

TESTS OF BIOACCUMULATION MODELS FOR PCBs: A STUDY OF YOUNG-  
OF-THE-YEAR BLUEFISH IN THE HUDSON RIVER ESTUARY

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by

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## ABSTRACT

A field-based study of the uptake of polychlorinated biphenyl compounds (PCBs) in young-of-the-year (YOY) bluefish (*Pomatomus saltatrix*) was initiated in the summer of 1993. Samples of YOY bluefish and their prey items, were taken several times over the summer and fall, starting upon the arrival of the bluefish into the Hudson River estuary, in late June. Whole body homogenates of two or three individuals were extracted with organic solvent and analyzed for PCBs, on a congener specific basis, by capillary gas chromatography with electron capture detection. Results showed that YOY bluefish began accumulating PCBs rapidly upon entry in the Hudson. Differences between congeners were seen in the pattern of accumulation over time, with the heavier molecular weight, more highly chlorinated congeners being bioaccumulated to a greater extent and responding more slowly to an increase in body mass. Differences in the patterns of accumulation of PCB congeners were also seen between bluefish and striped bass (an important prey species) with striped bass accumulating PCBs to a much lesser extent, and showing a dilution effect of increased body size not seen in the bluefish. Log plots of a calculated bioaccumulation factor (BAF) versus the log of the octanol-water partition coefficient revealed bluefish to be above equilibrium with dissolved water concentrations, implying that other modes of uptake besides partitioning into lipids from the water column (such as uptake from food) are important. This effect is not as dramatic as seen for other adult species of fish and may be due to the rapid growth rate of the YOY bluefish. Comparison of BAF values with values predicted by food chain models showed good agreement (to a first order approximation). Predicted bioconcentration values (BCFs) appeared to be sensitive to different estimates of uptake and excretion rates.

## TABLE OF CONTENTS

ABSTRACT .....	VII-3
I INTRODUCTION .....	VII-9
A. Partitioning Models .....	VII-9
B. The Thomann Food Chain Model .....	VII-11
II METHODS .....	VII-13
A. Field Collection .....	VII-13
B. Sample Preparation .....	VII-13
C. Instrumental Analysis .....	VII-16
D. Model Parameters .....	VII-17
III RESULTS .....	VII-19
A. Trends of Accumulation Over Time .....	VII-19
B. Equilibrium Partitioning .....	VII-27
C. Thomann Model .....	VII-27
IV. DISCUSSION .....	VII-32
V. CONCLUSION .....	VII-37
VI. RECOMMENDATIONS FOR FUTURE WORK .....	VII-38
VII. ACKNOWLEDGMENTS .....	VII-38
VIII. REFERENCES .....	VII-39

## LIST OF FIGURES

Figure 1.	Location of sampling sites along the Hudson River .....	VII-15
Figure 2.	Accumulation patterns of selected biphenyl congeners in YOY bluefish over time (wet wt. basis) .....	VII-23
Figure 3.	Accumulation patterns of selected biphenyl congeners in YOY bluefish over time (lipid wt. basis) .....	VII-24
Figure 4.	Accumulation patterns of selected biphenyl congeners in YOY bluefish versus mean wet weight .....	VII-25
Figure 5.	Accumulation patterns of selected biphenyl congeners in YOY striped bass over time .....	VII-26
Figure 6.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol water partition coefficient (log $K_{ow}$ ) for several PCB congeners. a) June 30 sample; b) July 28 sample; c) August 4 sample .....	VII-28
Figure 7.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol water partition coefficient (log $K_{ow}$ ) for several PCB congeners. a) August 11 sample; b) August 24 sample; c) September 11 sample .....	VII-29
Figure 8.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol-water partition coefficient (log $K_{ow}$ ) for several PCB congeners. Superimposed are predictions based on the on the Thomann model. a.) June 30 sample; b.) August 4 sample; c.) September 11 sample .....	VII-30
Figure 9.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol-water partition coefficient (log $K_{ow}$ ) for several PCB congeners for the August 4 sample. a) parameters $ku_1$ , $K_1$ and $\alpha_1$ ; b) parameters $ku_2$ , $K_2$ and $\alpha_2$ ; c) parameters $ku_3$ , $K_3$ and $\alpha_3$ .....	VII-33
Figure 10.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol-water partition coefficient (log $K_{ow}$ ) for several PCB congeners for the August 4 sample. a) parameters $ku_1$ , $K_1$ and $\alpha_1$ ; b) parameters $ku_2$ , $K_2$ and $\alpha_2$ ; c) parameters $ku_3$ , $K_3$ and $\alpha_3$ .....	VII-34

Figure 11.	Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol-water partition coefficient (log $K_{ow}$ ) for several PCB congeners for the September 11 sample. a) parameters $ku_1$ , $K_1$ and $\alpha_1$ ; b) parameters $ku_2$ , $K_2$ and $\alpha_2$ ; c) parameters $ku_3$ , $K_3$ and $\alpha_3$ .....	VII-35
Figure 12.	Plots of measured log BAF versus predicted log BAF values. a) fixed $\alpha$ value; b) $\alpha$ value that changes with log $K_{ow}$ .....	VII-36

## LIST OF TABLES

Table 1.	Instrumental conditions for PCB analysis .....	VII-16
Table 2.	Description of parameters used in Thomann's bioaccumulation model .....	VII-18
Table 3.	Parameter values used for the Thomann model .....	VII-19
Table 4.	Total PCBs (ng/g wet weight, as Arochlor) for young-of-the-year bluefish and striped bass homogenates collected at Croton Point .....	VII-20

## INTRODUCTION

The prediction of fish body burdens of persistent organic contaminants and the projection of changes in the body burdens into the future as the result of remedial actions are critical components of ecological risk assessments for areas that have been impacted by the input of anthropogenic pollutants. A pollutant, as defined by the International Commission for Exploration of the Sea, is a substance, when introduced into the marine environment, that results in "deleterious effects" such as "...harm to living resources; hazards to human health...(and) impairment of the quality for use of seawater and reduction of amenities" (from Libes, 1992). One obvious reason for the concern about present and future pollutant concentrations in fish is the human health risks due to consumption of contaminated fish. In the Hudson River, which serves as breeding grounds for such commercially important species as striped bass and American shad (Smith, 1988), a major concern has been concentrations of polychlorinated biphenyl compounds (PCBs) in edible-sized fish, as well as in the water column and sediments. Monitoring programs have documented declining levels of PCBs in fish tissue, the water column, and sediments with increasing time and distance from the main upriver source (Armstrong and Sloan; 1988, Sloan et al., 1991). Equally important is the study of the biological, chemical, and physical processes affecting pollutant uptake.

In the summer of 1993, a field-based study of uptake of PCBs in young-of-the-year (YOY) bluefish was initiated. We feel that this study was a valuable addition to the burgeoning literature on PCB uptake because it used field-based exposures on a rapid-growing YOY fish in a well studied system. Most exposure studies have been laboratory-based (Neely et al., 1974; Branson et al., 1975; Veith et al., 1979; Bruggeman et al., 1984; Oliver and Niimi, 1985; de Voogt et al., 1991) and have dealt with adults. These studies have provided valuable data, with which mathematical relationships between physical-chemical properties of organic compounds, such as the octanol-water partition

coefficient ( $\log K_{ow}$ ) and the rate of uptake of the compound, have been formulated (MacKay, 1982; Chiou, 1985; Hawker and Connell, 1986). Field-based studies have either collected and analyzed adults or have used concentration values from the literature to test the goodness-of-fit of these models (Thomann and Connolly, 1984; Oliver and Niimi, 1988; Thomann, 1989; Connolly, 1991). Our use of YOY bluefish can be thought of as an exposure study because they enter the Hudson River estuary from offshore, where PCB concentrations are much reduced, at a known time (Nyman and Conover, 1988; McBride and Conover, 1991). Exposure concentrations in waters and sediments of the Hudson River estuary have been characterized, especially for dissolved water concentrations, by a number of studies (Brown et al., 1985). Especially important to our study were measurements made in the lower estuary over the past two years (Brownawell et al., unpublished data). Data from this study can be used to further test the assumptions of these bioaccumulation models, as well as to examine the effects of growth rate and consumption rate, both of which are exceedingly high in YOY bluefish (Chiarella and Conover, 1990; Juanes et al., 1993).

#### A. Partitioning Models

Models which predict fish body burdens of hydrophobic organic compounds either assume equilibrium partitioning between the water column and the organism, or account for uptake and elimination kinetics of exchange between the organism and the external environment. In the following discussion the notation used in papers by Thomann (Thomann, 1989; Thomann et al., 1989; Thomann et al., 1991) was used, since this work had been applied to the fate of PCBs in the Hudson River estuary.

The simplest models are equilibrium partitioning models, which assume that uptake is related to the partitioning of a compound between the environment (water column or sediment) and the lipids of the organism. The partitioning of an organic compound into

an organism can be predicted by the degree of lipophilicity of the chemical compound as measured by its octanol-water partition coefficient,  $K_{ow}$  (Neely et al., 1974; Chiou et al., 1977; MacKay, 1982; Hawker and Connell, 1988; Chiou, 1985). The theoretical equilibrium partition coefficient,  $K_B$ , refers to the partitioning of a nonionic organic compound between aqueous and lipoidal phases in a closed system at equilibrium (Connell, 1990). The degree to which the partitioning of an organic chemical approaches an equilibrium condition can be investigated in open systems (i.e., the environment) by calculating bioaccumulation factors (BAFs) and bioconcentration factors (BCFs). The bioaccumulation factor (BAF) is defined by several authors, including Oliver and Niimi (1988) and Thomann (1989) as the ratio of the tissue concentration (ug/kg lipid weight) to water column concentration (in ug/L). When exposure is due solely to water, this ratio is called the bioconcentration factor (BCF), where  $BCF = v_w/c$ .  $v_w$  is the tissue concentration resulting from water uptake only (in ug/kg lipid), and  $c$  is the water concentration in ug/L (Thomann, 1989). These coefficients assume a steady state, but not necessarily an equilibrium condition. Tissue concentrations are expressed either on a wet weight basis (ug/kg wet weight) or on a lipid weight basis (ug/kg wet weight). Normalizing the bioaccumulation factor to lipid weight assumes that different species of similar lipid composition in different environments can be compared for their potential to bioaccumulate nonionic hydrophobic organic compounds.

Results from field studies have produced measured BAF values that exceed those predicted by equilibrium partitioning models (Oliver and Niimi, 1988; Thomann, 1989). These studies have concluded that uptake from food (trophic transfer) accounts for the elevated tissue concentrations.

