



# Historical sources of polychlorinated biphenyls to the sediment of the New York/New Jersey Harbor



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## HIGHLIGHTS

- PCBs measured in surface sediment and cores using method 1668 were analyzed with Positive Matrix Factorization.
- Several sources were identified including Aroclors and non-Aroclor PCBs.
- The Upper Hudson River contributed about half of the PCBs in a sediment core from the Lower Hudson River from 1950–2000.
- PCB 11 from pigments and PCB 209 from foundry wax, pigments, or titanium dioxide were identified as non-Aroclor sources.

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## ABSTRACT

Using dated sediment cores, polychlorinated biphenyl (PCB) congener concentrations in the New York/New Jersey Harbor and Lower Hudson River were investigated using Positive Matrix Factorization. Of the seven factors resolved, six represent Aroclors in various stages of weathering. Factor 1 resembles Aroclor 1242 and is consistent with the Upper Hudson River PCB signal associated with the General Electric capacitor plants near Hudson Falls, NY. This factor is the dominant source of PCBs in the upper layers of the sediment core collected in the Lower Hudson River. Factor 2 (Aroclor 1248) was the dominant PCB component in the core depths corresponding with around 1970, but it has decreased more rapidly since its peak (estimated half-life of about 5 years) than factor 1 (half-life of about 14 years), suggesting that PCBs from the Upper Hudson have delayed the recovery of the Harbor from PCB contamination. The seventh factor, comprised of PCBs 206, 208, and 209, was greatest in concentration in the deepest core slices and is thought to be associated with inadvertent production of PCBs during the manufacture of titanium dioxide and/or with foundry waxes containing PCBs. PCB 11, which is thought to be associated with the use of color organic pigments, was examined separately and was detected in sediment throughout the Harbor. Its maximum concentrations generally occurred at the same depth as the maximum total PCB concentrations, suggesting that PCB 11 concentrations decreased after the mid-1970s.

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## 1. Introduction

Polychlorinated biphenyls (PCBs) are a main contaminant of concern in the New York/New Jersey Harbor (“the Harbor”), which includes the Lower Hudson River (LHR) (TAMS Consultants and the Gradient Corporation, 1997; Steinberg et al., 2004). The LHR is defined as the tidal river from the Federal Dam in Troy, NY to the Battery at the southern tip of Manhattan, while the Harbor includes

all of the other tributaries to their heads of tide and includes the Raritan Bay (Fig. 1). A major source of PCBs to the Harbor is the Upper Hudson River (UHR) above the Federal Dam, which is the largest Superfund site in the US due to PCB contamination from two plants owned by General Electric (TAMS Consultants and the Gradient Corporation, 1997). In addition to this source, PCBs also entered the Harbor from the largest US city, New York, and its surrounding urban area as a result of urbanization and industrialization (Totten, 2005). Several attempts have been made to estimate and/or quantify the loads of PCBs to the Harbor. These loading estimates tabulated loads generally by multiplying flow rates (from rivers, wastewater treatment plants, etc.) by the concentrations of

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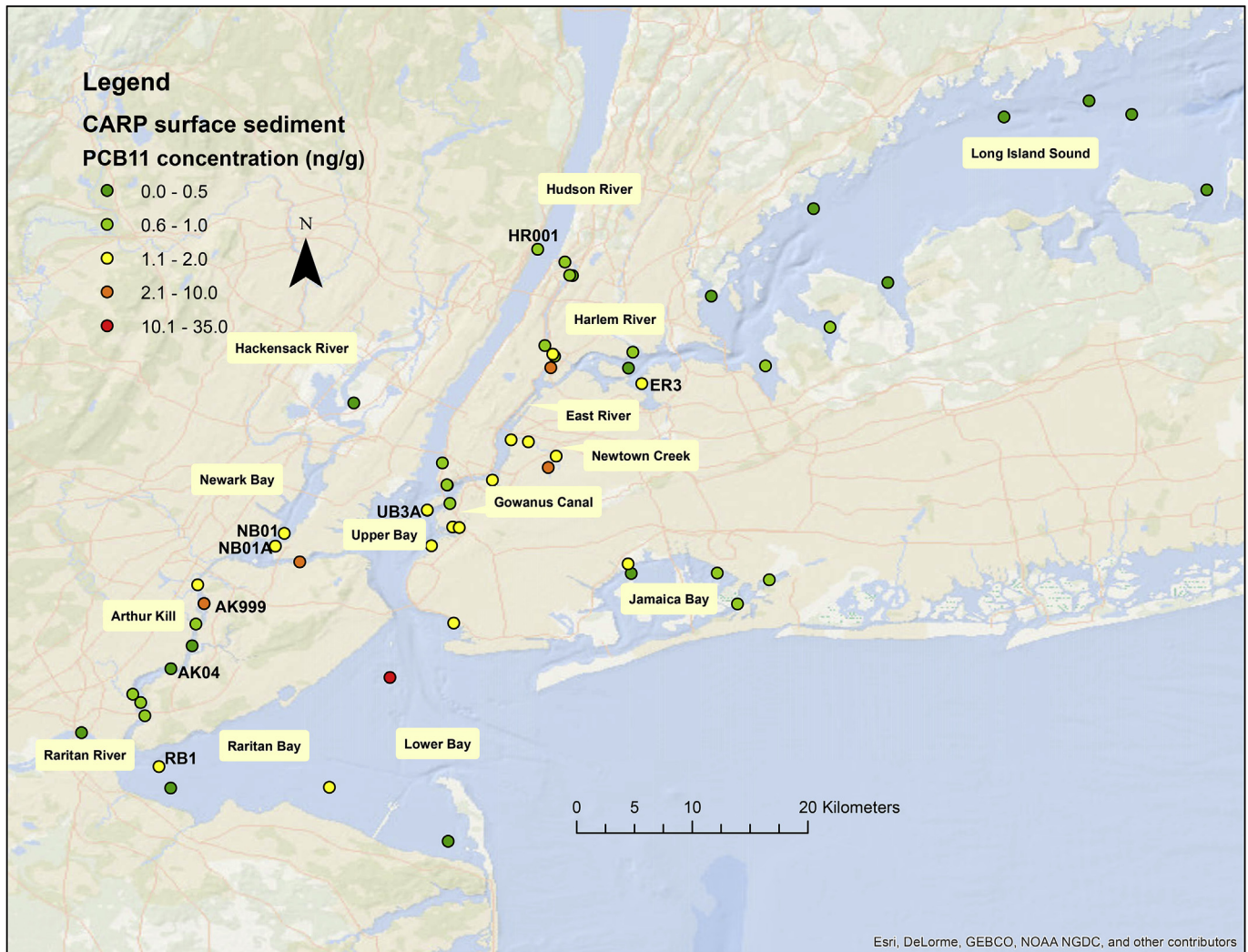


Fig. 1. Map of study area showing PCB 11 concentrations in surface sediment.

