

Distribution of polychlorinated biphenyls in the Newark Bay Estuary

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Abstract

As part of the NJ Toxics Reduction Workplan for NY/NJ Harbor, ambient water samples were collected at 15 locations along the tidal portions of the Hackensack, Passaic, Raritan, Rahway and Elizabeth Rivers, and in Newark Bay, the Arthur Kill, and Kill van Kull. A Trace Organics Platform Sampler was used to collect a total of 73 dissolved phase and 73 suspended sediment phase samples between June 2000 and May 2002. These samples were analyzed for spatial and wet versus dry weather trends in the 114 polychlorinated biphenyls (PCBs; modified USEPA Method 1668A). Mean total PCB concentrations at the sampling locations ranged between 3.45 and 56 ng/L. PCB homolog groups distribution patterns at the sampling locations are presented.

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1. Introduction

The presence of chlorinated organic compounds, including polychlorinated biphenyls (PCBs) in the marine environment is of international concern due to their persistence, toxicity, and bioaccumulation in the lipid tissue of marine biota [1]. PCBs are man-made organic compounds consisting of a biphenyl with a varying number of chlorine atoms substituted on the two six-carbon benzene rings. Ten different levels of chlorination (homolog groups) exist, mono (1) through deca (10), resulting in 209 distinct congeners. PCBs have low electrical conductivity and high resistance to heat and thermal degradation. As a result they were ideal for use as dielectric fluids in capacitors and transformers, and as hydraulic fluids and solvents [2]. Degradation of PCBs will occur very slowly through dechlorination. PCBs enter biota through ingestion of water or carbon matter, and readily bio-accumulate in the fatty tissue of organisms. PCBs were first manufactured commercially in 1929 [3] and were widely used until production was restricted or banned worldwide since the early 1970s [4]. PCBs have cycled throughout the global environment, and been found in fish, birds, and mammals in nearly all environments [3].

In response to problems resulting from toxic contamination of water, sediments, and biota, in the Newark Bay Estuary, the New Jersey Toxics Reduction Workplan for NY/NJ Harbor (NJTRWP) has been implemented by the NJ Department of Environmental Protection. The Newark Bay Estuary has typically been evaluated as a part of a much larger estuarine system referred to as the New York/New Jersey (NY/NJ) Harbor Estuary [5], which includes the lower Hudson River, New York Bay, Raritan Bay, and their tributaries. The principal tributaries to the Newark Bay Estuary include the Passaic, Hackensack, Elizabeth, and Rahway River and the tidal Arthur Kill. The net water flow from Newark Bay is toward New York Bay and Raritan Bay via the Kill van Kull and Arthur Kill [6]. Due to diurnal tidal fluxes into Newark Bay contaminants from the Hudson River, New York Bay and the Raritan Bay are carried into the Newark Bay Estuary [7]. Also persistent wind events of the east or west can disrupt the circulation in the Newark Bay Estuary [8]. The Newark Bay and its tributaries have been impacted by industrial and municipal activities since the early 1800s [5]. The major sources of PCB contamination in Newark Bay are PCBs originating from the Hudson River, where PCBs were released into the upper river from capacitor manufacturing operations at General Electric (GE) plants [9] beginning in 1947 and continuing through 1977, combined sewer overflows (CSOs) in the lower Passaic and the Kills. The fundamental goal of the NJTRWP is the consistent detection

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of toxic chemicals, including PCBs, for subsequent modeling studies and source trackdown. Past studies have shown that PCBs are not detected in the NY/NJ Harbor when conventional sampling methods are used. The New York State Department of Environmental Conservation developed the Trace Organics Platform Sampler (TOPS) to measure field concentrations of dilute organic pollutants [4,10]. The Stevens Institute of Technology-Trace Organics Platform Sampler (SIT-TOPS) has been developed to reliably measure trace ambient water column concentrations of organic contaminants in the dissolved and particulate phases in water with high suspended sediment concentrations [11]. In the SIT-TOPS, large volumes of water are drawn through two filters to obtain particulate phase organics, and a smaller portion of filtered water is then drawn through a series of two XAD-resin columns to adsorb the dissolved phase contaminants.

