
NOAA Technical Memorandum NMFS-F/NEC-83

# Organic Contaminants in Hepatic Tissues of Lobster and Flounder at the New York Bight "12-Mile" Sewage Sludge Dumpsite: 1987-88

U. S. DEPARTMENT OF COMMERCE
National Oceanic and Atmospheric Administration
National Marine Fisheries Service
Northeast Region
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Woods Hole, Massachusetts

July 1991

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# Organic Contaminants in Hepatic Tissues of Lobster and Flounder at the New York Bight "12-Mile" Sewage Sludge Dumpsite: 1987-88

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**Northeast Fisheries Science Center**
**Woods Hole, Massachusetts**

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## ABSTRACT

Analyses for organic contaminants in the hepatic tissues of winter flounder *(Pleuronectes americanus)* and American lobster *(Homarus americanus)* collected from the area of the "12-Mile" Sewage Sludge Dumpsite in the New York Bight apex at various times during and after the phased cessation of dumping revealed two statistically significant results ($P<0.05$). For each sampling period: (1) concentrations of polychlorinated biphenyls in flounder and lobster taken nearer the dumpsite were higher than in those from a reference area, and (2) lobster values were higher than those for flounder.


## INTRODUCTION

The concentrations of polynuclear aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), along with other anthropogenic contaminants, have been found to be higher in the sediments of the Christiaensen Basin (Figure 1) near waste dumpsites than at surrounding stations in the New York Bight apex (Boehm 1982). There are several routes by which contaminants enter the New York Bight apex (Gross 1976). Sewage sludge from the New York Metropolitan Area that was dumped (Figure 2) at the "12-mile" site is the main focus of this study. (The recent history of sewage sludge dumping is detailed below.) In addition, sediment dredged from the New York Harbor complex (about $10^7$ $m^3$ $yr^{-1}$; Mueller *et al.* 1976) is dumped at the dredge spoils site (Figure 1). Particularly relevant to the present study are PCBs that have accumulated in the sediments of the Hudson River and the harbor mainly as a result of discharge from the General Electric plants at Ft. Edward and Hudson Falls (Brown *et al.* 1985). In general, concentrations of PCBs in dredged material from the harbor complex range from nil to 10 mg/kg (Conner *et al.* 1979). Moreover, estuarine outflow probably carries PCBs on suspended sediment from the Hudson-Raritan Estuary, along with other contaminants and nutrients (Mueller *et al.* 1976). Finally, minor amounts of contaminants also enter the New York Bight by atmospheric deposition (*e.g.*, Gibson *et al.* 1979; Stanford and Young 1988).

After over 60 years of dumping of sewage sludge at the "12-mile" site (Figures 1 and 2), a phased cessation of dumping began in March 1986 when a site more than 100 miles from New York was brought into use. From June 1986 to July 1987, dumping at the "12-mile" dumpsite was conducted at a level of approximately 70-percent of the precessation rate, and all dumping was phased out by December 31, 1987. Termination of dumping provided a unique opportunity to conduct a very-large-scale, environmental perturbation experiment on the relation between sediment biogeochemistry, benthic community structure, and finfish distributions. The Northeast Fisheries Science Center (NEFSC) instituted a program (Environmental Processes Division, Northeast Fisheries Center 1988) to document environmental and biological conditions in and around this dumpsite as related to the cessation of sewage sludge dumping. This paper reports data on organic contaminant levels in livers of winter flounder *(Pleuronectes americanus)* and the hepatopancreas tissues of American lobster *(Homarus americanus)* collected on three cruises between June 1987 and November 1988.

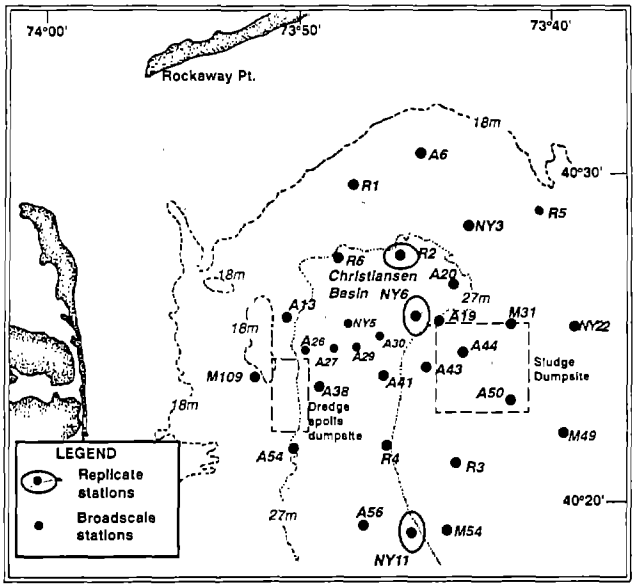


Figure 1. Location of stations sampled regularly during the "12-mile" dumpsite response study. (Stations in small type were sampled less frequently to monitor any contamination from proposed increases in dredged-material dumping.)

