bay. At the Wolfe’s Neck site monitoring is conducted for atmospheric concentrations and depositions of mercury, cadmium, fine particles, nitrogenous species and PAHs.

The University of Massachusetts Lowell (UML), Department of Environmental, Earth and Atmospheric Sciences, and Department of Chemistry, was contracted by CBEP to construct, deploy and measure atmospheric deposition of PAHs at Wolfe’s Neck. The contract was administered by the University of Southern Maine.

This report describes the measurement technique and the results of the two year’s measurements.

2. Experimental

2.1 The UML Wet/Dry PAH Collector

The basis of the UML wet/dry collector is an Aerochem-Metrics Corp., Bushnell, FL acid deposition collector. The original collector uses plastic buckets, one for wet, the other for dry deposition sampling. A precipitation sensor activates a shuttling lid. When precipitation falls, the wet bucket is open; during dry periods the dry bucket is open. In the UML collector, the buckets are replaced with funnels. Separate bases are used for the wet and dry collector, respectively. A photo of the UML wet/dry collector is presented in Figure 1.

During the contractual period two types of wet/dry collectors were used. In the first year, the so-called “old,” and in the second year the “new” collector was used. The major difference is that in the old collector a Solid Phase Extraction (SPE) cartridge was inserted in the field, and PAHs were extracted *in situ*. In the new collector, the aqueous samples flow directly into receiving bottles, and PAH extraction is done in the laboratory. The proper functioning of the SPE cartridge in the field posed technical problems which could not be solved without frequent personal attendance, thus the old collector was replaced with the new one. In the following we describe only the new collector.

Wet collector. The square shaped wet funnel is made of steel, the inside is coated with teflon, top area 0.078 m^2, about 8 L volume. It has a slanted bottom with a drain hole. All precipitation (rain or snow) falling during a collection period is collected in the wet funnel and flows immediately through the drain hole via a 1.25 cm ID teflon tube into a 20 L glass receiving bottle, located in the insulated bottom cabinet. A thermostated blower keeps the receiving bottle above freezing temperature, as well as directing warm air toward the bottom of the funnel, so that falling snow melts immediately.

Dry collector. The dry collector exposes an artificial water surface to dry deposition of particulate and vapor phase atmospheric pollutants. In this fashion, the dry collector simulates the uptake of pollutants onto a natural surface water, in this case, the dry deposition of PAHs onto the ocean surface. The dry funnel is also square shaped, teflon coated steel, 0.078 m^2 top area, 2.5 L volume. Purified water is continuously circulated through the funnel. Water is supplied from a 20 L glass bottle by a diaphragm pump via a flow meter at a rate of 1-1.5 mL/min. A teflon overflow tube with a 1 cm bore is inserted into the funnel drain hole, reaching to within 0.5 cm of the rim of the funnel. When the water reaches over the top of the overflow tube, it drains through the overflow tube and a 0.5 cm ID teflon tube into a receiving 20 L glass bottle. During hot summer days some evaporative losses of water may occur, such that the water level reaches only 0.5-1 cm below the overflow tube. The water level reaches its proper height during cooler periods and at night. During gusty wind conditions some spillage may occur over the rim of the funnel. Because of the square shape of the funnel, the surface area of the water is not affected significantly by the change of height of the water level. In a seven days sampling period 10-15 L water flows through the funnel into the receiving bottle. At the end of a sampling period, the residual water in the funnel is drained via a drain hole into the receiving bottle. A photo of the interior of the dry collector with the supply and receiving bottles, and the flow meter is shown in Figure 2. A top view of the funnel with the overflow tube is shown in Figure 3. The wavy surface of the water is caused by the vibrations of the diaphragm pump.

2.2 PAH Analysis

After a sampling period, the receiving bottles are brought to the laboratory for PAH analysis. First, 50 µL of a 1µg/mL standard solution of surrogate PAHs is added to both the wet and dry samples. The surrogates consist of 4 deuterated PAHs: acenaphthene-d10, anthracene-d10, chrysene-d12 and perylene-d12 (SUPELCO, Bellefonte, PA) in 100 mL aqueous solution. The concentration of the surrogates varies between 5-50 ng/L in the samples, depending on the sample volume. From the recovery rates of the surrogates we estimate the recovery rates of the corresponding ambient PAHs during the analytical process, including extraction, elution, concentration and GC. Table 1 lists the 16 PAH species, their abbreviations, chemical structure, and the corresponding deuterated surrogates.

The content of the receiving bottles with the ambient suspended particulate and dissolved PAHs, and the surrogate PAHs, is transferred by a syphon method to a SPE cartridge. After complete syphoning, the receiving bottles are rinsed with purified water, and the water rinse is also transferred to the SPE cartridge.

The SPE cartridge is made of polyethylene, 20 mL volume, packed with 5 g ENVI-18, a C-18 type sorbent, supplied by SUPELCO. The packing is held in place by top and bottom frits (20 µm pore size), made of polypropylene. The ENVI-18 is conditioned first with methanol, then with water. Two aliquots of 10 mL each of methylene chloride (CH_2Cl_2) are used to elute the PAHs from the ENVI-18 sorbent. An internal standard, anthracene-d12, is added to the eluted solution. The internal standard serves as a volumetric marker. The eluted solution is concentrated to approximately 0.5 mL in a Kuderna-Danish concentrator. The remaining solution is further concentrated by cold nitrogen evaporation to about 50 µL. Two µL (i.e. about 4% of the original sample) are injected into a GC-MS. The precise level of concentration is determined by measuring the amount of retrieved internal standard, not by volumetry.

