Trends in Chlorinated Hydrocarbon Levels in Hudson River Basin Sediments

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Analyses of sections from dated sediment cores were used to establish geographic distributions and temporal trends of chlorinated hydrocarbon contaminant levels in sediments from natural waters of the Hudson River Basin. Radiometric dating was based primarily on the depth distribution of 137Cs in the cores and on the occurrence of detectable levels of 210Pb in surface sediment samples. Eighteen sampling sites included several along the main stem of the Hudson, its major tributaries, and components of the New York/New Jersey (NY/NJ) harbor complex. Drinking-water reservoirs were sampled to place upper limits on atmospheric inputs. Core sections were analyzed for polychlorinated biphenyls (PCBs), 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (DDT)-derived compounds, chlordane, and dioxins. Sediment concentrations of most contaminants at most sites have decreased significantly since the mid-1960s. The data provide a basinswide perspective on major point-source inputs of PCBs to the upper Hudson River and of 2,3,7,8-tetrachlorodibenzo-p-dioxin and DDT to the lower Passaic River. Evidence was found for significant but poorly characterized sources of PCBs and chlordane to the western NY/NJ harbor, and of highly chlorinated dioxins to the upstream sites on the main stem of the Hudson. The results indicate that analysis of dated sediment samples is a most effective and efficient monitoring tool for the study of large-scale geographic and temporal trends in levels of particle-associated contaminants.

Key words: Hudson sediments, contaminant levels, radionuclide dating, PCBs, dioxin, DDT, chlordane, 137Cs

Introduction

Over the past 20 years, we have collected several hundred sediment cores from natural waters in the Hudson River Basin together with the major tributaries to the New York/New Jersey (NY/NJ) harbor complex. Sampling sites included the main stem of the Hudson, its major tributaries, several lakes and reservoirs, and the western NY/NJ harbor including Newark Bay and the Hackensack and Passaic Rivers (Figure 1). Radionuclide analyses were used to provide dating information, and sections from cores with readily identifiable time horizons were analyzed for particle-associated pollutants to provide contaminant chronologies. Publications resulting from this work [e.g., Bopp et al. (1-4)] have generally focused on a specific location (western NY/NJ harbor, the main stem of the Hudson, Jamaica Bay, etc.) and often on a particular contaminant or group of contaminants (polychlorinated biphenyls [PCBs], dioxins, or trace metals).

In this paper, data are presented in a much broader geographical perspective—that of the entire Hudson basin together with major tributaries to the NY/NJ harbor complex. Several chlorinated hydrocarbon contaminants are considered including PCBs, dioxins, 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (DDT), and chlordane. A similar treatment of the trace metal data is being prepared. This perspective on contaminants in the Hudson provides insights useful to regulators and managers who must deal with diverse and complex issues ranging from food chain transport of these substances to humans, to the disposal of contaminated sediments dredged from the Hudson River and the NY/NJ harbor complex, to assessment of the impact of individual Superfund sites on the overall system.

Methods

The majority of sediment samples were collected with gravity or piston cores. A few of the more recent particle samples were suspended matter collected on precombusted quartz fiber filters using a large-volume in situ pumping system (5). Radionuclide analysis of sediment core sections to obtain dating information was performed by nondestructive γ-ray spectrometry (1-4). Extracts of dated core sections for PCB, chlorinated hydrocarbon pesticide, and dioxin analyses were prepared using Soxhlet extraction and column chromatography cleanup. Most of the PCB samples were analyzed following procedures previously described by Bopp et al. (1). Total PCBs are generally reported as the sum of 18 individual packed column peak components ranging from dichlorobiphenyls through decachlorobiphenyl (2). This was not the case for Jamaica Bay (Figure 1, site 10) where, because of interfering peaks on the chromatograms, PCB levels were estimated as described elsewhere (4). At main-stem Hudson site 1 (Figure 1), where microbially mediated in situ reductive dechlorination has greatly increased the abundance of mono- and dichlorobiphenyls, congenerspecific analysis based on New
York State Department of Environmental Conservation Method 91-11 (6) was used to determine total PCB concentrations. Some of the PCB data discussed are from samples collected as part of the U.S. Environmental Protection Agency's (U.S. EPA) ongoing reassessment of the Hudson River PCB problem under federal Superfund activities (7) and analyzed by a congener-specific method (8) similar to 91-11 (6). Chlorinated hydrocarbon pesticides were analyzed following procedures reported elsewhere (7). Levels of chlorodane are reported in terms of \( \gamma \)-chlorodane, which comprises about 15% of technical chlorodane (9). Levels of 1,1-dichloro-2,2-bis(\( \gamma \)-chlorophenyl)ethane (DDD), an anaerobic breakdown product of DDT, are used as an indicator of the levels of DDT-derived compounds. Dioxin analyses were conducted at Quanterra Environmental Services (West Sacramento, CA) or at New York State Department of Health Laboratory (Albany, NY) following U.S. EPA Method 1613 (10) or at the University of Nebraska following a similar procedure. All contaminant concentrations in sediment samples are reported on a dry weight basis.