## METHODS

### SAMPLING DESIGN

Animals were collected by otter trawl from the NEFSC's research vessel *Kyma* over the course of two weeks during the regular NEFSC surveys of the dumpsite area or in the week thereafter. Specimens were measured and weighed (except when sea conditions were too rough), and the sex determined. Dissections were made under HEPA (high-efficiency particulate air)-filtered air using dichloromethane-rinsed implements. The tissues were placed in dichloromethane-cleaned jars with Teflon-lined screw caps (from I-Chem Research, Hayward, California) and frozen at -20°C.

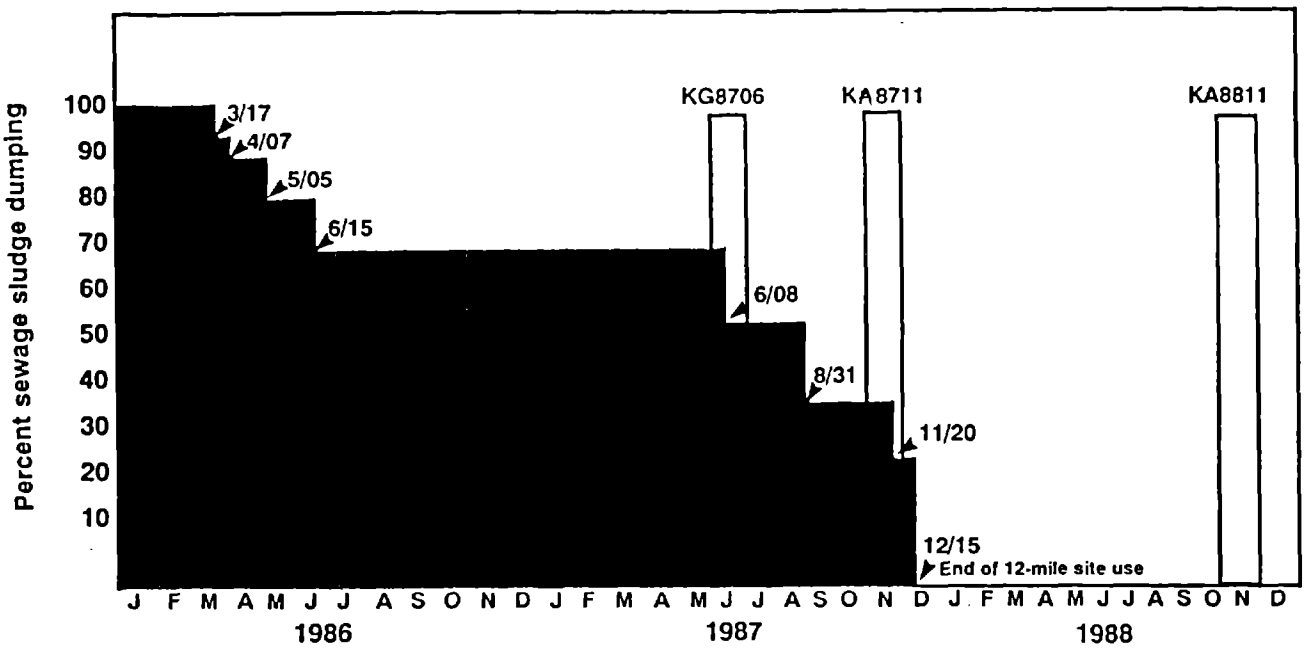


Figure 2. Negotiated phaseout schedule for sewage sludge dumping at the "12-mile" dumpsite, including winter flounder and American lobster sampling periods. (Data modified from E. Santoro, U.S. Environmental Protection Agency, 26 Federal Plaza, New York, NY 10278.)

The original sampling design (Table 1) called for the analysis of a total of 20 composites of 20 g each. This weight of tissue would require the liver from about five flounder or the hepatopancreas from about three lobsters. There were to be 10 composites at each of two stations (NY6, contaminated; NY11, reference; see Figure 1). Within stations, composites were to be allocated to consider spawning condition (pre- and post-).

The number of composites to be analyzed was reduced to 18 due to limited funds and difficulties in obtaining animals as specified in the original sampling design (Table 1). For example, lobsters were not found at station NY6 in June 1987, and flounder were not found at NY11 in November 1988. In addition, the numbers of lobsters that could be collected at stations NY6 and NY11 in November 1987 were only six and five, respectively. We therefore based the final scheme of analysis (Tables 2 and 3) on the following criteria:

1. All samples from station NY6 should be analyzed, on the grounds that if contamination were occurring from sewage sludge, it would probably be most evident at the station closest to the dumpsite.

2. Time series at stations that include pre- and post-cessation periods should be given priority, and interstation comparisons should be secondary.

3. For a species, equal numbers of animals should be included in each composite.

In the case of the November 1987 composite at station NY11, criterion No. 3 was violated, but we considered it preferable to uphold criterion No. 2.