2. Sampling strategy

The NJ Toxics Reduction Workplan for NY/NJ Harbor ambient water quality sampling program (water and suspended sediment quality measurements using SIT-TOPS and grab sampling techniques) used five fixed river head-of-tide locations (not discussed in this paper), 10 fixed sampling sites located along the banks of the tidally influenced part of the tributary rivers, and five shipboard Estuary locations (Fig. 1). The study area has been divided into two major survey areas:

- The Northern Tributaries, i.e. the Hackensack and Passaic Rivers, Newark Bay, and the Kill van Kull; and

- The Southern Tributaries, i.e. the Elizabeth, Rahway, and Raritan Rivers, and the Arthur Kill

This paper presents PCBs data from sampling surveys conducted between June 2000 and May 2002.

3. Methodology

Ambient water quality monitoring was performed using SIT-TOPS sampler units. A schematic of the SIT-TOPS units is shown in Fig. 2. Each SIT-TOPS unit is equipped with two types of filters and two XAD-resin columns in series:

STEVENS-TOPS Configuration for Tributary/Estuary Sampling

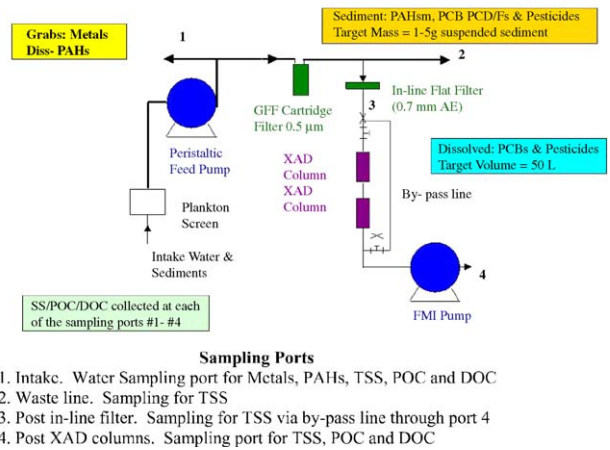


Fig. 2. Schematic diagram of the SIT-TOPS sampler.

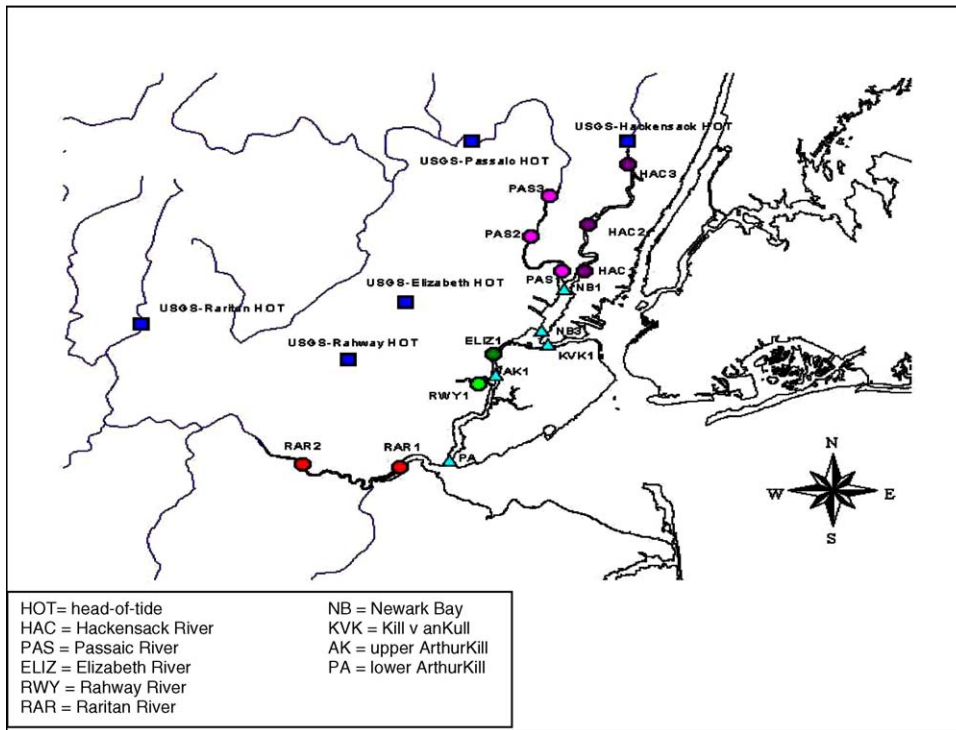


Fig. 1. Approximate locations of the NJTRWP water-quality sampling stations (circle: fixed sampling sites located on the banks of the tidally influenced part of the tributary rivers; triangle: shipboard estuary sampling locations; square: fixed head-of-tide sampling locations).