**Sediment Core Dating**

The depth distribution of \( ^{137} \text{Cs} \) in sediment cores provides important information on timing of sediment deposition. \( ^{137} \text{Cs} \), derived as fallout from atmospheric testing of nuclear weapons, first entered natural water systems on a global scale in significant amounts in the early 1950s. In cores with continuous and relatively rapid sediment accumulation (on the order of 0.5 cm/year or greater), maximum levels of fallout \( ^{137} \text{Cs} \) can be associated with years of peak fallout delivery (1963, 1964) (11–13). In undisturbed cores, the activity of \( ^{137} \text{Cs} \) decreases approximately exponentially toward the sediment–water interface (1–4). For many reasons, such cores are not commonly encountered in river systems. In many areas of the Hudson River, for example, there is little or no net deposition of sediment. In depositional environments such as coves and interper areas, biologic and physical mixing, dredging, and scour associated with large storms and other high discharge events can disturb the \( ^{137} \text{Cs} \) depth profile. Of the several hundred sediment cores we have collected in the Hudson River Basin, fewer than 40 exhibit the complete, undisturbed profile of \( ^{137} \text{Cs} \) activity with depth described above. In addition to global fallout, other sources of \( ^{137} \text{Cs} \) to specific regions of our study area include effluent from the Indian Point Nuclear Power Generating Facility on the lower Hudson and the Knolls Atomic Power Laboratory on the Mohawk River (Figure 1). In both cases, the known history of discharge provides additional dating information in sediments from nearby sampling sites (2,14).

A near-ideal depth profile of fallout \( ^{137} \text{Cs} \) activity in a sediment core from the Batten Kill, a major tributary of the upper Hudson (Figure 1, site 2) is shown in Figure 2. On the basis of three time horizons noted in Figure 2, approximate dates of deposition can be assigned to individual core sections. The net sediment accumulation rate in this core is about 0.6 cm/year. Water content ranged from 51 to 69% with no regular down core trend, indicating a fairly constant mass accumulation rate of about 0.3 g cm\(^{-2}\) year\(^{-1}\). Such areas of undisturbed, continuous, relatively rapid sediment accumulation that yield readily interpretable depth profiles of \( ^{137} \text{Cs} \) activity are most useful for studying temporal trends in contaminant levels. This paper focuses on samples from about 20 such cores that we have collected from 18 sites in the Hudson basin (Figure 1).

Confirmation that the uppermost section of the cores contains recently deposited sediment comes from analysis of \( ^{7} \text{Be} \), a cosmic ray-produced radionuclide with a half-life of 53 days. Detectable activity of \( ^{7} \text{Be} \) is confined to sediment samples containing a significant component of particles deposited within about a year prior to sample collection (2,3).

\( ^{7} \text{Be} \) analysis was also most useful at several sites in the NY/NJ harbor where extremely rapid deposition (sometimes exceeding 10 cm/year) and disturbance of the sediment column caused by human activities such as dredging are commonly encountered. These factors make collection of cores with continuous decade-long...
records of sediment accumulation most difficult. Under such circumstances, surface sediments from areas of rapid deposition, if they contain detectable $^7$Be, define a time horizon within about a year prior to sample collection.