A Hewlett-Packard model 5890 Series II/5971A GC-MS is used with imbedded library for identifying the analytes. The MS is used in the selected ion monitoring mode (SIM), which quantifies only the preselected PAH peaks. The SIM mode allows quick identification and quantitation of the preselected mass peaks. On the other hand, non-preselected peaks, e.g. substituted PAHs and other pollutants are not quantified. The column is a 30 m×0.25 mm ID PTE-5 (5% diphenyl - 95% dimethylpolysiloxane) capillary with a film thickness of 0.25 µm (SUPELCO). A 3-5 m uncoated deactivated silica capillary column (Restek Corp., Bellefonte, PA) is used as a guard column. Ultra pure (99.999%) helium is used as a carrier gas. A 4 mm ID goose neck glass inlet liner packed with glass wool is inserted into the injection port. The glass wool is deactivated with 5% dimethylpolysiloxane in toluene prior to use. The 2 µL concentrates are injected in the splitless mode at 1 min split delay. Injection temperature is 300^{o}C; oven temperature program starts at 35^{o}C, held for 20 min, increased to 100^{o}C at 20^{o}C/min, increased to 200^{o}C at 8^{o}C/min, finally increased to 300^{o}C at 5^{o}C/min, held for 20 min. Transfer line temperature is 300^{o}C.

The identity and subsequent retention time of each PAH is confirmed by the use of a standard PAH mix (EPA mix 525). Calibration of the ion peaks is performed by a five point linear regression analysis. The calibration is performed once every week. The minimum quantitation limit for most PAHs is 1-3 ng, although sub-nanogram peaks are discernible.

2.3 Quality Control - Quality Assurance

Basically, the analytical procedure is in consonance with EPA method 525 for determining organic pollutants in drinking water. Ultrapure water obtained with reverse osmosis and organic free filter (Barnstead/Thermolyne, Dubuque, IA) is used for preparing solutions, rinsing the bottles and funnels. Solvents are HPLC grade. NIST certified standards and surrogates are used. Laboratory glassware is rinsed with water and methanol, then baked at 300°C.When peak shapes deteriorate, the GC capillary column is cleansed or replaced. Two injections per sample are GC-MS analyzed and averaged.

In spite of the ultrapure water and HPLC grade solvents used, interfering peaks show up in the gas chromatogram at the preselected PAH mass peaks when the sample does not contain added PAHs. These interferences may be due to PAHs or their simulants leaching from glassware or from ENVI-18. The interferences are called blanks. They are particularly noticeable for the more volatile PAHs, such as naphthalene, fluorene and phenanthrene. The amount of blank is dependent on the volume of water that passes through the ENVI-18 cartridge. A calibration curve was produced of blank amounts vs. water volume. Blank amounts are subtracted from the measured PAH peaks depending on the volume of water used in each sample. The estimated error band of the analytical procedure is ±30%. Because the excessive amounts of the naphthalene peak found in the blanks, ambient naphthalene deposition was difficult to quantify. Therefore, no naphthalene data are reported.

2.4 Recovery Rates

For the EPA Mix 525 PAH solution in water, the SPE method yields recovery rates from 60-110% (SD ±15%). For the 4 deuterated surrogate PAHs, which are added to the retrieved samples, the recovery rates are lower, in the range of 46-74% (SD±12%). The relatively low recovery rates of the surrogates are attributed to several factors: (a) volatilization during the drying step, (b) adsorption on glass and tubing surfaces, (c) degradation during storage time, and most importantly, (d) breakthrough occuring in the SPE cartridge. The latter is dependent on the volume of water of the samples. The more water passes the SPE cartridge the more breakthrough may occur. Note that up to 20 L of water is passed through the SPE cartridge. The recovery rates of the surrogates are applied to estimate the recovery rates of the corresponding ambient PAHs.

3. Results and Discussion

Measurements started in March 1998 and ended in May 2000. Weekly samples of wet and dry deposition were taken, usually one week per month. In addition, in the middle of the seasons, four weekly samples were taken. When instrumentation failure occurred, a repeat sample was collected during the following week. In 1998/99 the old wet/dry collector was used, in 1999 for several weeks both the old and new collector were used simultaneously for comparison purposes. Since comparisons were satisfactory, later on in 1999 and in 2000, only the new collector was used. Altogether, wet samples were obtained for 32 weeks, and dry samples for 41 weeks. For 6 weeks two simultaneous wet samples and for 7 weeks two simultaneous dry samples were obtained, making the total count 38 wet and 48 dry samples.