- A baked 4-in. long, 0.5 μm nominal size retention Cartridge GFF filter (C-GFF) located in the discharge line of the peristaltic pump right after sampling port 1.
- A 142 mm diameter in-line 0.7 μm GF/F Whatman glass microfiber filter (“flat filter”) located before the XAD columns.
- Two XAD-resin columns connected in series between the in-line flat filter and the FMI pump. Each column contained approximately 125 g of pre-cleaned Supelco Amberlite XAD Type 2 resin, supplied as 20–60 mesh-size beads. The first of the two XAD columns was spiked with ^{13}C -labeled PCB field surrogates (FS) (80 ng each of PCB #31L, 95L, and 153L).

A minimum of 50 L of water must flow through the XAD columns to insure that sufficient mass of the organic compounds of interest will be trapped in these columns. The peristaltic feed pump flow rate is set at approximately 2000 mL/min. The flow rate of the FMI pump is restricted by breakthrough and is set at approximately 250 mL/min and is monitored every 30 min. The in-line filters are replaced as needed in order to maintain the flow rate in the FMI line above the 200 mL/min threshold value. Based on the above considerations, the duration of the sampling event is 4 h.

At the tidal tributary and estuary sites, each SIT-TOPS sampling event began approximately at high tide and had a duration of at least 4 h, in order to identify those tributaries/watersheds that are significant sources of suspended sediments and toxic chemicals into the Newark Bay. The tributaries were sampled during two storm (wet) events, and two low-flow discharge (dry) events. The wet weather river flow (storm) magnitude scale has been developed by USGS using data for the last 25 years of record for each river. Ambient water was continuously pumped through the SIT-TOPS during this sampling period. In addition, aqueous samples were taken at hourly intervals through the sampling ports at the intake, waste line, after the in-line flat filter, and after the XAD columns, and analyzed for suspended sediments (SS), particulate organic carbon (POC) and dissolved organic carbon (DOC). Dissolved phase PCBs were eluted from the XAD columns. Particulate phase PCBs were extracted from the set of SIT-TOPS filters. The samples were usually collected at a depth of 1.5 m below the water surface, since the tidal range during spring tide exceeds 1.5 m. In order to obtain a vertical profile at selected locations, samples were also collected at a depth of 1.5 m above the river/estuary bottom, in order to avoid sampling bottom sediments; these are noted with a “D” suffix (for example, NB1-D) to distinguish them from the surface samples (for example, NB1-S).

4. Analytical procedures

Table 1 lists the different PCB congeners measured in this work. A modified version of USEPA Method 1668A [12] was used to measure the sediment bound and dissolved concentrations of the 114 PCB congeners (Table 1). This method used high-resolution gas chromatography and mass spectrometry (HRGC/HRMS) with isotope dilution to analyze the TOPS filter samples. There are 114 PCB congeners selected by the NJ

Table 1
List of analyzed PCB congeners

Congener number	Substitutions and homolog group
	Monodichlorobiphenyl
3	4
	Dichlorobiphenyl
4	2,2'
5	2,3
8	2,4'
10	2,6
11	3,3'
15	4,4'
	Trichlorobiphenyl
16	2,2',3
17	2,2',4
18	2,2',5
19	2,2',6
20	2,3,3'
22	2,3,4'
25	2,3',4
26	2,3',5
27	2,3',6
28	2,4,4'
29	2,4,5
31	2,4',5
33C20	2',3,4
37	3,4,4'
	Tetrachlorobiphenyl
40	2,2',3,3'
42	2,2',3,4'
43	2,2',3,5
44	2,2',3,5'
45	2,2',3,6
46	2,2',3,6'
47	2,2',4,4'
48	2,2',4,5
49	2,2',4,5'
50	2,2',4,6
52	2,2',5,5'
53	2,2',5,6'
55	2,3,3',4
56	2,3,3',4'
59	2,3,3',6
60	2,3,4,4'
62C44	2,3,4,6
63	2,3,4',5
64C40	2,3,4',6
66	2,3',4,4'
70	2,3',4',5
74C70	2,4,4',5
75C44	2,4,4',6
77	3,3',4,4'
81	3,4,4',5
	Pentachlorobiphenyl
82	2,2',3,3',4
83	2,2',3,3',5
84	2,2',3,3',6
85	2,2',3,4,4'
86	2,2',3,4,5
87C86	2,2',3,4,5'
90	2,2',3,4',5
91	2,2',3,4',6
92	2,2',3,5,5'
95	2,2',3,5',6