## B. The Thomann Food Chain Model

Models developed by Thomann and Connolly (1984) and Thomann (1981, 1989) account for uptake from water as well as from food. These models take into account the position of an organism in the food chain by allowing a predator at level  $i$  to feed on prey, which are at level  $i-1$ . Important physiological parameters, such as growth rate (kg lipid/day), food consumption rate (kg (w) of prey/kg (w) of predator•day), and chemical assimilation efficiency (ug of chemical absorbed/ug of chemical ingested), and chemical excretion rate(ug/day) are either measured, or estimated from literature values. Models with these parameters have been applied in the Great Lakes (Thomann and Connolly, 1984; Connolly et al., 1992), New Bedford Harbor (Connolly, 1991) and the Hudson River (Thomann et al., 1989, 1991). The general predictive equation for the model at steady state is as follows:

$$dv_i/dt = k_{ui}c + \alpha_{i,i-1} \cdot C_{i,i-1} v_{i-1} - (K_i + G_i)v_i$$

where  $v_i$  = the concentration of the chemical in the predator due to uptake from water + food (in ug/kg lipid wt.).  $v_{i-1}$  is the lipid-normalized chemical concentration in the prey species, and  $c$  is the dissolved water concentration (ug/L). The second term includes parameters involved in the uptake from food, such as  $\alpha$ , the chemical assimilation efficiency (ug of chemical absorbed/ug of chemical ingested), and  $C_{i,i-1}$  the specific consumption rate (kg (lipid wt.) of prey/kg (lipid wt.) of predator•day). Excretion rate ( $K_i$ , day<sup>-1</sup>) and growth rate ( $G_i$ , in kg lipid/day) are constants that describe loss terms. Growth is not a true loss term, but a dilution effect, due to an increase in tissue mass relative to chemical mass. The first term on the right side of the equation is the contribution from water to the whole body concentration. The lipid-normalized chemical uptake rate,  $k_{ui}$ , has been estimated by Thomann (1989) by utilizing metabolic parameters such as ventilation volume and respiration rate, as well as a mass transfer efficiency. The general equation is  $k_u = VE/w_l p$ , where  $V$  = ventilation volume (L/day)

which is in turn a function of the respiration rate of the organism.  $W_l p$  is the lipid weight of the organism.  $E$  is the mass transfer efficiency of the chemical across the gills. Thomann estimates  $k_u$  as  $10^3(w^{-\gamma}/p)E$  where  $\gamma$  is a respiration-based parameter given a value of 0.2. The mass transfer efficiency, is based upon chemical uptake studies of Boese (1984) and K onemann et al. (1980) and varies with log  $K_{ow}$ . Rates of uptake and excretion have been determined in laboratory-based exposure studies by several investigators, including Neely (1979), K onemann et al. (1980) and Bruggeman et al. (1981).

Under steady state conditions, the solution to the above equation, for a predicted body burden concentration of a predator, at level  $i$  in the food chain is as follows:

$$v_i = \frac{N_w c + \alpha_{i,i-1} C_{i,i-1}}{K_i + G_i} v_{i-1}, \text{ where } N_w \text{ at steady state is the BCF.}$$

BAFs are predicted from the above equation as follows:  $BAF = v_i/c$ .

Also,  $N_w = k_u/K + G$ . Thus:

$$BAF = \frac{k_u}{K_i + G_i} + \frac{\alpha_{i,i-1} C_{i,i-1}}{K_i + G_i} v_{i-1}/c, \text{ where } v_{i-1} \text{ is the BCF for striped bass.}$$

Given the extremely rapid growth rates and consumption rates that have been measured for YOY bluefish (Conover, unpublished data), YOY bluefish should provide a sensitive test for some of the parameters in the Thomann model. Equally significant is the fact that exposure conditions are reasonably well controlled, since dissolved PCB concentrations have been measured a number of times in the lower estuary (Brownawell et al., unpublished data) and prey species have been identified and enumerated (Juanes et al., 1993; Buckel and Conover, unpublished data).

## METHODS

### A. Field Collection

Collections of YOY bluefish were made with 100 and 200 foot seine nets at several sites in the Hudson River (Figure 1). Samples were wrapped in solvent-rinsed aluminum foil and placed on dry ice and transported back to the laboratory, where they were stored frozen until analysis.

Water samples were taken in August, using the following procedure. Sixteen liters of surface water were sampled by deploying a solvent-rinsed stainless steel pressure cylinder from a small boat, approximately 300 yards off of Croton Point, in Haverstraw Bay. Back on shore, samples were filtered under positive pressure, using high purity nitrogen, through a Whatman glass fiber filter (14.2 cm, grade GF/F, Whatman International) placed in a stainless steel filter housing (Millipore Corporation, Massachusetts, USA). The filtered water was collected in four-liter amber glass bottles, each containing 100 ml of methylene chloride.

Although fish were collected from several stations at several time points over the summer and fall, due to time limitations emphasis was placed on analyzing samples from the Croton Point station in Haverstraw Bay (Figure 1).

### B. Sample preparation

Fish samples were removed from their foil wrapping and thawed at room temperature. Two to three fish were homogenized using a Brinkman polytron (Brinkman products, Switzerland) and a Virtis homogenizer (Virtis Company, Boston, MA). Because of their small size, 50 individuals were homogenized for the June 20 striped bass sample. For the bluefish, stomachs were removed before homogenization using solvent-rinsed stainless steel forceps and scissors. Also prior to homogenization, total length, fork length, and wet weight were measured for each individual.

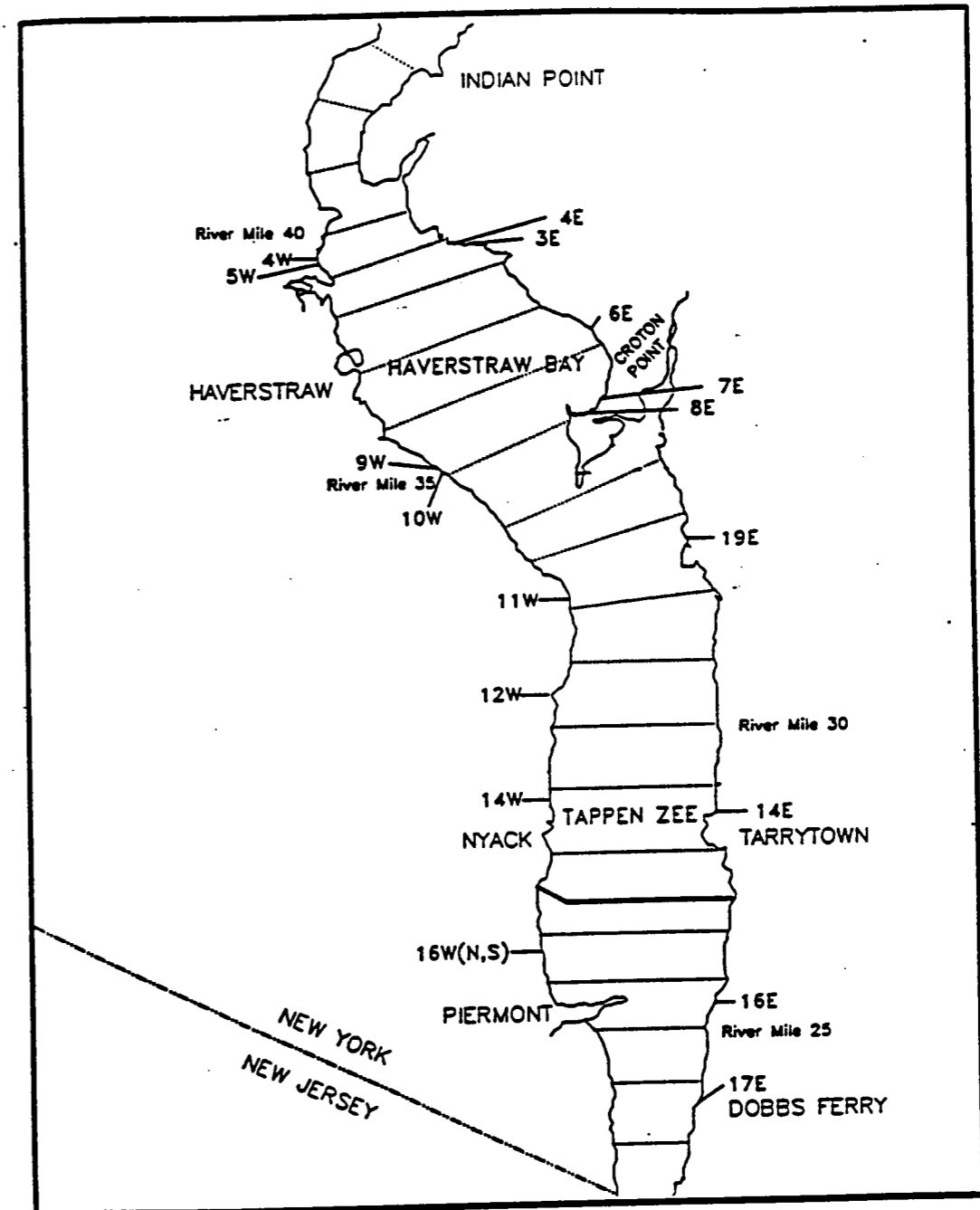


Figure 1. Location of sampling sites along the Hudson River. Station numbers correspond to sampling stations of the New York State Department of Environmental Conservation striped bass sampling program. The bluefish and prey analyzed in this study are from stations 7E and 8E (Croton Point).

Approximately four grams wet weight of homogenate was placed in a 50 ml centrifuge tube, surrogate standards added, and the sample was extracted three times with 25 ml of acetone using one of the homogenizers described above. Homogenization time for each extraction was three one-minute blendings (total blend time = 3 minutes per extraction). After being spun down in a table top centrifuge the acetone extracts were decanted from the centrifuge tube into a 1000 ml separatory funnel containing 500 ml of pre-extracted deionized water, saturated with NaCl. This aqueous extract was extracted three times with 50 ml of hexane. The hexane extracts were combined, and reduced in volume to 10 ml. Two ml were removed from each sample for lipid weight determination, and the remaining 8 ml were reduced in volume to one ml for column chromatography.

Each water sample, which consisted of approximately 16 liters (4 x 4 liter bottles) of water + methylene chloride, were poured into 4 liter separatory funnels for extraction. Samples were shaken for three minutes, left to settle, and the methylene chloride was drawn off from the bottom of the funnel. Each bottle was extracted twice more with 100 ml of methylene chloride, and the methylene chloride extracts were combined, dried over sodium sulfate and reduced in volume to one ml for column chromatography.

Fish extracts were separated from interfering lipid compounds with a column consisting of five grams of 5% deactivated florisol (60/100 mesh, Supelco, Inc., Bellefonte, PA), topped with two grams of 5% deactivated alumina (100-200 mesh, Bio Rad Laboratories, Richmond, CA). Both the florisol and the alumina were activated by combustion at 450°C overnight, and then deactivated by addition of 5% by weight of pre-extracted deionized water. Columns containing florisol plus alumina were further cleaned by eluting two times 50 ml of methylene chloride followed by two 50 ml rinses of hexane. The sample, at 1 ml volume, was charged onto the column, and a PCB-containing fraction was eluted, using 50 ml hexane. This fraction was reduced to

between one and two ml for instrumental analysis. For the water samples, a column consisting of seven grams of 5% deactivated silica gel (100-200 mesh, Bio Rad Laboratories, Richmond, CA) was used, following the same activation/deactivation cleaning and elution procedures described above.