PCBs found in those inputs. Various assessments by USEPA Thomann et al. (1989), Farley et al. (1999), the Contamination Assessment and Reduction Project (CARP) model (HydroQual, 2007), and Totten (2005) are all in agreement that USEPA that the UHR is responsible for about half of the total PCB load to the Harbor, followed by treated wastewater, stormwater, and combined sewer overflows (CSOs). These loading assessment are in agreement with fingerprinting of the water column data from the CARP, which suggested that a fingerprint similar to dechlorinated Aroclor 1242 that is dominant in the UHR constituted about 40% of PCBs in the water column in the core area of the LHR around 2000 (Rodenburg et al., 2011a). Circa 1980, Thomann et al. (1989) estimated that the UHR is the dominant source of PCBs to the Harbor, with a load of about 2500 kg/y. By the 1990s, Farley et al. (1999) estimated that the load had fallen to about 300 kg/y, and the fate model based on data collected by the Contamination Assessment and Reduction Project (CARP) estimated that the load was still about 300 kg/y around 2000 (HydroQual, 2007).

From 1999 to about 2003, the CARP (Contamination Assessment and Reduction Project (CARP), 2007b, a) measured PCBs and other contaminants in order to characterize water and sediment quality in the Harbor. The CARP collected cores as well as surface sediment,

allowing an assessment of the historical PCB concentrations with depth down the cores. The purpose of this investigation is to analyze the CARP data on PCBs in the sediment of the Harbor using advanced factor analysis methods to understand, and where possible quantify, contemporary and historical PCB sources to the Harbor by linking dated sediment core data and to historical inputs. This represents an independent check on the previous loading estimates cited above. Identification of past and present sources of PCBs can be used to assess impacts of any continuing inputs relative to legacy contamination and to estimate time scales of recovery toward background concentrations for different PCB sources. Distinguishing PCB sources allows for examination of the relative importance of the Upper Hudson River as a source of PCBs to the Harbor. This will inform assessment of the extent of the injury caused to the Harbor by the PCB inputs, and will therefore aid the Natural Resource Damage Assessment process for the UHR Superfund site, and will provide context for monitoring of the recovery post-remediation. The Hudson River system is representative of many urbanized waterways that received PCB inputs from diverse sources over an extended period, so understanding past and present PCB sources in the Harbor may aid efforts to protect other systems as well.

## 2. Materials and methods

Data were obtained via request to the Hudson River Foundation (HRF), which provides the data on CDs in several MS Access databases. For this work, the data from the CARP\_NY\_SED\_VIEWS.mdb was used.  $^{137}\text{Cs}$  measurements in the sediment cores were provided by the HRF as a MS Excel spreadsheet upon request. All of the methods and quality assurance pertaining to the CARP program are available in the CARP documentation ([carpweb.org](http://carpweb.org)) (Contamination Assessment and Reduction Project (CARP), 2007b, a). The CARP collected both surface sediment (0–10 cm) and deeper sediment cores in 2000 and 2001. Surficial sediment samples were collected using the following sampling devices: standard stainless steel Ponar for most surface samples; stainless steel box corer for both surficial and short cores generally less than 40 cm; Smith-McIntyre sampler for some surface samples; modified Van-veen sampler for some surface samples; and electric vibrocorer for deeper sediment cores. Radio dating samples were collected from extruded cores and placed in zip-lock bags marked with the site ID and depth. These samples were submitted to Rensselaer Polytechnic Institute (RPI) for analysis. All 209 PCB congeners were analyzed in 172 peaks by USEPA method 1668A (USEPA, 1999) using the SPB-octyl gas chromatography column and high resolution mass spectrometry.

### 2.1. Positive matrix factorization (PMF) methods

Of the 172 peaks detected, 90 peaks representing 136 congeners were used in the initial PMF analysis. These 90 were chosen to match the 90 used in our previous study of PCB fingerprints in the water column of the Harbor, which also utilized CARP data (Rodenburg et al., 2011a). Based on the results of the initial PMF runs, four peaks out of this 90 were discarded for the final data analysis: PCBs 11, 194, 195, and 196. PCB 11 was discarded because it was not well described by the initial PMF results and introduced instability into the solution, making it difficult to identify the correct number of factors. PCB 11 is often not well-described by PMF models (Du et al., 2008; Rodenburg et al., 2012) because it is produced inadvertently during the synthesis of diarylide yellow pigments (Litten et al., 2002a), and therefore arises from a different source than virtually all other PCB congeners. PCBs 194, 195, and 196 were also not well described in initial model runs, and since these congeners are not particularly useful in identifying PCB sources, they were discarded.

The final data matrix therefore contained 86 peaks comprising 132 congeners measured in 204 samples (17544 data points). A listing of all sediment cores is provided in Supporting Information, Tables S–1. Of these, 874 (4.98%) were below detection limit (BDL). For the concentration matrix, values BDL were replaced with one-half of the limit of detection (LOD) (Rodenburg et al., 2014, 2015a, 2015c). The LOD matrix was constructed from the actual LODs for each congener in each sample, which were provided in the MS Access database and ranged from 0.000116 to 0.28 ng/g dry weight. Surrogate recoveries ranged from 17% to 294%. The relative standard deviations (RSDs) of the surrogate recoveries, which ranged from 13% to 41%, were used as the uncertainty matrix (Rodenburg et al., 2011b, 2012; Praipipat et al., 2013; Rodenburg and Meng, 2013; Rodenburg et al., 2014, 2015a, 2015c). The data set was analyzed using PMF2 (Paatero and Tapper, 1994).

## 3. Results

PCBs were detected in virtually all sediment samples, even in the deepest layers of the sediment cores. Total PCB concentrations ( $\Sigma_{132}\text{PCB}$ ) in all samples (surface and core) ranged from 2.3 to

36,100 ng/g dry weight. The lowest concentrations were found in the deepest layers of the cores. In surface sediment, the  $\Sigma_{132}\text{PCB}$  concentrations ranged from 28.5 ng/g dw to 36,100 ng/g dw.