## ANALYTICAL METHODS

### Analyte Extraction

A procedure based on a combination of three methods (Environmental Protection Agency 1982, 1985, 1986) was employed for the sequential analysis of PAHs, PCBs, 2,3,7,8-tetrachlorodibenzodioxin (TCDD), and 2,3,7,8-tetrachlorodibenzofuran (TCDF). Approximately 10 g (wet weight) of the tissue homogenized with dry ice was spiked with surrogates and internal standards, and then saponified in 40-percent aqueous potassium hydroxide in a reflux apparatus for 60 min. The solution was allowed to stand overnight to ensure complete saponification. The solution was then diluted with 50-percent aqueous ethanol and extracted with one 25-ml portion and three 15-ml portions of hexane. The combined hexane extract was washed with 10 ml of water to remove the excess base, and then evaporated to a final volume of 1 ml.

### PAH and PCB Analyses

To remove lipid-specific and other matrix interferences, the hexane extract was eluted through a column

Table 1. Number of composites to be analyzed for organic contaminants, according to the original sampling design, on a station, species, and spawning condition basis

| Station | Winter Flounder | | American Lobster | |
|---|---|---|---|---|
| | Pre-spawn | Post-spawn | Pre-spawn | Post-spawn |
| NY6 | 3 | 2 | 2 | 3 |
| NY11 | 3 | 2 | 2 | 3 |

Table 2. Number of composites actually analyzed for organic contaminants on a species, sampling period, and station basis

| Species | June 1987 | | November 1987 | | November 1988 | |
|---|---|---|---|---|---|---|
| | NY6 | NY11 | NY6 | NY11 | NY6 | NY11 |
| Winter flounder | 2 | 2 | 2 | - | 2 | - |
| American lobster | - | 2 | 2 | 2 | 2 | 2 |

containing 20 g of silica gel with 300 ml of hexane. The eluant was concentrated to a final volume of 100 µl, and sequentially analyzed by gas chromatography - mass spectrometry (GC-MS) for PAHs and PCBs. A quality control (QC) matrix spike was prepared by adding known amounts (1000 ng) of PAH and PCB analytes to lobster composite No. 1 from June 1987. The recovery for PCB mixture Aroclor 1260 was 76 percent, while recoveries for PAHs ranged from 0 to 28 percent. In addition, QC blanks showed contamination with naphthalene, 1-methyl naphthalene, and 2-methyl naphthalene.

## TCDD and TCDF Analyses

The extract remaining after the PAH and PCB analyses was subjected to further purification as follows:

1. The analyte extract was diluted to 100 ml with hexane and then washed with four 10-ml portions of concentrated sulfuric acid and finally with 10 ml of water. The hexane layer was concentrated to 1 ml.

2. This extract was loaded on a column packed (from the bottom) with 1 g of neutral silica gel, 2 g of 33-percent NaOH-silica gel, 1 g of neutral silica gel, 20 g of 44-percent sulfuric acid-silica gel, and 2 g of neutral silica gel. The column was eluted with 150 ml of hexane, and the eluant evaporated to near dryness.

3. A column packed with 1.5 g of 10-percent silver nitrate-silica was loaded with the hexane extract from step No. 2. It was eluted with 30 ml of hexane, and the eluant was passed through a second column packed with 5 g of basic alumina. The latter was washed with 50 ml of 50-percent carbon tetrachloride-hexane. The chlorinated dioxins and chlorinated dibenzofurans were eluted from it with 60 ml of 50-percent dichloromethane-hexane, and the eluant was evaporated to about 0.5 ml.

4. A pasteur pipette packed with 18-percent Carbopack C on Celite 545 (1:4.5 mixture of Carbopack C, 80-100 mesh, and Celite 545, activated at 130°C for 6 hr) was prewashed with 2 ml of toluene, 1 ml of 75:20:5 dichloromethane-methanol-benzene, 1 ml of 1:1 cyclohexane-dichloromethane, and 2 ml of hexane. The column was loaded with the hexane extract obtained from step No. 3. It was then washed with two 1-ml portions of hexane, 1 ml of 1:1 cyclohexane-dichloromethane, and 1 ml of 75:20:5 dichloromethane-methanol-benzene. The TCDD-TCDF fraction was collected by elution with 2 ml of toluene and analyzed by GC-MS.

### *GC-MS Analyses*

The analytes were separated on a DB-5, fused-silica, open-tubular capillary column (from J & W Scientific, Folsom, California; 60-m x 0.25-mm I.D. x 0.25-µm film thickness) by using a Hewlett-Packard 5890 gas chromatograph. Helium was used as a carrier gas at a linear velocity of 24 cm/sec. The column oven temperature was programmed from 150 to 340°C at a rate of 4°C/min. The samples were introduced in a splitless injection mode. The injector and the interface temperatures were each set at 250°C. The PAHs were analyzed by electron impact mass spectrometry in the scan mode (total ion chromatogram), while the PCBs (Table 4), 2,3,7,8-TCDD (m/z = 259, 320, 322), and 2,3,7,8-TCDF (m/z = 243, 304, 306) were ana-