Congeners 29 (2,4,5 trichlorobiphenyl) and 143 (2,2',3,4,5,6' hexachlorobiphenyl, Ultra Scientific, North Kingstown, Rhode Island) were added as surrogate standards at the beginning of the procedure, and octachloronaphthalene (OCN, Ultra Scientific) was added as an external standard prior to injection.

### C. Instrumental Analysis

Samples were injected onto a Hewlett-Packard 5890A gas chromatograph fitted with a 30 m DB-5 fused silica capillary column (J&W Scientific, Cupertino, California) and an electron capture detector. Instrumental conditions and temperature programming are listed in Table 1. Samples were quantified as total PCBs based upon a mixture of three Arochlor standards (Arochlors 1242, 1254 and 1260), and on a congener-specific basis. Quantitation was based on the surrogate standards CB 29 and 143, using peak areas. Recoveries of the surrogate standards were quantified based upon the OCN. Relative response factors of individual congeners were monitored using congener mixes obtained from the National Research Council of Canada.

Table 1. Instrumental conditions for PCB Analysis

Injection Port Temperature:	375°C
Detector Temperature	375°C
Column Flow Rate	3 mls/min
Carrier Gas:	Hydrogen
Analytical Column	30 m DB-5, 37 um ID, 25 mm film thickness
Temperature Program:	40°C for 2 min., ramp to 120°C @ 30°C/min, 120°C to 240°C @ 2°C/min, hold for 10 min.

D. Model Parameters.

Table 2 lists the sources of the parameters used in the Thomann model. The actual values used in the model calculations for these parameters are listed in Table 3. Weight specific growth rates and consumption rates were taken from estimates derived from laboratory studies performed, at different temperatures by Jeff Buckel for YOY bluefish (unpublished data). Body burden concentrations of PCBs, as well as lipid weights of predator and prey, were determined by the methods described above. For dissolved concentrations, a mean of two samples taken during this study along with four other samples from Haverstraw Bay, collected between 1991 and 1993 was used. The uptake rate,  $k_u$ , was determined three different ways. The first way, was after Thomann (1989), who estimated uptake based on body size as a measure of the respiration rate, and uptake efficiencies as a function of  $\log K_{ow}$ , based on experimental data. The second and third estimates of  $k_u$  were from Hawker and Connell (1985, 1986), who established linear regressions for  $k_u$  and  $K$  (excretion rate) versus  $\log K_{ow}$  for fish, molluscs, and other species, using laboratory experimental results cited in the literature for chlorinated hydrocarbons, including PCBs. Chemical assimilation efficiencies ( $\alpha$ ) were based on two estimates made by Thomann. The first was simply a constant value (0.5) and the second varied with PCB homologues (Thomann, 1989; Thomann et al., 1991). The Thomann model was tested on three different dates, June 30, August 4, and September 11. Striped bass, caught on these dates along with the bluefish, were used as a model prey species.

Table 2. Description of parameters used in Thomann's bioaccumulation model

**$k_u$  = uptake rate (L/day•kg wet wt) - 3 ways of estimating:**

1) Source: Thomann (1989)

$k_u$  as a function of  $K_{ow}$  and wet wt.

$k_u = 10^3(w^{-\gamma/p})E$ , where  $\gamma$  is a respiration parameter

$E$  varies with  $\log K_{ow}$ , based on laboratory-based exposure studies, as follows:

$\log E = -2.6 + 0.5 \log K_{ow}$  for  $\log K_{ow} = 2-5$

$E = 0.8$  for  $\log K_{ow} = 5-6$

$\log E = 2.9 - 0.5 \log K_{ow}$  for  $\log K_{ow} = 6-10$ .

2, 3) Source: Hawker and Connell (1985, 1986)

$k_u$  is based upon regression equations, derived from laboratory-based exposure studies.

2)  $\log k_1 = 0.18 \log K_{ow} + 1.98$

3)  $\log k_1 = 0.34 \log K_{ow} + 1.01$

Note: for 2 and 3,  $k_u = k_1/(p_i)$

**$K$  = chemical excretion rate: (day<sup>-1</sup>)**

1) Source: Thomann (1989) Calculated from model:  $K = k_u/K_{ow}$

2,3) Source: Hawker and Connell (1985, 1986) - Regressions derived from laboratory experiments.

2)  $\log k_2 = -0.41 \log K_{ow} + 1.47$

3)  $\log 1/k_2 = 0.66 \log K_{ow} + 0.43$

Note: for 2 and 3,  $K = k_2/(p_i)$

**$\alpha$  = chemical assimilation efficiency (ug chemical absorbed/ug chemical ingested):**

Source: Thomann, 1989 - 2 estimates

1)  $\alpha$  is fixed at a value of 0.5

2)  $\alpha$  varies with  $\log K_{ow}$

for Cl2 homologues:  $\alpha = 0.71$

for Cl3 homologues:  $\alpha = 0.8$

for Cl4 homologues:  $\alpha = 0.8$

for Cl5 homologues:  $\alpha = 0.63$

for Cl6 + homologues:  $\alpha = 0.35$

**$C'_{i,i-1}$  = Specific Consumption Rate (kg(w) of prey/kg(w) predator•day)**

Source: J. Buckel (unpublished data)

21°C:  $\log$  consumption rate =  $-1.17 (\log \text{ wet weight}) + 0.198$  (September sample)

27°C:  $\log$  consumption rate =  $-0.778 (\log \text{ wet weight}) + 0.112$  (June and August samples)

Note:  $C'_{i,i-1} = C'_{i,i-1}/(p_{i-1}/p_i)$

**$G$  = Weight specific Growth rate (day<sup>-1</sup>)**

Source: J. Buckel (unpublished data)

21°C:  $\log$  growth rate =  $-1.13 (\log \text{ wet weight}) + 1.747$  (September sample)

27°C:  $\log$  growth rate =  $-0.862 (\log \text{ wet weight}) + 1.625$  (June and August samples)

Table 3. Parameter values used for the Thomann model

Sample Date	Mean Wet Wt. (g)	Lipid Wt. (%)	Growth Rate	Consumption Rate
6/30/93	3.05	1.9	0.162	0.545
8/4/93	10.2	1.4	0.057	0.213
9/11/93	39.6	3.2	0.004	0.021

III. RESULTS

A. Trends of Accumulation Over Time

Recoveries of internal standards ranged between 75-100%, with a mean of  $84.5 \pm 8.3\%$ . Blank concentrations of total PCBs (normalized to a mean homogenate wet weight) ranged from 18.3 to 29.9 ng/g and never exceeded 3% of sample values.

Results are presented for two water samples, YOY bluefish from six dates, and YOY striped bass from three dates. Preliminary results from the 1993 survey of stomach contents of YOY bluefish showed that YOY striped bass had been a major component in the diet of YOY bluefish, at least through August (J. Buckel, personal communication). Total PCBs are presented in Table 4 for all samples analyzed to date. The two water samples showed good agreement, with total dissolved PCB concentrations of 10.8 and 33.3 ng/liter, which bracketed results from waters sampled previously near this location (Table 3). The main difference between these current samples and previous samples was the absence of octa - and nona-chlorinated biphenyl congeners, which are at very low concentrations in the other samples.

Table 4. Total PCBs (ng/g wet weight, as Arochlor) for young-of-the-year bluefish and striped bass homogenates, collected at Croton Point, and YOY bluefish collected at Great South Bay and water samples taken at Haverstraw Bay.

Collection Date	No. of Individ.	Mean Wet Wt. (g)	Lipid Wt. (% wet)	Conc. wet wt. (ng/g)
YOY Bluefish, Croton Point				
6/30/93	4	3.05	1.9	1,160
7/28/93	2	30.6	1.5	1,280
8/4/93	3	10.2	1.4	1,090
8/4/93 Large	2	99.9	2.0	1,140
8/11/93	3	17.2	1.4	1,870
8/24/93	3	29.4	1.3	1,090
9/11/93	3	39.6	3.2	1,060
Striped Bass, Croton Point				
6/30/93	25	0.31	1.8	1360
8/4/94	4	4.00	2.4	1530
9/11/93	2	9.72	1.6	795
YOY Bluefish, Great South Bay				
6/20/93	7	8.00	0.5	344
Water Samples, Haverstraw Bay				
Collection Date	No. of Liters			Conc. ng/l
8/4/93	17.0			10.8
8/4/93	16.9			33.3
9/92	16.7			20.3
5/92	15.4			14.0
12/91	23.7			17.0
10/91	8.97			31.8

There was a rapid uptake of PCBs for YOY bluefish almost immediately upon entering the estuary (6/30 sample, Table 2). Whole body concentrations of total PCBs on a wet weight basis remained fairly constant over the summer, despite large differences in body

size. Concentrations of total PCBs ranged from 1100 - 1280 ng/g wet weight, with the exception of the sample from 8/11. For each homogenate, individuals of the same size and weight were chosen (mean  $\pm$  5% wet weight). Especially noteworthy is the similarity of concentrations between homogenates of fish of two very different sizes sampled on 8/4. The larger 8/4 fish were sampled off Croton Point in a gill net, and their identity as YOY fish was verified by examining their scales (Jeff Buckel, personal communication).