Table 1 (Continued)

Congener number	Substitutions and homolog group
97C86	2,2',3',4,5
99C83	2,2',4,4',5
101C90	2,2',4,5,5'
104	2,2',4,6,6'
105	2,3,3',4,4'
109	2,3,3',4,6
110	2,3,3',4',6
114	2,3,4,4',5
118	2,3',4,4',5
119C109	2,3',4,4',6
120	2,3',4,5,5'
123	2',3,4,4',5
124	2',3,4,5,5'
126	3,3',4,4',5
128	2,2',3,3',4,4'
132	2,2',3,3',4,6'
134	2,2',3,3',5,6
135	2,2',3,3',5,6'
136	2,2',3,3',6,6'
137	2,2',3,4,4',5
138	2,2',3,4,4',5'
141	2,2',3,4,5,5'
142	2,2',3,4,5,6
146	2,2',3,4',5,5'
147	2,2',3,4',5,6
149C147	2,2',3,4',5',6
151C135	2,2',3,5,5',6
153	2,2',4,4',5,5'
154C135	Hexachlorobiphenyl
156	2,2',4,4',5,6'
157	2,3,3',4,4',5
158	2,3,3',4,4',5'
166	2,3,3',4,4',6
167	2,3,4,4',5,6
168C153	2,3',4,4',5,5'
169	3,3',4,4',5,5'
170	Heptachlorobiphenyl
171	2,2',3,3',4,4',5
172	2,2',3,3',4,4',6
174	2,2',3,3',4,5,5'
177	2,2',3,3',4,5,6'
178	2,2',3,3',4',5,6
179	2,2',3,3',5,5',6
180	2,2',3,3',5,6,6'
183	2,2',3,4,4',5,5'
185	2,2',3,4,4',5',6
187	2,2',3,4,5,5',6
189	2,2',3,4',5,5',6
190	2,3,3',4,4',5,5'
190	2,3,3',4,4',5,6
191	2,3,3',4,4',5',6
194	Octachlorobiphenyl
195	2,2',3,3',4,4',5,5'
196	2,2',3,3',4,4',5,6
198	2,2',3,3',4,4',5,6'
198	2,2',3,3',4,5,5',6
BZ-201	2,2',3,3',4,5,5',6'
199C198	2,2',3,3',4,5,6,6'
BZ-200	2,2',3,3',4,5',6,6'
203	2,2',3,4,4',5,5',6
205	2,3,3',4,4',5,5',6

Table 1 (Continued)

Congener number	Substitutions and homolog group
206	Nonachlorobiphenyl
207	2,2',3,3',4,4',5,5',6
208	2,2',3,3',4,4',5,6,6'
208	2,2',3,3',4,5,5',6,6'
209	Decachlorobiphenyl
209	2,2',3,3',4,4',5,5',6,6'

Note: C indicates co-elution with PCB congener.

Toxics Reduction Workplan for measurement, 73 congeners did not co-elute with other congeners, 15 congeners co-eluted with congeners in the list, and 26 co-eluted with congeners outside of the list. The detection limit achieved for each PCB congener varied among the samples, but was approximately 180 pg for the dissolved phase and 600 pg for the suspended phase. As mentioned in Section 3 a minimum of 50 L of water must flow through the XAD columns, which translated into an achieved detection limit of 3.6 pg/L for each PCB congener in the dissolved phase. The sample size for the suspended phase (mass of sediment collected on the filters) also varied, but a target sediment mass of 5 g was established. This translates into a detection limit of approximately 120 pg/g. Mean SS at the sample sites ranged between 7.5 and 65 mg/L; thus, 77–667 L would be needed to collect the target mass of 5 g sediment.