Dating of some of the sediment cores was attempted using  $^{137}\text{Cs}$ , which was measured only in a small subset of the cores.  $^{137}\text{Cs}$  is a product of fallout from the testing of nuclear weapons, with its first appearance in a core from a depositional environment corresponding with about 1954 and the peak concentration corresponding with about 1964 (Olsen et al., 1981). Within the limited number of cores with  $^{137}\text{Cs}$  measurements, only a few had concentration profiles that were sufficiently depositional to be dated accurately. This is consistent with previous studies in the Hudson that have found nearly uniform concentration or otherwise uninterpretable  $^{137}\text{Cs}$  profiles in regions where bed sediment remobilization by tidal currents or discharge events disrupts the continuous depositional record (Bopp and Simpson, 1989; Klingbeil and Sommerfield, 2005). Only one core from the East River near Flushing Bay (ER3) displayed an interpretable  $^{137}\text{Cs}$  profile and also was analyzed for PCBs. In this core, the first detection of  $^{137}\text{Cs}$  occurred at the core slice from 64 to 68 cm, implying an accumulation rate of 1.4 cm/y. The  $^{137}\text{Cs}$  maximum occurred in the core slice from 40 to 44 cm, implying an accumulation rate of 0.9 cm/y. The  $^{137}\text{Cs}$  maximum was only slightly higher than the  $^{137}\text{Cs}$  concentration in the surrounding slices, suggesting that the  $^{137}\text{Cs}$  onset was a more reliable indicator of age, as was found in a study of accumulation rates in the LHR near HR001 (Fig. 1) (Klingbeil and Sommerfield, 2005). One other core displayed an interpretable  $^{137}\text{Cs}$  profile, although PCBs were not measured in it. In this core from the Arthur Kill (AK001), the  $^{137}\text{Cs}$  onset corresponded with an accumulation rate of 1.4 cm/y and the  $^{137}\text{Cs}$  peak with an accumulation rate of 0.6 cm/y. The rates derived from the CARP cores were consistent with previous estimates of accretion rates in the LHR and Harbor based on  $^{137}\text{Cs}$  of 0.7–1.5 cm/y (Klingbeil and Sommerfield, 2005).

Unfortunately,  $^{137}\text{Cs}$  was not measured in the majority of sediment cores. To estimate the age of the sediment layers at these locations we assume that the maximum  $\Sigma\text{PCB}$  concentration occurred around 1973. PCB production in the US was at its maximum in 1970, and presumably the maximum release of PCB closely followed this date (Agency for Toxic Substances and Disease Registry (ATSDR), 2000). In 1973, the removal of the Fort Edward Dam also resulted in a huge influx of sediment contaminated with PCBs from the Upper Hudson River. In 1986, Ayres and Rod (Ayres and Rod, 1986) noted that maximum PCB concentrations in dated sediment cores from the NY/NJ Harbor corresponded with 1973, and that “The benchmark year of 1973 revealed such a clear PCB peak on all records that it can be used as the reference point to normalize the curves for earlier and later years.” By assuming that the highest PCB concentrations in each sediment core represent 1973, sediment accumulation rates can be estimated for 14 cores in which the maximum  $\Sigma\text{PCB}$  concentration is found in a layer above the deepest sampled layer. By this method, calculated sediment accumulation rates ranged from 0.36 cm/y to 2.3 cm/y. The accumulation rate across the 14 cores averaged  $1.12 \pm 0.55$  cm/y. The dating method is not perfect: when these accumulation rates were used to estimate the age of the core slices in each of the 14 cores, several slices with significant PCB concentrations are calculated to have deposited prior to 1900, which is highly unlikely. However, the imputed deposition rates were similar to those measured by other researchers in various areas of the Harbor. Bopp et al. (1991) presented  $^{137}\text{Cs}$  profiles for three cores collected in the Harbor in 1985–1986, from which we calculate accumulation rates of approximately 1.4 cm/y for the Upper Passaic River, 1.7 cm/y for the Lower Passaic River, and 1.2 cm/y for Newark Bay. Ferguson et al. (2003) reported sedimentation rates in Jamaica Bay from cores

collected in 1988 and 1996 of 1.4 cm/y and 0.92 cm/y, respectively. Klingbeil et al. (Klingbeil and Sommerfield, 2005) likewise reported sedimentation rates of about 1.1–1.5 cm/y in the LHR. The Contamination Fate, Transport and Bioaccumulation (CFTB) Model (Contamination Assessment and Reduction Project (CARP), 2007b) constructed for the NY/NJ Harbor compiled a detailed solids mass balance and predicted rates of deposition in model segments throughout the Harbor that were broadly consistent with the deposition rates calculated for the CARP sediment cores. For the ER3 core, the  $^{137}\text{Cs}$  dating implied a sedimentation rate of 0.9–1.4 cm/y, while the PCB maximum suggested a deposition rate of 0.7 cm/y (See Supporting information Figure S-1).

The core collected at site HR001 is particularly important because it is one of the deepest cores (maximum depth 2.28 m) and because it is located in the main stem of the LHR, so it can be used to estimate PCB inputs to the Harbor from the UHR.  $^{137}\text{Cs}$  was not measured in this core. The deposition rate of this core estimated from the maximum  $\Sigma_{132}\text{PCB}$  concentration was 1.1 cm/y. When this accumulation rate is used to calculate the ages of the core slices, the first detection of significant quantities of PCBs (110 ng/g dw) occurred around 1958, which is consistent with previous cores from the Harbor and LHR (Bopp and Simpson, 1989; Bopp et al., 2006). In deeper slices, just 1–5 ng/g dw PCBs were detected.

### 3.1. PCB 11

The main focus of the PMF investigation was apportioning Aroclor-type PCB sources. However, several PCB congeners are associated with non-Aroclor sources. PCB 11 in particular is thought to arise primarily from the production and use of organic pigments (Litten et al., 2002b; Rodenburg et al., 2010b; Rodenburg et al., 2015b). Because it was not routinely measured before method 1668 (USEPA, 1999) was widely adopted, published accounts of PCB 11 concentrations in sediment cores are rare. Hu et al. (2011) observed that PCB 11 concentrations in sediment cores from the Great Lakes peaked in the 1950s, declined, and then peaked again in the 1970s or in the early 1980s.

Since PCB 11 was not included in the PMF analysis, it is examined separately (Fig. 1). PCB 11 was detected in all surface sediment samples at concentrations ranging from 0.22 ng/g dw to 150 ng/g (ppb), representing on average about 0.7% of  $\Sigma_{132}\text{PCBs}$ . In some deeper layers of sediment cores, PCB 11 was BDL, but it was detected in sediment layers as deep as 1.4 m. PCB 11 concentrations were not correlated with  $\Sigma_{132}\text{PCB}$  concentrations. In the vast majority of cores, PCB 11 concentrations were greater in deeper layers than in the surface sediment, suggesting that PCB 11 loads to the Harbor have decreased over the last ~40 years. Diarylide yellow pigments were manufactured for many years at a number of facilities that discharged process waste indirectly to the Harbor, mostly via the Passaic Valley Sewerage Commission (PVSC), which is the largest water treatment plant by flow into the Harbor (Litten et al., 2002b). According to CARP data, in 2002 PCB 11 was the dominant congener in the effluent of the PVSC (Litten et al., 2002b). The second-highest concentration of PCB 11 detected in surface sediment occurred near the PVSC outfall at site LB1. (Although PVSC is located in Newark, NJ, its outfall pipe travels under the Bayonne Peninsula to emerge in the Upper Bay.) The highest surface concentration of PCB 11 was detected in the Arthur Kill at site AK999, where it comprised about 6% of  $\Sigma\text{PCBs}$ . In the ER3 core, PCB 11 concentrations were greatest (12 ng/g dw) in the same core slice containing the maximum  $\Sigma_{132}\text{PCBs}$  (i.e. 1973). PCB 11 was detectable at 1 ng/g dw in the deepest part of this core (0.48–8 m, 1930s).