Trends in accumulation of individual congeners in YOY bluefish are presented in Figures 2, 3, and 4. Concentrations were calculated on a wet weight basis (Figure 3a, 3b, 3c) and on a lipid weight basis (Figure 4a, 4b, 4c), and then normalized to the first sample date (considered to be  $t_0$ ), in order to be able to view several congeners with different concentrations on the same graphs. Thus a value of 3 for congener 26 signifies a threefold increase in concentration (on a wet weight basis) over  $t_0$ . Differences between homologue groups (congeners with the same number of chlorine atoms) can be seen when representative congeners are plotted over time (Figure 2). Concentrations of the trichlorobiphenyl (C13) and tetrachlorobiphenyl (C14) congeners remained relatively constant until day 72, when the C13 congeners increased (Figure 2a). The heavier molecular weight congeners (C15 and above) increased steadily with time and then remained constant, or decreased on the last date. This trend, plus the fact that the greatest increase over day 0 is seen for the C18 and C19 homologues (Figure 2c), was indicative of the bioaccumulation of the heavier molecular weight compounds. The increase in the C13 congeners on 9/11 may be a response to the increase in lipid stores in the fish, as reflected by the higher sample lipid weight (Table 2). The reasons this response was not seen in the other congeners may be due to the slower time to equilibration, due to greater resistance to uptake and excretion. Plotting these concentrations on a lipid-normalized basis showed similar trends (Figure 3a, 3b, 3c), except that on 9/11 the C13 congeners

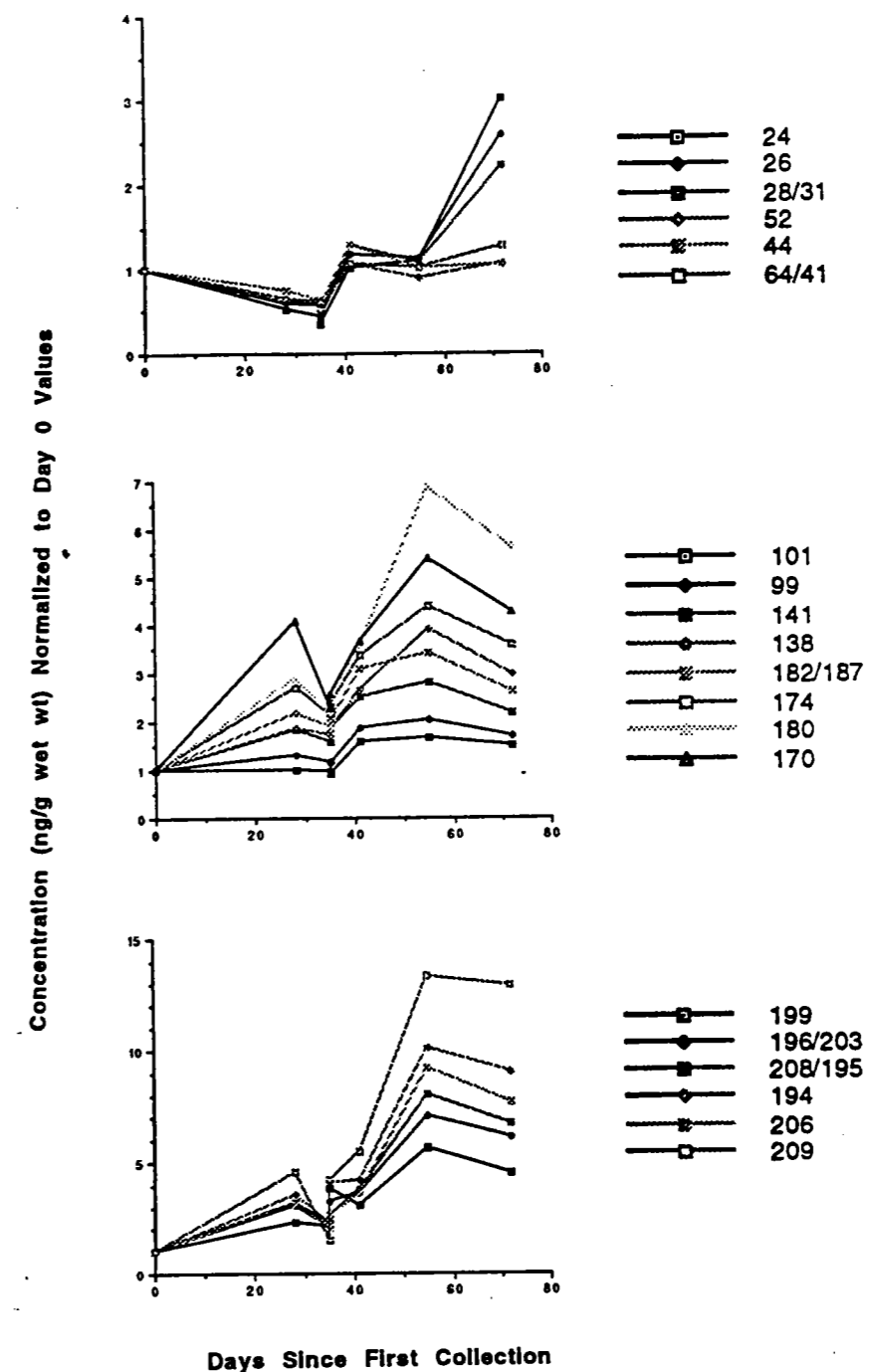


Figure 2. Accumulation patterns of selected biphenyl congeners in YOY bluefish over time. Concentrations are on a wet weight basis (ng/g wet), and are normalized to the first collection date (6/30/93). (a) trichloro (24,24,28,31) and tetrachloro (52, 44, 64/41) biphenyl congeners; (b) pentachloro (101, 99), hexachloro (141, 138) and heptachloro (182/187, 174, 180, 170) biphenyl congeners; (c) octachloro (CBs 199, 196, 203, 195) and nonachloro (208, 206, 209) biphenyl congeners.

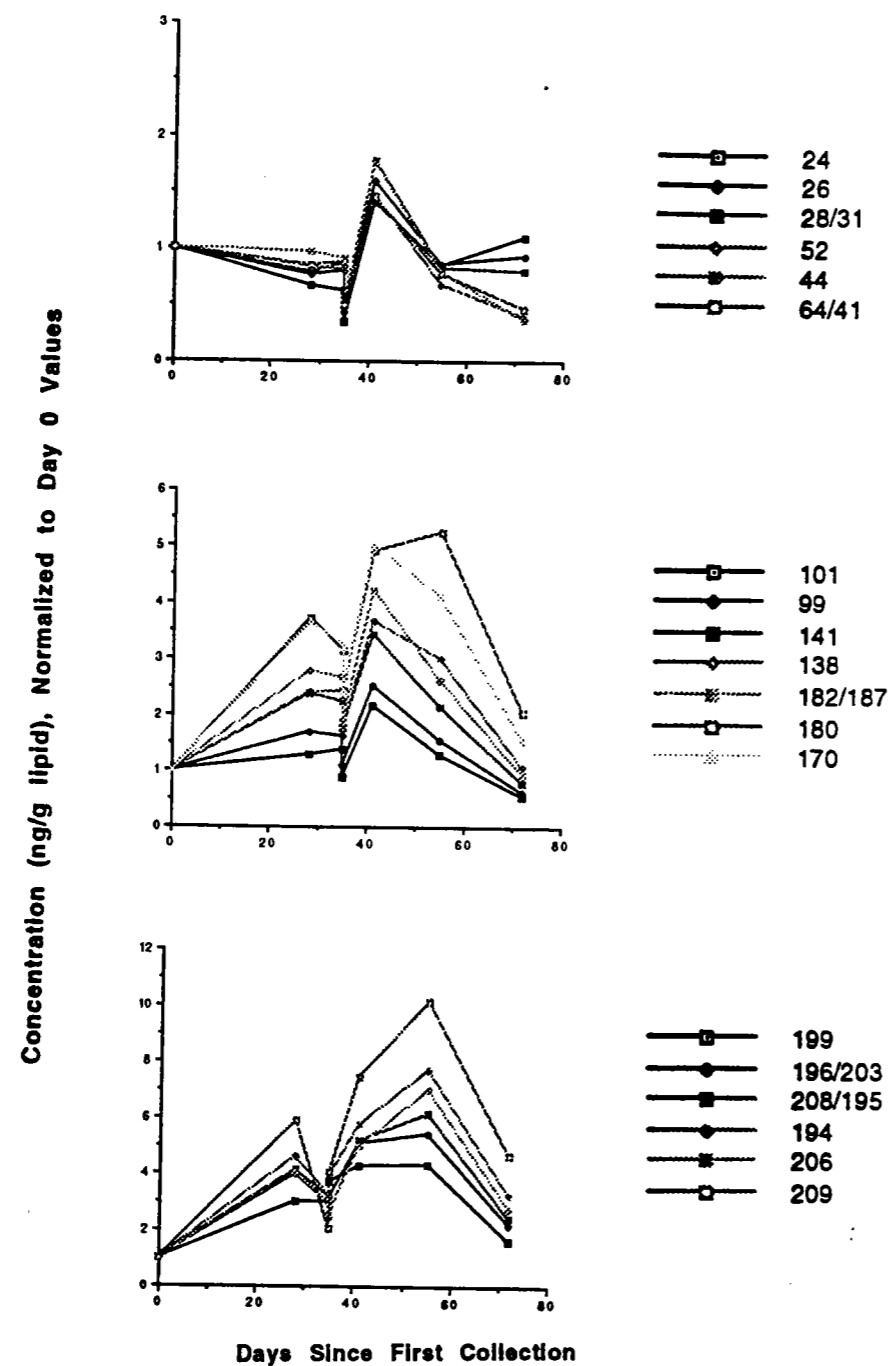


Figure 3. Accumulation patterns of selected biphenyl congeners in YOY bluefish over time. Concentrations are on a lipid weight basis (ng/g lipid), and are normalized to the first collection date (6/30/93). (a) trichloro (24,24,28,31) and tetrachloro (52, 44, 64/41) biphenyl congeners; (b) pentachloro (101, 99), hexachloro (141, 138) and heptachloro (182/187, 174, 180, 170) biphenyl congeners; (c) octachloro (CBs 199, 196, 203, 195) and nonachloro (208, 206, 209) biphenyl congeners.