Table 3. Characteristics of specimens [*i.e.*, animal no., length (mm), weight (g), and sex (M/F)] used for organic contaminant analysis on a species, station, cruise/sampling period, and composite basis

| Species | Station | Cruise No./ Sampling Period | Composite No. | Species Characteristics | | | |
|---|---|---|---|---|---|---|---|
| | | | | Animal (no.) | Length (mm) | Weight (g) | Sex (M/F) |
| Winter flounder | NY6 | KG 8706/ Jun. 1987 | 1 | 2 | 300 | - | F |
| | | | | 3 | 335 | - | F |
| | | | | 4 | 335 | - | F |
| | | | | 8 | 222 | 153 | F |
| | | | | 11 | 235 | 177 | F |
| | | | 2 | 1 | 257 | - | M |
| | | | | 6 | 233 | - | M |
| | | | | 7 | 220 | 133 | M |
| | | | | 9 | 340 | 594 | F |
| | | | | 10 | 266 | 228 | F |
| | | KA 8711/ Nov. 1987 | 1 | 1 | 252 | 205 | F |
| | | | | 3 | 222 | 125 | F |
| | | | | 9 | 344 | 419 | F |
| | | | | 11 | 266 | 241 | M |
| | | | | 13 | 273 | 255 | F |
| | | | 2 | 5 | 220 | 130 | F |
| | | | | 6 | 234 | 160 | F |
| | | | | 10 | 334 | 430 | M |
| | | | | 12 | 306 | 410 | F |
| | | | | 16 | 245 | 173 | F |
| | | KA 8811/ Nov. 1988 | 1 | 3 | 252 | 180 | F |
| | | | | 4 | 260 | 215 | F |
| | | | | 10 | 245 | 196 | F |
| | | | | 21 | 206 | 105 | M |
| | | | | 26 | 288 | 315 | M |
| | | | 2 | 5 | 296 | 318 | F |
| | | | | 7 | 278 | 270 | F |
| | | | | 8 | 253 | 225 | M |
| | | | | 22 | 275 | 290 | F |
| | | | | 28 | 277 | 280 | F |
| | NY11 | KG 8706/ Jun. 1987 | 1 | 1 | 300 | - | F |
| | | | | 2 | 298 | - | F |
| | | | | 12 | 210 | - | F |
| | | | | 18 | 287 | - | F |
| | | | | 19 | 288 | - | F |
| | | | 2 | 6 | 225 | - | F |
| | | | | 7 | 203 | - | M |
| | | | | 10 | 307 | - | F |
| | | | | 17 | 315 | - | M |
| | | | | 20 | 255 | - | M |
| American lobster | NY6 | KA 8711/ Nov. 1987 | 1 | 1 | 65 | 180 | M |
| | | | | 3 | 74 | 220 | M |
| | | | | 4 | 46 | 95 | M |
| | | | 2 | 2 | 61 | 180 | M |
| | | | | 5 | 62 | 205 | M |
| | | | | 6 | 58 | 155 | F |
| | | KA 8811/ Nov. 1988 | 1 | 1 | 83 | 400 | F |
| | | | | 26 | 66 | 198 | F |
| | | | | 27 | 67 | 245 | F |
| | | | 2 | 4 | 69 | 263 | F |
| | | | | 13 | 75 | 275 | F |
| | | | | 17 | 79 | 300 | M |

Table 3. Continued.

| Species | Station | Cruise No./ Sampling Period | Composite No. | Species Characteristics | | | |
|---|---|---|---|---|---|---|---|
| | | | | Animal (no.) | Length (mm) | Weight (g) | Sex (M/F) |
| American lobster | NY11 | KG 8706/ Jun. 1987 | 1 | 2 | 68 | - | M |
| | | | | 4 | 72 | - | F |
| | | | | 9 | 81 | - | F |
| | | | 2 | 6 | 70 | - | F |
| | | | | 8 | 71 | - | M |
| | | | | 20 | 70 | - | F |
| | | KA 8711/ Nov. 1987 | 1 | 1 | 90 | 250 | M |
| | | | | 2 | 100 | 335 | F |
| | | | | 4 | 70 | 301 | M |
| | | | 2 | 3 | 79 | 295 | M |
| | | | | 5 | 73 | 288 | F |
| | | KA 8811/ Nov. 1988 | 1 | 10 | 81 | 370 | M |
| | | | | 11 | 59 | 170 | M |
| | | | | 25 | 61 | 114 | F |
| | | | 2 | 6 | 70 | 260 | F |
| | | | | 12 | 77 | 350 | F |
| | | | | 30 | 68 | 275 | M |