PCB 11 was first detectable in the HR001 core in the 0.3–0.4 m slice, which based on the  $\Sigma_{132}\text{PCB}$  dating corresponds with 1961–1971. The maximum PCB 11 concentration of 18 ng/g dw

occurred at the same depth as the maximum  $\Sigma\text{PCB}$  concentration (0.24–0.3 m), i.e. in the early 1970s. The PCB 11 concentration in the surface of this core was 4 ng/g dw. In general, maximum PCB 11 concentrations occurred at the same depths as the maximum  $\Sigma\text{PCB}$  concentrations. The overall decline in PCB 11 concentrations may be a result of the implementation of the Toxic Substances Control Act, which was enacted in 1976 and set limits on the amount of inadvertent PCBs that could be present in pigments (USEPA). Declines in PCB 11 concentrations could also be a by-product of overall reduction in water pollution resulting from the construction of at least 40 wastewater treatment plants in the Harbor area in the 1960s and 1970s (Interstate Environmental Commission, 2001).

### 3.2. PMF factors

The optimal number of factors from PMF analysis was determined to be seven (Fig. 2), based on several lines of evidence. First, seven factors produced a stable solution, with nine seed runs of the solution agreeing to within 1.35% relative standard deviation. Second, in a multiple linear regression of the measured  $\Sigma_{132}\text{PCBs}$  (y) versus the G matrix (x's), the coefficients were positive and significant ( $p < 0.05$ ) for all seven factors. Third, the factors were independent of each other. Fourth, the solution was interpretable.

Factor 1 was similar to Aroclor 1242 ( $R^2 = 0.60$ ), and comprised 14% of the PCB mass across the entire data set with the highest concentrations being found in the Lower Hudson River and Upper Bay (see below and Supporting information figure S-2). This factor was also similar ( $R^2 = 0.57$ ) to the PMF factor isolated from the water column CARP data (Rodenburg et al., 2011a) that represented the UHR General Electric PCB source. The main difference was in the amount of PCB 4, which is more abundant in the UHR water column samples than in Aroclor 1242 or in factor 1. Microbial dechlorination is a major source of PCB 4, resulting in increased PCB 4 in the UHR water column relative to the original Aroclor 1242 fingerprint (Brown et al., 1984). Water column data collected under the CARP program showed that PCB 4 concentrations decreased by about a factor of 3 over the 140 miles from the UHR north of Troy to the area south of the Haverstraw Bay/Tappan Zee, while the concentrations of higher molecular weight PCB congeners remained unchanged. The water column data indicate that PCB 4 volatilizes extensively or is aerobically degraded during transport down the river, consistent with its reduced prevalence in the bed sediment factor 1 of the LHR.

The PMF program did not isolate any other factor that resembles Aroclor 1242, suggesting that it could not discriminate between the UHR fingerprint and Aroclor 1242 used in the Harbor region. However, factor 1 was not very abundant in areas that are highly urbanized/industrialized but not well connected hydrodynamically to the UHR, such as the Arthur Kill and the Long Island Sound (see “Spatial distribution of factors” below and figure S-2 of supporting information). In the nearby Delaware River, no factor resembling Aroclor 1242 was isolated during PMF analysis of the PCB data from the sediment (Praipipat et al., 2013), suggesting that release of Aroclor 1242 was not widespread across the urban/industrial areas of the Mid-Atlantic states (perhaps because it was primarily used in closed applications such as transformers (Agency for Toxic Substances and Disease Registry (ATSDR), 2000)). Input of contaminants from the UHR remained a massive and continuing source of Aroclor 1242 and its degradation products over the several decades since PCB use urban/industrial areas declined (TAMS Consultants and the Gradient Corporation, 1997; Totten, 2005). Remedial dredging in the UHR conducted from 2009 to 2015 (i.e. after CARP sampling) removed an estimated 146 metric tons of PCBs from the UHR sediment (Parsons, 2016) much of it in the form of dense non-aqueous phase liquid (DNAPL) (The Louis Berger Group, Inc, 2010). These two lines of evidence (i.e. that factor 1 is