3.2 Wet Deposition

Wet deposition rates for the 38 samples are presented in Table 2. Wet samples were analyzed only when more than 250 mL of precipitation was collected during the week of sampling. The top line lists the end date of the sampling period; the next line indicates whether the old and new collector was used, respectively. The third line indicates the total precipitation in cm during the week. The left column lists the 16 PAH compounds analyzed . The units are ng m^{-2}. NQ means not-quantified, i.e. the GC-MS peaks were either absent or too small to be accurately quantified. Note that blanks were subtracted from the raw data, and recovery rates of the deuterated surrogates were applied to the corresponding PAHs in order to calculate their concentrations in the samples. The bottom line gives the deposition rate for the sum of 16 PAH species, also in units of ng m^{-2}. The range is from 313 to 5663 ng m^{-2}, with an average of 1600 ng m^{-2}. The amount of rainfall varied between 0.38 to 31.5 cm/week. There is no clear correlation between amount of rainfall and amount of deposition; in fact, often small amounts of precipitation bring more PAH wet deposition than large amounts of precipitation. For example, the 0.84 cm precipitation event brought 4071ng m^{-2} wet deposition, the 31.5 cm precipitation event, 2161 ng m^{-2} .

The average wet deposition of the sum of 16 PAH species *per cm* of precipitation is 831 ng m^{-2} cm^{-1}. The normal annual rainfall at Portland, ME is 109 cm. Multiplying 831 ng m^{-2} cm^{-1} by 109 cm, the estimated annual wet deposition of the sum of 16 PAH species at Wolfe's Neck in the 1998-2000 period is 91 µg m^{-2} yr^{-1}. During the same period, under the sponsorship of the MIT College Seagrant, our team measured wet and dry deposition at Nahant, MA, on the shore of Massachusetts Bay. The estimated annual wet deposition of the 16 PAH species at Nahant is 78.5 µg m^{-2} yr^{-1}, somewhat less than that of Wolfe’s Neck. We interpret this finding as follows. Wet deposition originates from regional, not local sources. Upper air trajectories bring pollutants, including PAHs, from distant sources, hundreds to a thousand km away. Clouds incorporate the pollutants into their droplets. When the clouds precipitate over the monitoring site, the pollutants are brought down as wet deposition. Apparently, upper air trajectories and precipitating clouds bring somewhat more wet deposition to Wolfe’s Neck than to Nahant.

The average wet deposition during the heating season (15 Oct-15 Apr) is 872 ng m^{-2} cm^{-1}; during the non-heating season (15 Apr-15 Oct), 790 ng m^{-2} cm^{-1}. Evidently, during the heating season more PAHs are emitted than during the non-heating season due to commercial and residential space heating by fossil fuel combustion.

The average annual composition of PAH species in wet deposition is presented in the pie chart of Figure 4. The average composition of the wet samples is: phenanthrene 27%, fluoranthene 13%, pyrene 10.5%, fluorene 9%, benzo(b and k)fluoranthene 9.5%, chrysene 4%, indeno(1,2,3,-c,d)pyrene 4%, benzo(g,h,i)perylene 4% and others 19%.

3.1 Dry Deposition

Over the monitoring period, dry deposition samples were obtained for 41 weeks. During 7 weeks samples were obtained simultaneously by the old and new collector, so that 48 dry samples were obtained in total. Table 3 presents the results in units of nanograms per square meter *per hour*. The unit per hour was chosen because in such a fashion it is easier to average the data and extrapolate to yearly deposition rates. The dry deposition of the sum of 16 PAH species is listed in the bottom line.

The deposition rates varied considerably from sample to sample, ranging from a low of 0.9 ng m^{-2} hr^{-1} (average of two simultaneous samples obtained by the old and new collector) to a high of 34.7 ng m^{-2} hr^{-1}, with an average of 9.3 ng m^{-2} hr^{-1}. Multiplying by 8760, the number of hours in a year, the estimated annual dry deposition of the sum of 16 PAH species in the 1998-2000 period is 81.5 µg m^{-2} yr^{-1}. In the same period, the estimated annual dry deposition of the 16 PAH species at Nahant, MA is 832 µg m^{-2} yr^{-1}, about 10 times as much. Clearly, Nahant receives much more dry deposition than Wolfe's Neck. This is interpreted as follows. Nahant is about 16 km from Downtown Boston and about 10 km from Logan International Airport, and only a few km from the industrial suburbs of Lynn, Saugus and Salem. The large emission rates from these sources cause high dry deposition rates of PAHs at Nahant. Wolfe's Neck is about 30 km from the metropolitan Portland area and Portland International Airport, and there are no industrial suburbs in the vicinity of the monitoring site. Altogether, the metropolitan Portland area has a smaller PAH emission rate than the metropolitan Boston area, and the Portland emission sources are at a greater distance to Wolfe's Neck than the Boston sources to Nahant, explaining the low dry deposition rate of PAHs at Wolfe's Neck.

The average dry deposition during the heating season (15 Oct-15 Apr) is 10.4 ng m^{-2} hr^{-1}, during the non-heating season 8.2 ng m^{-2} hr^{-1}. Just as with wet deposition, dry deposition is higher in the heating than in the non-heating season on account of greater use of fossil fuels and wood for space heating in the winter months.