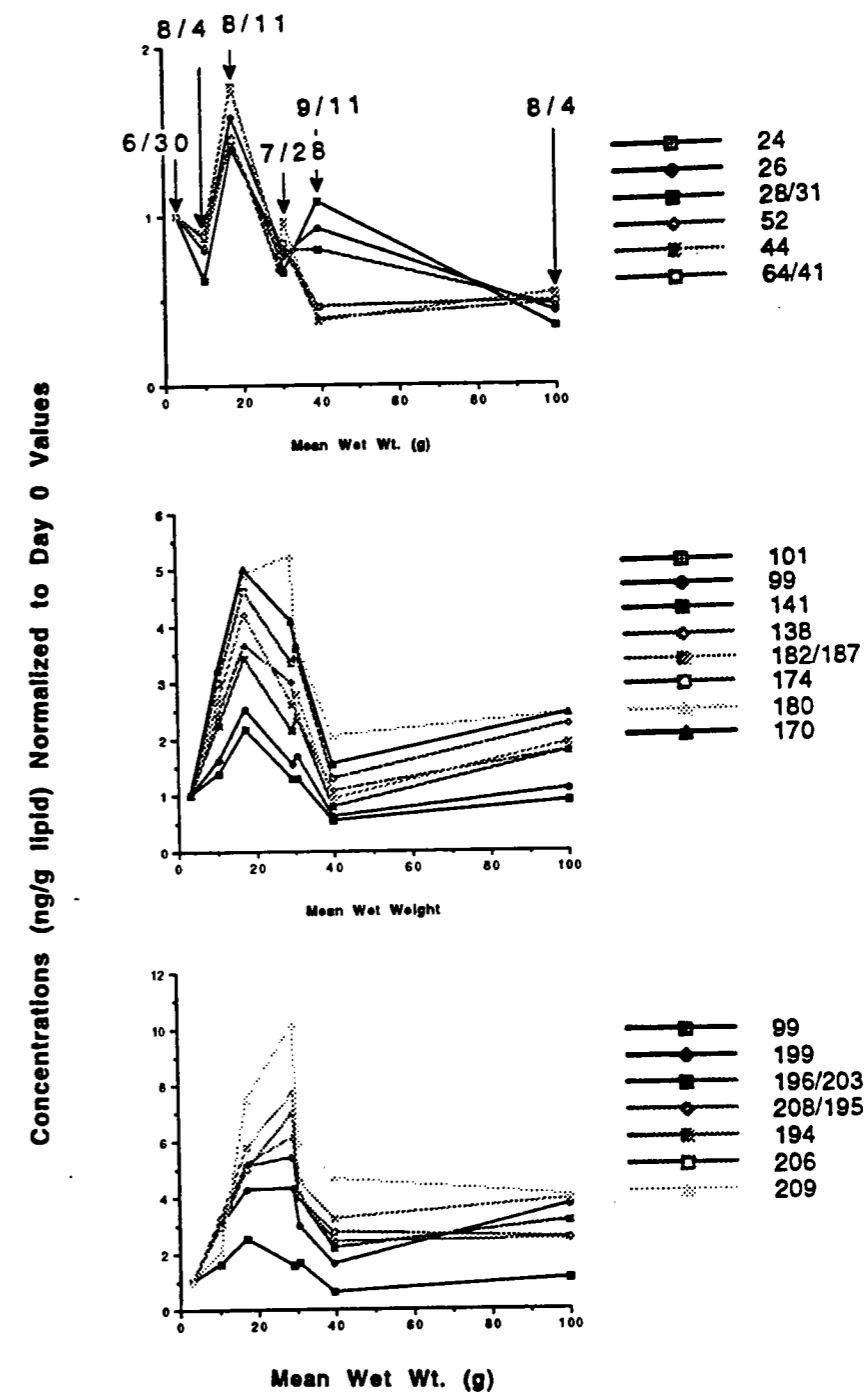


Figure 4. Accumulation patterns of selected biphenyl congeners in YOY bluefish versus mean wet weight of the individuals comprising the homogenate. Concentrations are on a lipid weight basis (ng/g lipid), and are normalized to the first collection date (6/30/93). (a) trichloro (24,24,28,31) and tetrachloro (52, 44, 64/41) biphenyl congeners; (b) pentachloro (101, 99), hexachloro (141, 138) and heptachloro (182/187, 174, 180, 170) biphenyl congeners; (c) octachloro (CBs 199, 196, 203, 195) and nonachloro (208, 206, 209) biphenyl congeners.

appeared to have reached an equilibrium with the increased lipid stores, while the other congeners, having longer equilibration times, showed a dilution effect (decreased concentration). Plotting lipid normalized concentrations against increasing body weight produced a more complex pattern (Figure 4a, 4b, 4c). There seemed to be no clear trend with increasing size, as evidenced by the similar concentrations of the two 8/4 samples. The length of time of exposure appeared to be an important factor controlling body burden concentrations. Again, the Cl3 congeners responded differently than the other congener classes, and that the greatest increase over initial values was seen with the Cl8 and Cl9 congeners.

Total PCBs in YOY striped bass were different than for bluefish, showing an increase in concentration and then a decrease on the last sampling date (Table 4). The increase and subsequent decrease followed the trend of the lipids concentration. The trends seen in the individual congener concentrations (ng/g lipid) over time were also different (Figure 5a, 5b, 5c). Unlike bluefish, the Cl8 and Cl9 congeners did not decrease in concentration on the last sampling date. This could be because unlike the bluefish, no large increase in lipid concentration was seen at this time (Table 2). The pattern of the Cl3 and Cl4 congeners showed a slight decline (roughly 0.6 of original concentrations), which could be the diluting effect of growth, as described by Thomann (1989). The more highly chlorinated biphenyl congeners (Cl5 - Cl9) were being accumulated, to a factor of two and three times the initial concentration. It is interesting to note that in YOY striped bass there was at the most, a threefold increase in concentration for the more highly chlorinated congeners over time zero, as opposed to the greater than tenfold increase in the Cl8 and Cl9 congeners seen for YOY bluefish.

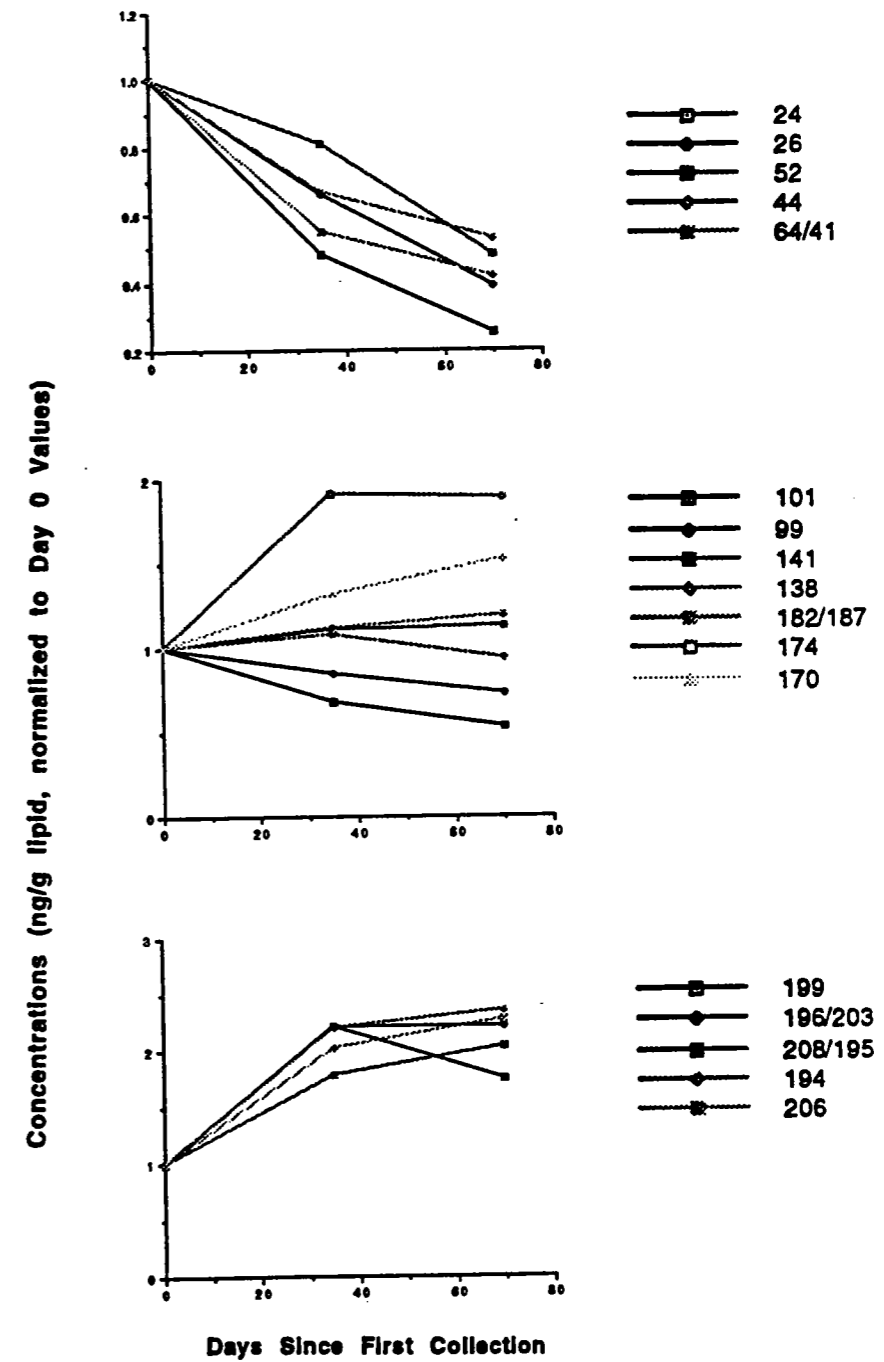


Figure 5. Accumulation patterns of selected biphenyl congeners in YOY striped bass over time. Concentrations are on a lipid weight basis (ng/g lipid), and are normalized to the first collection date (6/30/93). (a) trichloro (24,24,28,31) and tetrachloro (52, 44, 64/41) biphenyl congeners; (b) pentachloro (101, 99), hexachloro (141, 138) and heptachloro (182/187, 174, 180, 170) biphenyl congeners; (c) octachloro (CBs 199, 196, 203, 195) and nonachloro (208, 206, 209) biphenyl congeners.

### B. Equilibrium Partitioning

In Figures 6 and 7, log BAF (on a wet weight basis) versus log  $K_{ow}$  is plotted for 30 PCB congeners over several different collection dates for YOY bluefish. Log BAFs were calculated using mean dissolved concentrations from six water samples collected between 1990 and 1993, in Haverstraw Bay. The tissue concentrations were calculated on a lipid weight basis, and the resulting BAFs were compared to a theoretical prediction based on equilibrium partitioning between fish lipids and water derived by Chiou (1985, dashed line), which has the equation:  $\log BCF = 0.893(\log K_{ow}) + 0.607$ . The solid line is the best fit line through the data. The  $r^2$  values for the best fit line were fairly high and ranged from 0.85 - 0.92. Overall, the slopes of the best fit line and the theoretical line were similar, with the exception of the June 30 sampling. On 6/30, the BAFs for the higher molecular weight congeners were lower than predicted by the equilibrium line, indicating that perhaps at this early date where exposure time had not exceeded 10 days, these heavier compounds had not yet reached steady state. For the next four dates, the data lie above the predicted line. The deviation between the slopes of the best fit line and the theoretical line can be seen again, on 8/24, due to the higher molecular weight compounds lying farther above the predicted line than the lower molecular weight compounds. On 9/11, the data fell very close to the theoretical line, suggesting the possibility of equilibrium partitioning between the fish and dissolved PCBs.