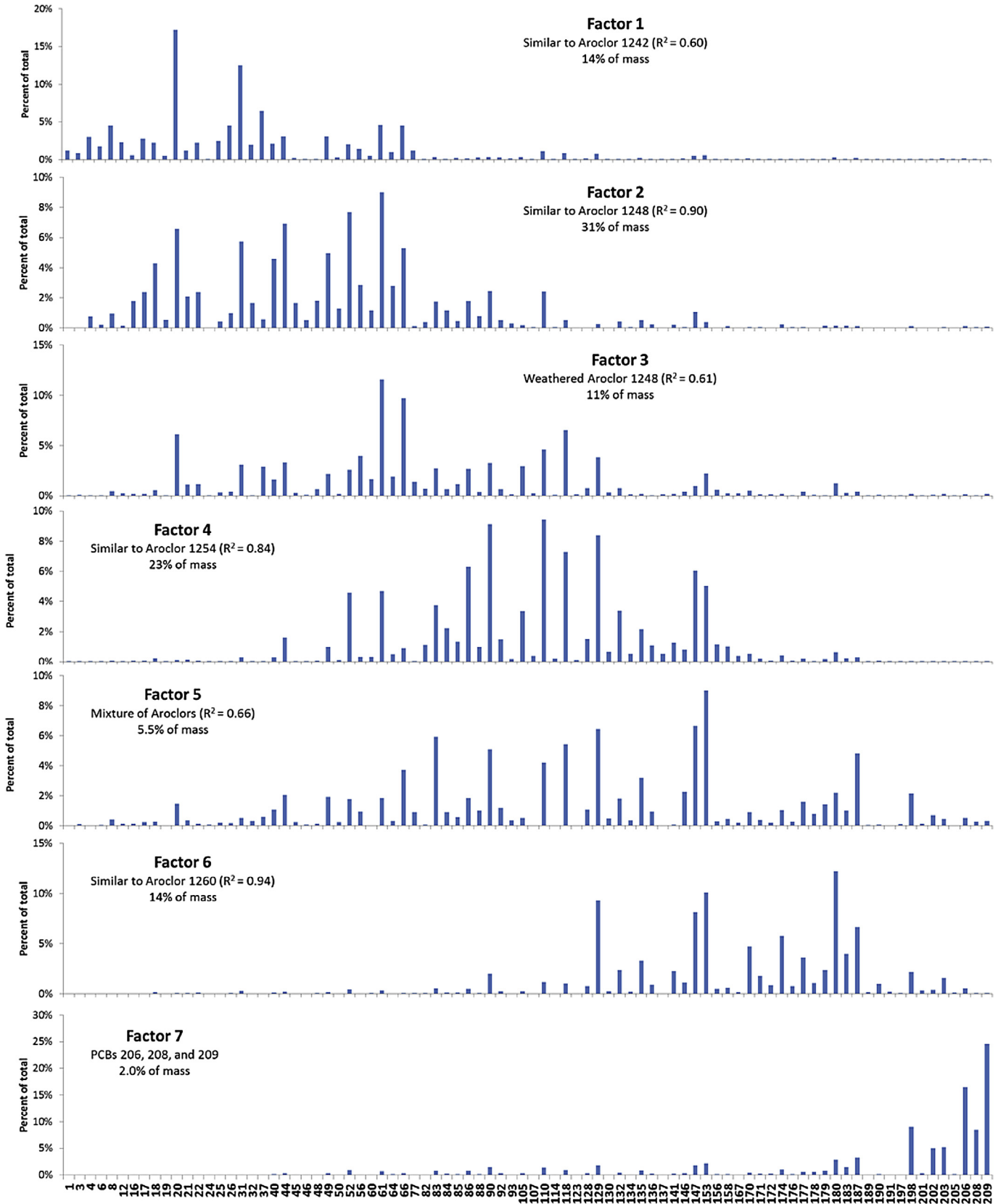


Fig. 2. Congener profiles of the seven resolved factors (weight % contribution to the sum of PCBs plotted versus IUPAC congener number).

concentrated in areas of the NY/NJ Harbor that are hydrodynamically connected to the UHR and that other regions, including the

nearby Delaware River, do not display large amounts of Aroclor 1242) suggest that the vast majority of factor 1 is related to inputs

from the UHR, not local use of Aroclor 1242 in the Harbor.

The other main Aroclors (1248, 1254, and 1260) were represented by Factors 2, 4, and 6, respectively. The correlations ( $R^2$ ) between these factors and their corresponding Aroclor congener fingerprints were 0.84 or greater, suggesting that these factors represent relatively unweathered Aroclors. Factors 2, 4, and 6 comprised 31%, 23%, and 14% of the PCB mass across the entire data set and were therefore some of the dominant PCB constituents in the Harbor.

Factors 3 and 5 represented weathered Aroclor patterns. Factor 3 somewhat resembled Aroclor 1248 ( $R^2 = 0.61$ ). The best fit for this factor as a mixture of Aroclors was a 74/17/8 mixture of Aroclors 1248, 1254, and 1260, respectively, but this best fit mixture only resulted in a  $R^2$  of 0.66, suggesting that this factor represented weathered 1248 or a weathered mix of low molecular weight Aroclors. Factor 3 comprised 11% of the PCB mass across the entire data set. Factor 5 was best described as a 3/11/35/45 mix of Aroclors 1242, 1248, 1254, and 1260, but again the  $R^2$  value for this best match was lower (0.67). Factor 5 therefore represented a weathered mixture of high molecular weight Aroclors, comprising 5% of the PCB mass across the entire data set. Factor 5 was most prevalent in the Long Island Sound and may therefore represent a unique source in that region (see below).

Factor 7 was dominated by PCBs 206, 208, and 209, and comprised just 2.0% of the PCB mass across all samples. In the Delaware River, these congeners have been tied to the production of  $TiCl_4$ , which is subsequently used to produce  $TiO_2$  (Du et al., 2008; Rodenburg et al., 2010a; Praipipat et al., 2013). It is also possible that factor 7 in deep core layers was related to foundry wax castings, which used high molecular weight PCB formulations (Durfee, 1976; Erickson and Kaley, 2011). Potential sources relating to Factor 7 are discussed in more detail in the Supporting Information.

### 3.3. Comparison of water and sediment fingerprints

Given the strong linkage between suspended sediment in the water column and surface bed sediment, the fingerprints of the major PCB sources should be similar in both compartments. A previous PMF analysis of PCB fingerprints in the water column (dissolved plus particulate) revealed seven fingerprints (here denoted W1 through W7) (Rodenburg et al., 2011a). In these samples, the particulate phase accounted for an average of 56% of the total PCBs. Six of the seven sediment factors in this study had a corresponding match among the water factors (Supporting Information figure S-3). Only factor 7, which was dominated by PCBs 206, 208, and 209, did not have match among the water factors. This is not surprising given that factor 7 was most prevalent in deeper core slices, suggesting that it was an important PCB source many years ago, not in 1998–2001 when the water samples were collected. Factor 5 somewhat resembled ( $R^2 = 0.61$ ) factor W5, but this correlation was probably not meaningful because W5 was dominated by PCB 11, a congener that was not included in the sediment PMF analysis. Nevertheless, it is reassuring to note that the PMF analysis of PCB fingerprints in the water column and sediment were generally in agreement about the identity and relative weighting of the PCB sources in the Harbor. Lower molecular weight fingerprints were generally more abundant in the water column than in the sediment (Fig. 3). This is consistent with the greater partitioning of lower molecular weight congeners into the dissolved phase based on their hydrophobicity and was observed in the CARP data. In the Delaware River, PMF analysis of PCB fingerprints in the water column and sediment revealed a similar trend (Praipipat et al., 2013).

### 3.4. Spatial distribution of factors

The spatial variations in the various PCB factors derived from water and surface sediment are shown in Supporting Information Figures S-2 and S-3. In the HR001 core in the LHR, Factor 1 was most prevalent, consistent with its correlation with Aroclor 1242 and the UHR source. Factor 1 was also prominent in cores from the Upper and Lower Bay, which are directly seaward of the LHR. In the surface sediment from core HR001, factor 1 comprised 57% of the sum of PCBs, and it was about 30% of the sum of PCBs in the surface sediment in the Upper Bay cores. This prevalence is consistent with estimates that the UHR is the source of about half the PCBs entering the Harbor at the time of the CARP sampling (Totten, 2005; Contamination Assessment and Reduction Project (CARP), 2007b) although the discussion above about the prevalence of the various factors in water versus sediment suggests that the UHR PCB source is proportionately more important in the water column than in the sediment.