The average composition of dry deposition is presented in the pie chart of Figure 5.The average composition of the dry samples is: fluoranthene 22%, pyrene 18%, acenaphthylene 12%, benzo(b and k)fluoranthene 11.5%, benzo(a)pyrene 6%, chrysene 6%, anthracene 4%, benzo(g,h,i)perylene 4%, indeno(1,2,3,-c,d)pyrene 4% and others 12.5%. The different composition of wet and dry samples indicates different sources, solubility in cloud droplets, and different pathways for PAHs in wet and dry deposition, respectively.

4.Conclusions and Recommendations

Dry and wet deposition of 16 PAH species has been successfully measured in 1998-2000 with the UML wet/dry collector at the Casco Bay Estuary Project monitoring site at Wolfe's Neck, near Freeport, ME. Weekly samples of wet and dry deposition have been obtained in the field and analyzed for PAHs in the laboratory. Wet samples were obtained for 32 weeks, with 6 weeks of simultaneous sampling by the old and new collector = 38 wet samples. Dry samples were obtained for 41 weeks, with 7 weeks of simultaneous sampling by the old and new collector = 48 samples.

The data indicate wide variability of wet deposition from week to week. However, no clear correlation exists between wet deposition rates of PAHs and amount of precipitation. There is more wet deposition in the heating season (15 Oct-15 Apr) than in the non-heating season (15 Apr-15 Oct). The estimated annual wet deposition at Wolfe's Neck is 91 µg m^{-2} yr^{-1}. This compares to the estimated wet deposition of 78.5 µg m^{-2} yr^{-1} at Nahant on Massachusetts Bay. Apparently, upper air trajectories and precipitating clouds bring somewhat more wet deposition to Wolfe's Neck than to Nahant.

Dry deposition also varies from week to week, with greater deposition in the heating season than in the non-heating season. The estimated annual dry deposition at Wolfe's Neck is 81.5 µg m^{-2} yr^{-1}, far less than at Nahant, 832 µg m^{-2} yr^{-1}, indicating that dry deposition is due to local emission sources, which are far fewer and more distant around Wolfe's Neck than around Nahant.

It is recommended that the measurements at Wolfe's Neck be extended for a longer period and on a continuous rather than intermittent basis, in order to obtain empirical rather than estimated annual and seasonal wet and dry deposition rates of PAHs. For a proper assessment of atmospheric loading of PAHs to Casco Bay, a monitoring site closer to the metropolitan Portland area should be selected, such as Mackworth Island.

5. References

[1] Kennicutt M. C., Wade T. L., Presley B. J., Requejo J. M. and Denoux G. J. (1993) Sediment contaminants in Casco Bay: Inventories, sources and potential for biological impact. *Envir. Sci. Technol*. **28,** 1-15.
[2] US EPA (1997) Deposition of toxic air pollutants to the Great Waters. 2nd Report to Congress. EPA-749/R-97/001B, US Environmental Protection Agency, Research Triangle Park, North Carolina.
[3] CBEP, Casco Bay Estuary Project: Fact Sheet, University of Southern Maine, POB 9300, Portland, ME 04104 (1999).

Table 1. The 16 PAH species analyzed, their abbreviations, chemical structure, and the corresponding deuterated surrogates used to quantify their recovery rates.

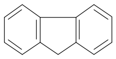
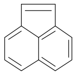
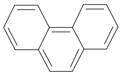
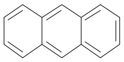
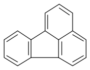
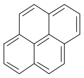
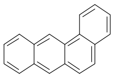
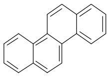
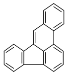
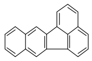
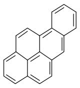
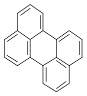
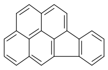
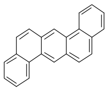
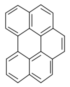
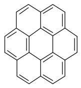
Abbreviation	PAH	Surrogate	Structure
ACN	Acenaphthylene	Acenaphthene-d_{10}	
FLU	Fluorene		
PHE	Phenanthrene	Anthracene-d_{10}	
ANT	Anthracene		
FLA	Fluoranthene	Chrysene-d_{12}	
PYR	Pyrene		
BaA	Benzo(a)anthracene		
CHR	Chrysene		
BbF	Benzo(b)fluoranthene	Perylene-d_{12}	
BkF	Benzo(k)fluoranthene		
BaP	Benzo(a)pyrene		
PER	Perylene		
I1P	Indeno(1,2,3-c,d)pyrene		
DaA	Dibenz(a,h)anthracene		
BgP	Benzo(g,h,i)perylene		
COR	Coronene		