Table 4. SIM ions used in the GC-MS analyses of PCBs

| Group No.: | 1 | | 2 | | 3 | | 4 | | 5 | |
|---|---|---|---|---|---|---|---|---|---|---|
| Start-stop (min): | 5.00-15.94 | | 15.94-21.14 | | 21.14-24.35 | | 24.35-31.41 | | 31.41-38.00 | |
| | m/z | Dwell | m/z | Dwell | m/z | Dwell | m/z | Dwell | m/z | Dwell |
| | 152 | 50 | 186 | 50 | 254 | 50 | 240 | 50 | 356 | 50 |
| | 153 | 50 | 188 | 50 | 256 | 50 | 241 | 50 | 358 | 50 |
| | 186 | 50 | 220 | 50 | 288 | 50 | 288 | 50 | 360 | 50 |
| | 187 | 50 | 222 | 50 | 290 | 50 | 290 | 50 | 390 | 50 |
| | 188 | 50 | 254 | 50 | 310 | 50 | 322 | 50 | 392 | 50 |
| | 189 | 50 | 255 | 50 | 312 | 50 | 324 | 50 | 394 | 50 |
| | 190 | 50 | 256 | 50 | 322 | 50 | 326 | 50 | 424 | 50 |
| | 220 | 50 | 258 | 50 | 323 | 50 | 356 | 50 | 425 | 50 |
| | 221 | 50 | 288 | 50 | 324 | 50 | 357 | 50 | 426 | 50 |
| | 222 | 50 | 290 | 50 | 326 | 50 | 358 | 50 | 428 | 50 |
| | 224 | 50 | 292 | 50 | 328 | 50 | 360 | 50 | 430 | 50 |
| | 255 | 50 | 294 | 50 | 357 | 50 | 362 | 50 | 432 | 50 |
| | 256 | 50 | 310 | 50 | 358 | 50 | 391 | 50 | 462 | 50 |
| | 258 | 50 | 312 | 50 | 360 | 50 | 392 | 50 | 464 | 50 |
| | 290 | 50 | 324 | 50 | 362 | 50 | 394 | 50 | 466 | 50 |
| | 292 | 50 | 326 | 50 | 392 | 50 | 396 | 50 | 496 | 50 |
| | 294 | 50 | 328 | 50 | 394 | 50 | 398 | 50 | 498 | 50 |
| | 310 | 50 | 358 | 50 | 396 | 50 | 428 | 50 | 499 | 50 |
| | 312 | 50 | 360 | 50 | 398 | 50 | 430 | 50 | 500 | 50 |
| | | | 362 | 50 | | | 432 | 50 | 502 | 50 |

lyzed by the electron impact mass spectrometry in the select ion monitoring (SIM) mode by using a Hewlett-Packard 5996 mass selective detector with a direct capillary interface. The relative electron multiplier voltage was 2500, and the emission current was 300 μA. The analytical detection limit is defined as five times the variation of the baseline, which is functionally equivalent to three times the mean standard deviation. The limit of detection for each analyte in each sample takes into account differences in the weight of sample analyzed.

Table 5. Concentrations (average of two composites) of organic contaminants (µg/kg, wet weight) and lipid content (percent) on a species, cruise, and station basis

| Species | Analyte | KG 8706 | | KA 8711 | | KA 8811 | |
|---|---|---|---|---|---|---|---|
| | | NY6 | NY11 | NY6 | NY11 | NY6 | NY11 |
| Winter flounder | Lipid | 23.0 | 34.0 | 18.0 | | 22.6 | |
| | Dichlorobiphenyl | 2.5[a] | ND[b] | ND | | 0.6[a] | |
| | Trichlorobiphenyl | 42.2 | 18.8 | 13.6 | | 24.1 | |
| | Tetrachlorobiphenyl | 194.5 | 95.1 | 58.4 | | 113.6 | |
| | Pentachlorobiphenyl | 410.5 | 253.0 | 199.9 | | 267.5 | |
| | Hexachlorobiphenyl | 199.0 | 154.0 | 111.3 | | 144.0 | |
| | Heptachlorobiphenyl | 70.4 | 66.0 | 40.6 | | 52.7 | |
| | Octachlorobiphenyl | 15.5 | 14.1 | 9.2 | | 12.0 | |
| | Nonachlorobiphenyl | 2.2 | 6.0 | 1.9[a] | | 2.0 | |
| | Decachlorobiphenyl | ND | 3.33[a] | ND | | ND | |
| | Total PCBs | 936.7 | 610.1 | 435.0 | | 616.3 | |
| | Fluoranthene | ND | ND | ND | | ND | |
| | Pyrene | ND | ND | ND | | ND | |
| American lobster | Lipid | | 49.9 | 24.1 | 31.2 | 35.7 | 34.8 |
| | Dichlorobiphenyl | | 17.7 | 21.9 | 6.9 | 4.4 | 1.7 |
| | Trichlorobiphenyl | | 56.2 | 56.5 | 47.7 | 24.1 | 23.4 |
| | Tetrachlorobiphenyl | | 247.5 | 327.0 | 256.5 | 130.7 | 118.2 |
| | Pentachlorobiphenyl | | 549.0 | 721.5 | 624.5 | 340.5 | 284.0 |
| | Hexachlorobiphenyl | | 340.5 | 467.0 | 391.0 | 218.5 | 175.1 |
| | Heptachlorobiphenyl | | 161.0 | 286.5 | 154.5 | 103.1 | 69.1 |
| | Octachlorobiphenyl | | 39.0 | 37.7 | 39.2 | 26.7 | 19.2 |
| | Nonachlorobiphenyl | | 12.9 | 22.4 | 12.0 | 5.7 | 3.8 |
| | Decachlorobiphenyl | | 6.9[a] | 12.4 | 13.7 | 5.1[a] | 5.1[a] |
| | Total PCBs | | 1430.6 | 1952.8 | 1546.0 | 858.7 | 699.5 |
| | Fluoranthene | | ND | 17.6 | ND | 1.7[a] | ND |
| | Pyrene | | 50.9 | 77.8 | 51.0 | 16.8 | ND |

[a] Consists of one value above the limit of detection and one measurement below which was taken as zero.
[b] ND = not detected at the analytical and sample detection limit.