Factor 1 was also prominent in cores from areas that are not immediately adjacent to the LHR, such as Newark Bay (NW01, NW01A), Jamaica Bay, Raritan Bay, and Newtown Creek in the East River. The UHR is the primary source of fine sediment for the entire NY Harbor region, and estuarine and tidal processes redistribute fine sediment and associated contaminants throughout the system. During high river discharge periods such as the spring freshet, sediment load from the UHR increases and the sediment fluxes are from the LHR to the Harbor (Geyer et al., 2001). Extreme river discharge events can even more dramatically increase both sediment supply and the seaward sediment transport. For example, two tropical storms in 2011 moved an estimated 500,000 tons of sediment from the LHR to the Harbor within a few weeks (Ralston et al., 2013). Sediment delivered to the Harbor during high discharge periods deposits temporarily, and then during lower discharge periods is moved back up the LHR as well as to other parts of the Harbor region by the estuarine circulation and tidal dispersion. For example in Newark Bay, sediment fluxes coming from the Upper Harbor, and ultimately from the LHR, were six times greater than the local sediment inputs from the Passaic and Hackensack Rivers combined (Sommerfield and Chant, 2010). A previous assessment of PCB sources for Newark Bay did not identify the UHR as a primary source (Saba and Su, 2013), but did find that a PMF factor similar to Aroclors 1242 and 1248 was dominant in the region near our Newark Bay cores (NWB01 and NWB01A). Redistribution of sediment from LHR also likely contributes to the prevalence of factor 1 in cores from Newtown Creek, Jamaica Bay, and the Harlem River. These locations are also highly impacted by wastewater treatment and CSO discharges, so factor 1 may also include weathered Aroclor 1242 arising from to microbial dechlorination of PCBs in sewers and landfills (Rodenburg et al., 2012).

Factors 2, 3, and 4 all had similar spatial distributions, being highest in concentration and the largest contributors to PCB mass throughout the Harbor, East River, and Arthur Kill. These factors represent the most widely used Aroclor formulations and these locations have some of the highest population densities and receive inputs from treated sewage, urban stormwater runoff, and combined sewer overflows, which are thought to be some of the dominant sources of PCBs to the Harbor, after the UHR (Totten, 2005).

Concentrations of factor 5 (weathered high MW Aroclors) were spread relatively evenly across the entire study area, including Long Island Sound. As a percent of total PCBs, factor 5 was the dominant PCB source in the Long Island Sound. Bedard et al. (Bedard and May, 1996; VanDort et al., 1997) have examined congener fingerprints from the Woods Pond Superfund site in the Housatonic River basin, and concluded that the Aroclors 1254 and 1260 originally released

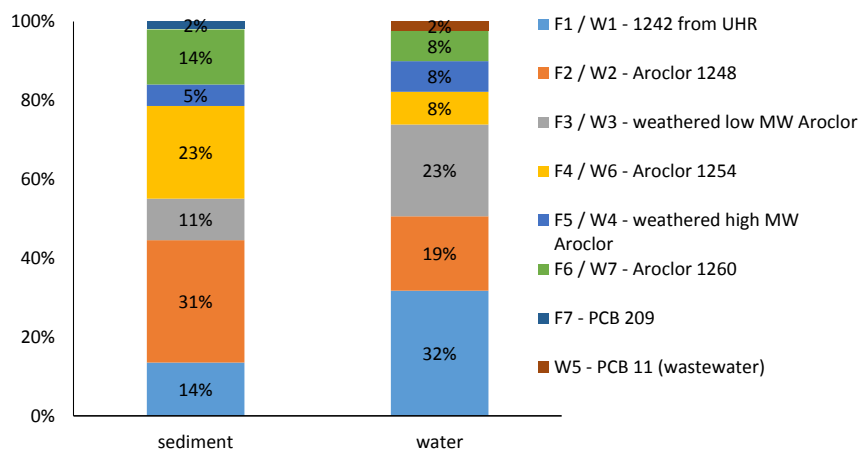


Fig. 3. Distribution of sediment factors (F1 through F7) and their corresponding water column factors (W1 through W7) in water and sediment.

in this system have undergone measurable microbial dechlorination, giving rise to a PCB signal in the Housatonic River that resembles these Aroclors but is altered to include more lower molecular weight PCB congeners. The PMF analysis suggests a distinct source of PCBs to the Long Island Sound different from the NY Harbor region, perhaps representing contributions from the Housatonic.

Factor 6 (Aroclor 1260) was similar to factor 2, 3, and 4 in that its highest concentrations occurred in the central Harbor area. However, factor 6 was also high in Jamaica Bay, where it dominated as a percent of total PCBs. Treated wastewater and combined sewer overflows are likely the primary source of PCB inputs to Jamaica Bay (New York City Department of Environmental Protection, 2007). Previous studies have shown that sediment PCB concentrations in Jamaica Bay decreased by ten-fold from the peak around 1970 to the mid-1980s with reduced usage and improved treatment methods (Bopp et al., 1993).

Factor 7 (PCBs 206, 208, and 209) was not particularly prevalent in any of the surface sediments. It was most important in deep layers of cores collected at UB3A (Upper Bay), ARK04 (Arthur Kill), and RB1 (Raritan Bay). As noted above, there are two likely sources of these congeners in the Harbor: TiO<sub>2</sub> production and waxes used in the lost-wax casting process. These are explored in more detail in Supporting Information.

### 3.5. Depth profiles of the factors

Here we focus on four cores of significance to the Harbor. The ER3 core is discussed in detail because it has the most reliable dating. The HR001 core is discussed because it lends insight into the magnitude of the UHR source relative to other sources of PCBs in the Harbor. Finally, cores from Newark Bay (NWB01 and NWB01A) are discussed because they lend insight into sources of PCBs to this area, which is of high concern in the Harbor.