TABLE 2. Dry deposition of PAHs at Wolfe's Neck, 1998-2000. Units ng m-2 hr-1

PAHs	17 Mar 98	7 Apr 98	14 Apr 98	21 Apr 98	5 May 98	12 May 98	2 Jun 98	9 Jun 98	14 Jul 98	11 Aug 98	8 Sep 98	13 Oct 98	10 Nov 98	18 Dec 98	12 Jan 99	9 Feb 99	22 Jun 99
	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	New
ACN	0.31	0.16	0.47	0.81	0.22	0.15	0.27	0.11	NQ	0.24	0.24	NQ	0.71	1.81	0.44	0.35	2.27
FLU	0.95	NQ	NQ	NQ	0.35	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	1.87	NQ	NQ
PHE	0.31	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	2.85	NQ	4.05
ANT	0.93	0.04	0.14	0.05	0.22	0.22	0.24	0.32	NQ	0.28	0.43	NQ	0.25	0.00	0.22	0.13	3.03
FLA	3.57	0.76	1.94	1.78	0.85	0.83	1.20	1.91	2.67	2.33	0.94	1.88	2.55	3.77	2.71	4.50	0.42
PYR	1.71	0.43	1.46	1.00	0.55	0.64	0.75	1.15	8.38	2.97	1.50	4.85	1.92	2.20	1.78	3.72	0.94
BaA	0.49	0.14	0.54	0.38	0.14	0.12	0.15	0.14	0.14	0.08	0.29	0.19	0.13	0.22	0.62	1.07	0.16
CHR	0.56	0.25	0.96	0.85	0.40	0.19	0.30	0.22	0.42	0.26	0.54	0.06	0.41	0.51	1.01	2.12	0.37
BbF	0.82	0.22	1.07	0.94	0.83	0.28	0.24	0.22	NQ	0.18	0.74	NQ	0.26	0.46	1.89	3.20	0.21
BkF	0.70	0.22	0.60	0.99	NQ	NQ	NQ	NQ	0.03	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
BaP	0.99	0.12	0.73	0.61	0.45	NQ	NQ	NQ	NQ	NQ	0.41	NQ	0.07	0.19	0.77	1.36	0.38
PER	0.39	0.29	0.23	0.22	0.24	0.11	0.29	0.30	0.61	0.05	0.34	0.12	0.04	0.09	0.68	0.34	NQ
I1P	0.58	0.21	0.87	0.70	0.20	0.09	0.04	0.05	0.06	NQ	0.39	0.18	0.12	0.17	0.80	1.19	NQ
DaA	0.40	NQ	0.27	0.28	NQ	NQ	NQ	NQ	NQ	NQ	0.18	NQ	0.03	NQ	0.31	0.38	0.41
BgP	0.50	0.18	0.74	0.74	0.22	0.10	0.04	0.07	NQ	0.02	0.33	0.12	NQ	0.13	0.70	0.99	NQ
COR	0.27	0.16	0.49	0.62	NQ	NQ	NQ	NQ	NQ	NQ	0.26	0.20	0.02	0.25	0.92	0.78	0.39
Total	13.48	3.18	10.51	9.95	4.68	2.73	3.53	4.49	12.30	6.41	6.59	7.59	6.50	9.80	17.57	20.12	12.65

Table 2. Continued

PAHs	15 Jul 99		22 Jul 99	29 Jul 99	5 Aug 99		12 Aug 99		19 Aug 99		26 Aug 99		2 Sep 99		7 Oct 99	14 Oct 99	21 Oct 99
	Old	New	Old	New	Old	New	Old	New	Old	New	Old	New	Old	New	New	New	New
ACN	NQ	1.61	1.42	0.52	0.58	0.56	0.82	0.19	0.75	NQ	0.69	0.35	0.52	0.29	1.58	1.53	1.07
FLU	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	0.03	NQ	NQ	NQ	NQ	NQ	NQ
PHE	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
ANT	NQ	NQ	0.27	0.65	NQ	NQ	NQ	NQ	NQ	NQ	NQ	0.33	0.17	0.51	0.15	0.11	0.14
FLA	0.71	0.24	0.64	2.14	0.73	0.13	0.54	0.29	0.29	NQ	0.76	0.14	0.28	0.48	0.71	0.71	2.40
PYR	1.16	0.36	0.62	1.70	0.38	0.18	0.29	0.07	NQ	0.01	0.37	0.14	0.04	0.55	0.43	0.27	5.49
BaA	NQ	NQ	NQ	0.33	0.02	0.01	0.06	0.03	NQ	0.05	0.41	0.07	NQ	0.02	NQ	NQ	NQ
CHR	NQ	0.04	0.04	0.83	NQ	NQ	NQ	0.06	NQ	NQ	0.69	0.04	NQ	0.03	NQ	NQ	NQ
BbF	0.17	0.15	0.04	1.69	0.01	0.19	0.05	0.38	0.04	0.26	0.78	0.09	NQ	NQ	NQ	NQ	NQ
BkF	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	0.89	0.31	NQ	0.12	NQ	NQ	NQ
BaP	0.25	0.14	0.14	0.81	0.25	0.25	0.25	0.32	NQ	0.38	0.90	0.23	NQ	NQ	NQ	NQ	0.02
PER	NQ	NQ	0.07	0.29	NQ	0.14	NQ	0.16	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
I1P	NQ	NQ	NQ	0.43	NQ	NQ	NQ	NQ	NQ	NQ	1.36	0.05	NQ	NQ	NQ	NQ	NQ
DaA	0.12	0.11	NQ	0.31	NQ	NQ	0.26	NQ	NQ	NQ	1.53	0.33	NQ	NQ	0.05	NQ	NQ
BgP	0.09	NQ	0.04	0.71	NQ	NQ	NQ	NQ	NQ	NQ	1.39	0.12	NQ	NQ	NQ	NQ	NQ
COR	0.25	0.12	0.23	0.67	NQ	NQ	NQ	0.20	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	0.06
Total	2.76	2.78	3.50	11.07	1.97	1.46	2.28	1.70	1.08	0.69	9.82	2.20	1.02	1.99	2.93	2.62	9.18