### C. Thomann Model

Figure 8 compares my results with the predictions of the Thomann model for the three dates where striped bass were available. Uptake rate ( $k_u$ ) and excretion rate  $K$  were estimated, via the Thomann (1989) model; a fixed chemical assimilation efficiency ( $\alpha$ ) of 0.5 was used (Table 2). As can be seen, the model predictions of BAF matched the data fairly well, to a first approximation. The model predictions of BCF underestimated

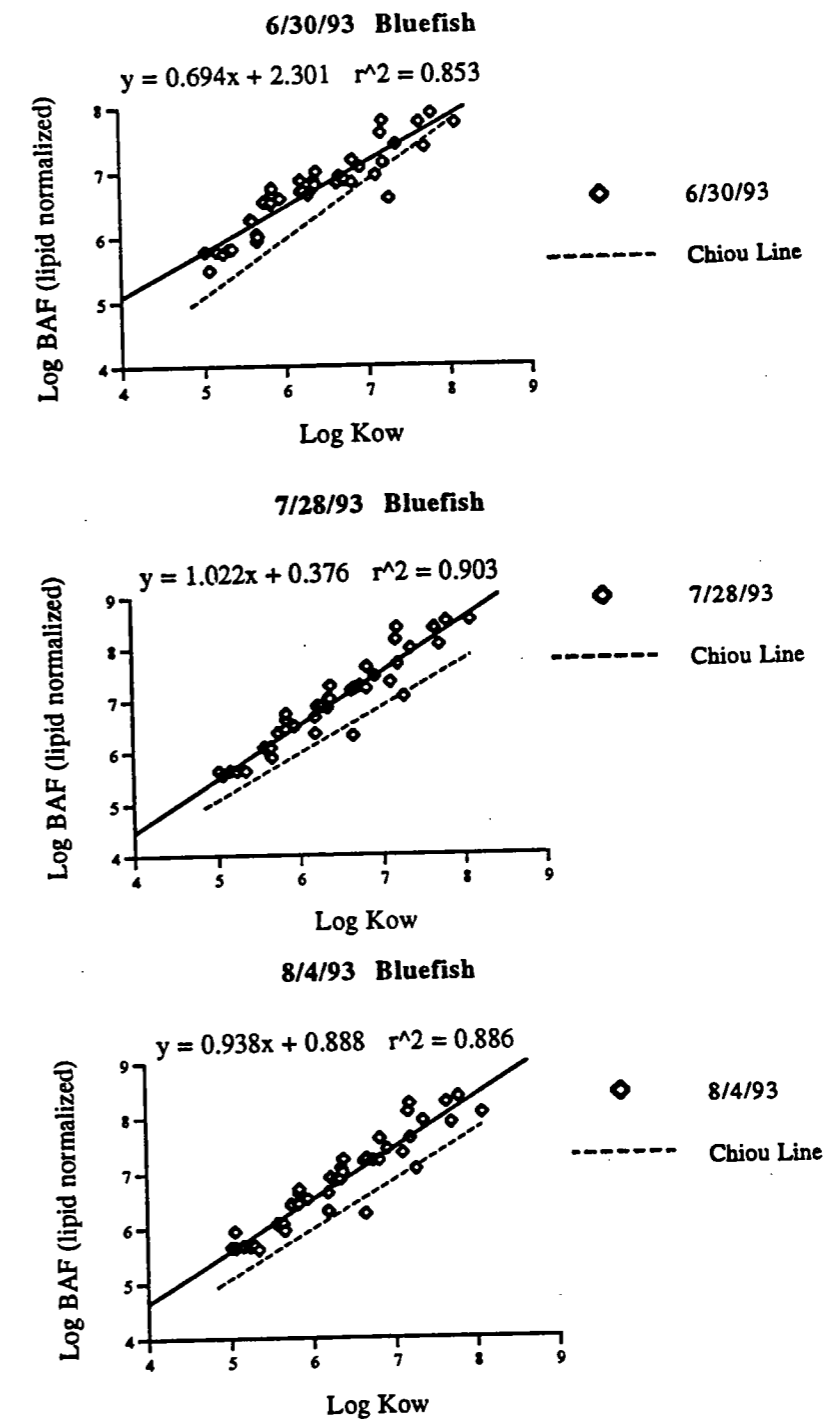


Figure 6. Plots of the log bioaccumulation factor versus the log of the octanol-water partition coefficient (log  $K_{ow}$ ) for several PCB congeners. BAF = organism concentration (ng/g lipid wt.)/water concentration (ng/Liter). Values of Log  $K_{ow}$  are from Hawker and Connell (1986). (a) June 30 sample; (b) July 28 sample and (c) August 4 sample. Superimposed is the theoretical line of Chiou (1985) (see text for the equation of the line).

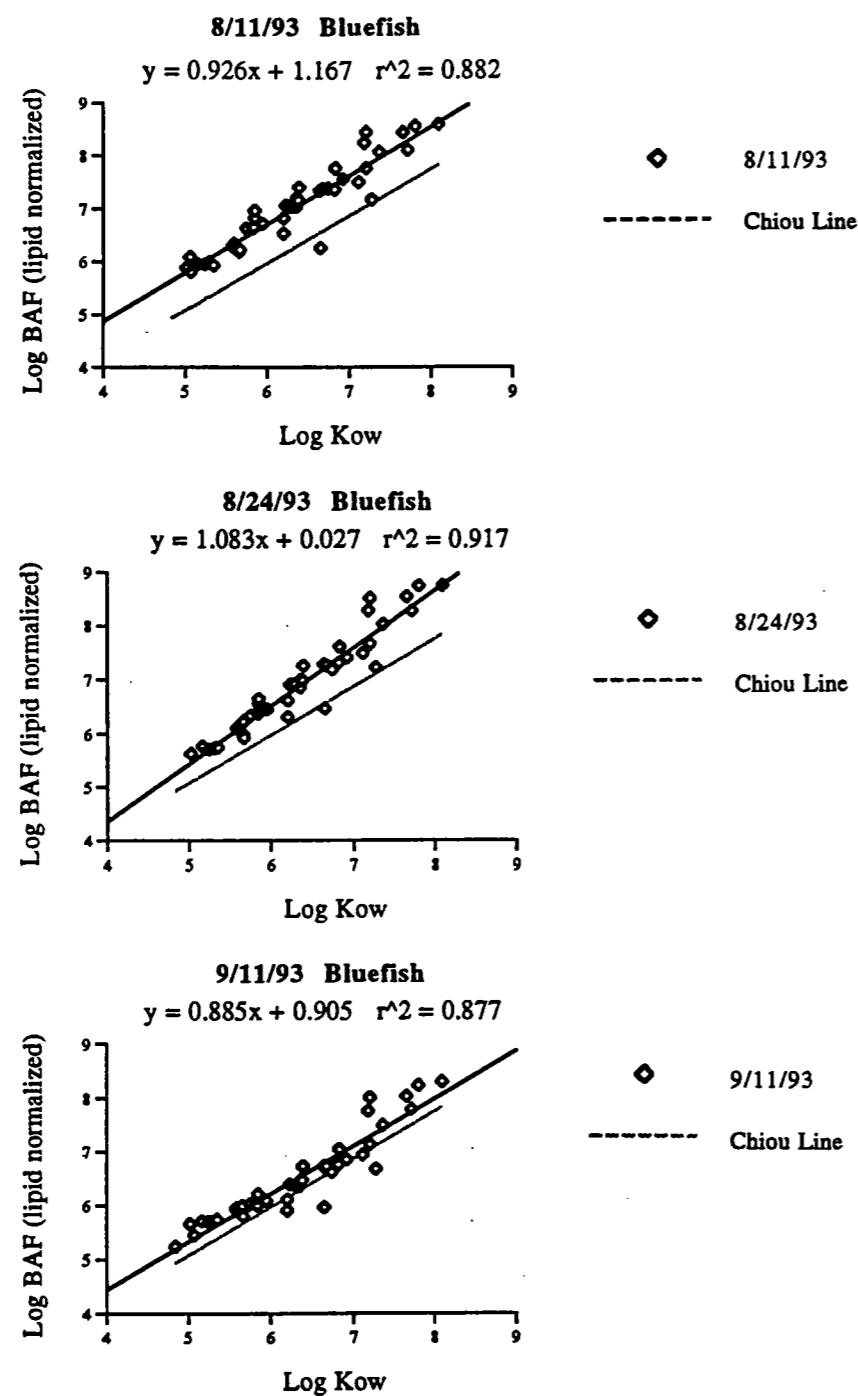


Figure 7. Plots of the log bioaccumulation factor (log BAF) versus the log of the octanol-water partition coefficient (log Kow) for several PCB congeners. Values of Log Kow are from Hawker and Connell (1986). (a) August 11 sample; (b) August 24 sample and (c) September 11. Superimposed is the theoretical line of Chiou (1985) (see text for the equation of the line)

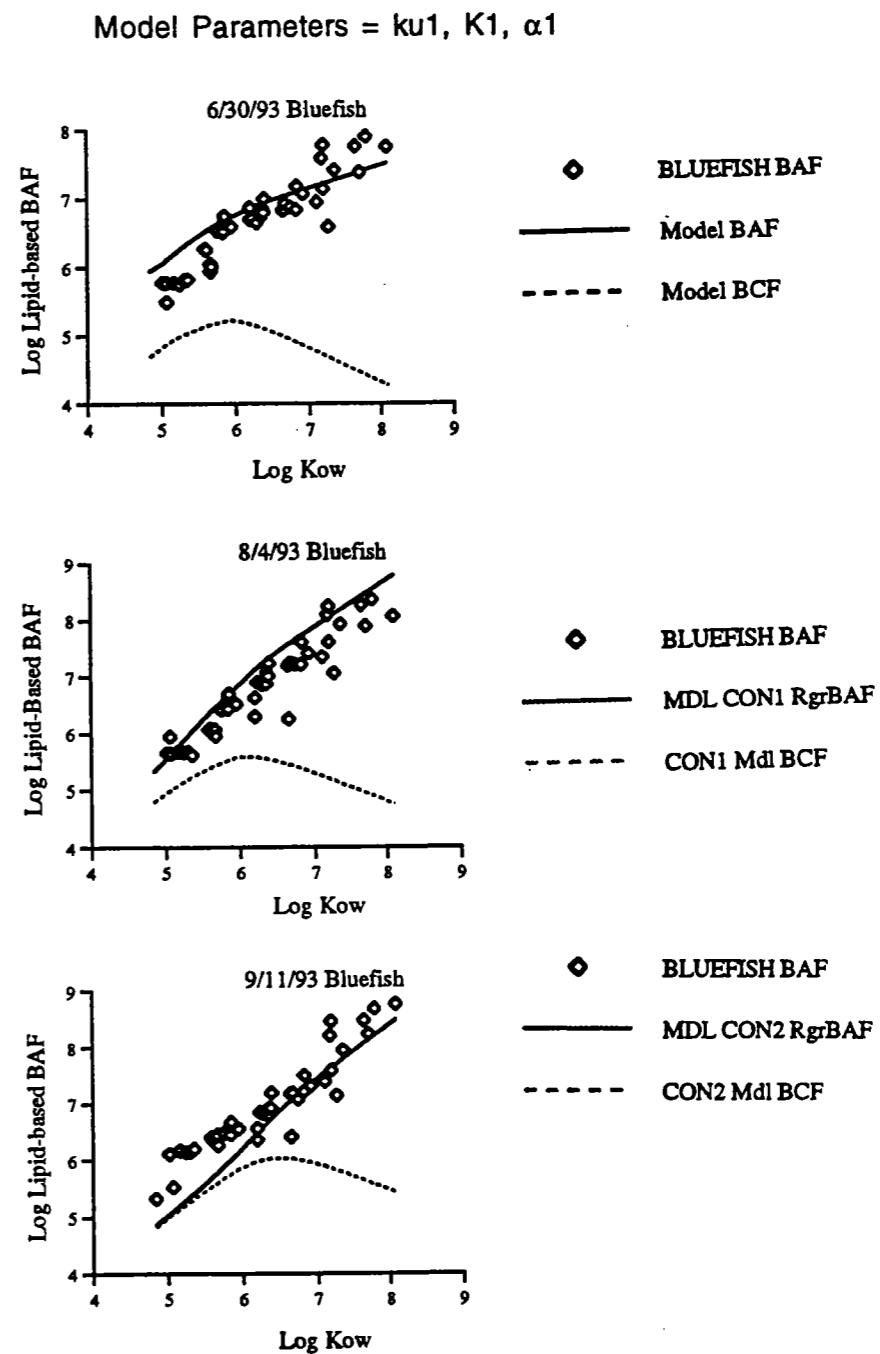


Figure 8. Plots of log bioaccumulation factor versus log of the octanol-water partition coefficient (log Kow) for several PCB congeners. Superimposed are predictions based on the Thomann model for BAF (solid line) and BCF (dashed line). (a) June 30 sample; (b) August 4 sample and (c) September 11 sample.