### 3.6. ER3 core

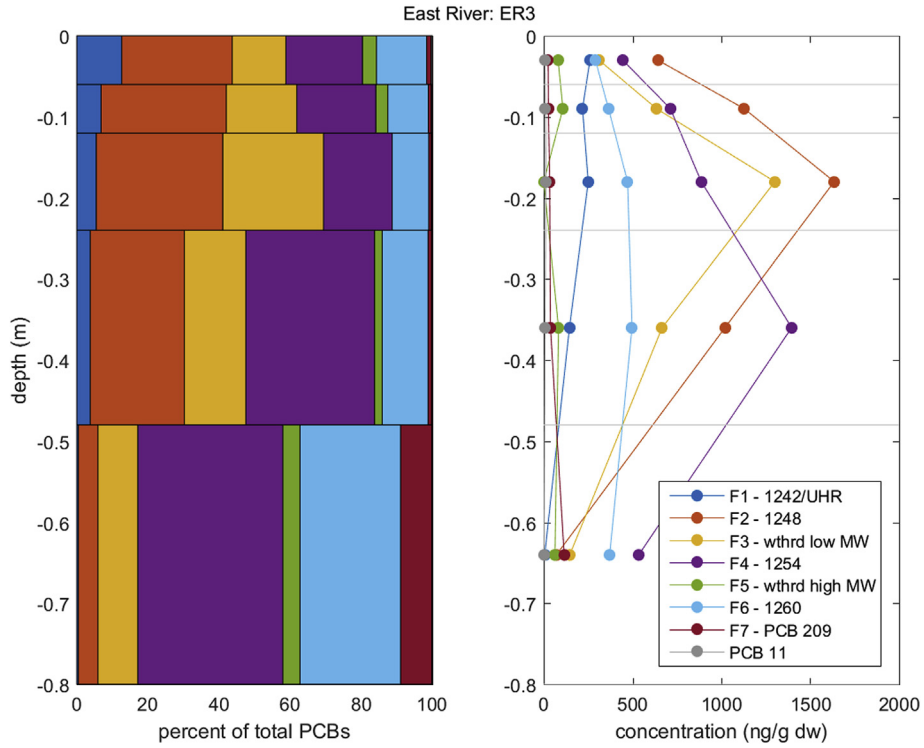
The one core with an interpretable <sup>137</sup>Cs profile was ER3. In the deepest slices of this core (0.48–0.8 m, 1930s), factor 7 (PCBs 206, 208, and 209) was discernable (Fig. 4). This deepest layer corresponded with increasing <sup>137</sup>Cs up the core from background levels, suggesting it is consistent with the increased PCB usage in the 1950s and early 1960s. In the layer from 0.24 to 0.48 m, corresponding to the 1960s, factor 4 (Aroclor 1254) was the dominant source of PCBs (Fig. 4). In more recent layers, factor 2 (Aroclor 1248)

was the dominant source of PCBs.

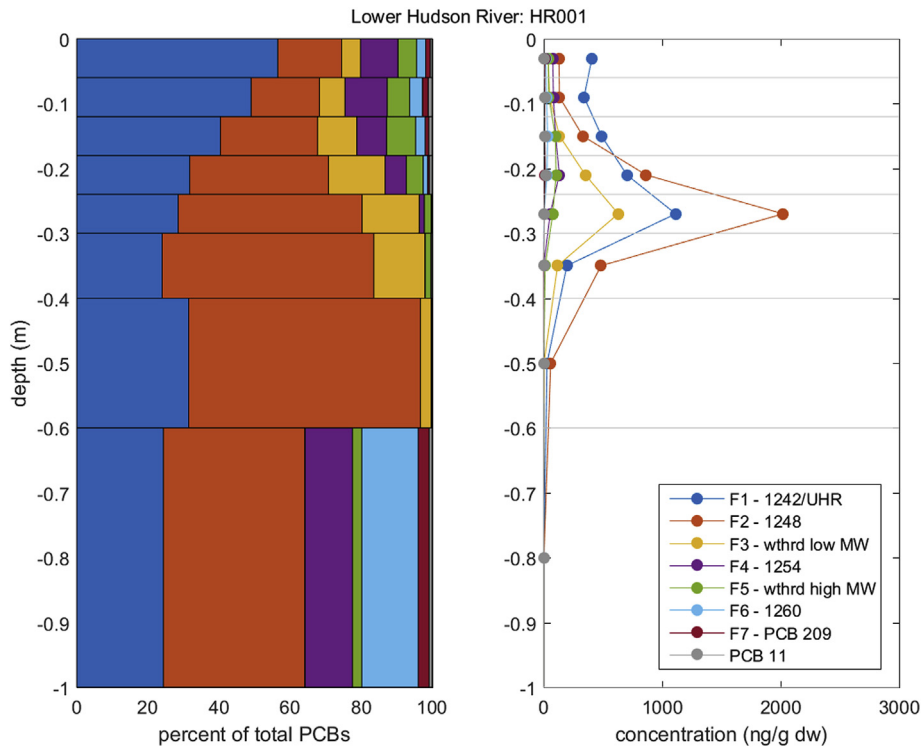
### 3.7. HR001 core

In core HR001 the highest concentration of the sum of PCBs occurred during the in the core slice from 0.24 to 0.3 m (Fig. 5). Factors 1, 2, and 3 generally declined in concentration from that depth to the surface. All of the other factors also decreased toward the surface, but from maximum concentrations in shallower slices. Factor 1, which is thought to be related to UHR contamination, diminished in relative importance with depth in the HR001 core. At the surface it comprised 57% of Σ<sub>132</sub>PCB, but in the slice containing the PCB maximum it accounted for only 29%. At the General Electric plants in the UHR, the primary PCB formulation used in the manufacture of capacitors changed over time. Aroclor 1254 was predominant from 1946 to 1955, Aroclor 1242 was predominant from 1955 to 1971, and Aroclor 1016 was predominant from 1971 until the plants switched to non-Aroclor fluids around 1977. GE estimates that at least 80% of PCBs discharged were Aroclor 1242, with lesser amount of Aroclors 1016, 1221, and 1254 (TAMS Consultants Inc. et al., 2000). The HR001 core indeed shows relatively little contribution from factor 4 (Aroclor 1254) from the surface (2001) to a depth of 0.6 m (i.e. 1940s). Deeper than this (0.6–2 m, i.e. before 1940), factor 4 comprised a larger fraction of the sum of PCBs, but the absolute concentrations were less than 1 ng/g.

Factors 2 (Aroclor 1248) and 3 (weathered low MW Aroclors) had maximum concentration in the same slice as factor 1, but the subsequent declines have been faster for factors 2 and 3 than for factor 1. A regression of the natural log of the factor concentration versus depth (as a surrogate for time) suggests that factors 1, 2, and 3 all displayed significant ( $p < 0.05$ ) declines since 1973, but factors 2 and 3 have been declining significantly faster than factor 1. Using the core dating to convert the rate of decline (in meters of depth) to a half-life suggests that factor 1 is decreasing with a half-life of 14 years, versus about 5 years for factors 2 and 3. This is consistent with the sediments of the UHR acting as a large reservoir of PCBs that have continued to release factor 1 long after other major sources have been eliminated by regulatory actions. In contrast, factors 2 and 3 likely came from direct discharges that were curtailed immediately after the passage of the TSCA. A similar assessment of decreasing PCB concentrations toward the surface of a core collected in 1986 from the freshwater tidal Hudson (143 km from the Battery) estimated a recovery time scale of about 3.5 years (Bopp and Simpson, 1989). The PCB load flowing over the Troy Dam



**Fig. 4.** Prevalence of the 7 PMF-resolved factors and PCB 11 in the ER3 core from the East River. Right panel shows concentrations of the PMF-resolved factors with depth and imputed age. Left panel shows the relative contribution of each factor to the sum of PCBs in each core slice.