Table 2. Continued

PAHs	28 Oct 99		4 Nov 99	3 Feb 00	10 Feb 00	17 Feb 00	24 Feb 00	2 Mar 00	9 Mar 00	6 Apr 00	13 Apr 00	20 Apr 00	27 Apr 00	4 May 00
	Old	New	New	New	New	New	New	New	New	New	New	New	New	New
ACN	1.02	0.59	0.38	0.58	0.75	0.87	2.61	3.37	1.08	1.17	3.39	3.14	5.54	1.67
FLU	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
PHE	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
ANT	0.23	0.22	0.12	NQ	0.18	NQ	0.38	NQ	NQ	0.42	NQ	0.92	2.88	2.45
FLA	0.67	0.64	0.80	0.81	3.38	1.39	1.19	1.78	1.38	5.72	6.68	6.21	2.32	6.68
PYR	0.39	0.30	0.28	NQ	1.59	0.85	0.49	0.69	0.63	3.26	3.97	3.51	1.24	4.28
BaA	0.18	0.04	NQ	NQ	0.10	0.05	0.11	0.07	0.03	0.97	1.29	1.15	0.33	1.45
CHR	0.31	0.05	NQ	0.03	0.78	0.44	0.47	0.38	0.42	1.72	2.17	1.78	0.59	2.55
BbF	0.37	NQ	0.03	0.07	1.03	0.76	0.65	0.42	0.40	3.77	6.03	4.06	1.83	6.28
BkF	0.62	0.01	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ
BaP	0.53	0.11	0.07	NQ	0.48	0.23	0.33	NQ	NQ	1.93	3.35	2.63	1.09	2.82
PER	0.18	NQ	NQ	NQ	0.22	0.16	NQ	NQ	NQ	0.73	0.76	0.65	0.33	0.59
I1P	0.42	NQ	NQ	NQ	NQ	NQ	0.11	NQ	NQ	1.22	1.90	1.53	0.39	1.82
DaA	0.81	0.13	NQ	NQ	0.19	0.14	0.30	NQ	NQ	0.88	1.05	0.98	0.39	0.97
BgP	0.52	NQ	NQ	NQ	NQ	0.11	0.22	0.07	0.11	1.35	2.13	1.69	0.54	2.05
COR	NQ	NQ	0.06	NQ	0.18	0.19	NQ	NQ	NQ	0.87	1.31	1.15	0.60	1.11
Total	6.24	2.10	1.74	1.49	8.88	5.19	6.85	6.79	4.05	24.01	34.03	29.39	18.06	34.70

TABLE 3. Wet deposition of PAHs at Wolfe's Neck, Maine 1998-2000 (ng m-2)

PAHs	7-Apr-98	21-Apr-98	5-May-98	12-May-98	2-Jun-98	9-Jun-98	14-Jul-98	11-Aug-98	13-Oct-98	18-Dec-98	12-Jan-99	9-Feb-99	15-Jul-99		22-Jul-99	29-Jul-99	12-Aug-99
	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	Old	New	Old	Old	Old
Preciptation (cm)	1.60	6.10	3.25	5.72	3.43	7.11	0.66	0.70	29.72	0.84	2.92	31.50	0.53		1.32	1.12	3.05
ACN	13.5	39.3	15.0	15.5	44.8	109.0	68.7	N/Q	119.6	122.7	312.3	97.6	N/Q	8.98	N/Q	19.3	17.6
FLU	N/Q	N/Q	N/Q	149.3	N/Q	1338.2	202.4	564.5	290.7	N/Q	332.8	N/Q	36.67	51.45	33.99	53.08	79.64
PHE	N/Q	144.6	N/Q	280.0	N/Q	2194.0	168.4	1490.7	989.6	577.8	1805.8	381.1	195.07	201.97	92.82	92.60	234.02
ANT	N/Q	14.9	38.4	N/Q	33.7	217.9	63.8	126.6	91.5	N/Q	40.7	17.3	N/Q	N/Q	112.24	40.65	N/Q
FLA	76.2	329.6	121.7	89.5	88.2	130.4	87.2	163.6	221.7	635.9	992.5	348.2	N/Q	26.75	N/Q	N/Q	37.53
PYR	50.7	209.2	91.5	78.1	84.0	196.8	106.4	63.4	168.7	1848.4	534.9	352.8	N/Q	N/Q	N/Q	N/Q	12.58
BaA	N/Q	64.2	32.2	29.0	32.6	19.6	22.1	24.9	40.6	77.6	130.5	130.3	N/Q	N/Q	N/Q	N/Q	32.40
CHR	39.0	148.7	50.0	13.4	65.2	14.4	31.7	N/Q	72.7	98.9	238.5	97.6	N/Q	N/Q	N/Q	N/Q	6.45
BbF	54.2	170.7	123.2	19.4	142.5	N/Q	N/Q	9.7	93.4	178.0	401.6	295.8	N/Q	9.75	N/Q	N/Q	82.70
BkF	54.2	127.2	N/Q	N/Q	45.5	N/Q	N/Q	16.8	N/Q	40.2	N/Q	N/Q	N/Q	N/Q	N/Q	N/Q	N/Q
BaP	40.1	110.9	45.7	8.6	40.6	N/Q	N/Q	N/Q	30.4	74.7	127.4	121.5	35.19	33.58	34.30	N/Q	54.82
PER	85.3	40.6	63.7	N/Q	151.9	N/Q	148.3	94.5	30.1	84.7	15.3	N/Q	N/Q	19.18	20.61	N/Q	30.86
I1P	60.4	171.8	25.2	35.7	31.2	25.3	9.3	N/Q	82.6	78.0	198.1	108.0	N/Q	N/Q	N/Q	N/Q	N/Q
DaA	N/Q	56.3	N/Q	N/Q	N/Q	N/Q	N/Q	21.4	67.2	88.1	93.6	75.2	42.63	28.08	N/Q	N/Q	56.74
BgP	56.8	155.8	19.6	35.1	16.3	41.3	5.9	N/Q	64.6	57.8	155.7	48.5	3.30	N/Q	N/Q	N/Q	35.85
COR	56.7	157.4	N/Q	N/Q	10.5	N/Q	N/Q	N/Q	75.8	108.5	283.9	87.0	N/Q	31.05	N/Q	N/Q	44.81
Total	586.94	1941.16	626.25	753.62	787.06	4286.74	914.04	2576.15	2438.92	4071.19	5663.55	2160.88	312.85	410.79	293.97	205.67	726.04