the data by one to several log units; this is inconsistent with the equilibrium model predictions of BCF (Figures 6 and 7), which seldom varied by more than 0.5 log units from the measured BAF values. It can also be seen that the model BCFs (which equal  $k_U/K+G$ ) bent sharply downward beyond a value of  $\log k_{OW}$  of 6, which is due to decreasing mass transfer efficiencies (E), at high  $\log k_{OW}$ s. The sensitivity of the BCF prediction to growth rate also could be seen. Over the three dates, the difference between the predicted BAF and BCF became less as growth rates decreased.

### DISCUSSION

One of the main observations of this study is that accumulation patterns of the individual congeners over time behaved differently than the sum of the total PCBs. Although the total PCB concentrations varied less than a factor of two over the entire study, individual PCB congeners varied by as much as a factor of 15 on a wet weight basis and 12 on a lipid weight basis. Dividing by the lipid weight did not change the overall accumulation pattern, except for the last date, when lipid-normalized concentrations decreased, rather than remaining constant. This may reflect the influence of growth, as a dilution factor, on this date. It is also interesting to note that the Cl3 congeners behave differently than all of the other congeners on this date, and may be responding to the increase in lipids more quickly than the more highly chlorinated compounds.

It is difficult to say much about the striped bass accumulation patterns, since there are only three time points. However, there appears to be a difference between the patterns of the Cl3 and Cl4 congeners, which show a continuing decrease, and the higher molecular weight congeners, which increase after the first date and remain at a steady concentration.

The data show that measured BAFs are above values predicted from uptake from water alone due to partitioning into lipids or by the food chain model. Since equilibrium

partitioning accounts for partitioning into lipids only, it is possible that the observed BAFs could be due to partitioning into other biochemical pools in the organisms. Another possibility, accounted for in the food chain models, is that dietary intake is an important route for bioaccumulation. Oliver and Niimi (1988) found  $\log$  BAF values to be elevated one or more log units above values predicted by equilibrium partitioning for salmonid fish in Lake Ontario. They ascribed these elevated values to a dietary source. The differences in this study between equilibrium predictions of BCFs and actual BAFs were only approximately 0.5 log units. It must be pointed out, however that the BAFs calculated by Oliver and Niimi (1988) were on a wet weight basis rather than a lipid weight basis. In this study, it appears as if a steady state situation is reached by 9/11, when the theoretical line and the best fit line merge (Figure 7c). This explanation is not supported by the predictions of the Thomann model which, for 9/11 has values above the model BAF line for congeners with  $\log k_{OW}$ s between 5 and 6, and 7 and 8. Differences between predicted and measured BAFs could be due to the inappropriateness of using of striped bass as a model prey. Juanes et al. (1993) found that in 1992, striped bass only made up 14% of the diet of bluefish in this size range.

To examine the effects of the different estimates of uptake and excretion rates on the model, results for the three dates were plotted, in Figures 9-11. It can be seen that uptake and growth rates affect the predicted BCF values greatly, and the predicted BAF lines, only slightly. The difference between using a fixed  $\alpha$  versus a changing  $\alpha$  value does not seem to affect BAF predictions, as seen in Figure 12. Therefore, it can be seen that accurate estimates of uptake and excretion are very important in using the Thomann model to predict BCFs. The uncertainty in these estimates, along with the chemical assimilation efficiency, could in part explain the large discrepancy between the equilibrium partitioning predictions and BCF predictions from the Thomann model.

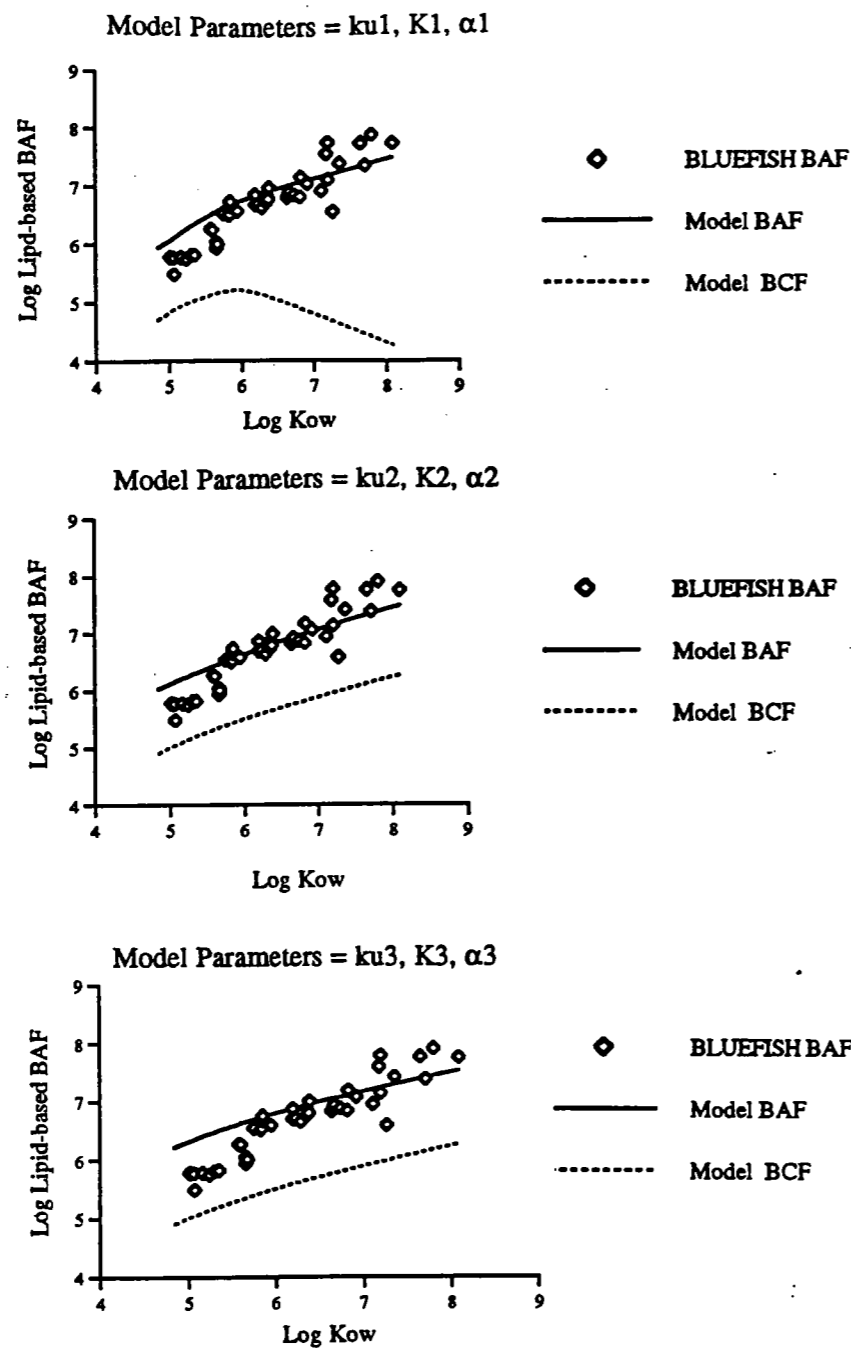


Figure 9. Plots of log bioaccumulation factor versus log of the octanol-water partition coefficient (low  $K_{ow}$ ) for several PCB congeners, for the June 30 sample. Superimposed are predictions based on the Thomann model for BAF (solid line) and BCF (dashed line). (a) parameters  $ku_1, k_1$  and  $\alpha_1$  from Thomann, 1989. (b) parameters  $ku_2$  and  $K_2$  from Connell (1990) and  $\alpha_1$  from Thomann (1989) (c) parameters  $ku_3$  and  $K_3$  from Connell (1990) and  $\alpha_1$  from Thomann (1989).