The sample data was subjected to a preliminary quality assurance verification review. In addition, because of the nature of the samples and the trace concentrations expected for the target analytes, a “maximum blank” approach was developed to assess the impact of background contamination on the usability of the sample data. For each survey, one (1) Equipment and one (1) Field Blank for each sampling location were prepared and analyzed in the same manner as their associated samples. At least one (1) Laboratory Method Blank was also prepared and analyzed. That blank having the largest value (the “maximum blank”) was used to assess the effect of background contamination on the sample data for that survey. In order for a sample result to be usable, it must have been at least three times (3×) greater than the “maximum blank”. No other blank correction was performed on the sample data. Congeners in the dissolved phase that were repeatedly removed by blank elimination included PCB congeners 3, 8, 11, and 194. The suspended phase was affected very little by blank contamination.

5. Results

5.1. Spatial variations

Tables 2–4 show the dissolved (ng/L), suspended (ng/g) and total (ng/L) PCB concentrations, respectively, from all sampling stations during all sampling events. From the recorded dissolved PCB concentrations that pass the blank contamination test on average 29% of the total PCB concentration in the water column is in the dissolved phase. As seen in Table 2 the total PCB concentrations in the dissolved phase do not vary significantly in the study area. The Saline Aquatic Health Water Quality Crite-

Table 2
Total dissolved PCB concentrations (ng/L) at all of the NJTRWP sampling stations during all sampling events

		6/20–22/00	12/13–15/00	3/13–15/01	4/12/2001	4/25/2001	5/15/2001	5/24/2001	10/17–19/01	10/3/2001	11/6/2001	3/12–14/02	3/27/2002	5/14/2002	Mean	S.D.	%S.D.
Passaic River	PAS1	7.01	4.05	3.29					4.41			3.56			4.46	1.49	33
	PAS2		3.22	2.13					9.58			5.54			5.12	3.30	64
	PAS3		0.94	1.39					6.63			7.97			4.23	3.59	85
Newark Bay	NB1-S	9.77	3.25	2.84					4.51			3.26			4.73	2.89	61
	NB1-D	8.65	2.18	4.73					4.09			1.77			4.28	2.74	64
	NB3		5.27	2.55					2.99			2.29			3.276	1.3	42
Arthur Kill	AK1-S		2.55	2.27										4.22	3.01	1.05	35
	AK1-D					3.46		3.84			4.06				3.79	0.30	8
Perth Amboy	PA-S		1.72	1.55				1.59				2.28			1.78	0.34	19
	PA-D					1.92		2.08				2.52			2.18	0.31	14
Hackensack River	HAC1	5.96	2.99	2.86					3.29			2.35			3.49	1.42	41
	HAC2		3.59	6.33					6.97		5.18				5.52	1.49	27
	HAC3		3.77	5.00					3.27			4.78			4.20	0.82	20
Kill van Kull	KVK001		2.35	3.22					3.40			2.33			2.83	0.57	20
Raritan River	RAR1-S				2.17		1.87			2.46			1.08		1.89	0.60	32
	RAR2				1.13		1.49			2.14			1.08		1.46	0.49	34
Rahway River	RHWY1					4.29		5.61			4.50			4.68	4.77	0.58	12
Elizabeth River	ELIZ1					2.88		3.62			3.90			2.69	3.27	0.58	18

Table 3
Total suspended PCB concentrations (ng/g) at all of the NJTRWP sampling stations during all sampling events

		6/20–22/00	12/13–15/00	3/13–15/01	4/12/2001	4/25/2001	5/15/2001	5/24/2001	10/17–19/01	10/3/2001	11/6/2001	3/12–14/02	3/27/2002	5/14/2002	Mean	S.D.	%S.D.
Passaic River	PAS1	1015	629	794					610			1345			879	307	35
	PAS2		256	831					982						689	383	56
	PAS3		1318	168					893			819			799	475	59
Newark Bay	NB1-S	725	1023	590					690			1275			861	282	33
	NB1-D	966	466	544					669			926			714	224	31
	NB3		1049	414					461			678			650	290	45
Arthur Kill	AK1-S		817	602										1219	879	313	36
	AK1-D					807		495			948				750	232	31
Perth Amboy	PA-S		460	826				1303				787			844	348	41
	PA-D					757		520			1033				770	257	33
Hackensack River	HAC1	583	41	1119					657			1202			720	468	65
	HAC2		610	762					1148			1161			921	277	30
	HAC3		440	524					1001			418			596	274	46
Kill van Kull	KVK001		999	518					257			670			611	310	51
Raritan River	RAR1-S				352		724			409			419		476	168	35
	RAR2				169		503			349			218		310	149	48
Rahway River	RHWY1					1353		659			2099			1888	1500	643	43
Elizabeth River	ELIZ1					4171		1844			1272			1378	2166	1359	63