Table 3. Continued

PAHs	19-Aug-99		26-Aug-99		02-Sep-99		7-Oct-99	14-Oct-99	21-Oct-99	28-Oct-99		04-Nov-99		13-Jan-00	3-Feb-00	17-Feb-00
	Old	New	Old	New	Old	New	New	New	New	Old	New	Old	New	New	New	New
Preciptation (cm)	2.29		0.74		0.38		2.79	0.69	3.76	6.35		2.36		2.92	1.78	3.94
ACN	N/Q	35.12	13.3	11.63	48.6	20.06	1.91	NQ	24.71	39.1	49.00	NQ	NQ	29.43	20.65	53.52
FLU	92.12	81.69	48.89	41.58	131.89	50.97	53.36	47.97	53.12	79.12	81.12	27.36	20.86	72.25	81.35	108.17
PHE	212.88	333.87	176.18	154.42	327.70	205.73	252.20	66.15	176.33	236.96	281.16	183.63	147.92	241.36	496.86	396.63
ANT	N/Q	N/Q	33.50	NQ	31.21	14.09	NQ	NQ	16.90	37.67	41.48	NQ	NQ	36.47	11.55	36.81
FLA	31.05	138.78	32.14	30.62	71.87	64.60	241.96	61.56	139.12	77.71	116.14	22.54	20.01	292.95	380.01	477.90
PYR	N/Q	67.08	41.90	12.59	20.72	23.27	123.49	43.33	73.37	61.72	73.63	17.83	14.85	187.51	152.02	169.22
BaA	25.11	42.21	24.87	101.56	93.16	50.54	99.86	26.81	26.34	30.13	40.00	NQ	15.17	61.12	51.98	41.09
CHR	N/Q	27.67	NQ	91.93	92.96	NQ	214.75	18.77	59.79	24.23	34.05	NQ	NQ	111.49	98.11	163.80
BbF	53.85	106.57	8.68	124.51	106.17	20.69	77.79	40.59	118.77	40.72	65.19	30.41	49.38	256.83	257.43	380.47
BkF	N/Q	N/Q	4.70	104.12	148.19	30.96	NQ	NQ	NQ	14.13	NQ	NQ	NQ	NQ	NQ	NQ
BaP	N/Q	85.83	21.55	110.26	109.78	37.77	NQ	25.63	28.19	26.15	51.39	NQ	5.36	114.48	95.42	139.74
PER	N/Q	32.44	NQ	NQ	NQ	NQ	NQ	21.21	NQ	NQ	NQ	NQ	NQ	40.04	41.20	NQ
I1P	N/Q	N/Q	NQ	118.35	177.59	39.19	30.19	17.35	34.74	10.62	NQ	11.11	17.15	89.64	87.71	122.36
DaA	N/Q	56.86	3.44	127.80	208.86	71.38	15.69	32.23	43.24	36.96	41.15	NQ	NQ	64.33	63.87	NQ
BgP	N/Q	40.28	NQ	126.78	194.09	56.68	20.85	22.82	37.39	34.04	34.54	NQ	NQ	94.94	87.44	129.03
COR	N/Q	N/Q	NQ	NQ	NQ	NQ	19.26	26.14	34.85	NQ	NQ	NQ	NQ	56.49	73.78	115.00
Total	415.01	1048.40	409.20	1156.15	1762.79	685.93	1151.32	450.55	866.86	749.29	908.85	292.88	290.70	1749.31	1999.38	2333.75