**Sampling Sites**

Our 18 sediment sampling sites in the Hudson River Basin, the locations of PCB inputs to the Hudson River Superfund site, and dioxin and DDT inputs from a lower Passaic River Superfund site are shown in Figure 1. The cores utilized in this study were all from depositional areas characterized by fine-grained (silt and clay sized) particle accumulation. The high affinity of the contaminants of interest for fine-grained particles makes these cores useful for comparisons of contaminant levels between sampling sites and for the identification and characterization of contaminant sources (2,3,14). Except in site 1 (Figure 1), where some core sections contained wood chips, organic matter content in individual cores typically varied over a factor of two or less, and we have found no correlation between contaminant levels and organic matter content (4). This indicates that the contaminant level chronologies at our sites are not strongly influenced by variations in sediment composition.

The Jamaica Bay (site 10) and Newtown Creek (site 9) sites were chosen as indicators of New York City wastewater inputs from primarily residential and more industrial areas, respectively. Over two-thirds of the freshwater inputs to Jamaica Bay consist of discharge from secondary sewage treatment plants and approximately 10% enter from combined sewer overflows (15). The Newtown Creek sampling site is in the middle of a tidal basin that receives, at its mouth, effluent from a secondary sewage treatment plant. The Newtown Creek water pollution control plant is among the largest sources of municipal wastewater to the NY/NJ harbor complex. In 1994, the discharge of 292 million gal comprised about 12% of the total municipal wastewater input (16). Wastewater inputs have long been recognized as significant sources of contaminants, including trace metals, PCBs, and chlorinated hydrocarbon pesticides, to NY/NJ harbor sediments (2,16,17).

Figure 2. The distribution of $^{137}$Cs activity with depth in a sediment core from Batten Kill Creek no. 5 (Figure 1, site 2) collected 26 October 1993. Error bars shown represent ± 1 sigma based on counting statistics.

For a wide range of contaminants, concentrations in NY/NJ harbor sediments from along the main stem of the Hudson (site 8) can be derived from linear mixing of upstream (site 5) and Newtown Creek (site 9) sediment concentrations (Table 1). For samples deposited in the mid-1980s, the calculated sewage contribution to NY/NJ harbor sediment contamination ranges from about 30 to 65%. This simple mixing model is also consistent with the organic matter content of the samples. Alternatively, the relative contaminant levels could simply reflect a dissolved contaminant load adsorbed more effectively by more highly organic particles. We would argue, however, that this is not the controlling process. Based on measurements of particle–water partitioning in Hudson River samples (18–20), most of the contaminants in Table 1 would be dominantly adsorbed at particles at the suspended particle concentrations that characterize the major transport events to our depositional sites.

The Ashokan and Muscoot Reservoirs (sites 6 and 7) are both part of the New York City municipal water supply system. Contamination in these systems should include contributions from atmospheric inputs and suburban, rural, and agricultural runoff, as well as any direct inputs. Because direct and runoff inputs of PCBs and dioxins to the reservoirs are expected to be small, these sampling sites are useful for determining a reasonably well-constrained upper limit on regional atmospheric inputs of these contaminants.

**Results and Discussion**

Basinwide data on the levels of total PCBs, 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD), octachlorodibenzo-p-dioxin (OCDD), $p,p'$-DDE, and $\gamma$-chlordane in sediments are presented in Figures 3 to 7. To simplify the presentation of temporal data, two time horizons are represented: the mid-1960s and the mid-1980s to mid-1990s. The later samples were either surface sediment samples with detectable activity of $^7$Be or suspended matter samples (sites 11 and 13). The year of collection at each site is indicated on each figure. These samples are, in general, the most recent samples that have been analyzed at each site. Mid-1960s samples correspond to core sections with peak activities of fallout $^{137}$Cs.

One of the most striking general characteristics of the data is the dramatic decline in levels of most contaminants at most sites between the mid-1960s and more recent samples. The improvement in PCB and chlorinated hydrocarbon levels in sediments along the main stem of the Hudson and in Jamaica Bay and in levels of 2,3,7,8-TCDD in sediments from western NY/NJ harbor have been discussed in detail elsewhere (2–4,21). These trends reflect the success of efforts to limit inputs of contaminants to natural water systems through restrictions on the use of specific compounds, upgrades of municipal sewage treatment facilities, control of