Table 6. Concentrations (average of two composites) of selected organic contaminants (µg/kg, wet weight), normalized by percent lipid content, on a species, cruise, and station basis

| Species | Analyte | KG 8706 | | KA 8711 | | KA 8811 | |
|---|---|---|---|---|---|---|---|
| | | NY6 | NY11 | NY6 | NY11 | NY6 | NY11 |
| Winter flounder | Tetrachlorobiphenyl/lipid | 8.5 | 3.3 | 3.0 | | 4.9 | |
| | Pentachlorobiphenyl/lipid | 18.0 | 8.7 | 10.0 | | 11.8 | |
| | Hexachlorobiphenyl/lipid | 9.1 | 5.3 | 5.6 | | 6.5 | |
| | Fluoranthene/lipid | ND[a] | ND | ND | | ND | |
| | Pyrene/lipid | ND | ND | ND | | ND | |
| American lobster | Tetrachlorobiphenyl/lipid | | 6.1 | 16.0 | 8.3 | 3.9 | 3.6 |
| | Pentachlorobiphenyl/lipid | | 13.3 | 34.1 | 20.1 | 10.2 | 8.6 |
| | Hexachlorobiphenyl/lipid | | 8.3 | 21.1 | 12.6 | 6.3 | 5.3 |
| | Fluoranthene/lipid | | ND | 1.4[b] | ND | 0.04[b] | ND |
| | Pyrene/lipid | | 1.4 | 4.4 | 1.6 | 0.5 | ND |

[a] ND = not detected at the analytical and sample detection limit.
[b] Consists of one value above the limit of detection and one measurement below which was taken as zero.

Table 7. Detection limits (µg/kg, wet weight) of 2,3,7,8-TCDD and 2,3,7,8-TCDF on a species, composite, cruise, and station basis

| Species | Analyte | Composite No. | KG 8706 | | KA 8711 | | KA 8811 | |
|---|---|---|---|---|---|---|---|---|
| | | | NY6 | NY11 | NY6 | NY11 | NY6 | NY11 |
| Winter flounder | TCDD | 1 | <0.63 | -[a] | <2.10 | | <0.19 | |
| | | 2 | <0.93 | <1.50 | <1.80 | | <0.48 | |
| | TCDF | 1 | <0.82 | <3.30 | <0.98 | | <0.52 | |
| | | 2 | <0.73 | <0.81 | <0.85 | | <0.52 | |
| American lobster | TCDD | 1 | | <1.20 | <0.65 | <0.57 | -[a] | <1.10 |
| | | 2 | | <0.60 | <0.61 | <0.88 | <1.00 | <2.30 |
| | TCDF | 1 | | <0.82 | <0.93 | <0.59 | <8.50 | <0.78 |
| | | 2 | | <0.75 | <0.84 | <0.88 | <1.10 | <1.80 |

[a] Detection limit could not be calculated.

## RESULTS

The concentrations of analytes are summarized in Tables 5 and 6 as the average of each pair of composites. The following trends can be discerned:

1. For any sampling period, nearly all PCB values were higher in both flounder and lobster from station NY6 than those from station NY11. In a few instances, for a few compounds present in concentrations near the limit of detection, this was not the case. For example, for nonachlorinated biphenyls in winter flounder collected in June 1987, the station NY6 concentration was determined as 2 µg/kg, while at station NY11 it was 6 µg/kg. Analytical and compositing variances probably make these differences insignificant on a parametric basis.

2. For a given sampling period, all PCB concentrations in lobster were higher than in flounder (with only one exception in which they were equal).

3. Concentrations of PCBs in lobsters were approximately equal during both sampling periods in 1987 (June and November) and lowest in November 1988, 11 months after dumping ceased at the "12-mile" site.

4. Concentrations of PCBs in flounder were highest during the first sampling in spring 1987, and, in contrast to lobsters, lowest in November 1987.

5. When contaminant concentrations are normalized to lipid content (Table 6), the above results are basically unchanged. However, the time-course decrease in lobster PCB content (result No. 3 above) appears to be more pronounced.

Wilcoxon's signed-rank test for comparing two populations using paired data indicates that results No. 1 and No. 2 above are statistically significant ($P<0.05$).

Since recoveries for PAHs were low and contamination with certain compounds occurred in blanks, results for PAHs must be considered tentative. In addition, most PAH concentrations in lobster tissue and all PAH concentrations in flounder livers were below the limit of detection of the method. Those PAH values above the detection limit (fluoranthene and pyrene) are included in Tables 5 and 6. They follow the patterns of the PCB levels.

In no sample was the dioxin or dibenzofuran concentration above the detection limit. For TCDD, detection limit values ranged from <0.19 to <2.3 µg/kg; while for TCDF, values ranged from <0.52 to <8.5 µg/kg (Table 7).