**Fig. 5.** Prevalence of the 7 PMF-resolved factors and PCB 11 in core HR001 in the Lower Hudson River near the Harlem River. Right panel shows concentrations of the PMF-resolved factors with depth and imputed age. Left panel shows the relative contribution of each factor to the sum of PCBs in each core slice. Total depth of core was 2.28 m. Deeper layers are not shown.

from the UHR to the LHR/Harbor declined by about an order of magnitude during the 1980s, but during the 1990s there was little

or no decline in this load (U.S. EPA, 2002). These changes in loads are consistent with a faster decline in PCB sediment concentrations



observed by Bopp et al. versus the slower decline observed in this study.

### 3.8. NWB01 and NWB01A cores

The core from NW01 (Fig. 6) was deep enough that PCBs in the deepest core slice were barely detectable. The bottom slice extended from 0.48 to 0.84 m and would have been deposited prior to about 1930 based on the PCB maximum ( $^{137}\text{Cs}$  was not measured in either of these cores).  $\Sigma\text{PCB}$  concentrations in the next slice (0.48–0.36 m, i.e. 1940s) were just 32 ng/g dw. In the subsequent slice (0.24–0.36 m, i.e. 1960s) the  $\Sigma\text{PCB}$  concentrations rose dramatically to 494 ng/g dw, with factor 2 (Aroclor 1248) dominating the sources followed by factor 3 (weathered low MW Aroclors).

In core NW01A (see supporting information for data), factor 2 (Aroclor 1248) dominated in most layers. It declined in concentration from slice 0.22–0.52 m (about 1980) upwards, but concentrations of the other factors remained largely constant throughout the core with the exception of factor 4 (Aroclor 1254). Factor 4 dominated in only one slice (0.06–0.12 m, 1990s) and its concentrations were largely uniform throughout the rest of the core. Dating of this core was highly uncertain since  $^{137}\text{Cs}$  was not measured and PCBs had two maxima.

Saba and Su (2013) conducted a similar PMF analysis of PCB fingerprints in Newark Bay using data from the Remedial Investigations for the Newark Bay Study Area and the Passaic River. In those studies, PCBs were measured using method 1668A, but on a DB-5 chromatography column, leading to a different co-elution pattern than observed in the present work. In addition, they used a slightly different list of congeners in their PMF model. As a result, it is not possible to compare directly their factors with ours. Like us, Saba and Su found a fingerprint (their F2) containing PCBs 206, 208, and 209, which comprised 14% of the PCBs in their data set. That

factor also contained more of the other Aroclor-type congeners, which may explain why their F2 accounted for more mass in Newark Bay (14%) than in our factor 7 (3%). Their factor F3, which they attributed to Aroclors 1242 and 1248, was most similar to our factor 1. It accounted for 16% of the PCBs in their data set, similar to our factor 1 which constituted 22% of  $\Sigma\text{PCBs}$  in the surface of core NWB01 and 16% of  $\Sigma\text{PCBs}$  in the surface of nearby core NWB01A. Although the absolute concentrations of PCBs have declined since their peak in the 1970s (i.e. 0.12–0.24 m), the relative contributions of the 7 factors have remained fairly constant over this period.

## 4. Conclusions

The PMF results suggest that the dominant PCB sources to the Harbor have changed over time. In the early years before and during World War II, foundry wax and/or titanium dioxide manufacture introduced high molecular weight PCBs to the Harbor (Factor 7). After World War II, Aroclors became the dominant sources of PCBs. Unlike the nearby Delaware River, where Aroclor 1242 is not prevalent, Aroclor 1242 (factor 1) was an important component in the NY/NJ Harbor due to inputs from the UHR. Analysis of core HR001 suggests that Aroclor 1242 (factor 1) from the UHR has been the source of between 30% and 50% of the PCBs in the LHR during the period from about 1970 to 2000, and that PCBs from this source have dispersed throughout the Harbor, including Newark Bay. Furthermore, the relative importance of the UHR source increased from 1973 to 2000 because inputs from local PCB sources were declining more rapidly. Previous assessments suggested similar levels of PCB inputs from local and UHR to the Harbor in the 1980s, but that the UHR was a more dominant source (75%) in the 1970s (Bopp and Simpson, 1989). The PMF results suggest a more pronounced local source during the period of maximum PCB production and use, but that by the time the CARP data was collected (2000–2001) the UHR had become the dominant source

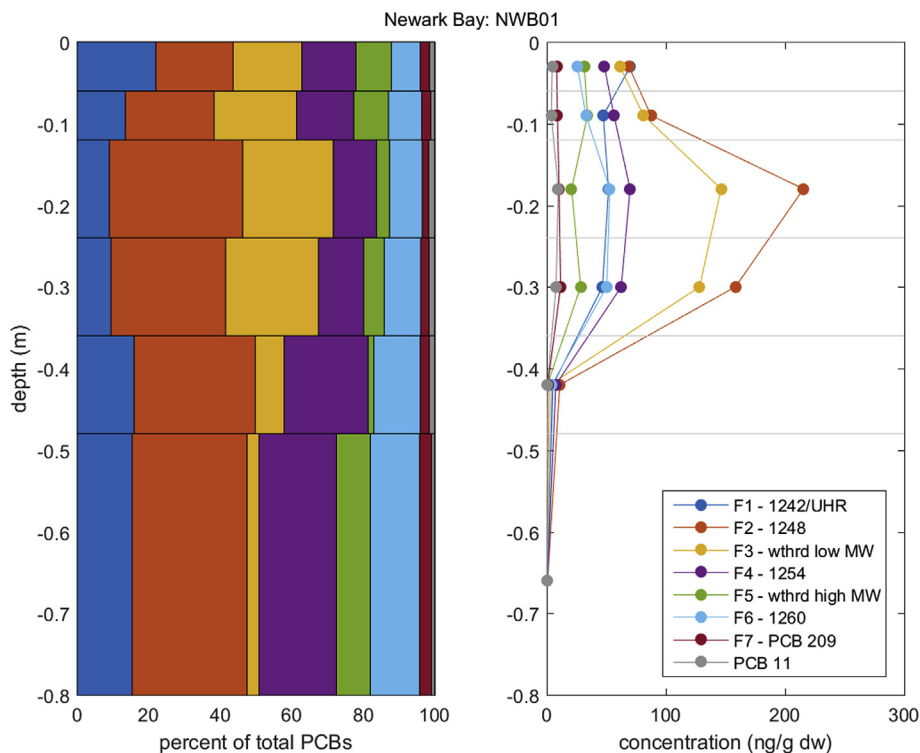


Fig. 6. Prevalence of the 7 PMF-resolved factors and PCB 11 in core NWB01 in Newark Bay. Right panel shows concentrations of the PMF-resolved factors with depth and imputed age. Left panel shows the relative contribution of each factor to the sum of PCBs in each core slice.

of continuing input. It remains to be seen how the removal of 146 metric tons of PCBs from the UHR sediment by dredging (Parsons, 2016) will impact PCB concentrations in the sediment of the LHR and NY/NJ Harbor.

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### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.chemosphere.2016.11.096>.

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