Table 4
Total PCB concentrations (ng/L) at all of the NJTRWP sampling stations during all sampling events

	6/20–22/00	12/13–15/00	3/13–15/01	4/12/2001	4/25/2001	5/15/2001	5/24/2001	10/17–19/01	10/3/2001	11/6/2001	3/12–14/02	3/27/2002	5/14/2002	Mean	SD	%SD
PAS1	46.71	24.22	10.80					45.45			16.27			28.69	16.58	58
PAS2		15.23	6.06					40.82						20.70	18.01	87
PAS3		4.00	1.69					40.99			22.52			17.30	18.34	106
NB1-S	17.24	14.75	6.46					17.04			8.68			12.83	4.97	39
NB1-D	19.39	9.87	9.66					12.80			7.34			11.81	4.66	39
NB3		11.61	5.93					7.30			5.79			7.66	2.72	36
AK1-S		14.19	8.48		9.95		8.01			9.63		13.89		12.19	3.21	26
AK1-D														9.20	1.04	11
PA-S		5.90	11.81		6.29		14.32		5.62					9.41	4.34	46
PA-D							9.84		5.56					7.23	2.29	32
HAC1	18.61	4.17	13.04					17.44			6.69			11.99	6.40	53
HAC2		14.28	13.82					29.11			10.30			16.88	8.35	49
HAC3		26.35	18.52					30.85			17.84			23.39	6.30	27
KVK001		14.65	8.56					7.22			7.84			9.57	3.43	36
RAR1-S				7.26		4.62			3.63			7.40		5.72	1.89	33
RAR2				2.17		4.99			4.24			2.40		3.45	1.38	40
RHWY1					19.74		72.13			15.43		116.65		55.99	47.96	86
ELIZ1					32.95		46.23			2.78		11.16		23.28	19.89	85

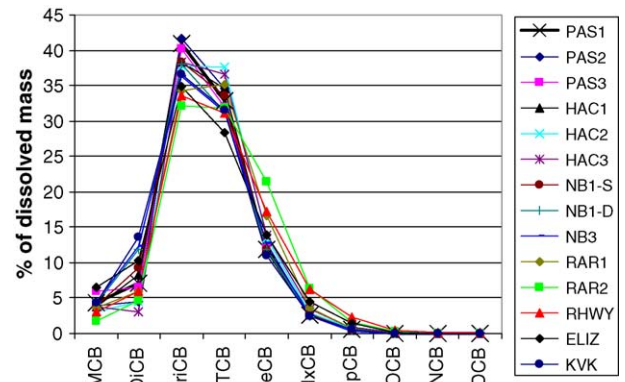


Fig. 3. Mean dissolved PCB congener distribution at each NJTRWP sampling station.

riation of 170 pg/L is exceeded in most stations, whereas the Saline Aquatic Chronic Criterion of 30,000 pg/L is not exceeded at any sampling station. Total dissolved PCB concentrations tend to be lower in the Raritan River stations (RAR1 and RAR2) and in the Perth Amboy stations (PA). At all other stations the mean total dissolved PCB concentration are measured to be higher than 3 ng/L. The ELR Water Quality Criterion of 23 ng/g and the EMR Water Quality Criterion of 180 ng/g are exceeded in all stations. The highest suspended phase concentrations are measured in the Elizabeth River (Station ELIZ1) with a mean concentration of 2166 ng/g and the Rahway River (Station RHWY1) with a mean concentration of 1500 ng/g. In all other station the mean measured total suspended concentrations do not exceed 1000 ng/g. Concentrations in the tributaries show a higher variability than concentrations in the estuarine locations.