## DISCUSSION

We found that contaminants in tissue were higher in animals collected nearer the dumpsite than in animals from the reference station farther from the dumpsite. This statistically significant difference in contaminant burdens between the stations may reflect a rapid response to environmental exposure. On the other hand, the result may be complicated by our having sampled groups of animals with different contaminant exposure histories due to differences in migration at the two sites.

The migration of animals is expected to be one of the largest sources of variation in the concentrations of contaminants in animal tissue from a given site. Since pollution gradients in the New York Bight apex exist on the scale of kilometers, a relatively uncontaminated animal could be captured shortly after entering a contaminated site, or a contaminated animal could enter a relatively unpolluted area and be captured. On a larger scale, lobsters migrate into the New York Bight apex from offshore in the summer and leave for the offshore areas when water temperature drops in winter. This picture is compounded further by the existence of a "coastal" population that is resident in the area (Uzmann *et al.* 1977). Similarly, the seasonal migration patterns of winter flounder should affect the contaminant burdens ob-

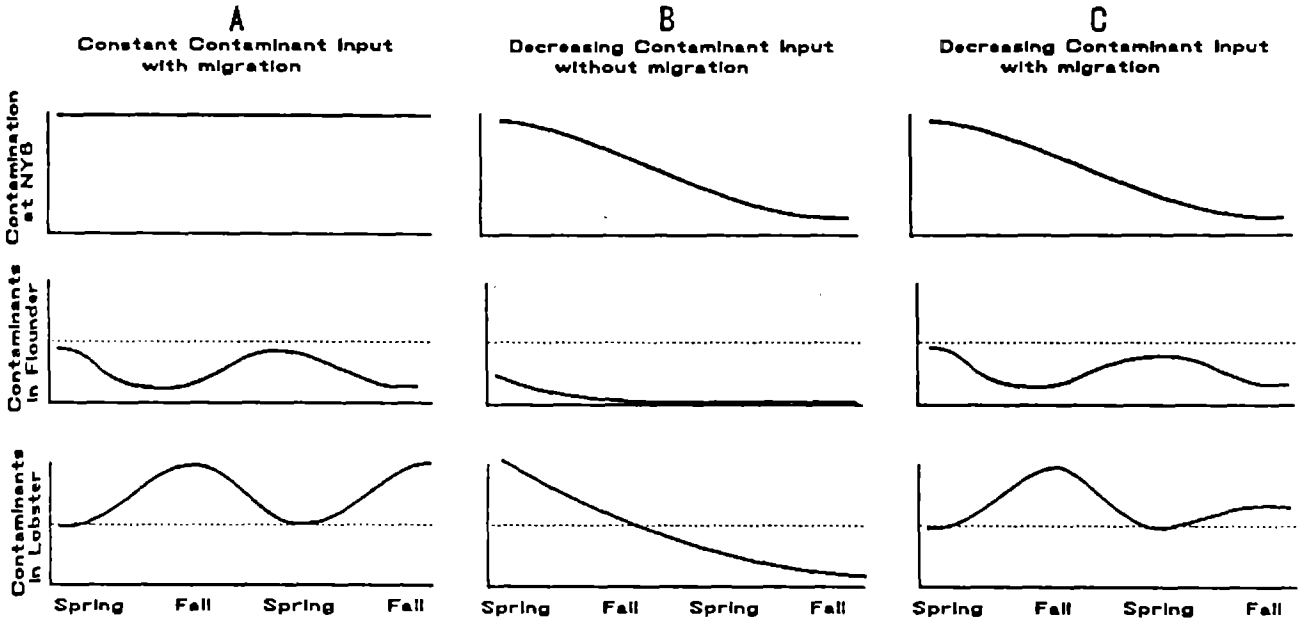


Figure 3. Hypothetical contaminant loadings and resulting contaminant burdens in winter flounder and American lobster in the New York Bight apex under different migration assumptions. (See text for detail.)

served. Winter flounder migrations have been followed in this study (Valdes 1988). The fish leave the estuary for the New York Bight apex in spring and return to the estuary in winter. Thus, lobsters arriving in the apex are likely to have relatively low contaminant burdens and be entering a more contaminated environment, while the opposite is likely the case for flounder.

We have constructed a hypothetical model based on these considerations (Figure 3), assuming that contaminant body burdens are functions of: (1) length of exposure time, (2) level of environmental contamination, and (3) species-specific elimination rate. In Figure 3A, we show the hypothetical body burdens for the case where environmental contamination in the New York Bight apex is constant. Variation in body burdens over time would then fluctuate as a result of the relative exposure intensity which is assumed to be highest in the estuary for the winter flounder and highest in the apex for the lobster.

Our objective in this study was to document environmental and biological conditions in and around the "12-mile" dumpsite related to the cessation of sewage sludge dumping. Therefore, we also sampled across another gradient: the presumed temporal decrease in environmental contamination following cessation of dumping. To help understand how this gradient may be expected to affect contaminant burdens in flounder and lobsters in the New York Bight apex, two more conditions are included in our model. In the middle panel (Figure 3B), we show the expected effect on contaminant burdens of decreasing environmental contamination if the animal were not free to migrate (for example if they were confined in cages). In the final panel (Figure 3C), we combine the expected effects of migration and decreasing environmental contamination. Under conditions of decreasing environmental exposure, the contamination in lobster would be expected to decrease, but since the flounder are still exposed to high concentrations in the estuary, decreases in their contaminant burdens would be expected to be minimal.