### Table 1. Contaminant levels in sediment samples deposited in the mid-1980s.

| Contaminant          | Newtown Creek, site 9 | NY/NJ harbor, site 9 | Kingston, NY site 5 |
|----------------------|------------------------|----------------------|---------------------|
| **Organic matter**   | 12.8                   | 9.5 (64%)            | 3.6                 |
| $p,p'$-DDE, ppb      | 62                     | 35 (53%)             | 6.2                 |
| $\gamma$-Chlordane, ppb | 28                    | 16 (57%)             | <1                  |
| **Total PCBs, ppm** | 1.91                   | 1.47 (81%)           | 0.79                |
| Copper, ppm          | 604                    | 207 (31%)            | 31                  |
| Lead, ppm            | 486                    | 223 (41%)            | 53                  |
| Zinc, ppm            | 174                    | 358 (35%)            | 145                 |
| Cadmium, ppm         | 10.5                   | 4.03 (31%)           | 1.12                |

*Values in parentheses represent the sewage contribution to contaminant levels in this sample, calculated as [(NY/NJ harbor – Kingston)/(Newtown Creek – Kingston)]×100. Based on weight loss on ignition at 375°C overnight.
point source inputs, and the remediation of Superfund sites.

**Polychlorinated Biphenyls**

Closer inspection of the PCB data (Figure 3) reveals other quite useful information. Extremely low levels in the mid-1960s Ashokan sample (site 6) indicate that atmospheric inputs of PCBs to the mid-Hudson basin are minor. Similar low levels were found in the other sections of the core that were analyzed, covering deposition from the early 1950s to 1981 (the date of core collection).

The extremely high mid-1960s level at site 1 reflects the direct influence of PCB inputs from General Electric capacitor plants located a few kilometers upstream (2). A significant drop in PCB levels occurs not only in the more recent sample from site 1, but also between this site and site 5 (Kingston, NY) on the lower Hudson. The downstream decrease of almost two orders of magnitude can be attributed to dilution of upper Hudson sediments with large amounts of relatively uncontaminated suspended matter delivered by the Batten Kill, the Mohawk River, and the other tributaries entering the Hudson between the two sites (14,22). A similar decrease was noted for cadmium derived primarily from a pigment manufacturing plant located upstream of site 1 (14).

Despite this dilution, detailed component analyses have indicated that between the early 1970s and 1989, roughly two-thirds of the PCB deposited in NY/NJ harbor sediments along the main stem of the Hudson (e.g., at site 8) were derived from inputs to the upper Hudson and subsequent downstream transport. The relative strength of this source has decreased over time, and since the mid-1980s, local NY/NJ metropolitan area sources, including discharge of municipal wastewater, appear to be responsible for the majority of PCB loading to the main-stem harbor sediments (2,14,23).

Downstream transport and inputs with discharges of municipal wastewater (Table 1; Figure 3, sites 9 and 10) have been the main focus of studies of PCB sources to the harbor. However, data for the western NY/NJ harbor show both mid-1960s and more recent PCB levels to be significantly higher than levels observed along the main stem of the Hudson at site 8 and at several other sites discussed elsewhere (14,21). These data indicate that there are major poorly characterized PCB sources located in the western NY/NJ harbor complex and in the Passaic River basin upstream of site 17.

**Dioxins**

Data on levels of 2,3,7,8-TCDD (Figure 4) also draw attention to the western NY/NJ harbor and the major contamination associated with the 80 Lister Avenue Superfund site (site 15), where 2,4,5-trichlorophenate (a well-known source of 2,3,7,8-TCDD contamination) and DDT were among the chemicals manufactured. Sediments in the western harbor, particularly in the lower Passaic River and Newark Bay, are among the world's most contaminated with respect to concentrations of 2,3,7,8-TCDD. Details of contamination of harbor sediments related to the 80 Lister Avenue site are the subject of a previous paper (3).
Relatively low levels of 2,3,7,8-TCDD are associated with wastewater inputs (Figure 4, sites 9 and 10) and atmospheric inputs to the mid-Hudson River Basin were not detectable in our reservoir sediment samples (Figure 4, sites 6 and 7). Also of note are the generally low levels in sediment samples from the upper Hudson basin. This is in sharp contrast to the levels of OCDD (Figure 5), which is associated primarily with combustion sources (24) and sources related to pentachlorophenol (25). The highest levels of OCDD are seen in the mid-1960s samples from the two sites furthest upstream along the main stem of the Hudson. At site 1, levels of over 50 ppb OCDD were found in samples deposited from about 1960 through the mid-1970s. Disposal of coal ash into the river is a plausible source.