5.2. PCB homolog groups: dissolved versus suspended concentrations

On average 84% of the total PCB mass in the dissolved phase is distributed among the tri-, tetra-, and penta- homolog groups with the maximum mass in the tri- and tetra- groups (Fig. 3). In all sampling stations 32–42% of the total PCB mass is in the tri-homolog group. In the Elizabeth (ELIZ1), Rahway (RHWY1), and Raritan (RAR2) River stations the PCB homolog concentrations are more evenly distributed with higher contributions from the hexa- homolog group. On the average 84% of the total PCB mass in the suspended phase is distributed among the tri-, tetra-, penta-, and hexa- homolog groups with the maximum mass in the tetra- and penta- groups (Fig. 4). In the Elizabeth (ELIZ1), Rahway (RHWY1), and Raritan (RAR2) River station the PCB homolog concentrations are more evenly distributed with higher contributions from the hepta- congener group.

Fig. 5 shows PCB homolog groups' distribution in the suspended phase at representative sampling stations during all events. The reason behind this analysis is to compare homolog distributions during wet versus dry events. Differences in homolog distributions between wet and dry events may indicate a PCB source during a wet event related to either

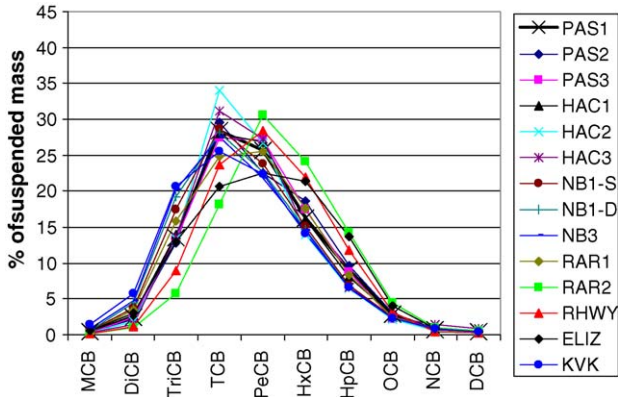


Fig. 4. Mean suspended PCB congener distribution at each NJTRWP sampling station.

a point discharge (e.g. combined sewer overflows) or a non-point discharge (e.g. river runoff). In the Passaic and Hackensack River stations there is little variability between wet and dry events. In the Elizabeth River the patterns are variable. There is a shift towards the higher homologs during the May 2001 Storm 7 event that may be indicative of a source. The May 2001 storm discharge had a significantly different homolog composition, with the hexa- and hepta-chlorinated compounds enriched and the di- and tri- homologs depleted compared with low-flow water. The same pattern is observed at the Raritan River station (RAR2) during the wet events, i.e. the hexa- and hepta-chlorinated compounds are enriched and the di-, tri- and tetra- homologs are depleted compared with low-flow water. There appears to be little difference in the homolog patterns at the NB1-S, NB1-D stations, indicative of a well-mixed estuary.