In light of the expected trends shown in Figure 3, we note several trends in the data of this study that, while not statistically significant, are interesting. The lobster data (Table 5) for most contaminants at station NY11 are consistent with both the pattern expected as a consequence of migration and that from pollution abatement. Lower values of contaminant concentration at the time of the first sampling in spring 1987 may be due to the shoreward migration of offshore animals with low body burdens. Low values at the end of the period covered by this paper may be due to the cessation of dumping.

For winter flounder, the highest values for most PCBs (Table 5) were found in the June 1987 samples, as might be expected because of one or both of two compounding reasons. Contaminant loading at the "12-mile" site was at the highest level (of the three sampling periods; see Figure 2) in June 1987, and animals migrating out of the estuary in spring likely had elevated body burdens. However, in contrast to the observations on lobsters, the lowest concentrations were not found in the samples collected the longest time after cessation (November 1988), but in November 1987 instead. It is possible that the proximity of station NY6 to the Hudson-Raritan Estuary, rather than its closeness to the "12-mile" dumpsite, may contribute to the observation that major organic contaminants are consistently higher in animals from station NY6 than station NY11. Future analyses of specimens (including animals from station R2; see Figure 1) will help to resolve this question by increasing the temporal and spatial distribution of observations.

There was a significant interspecific difference in PCB content which we assumed for the construction of the model might be due to species-specific differential net elimination rates. Similarly, in the few data for PAH concentrations (Table 5), values in flounder were all below the limit of

Table 8. Concentrations of total PCBs (μg/kg, wet weight) on a species, composite, cruise, and station basis

| Species | Composite No. | KG 8706 | | KA 8711 | | KA 8811 | |
|---|---|---|---|---|---|---|---|
| | | NY6 | NY11 | NY6 | NY11 | NY6 | NY11 |
| Winter flounder | 1 | 622 | 329 | 159 | | 760 | |
| | 2 | 1251 | 891 | 711 | | 473 | |
| American lobster | 1 | | 1015 | 1407 | 1491 | 646 | 1049 |
| | 2 | | 1846 | 2499 | 1601 | 1071 | 350 |

detection, while in 11 of 20 lobster composites, PAHs were detectable. This result is consistent with a biochemical model which holds that PAHs are not found in finfish, but are found in decapod crustaceans because of the less efficient hepatic metabolic system of the latter (*e.g.*, Stegeman 1981).

Guidelines for TCDD concentrations as set by the Food and Drug Administration (FDA) are 25 pg/kg for "limited" consumption, and 50 pg/kg for no consumption (Miller 1983). Since the limit of detection in this study was so high (190-2300 pg/kg; Table 7), and the values for all samples were below the limits of detection, we cannot determine from these data whether the fish and lobsters in the study contained concentrations of TCDD above the guidelines. However, the FDA action level for total PCBs in the edible portion of fish and shellfish of 2 μg/g (Food and Drug Administration 1984) is applicable to our results. The highest concentration of total PCBs we found was 2.5 μg/g in lobster composite No. 2 from station NY6 in November 1987 (Table 8). This value is about equal to the lowest mean values reported from Long Island Sound for the average of 20 lobsters (Chytalo 1989), and is well below the highest mean concentration she reports of 7.08 μg/g. However, composite No. 1 from the same sampling period and station contained only 1.4 μg/g, resulting in an average of 1.95 μg/g (Table 5) and suggesting the large variation among animals in our study. It appears, then, that the hepatopancreases of some of the lobsters in the area of the "12-mile" sewage sludge dumpsite were contaminated to a concentration above the FDA action level.

## SUMMARY

## CONCLUSIONS

1. For each of the three sampling periods, PCB values in both flounder and lobster were higher at the station nearer the dumpsite (NY6) than at the station relatively farther from the dumpsite (NY11).

2. For each of the three sampling periods, PCB concentrations in lobster were higher than in flounder.

## OBSERVATIONS REQUIRING ADDITIONAL ANALYSES AND EVALUATION

1. Concentrations of organic contaminants in lobsters were lowest in November 1988, perhaps associated with the cessation of dumping.

2. Concentrations of organic contaminants in flounder were highest in spring, perhaps associated with migrations to the sampling area in the New York Bight apex from the Hudson-Raritan Estuary.

3. Since detection limits were very high for 2,3,7,8-TCDD (range of <0.19 - <2.3 μg/kg) and 2,3,7,8-TCDF (range of <0.52 - <8.5 μg/kg), more tissue or a more sensitive analytical technique is required to discern spatial and temporal changes in these contaminants.

4. Extraction and cleanup procedures for PAH trace analysis from marine tissues need to be optimized to measure compounds that were below the detection limit in these samples.

5. PAH concentrations are consistent with a biochemical model which holds that PAHs are not found in finfish, but are found in decapod crustaceans because of the less efficient hepatic metabolic system of the latter (*e.g.*, Stegeman 1981).

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