High concentrations of OCDD are generally associated with elevated levels of hexa- and heptachlorinated dioxin congeners and with significant toxicity. This can be illustrated by calculating 2,3,7,8-TCDD equivalents for the dioxins found in the samples. Based on North Atlantic Treaty Organization toxic equivalency factors (26), the concentration of dioxins in the mid-1960s sample at sites 1 and 4 were 180 and 2100 ppt of 2,3,7,8-TCDD equivalents, respectively. These levels are comparable to the 2,3,7,8-TCDD equivalent concentrations of dioxins in the highly contaminated sediments of the western NY/NJ harbor. The toxic equivalents of dioxins in those later samples is dominated by 2,3,7,8-TCDD, whereas in the upstream Hudson samples, the 2,3,7,8-substituted hexa-, hepta-, and octachlorinated congeners accounted for over 85% of the toxic equivalents associated with dioxins.

In the reservoir samples, OCDD levels increased by more than an order of magnitude between sites 6 and 7 in both the mid-1960 and 1994 samples. The closer proximity of site 7 to New York City suggests that the atmospheric flux of OCDD may be a strong function of the distance from major combustion sources.

Chlorinated Hydrocarbon Pesticides

Data on \(p,p'-\text{DDD}\) concentrations are presented in Figure 6. The levels of \(p,p'-\text{DDD}\) are again greatest in sediments from the western NY/NJ harbor complex. Contamination in the lower Passaic (site 15) and in Newark Bay (sites 13 and 14) has been related to the history of DDT manufacture at the 80 Lister Avenue Superfund site and an exceptional high freshwater discharge event in the Passaic River in 1984 (3). The highest levels found in mid-1980s samples from sites 11 and 12 suggest the existence of a major local source of DDT near Staten Island. The high concentrations of \(p,p'-\text{DDD}\) at these sites are accompanied by similar levels of the parent compound, \(p,p'-\text{DDT}\). Twelve samples from the core at site 12 had an average ratio of \(p,p'-\text{DDT}\) to \(p,p'-\text{DDD}\) of 0.87, much higher than the average ratio of about 0.2, which was found in sediment and suspended matter samples from other sites in the lower Hudson basin (27). The occurrence of high relative concentrations of \(p,p'-\text{DDT}\) at sites 11 and 12 suggests a relatively fresh nearby source that could be related to improper dumping or disposal of the pesticide.

Levels of \(p,p'-\text{DDD}\) in several samples from the Ashokan Reservoir (site 6) were similar to those found in samples from the main stem of the Hudson near Kingston (site 5), consistent with the major sources to these sites being local application of DDT.
for agriculture and gypsy moth control. γ-Chlordane (Figure 7) was undetectable and less than a few parts per billion in sediment samples from the main stem of the Hudson near Kingston and in several samples from the Ashokan core. Data from sites 9 and 10 indicate that municipal wastewater inputs are an important source of chlordane to sediments of the NY/NJ harbor complex. However, the major source appears to be poorly characterized inputs to the Passaic and Hackensack Rivers and perhaps near Staten Island (sites 11 and 12).

**Conclusions**

Analysis of dated sediment core sections is a useful tool for identifying important sources of contaminants and temporal trends in contaminant levels in natural water systems. When applied over a large geographical area such as the Hudson River Basin, even a relatively limited number of measurements on carefully chosen samples can provide useful insights and perspective. From a broader perspective, based on a National Oceanic and Atmospheric Administration survey of sediments from approximately 200 sites (28), the western NY/NJ harbor is among our nation’s most contaminated coastal and estuarine systems with respect to each of the persistent contaminants discussed here.

Within the last year, sediment cores have been collected that should extend contaminant-level data at most of our sites to the mid-1990s and allow more detailed characterization of atmospheric inputs to the basin. The multicontaminant perspective, which already includes several trace metals, has recently been expanded to include analysis for saturated petroleum hydrocarbons and polycyclic aromatic hydrocarbons.

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