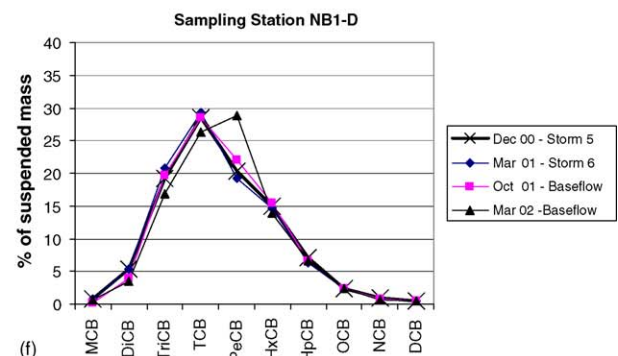
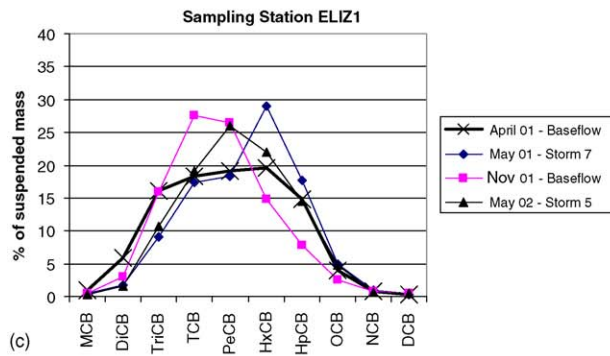
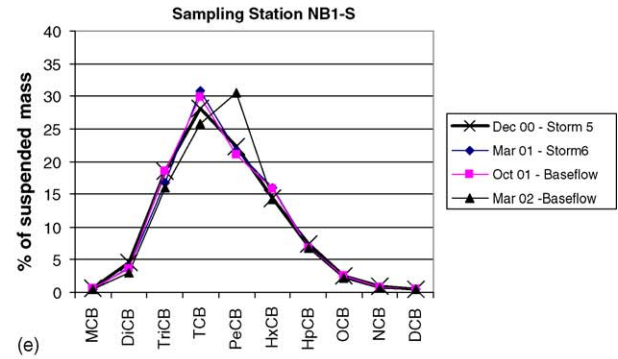
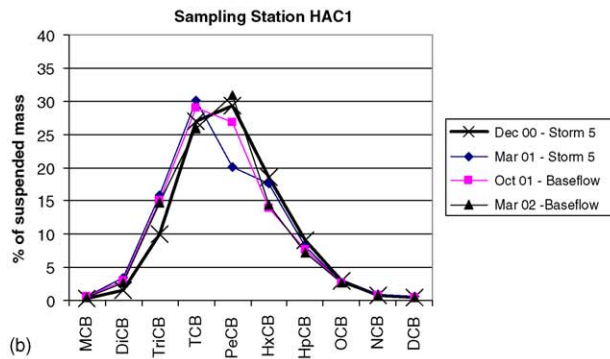
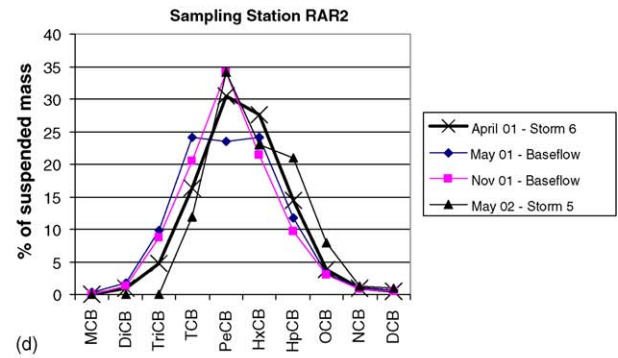
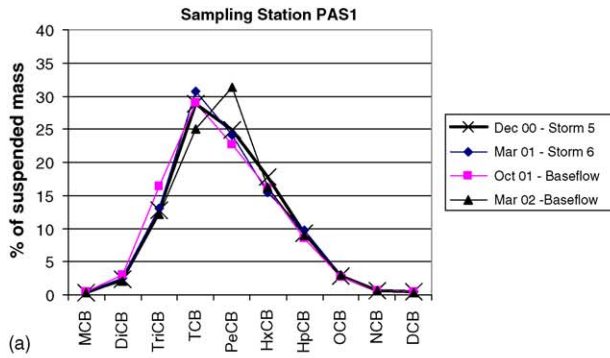


Fig. 5. Suspended PCBs congener distribution during all sampling events at sampling stations: (a) PAS1, (b) HAC1, (c) ELIZ1, (d) RAR2, (e) NB1-S, (f) NB1-D.

6. Conclusions

- Mean total PCB concentrations in the Newark Bay Estuary ranged between 3.45 and 56 ng/L. Mean total PCB concentrations in the dissolved phase ranged between 1.46 and 5.52 ng/L. Mean total PCB concentrations in the suspended phase ranged between 310 and 2166 ng/g.
- PCB homolog distributions in both the dissolved and the suspended phase show that the Southern Tributaries (Elizabeth, Rahway and Raritan River sampling stations) show a shift towards the higher homolog groups, a trend that may be indicative of a different PCBs source.
- There is little variability among shallow and deep stations (e.g. NB1-S versus NB1-D), which is indicative of a well-mixed estuary.
- There is little variability and no consistent wet versus dry event pattern in the suspended fraction data in all sampling stations with the exception of the Elizabeth and Rahway River stations, where there is high variability among events and a clear shift towards the more chlorinated homologs during wet events. This is an indication of the existence of a point or non-point source associated with storm events.

Concurrent with this work were studies that characterized conditions from combined sewer (CSO) and storm water outfalls (SWO) in the area [13]. The comparison of the PCB homolog patterns at the sampling stations with PCB homolog patterns at the CSOs and SWOs will indicate possible PCB sources. The different circulation patterns in NY/NJ Harbor Estuary due to diurnal tidal fluxes and persistent wind events, might explain the differences in homolog distributions among dry events.

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