Table 3. Continued

PAHs	24-Feb-00	2-Mar-00	6-Apr-00	13-Apr-00	27-Apr-00
	New	New	New	New	New
Preciptation (cm)	1.02	1.83	2.84	2.84	4.47
ACN	139.36	21.65	NQ	14.70	55.17
FLU	42.67	23.06	34.11	18.88	177.53
PHE	226.90	117.89	120.73	170.97	507.00
ANT	NQ	NQ	NQ	4.58	71.26
FLA	133.92	105.10	109.66	355.28	315.64
PYR	72.59	54.22	41.87	174.78	202.02
BaA	16.50	17.37	15.66	61.25	131.84
CHR	32.21	35.78	43.27	124.41	186.52
BbF	86.00	96.02	134.20	367.72	388.24
BkF	NQ	NQ	NQ	NQ	NQ
BaP	22.03	32.32	43.41	139.47	261.39
PER	NQ	NQ	NQ	40.00	NQ
I1P	40.06	39.17	51.01	129.81	144.46
DaA	39.72	NQ	NQ	79.94	NQ
BgP	43.48	43.02	61.16	146.26	181.36
COR	44.45	NQ	65.46	96.44	94.94
Total	939.90	585.60	720.53	1924.50	2717.37

Figure 1. The UML wet/dry collector

Figure 2. The interior of the dry collector

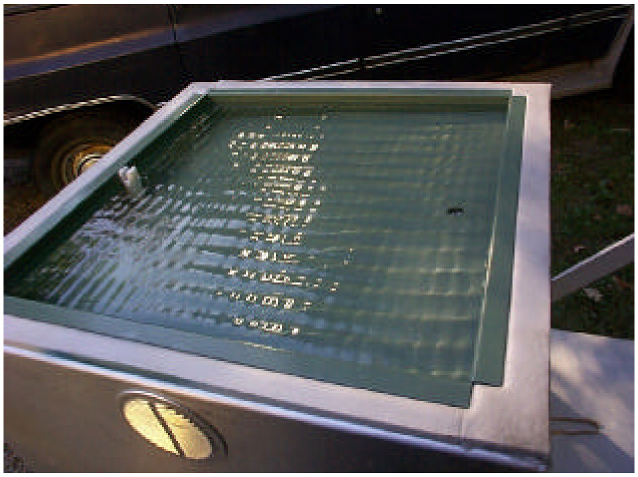


Figure 3. Top view of the dry collector

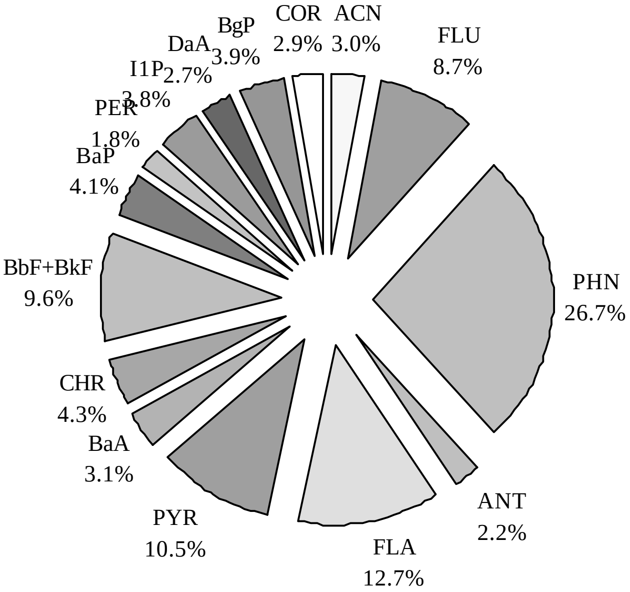


Figure 4. Average composition of PAH species in wet deposition

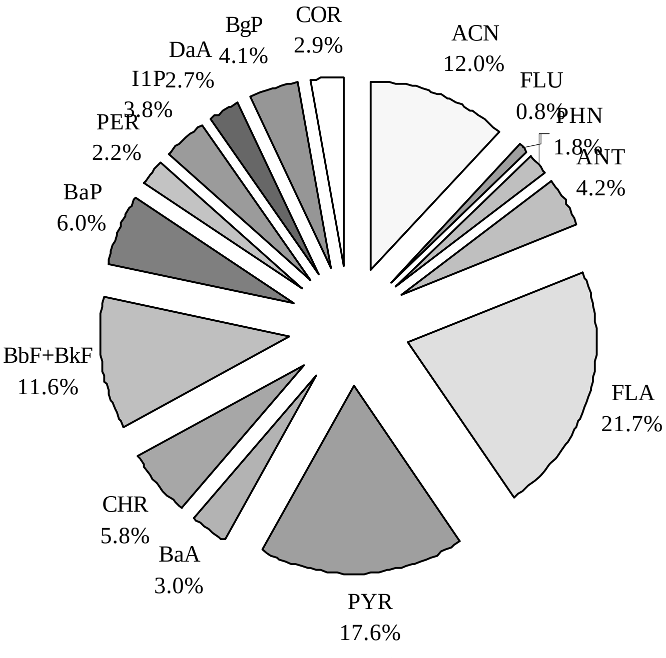


Figure 5. Average composition of PAH species in dry deposition