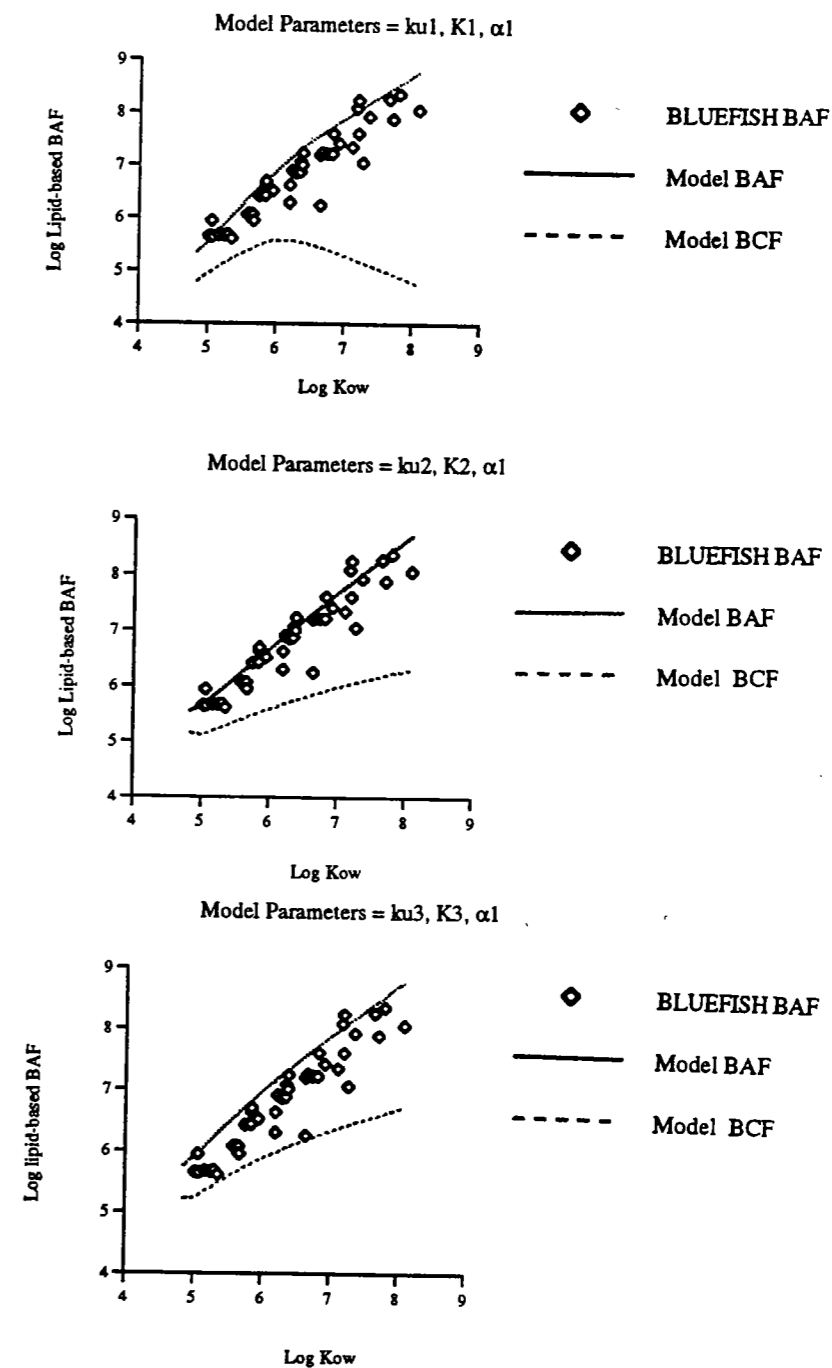


Figure 10. Plots of log bioaccumulation factor versus log of the octanol-water partition coefficient (low  $K_{ow}$ ) for several PCB congeners, for the August 4 sample. Superimposed are predictions based on the Thomann model for BAF (solid line) and BCF (dashed line). (a) parameters  $ku_1, k_1$  and  $\alpha_1$  from Thomann, 1989. (b) parameters  $ku_2$  and  $K_2$  from Connell (1990) and  $\alpha_1$  from Thomann (1989). (c) parameters  $ku_3$  and  $K_3$  from Connell (1990) and  $\alpha_1$  from Thomann (1989).

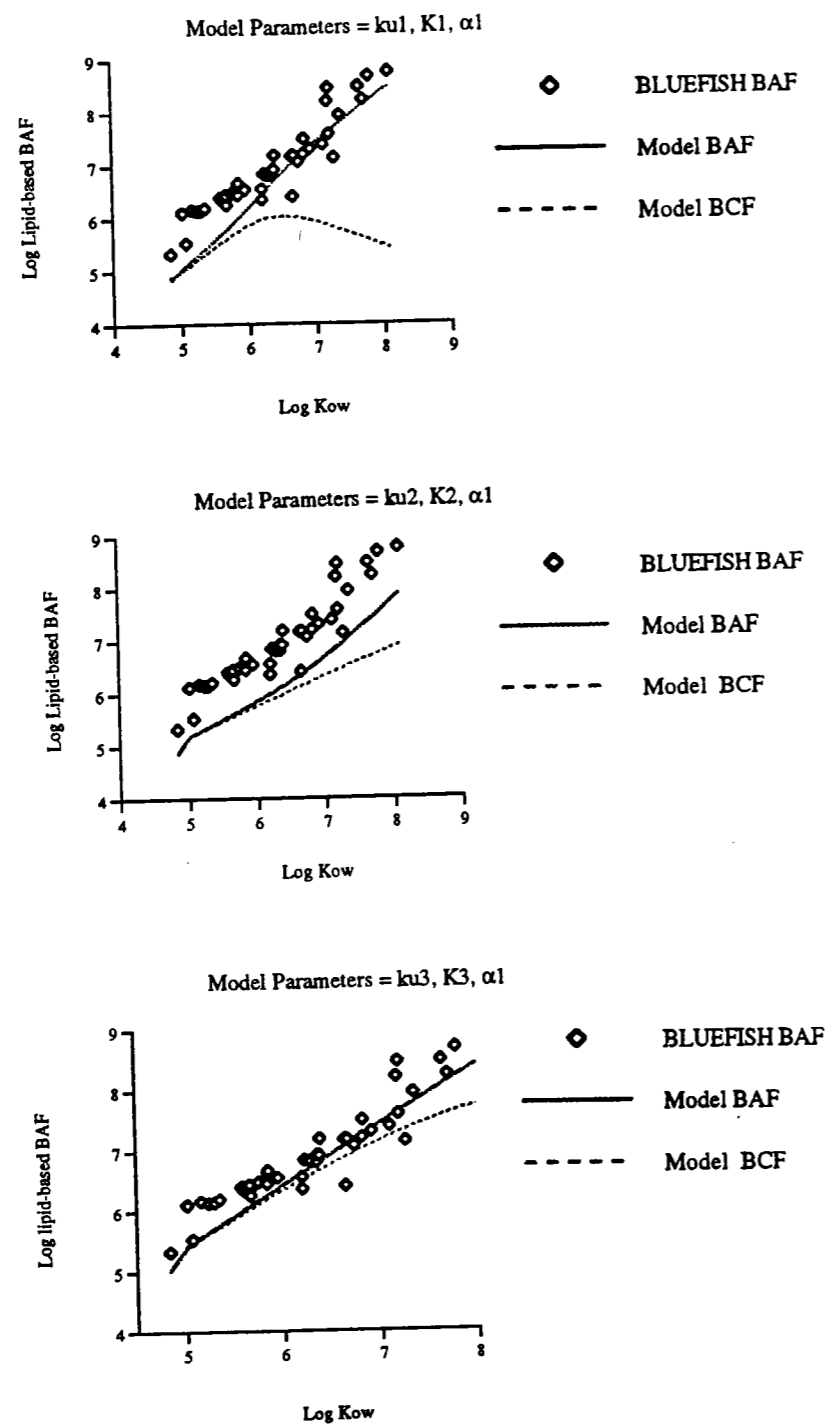


Figure 11. Plots of log bioaccumulation factor versus log of the octanol-water partition coefficient (low  $K_{ow}$ ) for several PCB congeners, for the September 11 sample. Superimposed are predictions based on the Thomann model for BAF (solid line) and BCF (dashed line). (a) parameters  $ku_1, k_1$  and  $\alpha_1$  from Thomann, 1989. (b) parameters  $ku_2$  and  $K_2$  from Connell (1990) and  $\alpha_1$  from Thomann (1989). (c) parameters  $ku_3$  and  $K_3$  from Connell (1990) and  $\alpha_1$  from Thomann (1989).

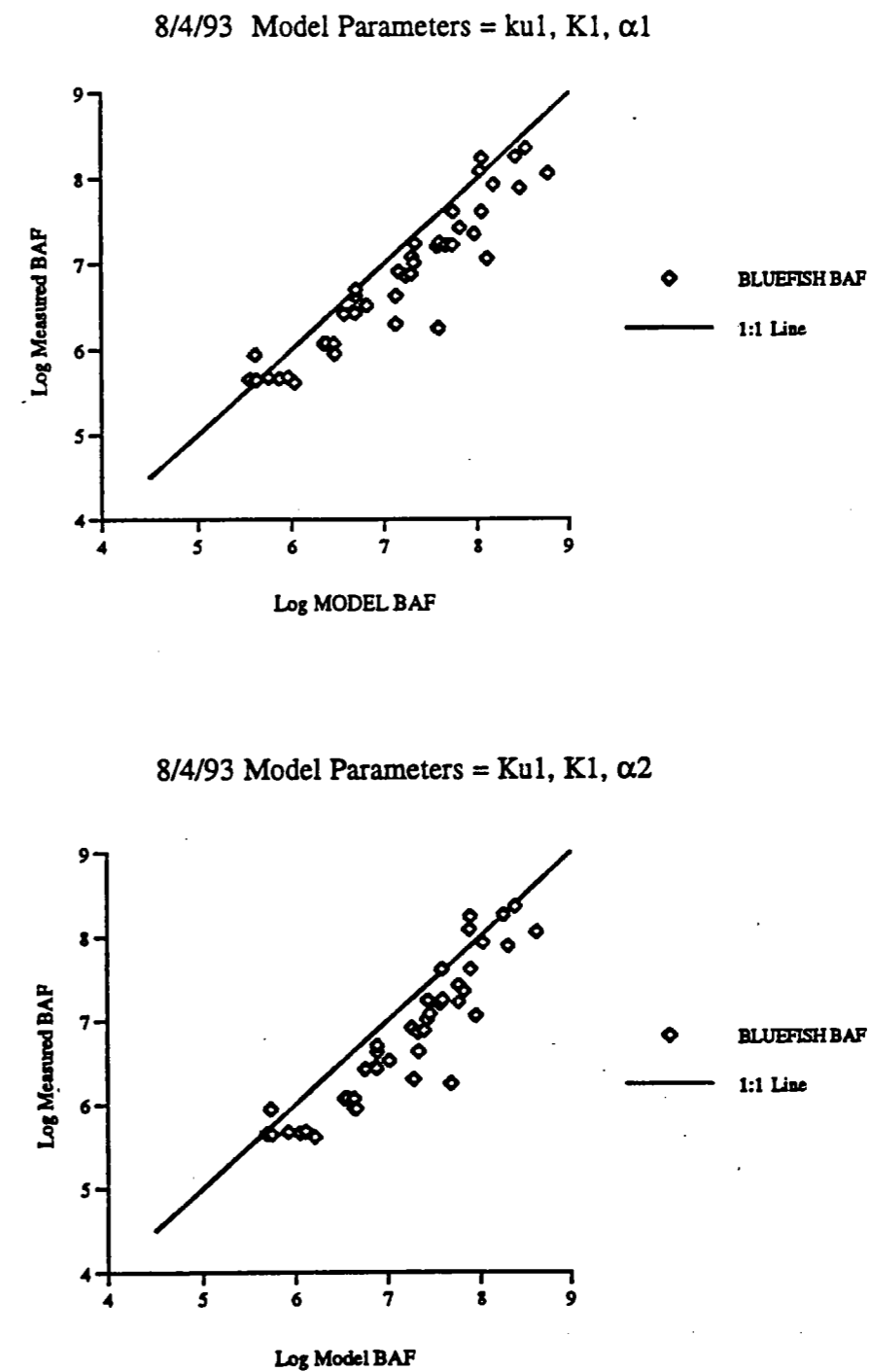


Figure 12. Plots of measured log BAF versus predicted log BAF values. The solid line represents the 1:1 relationship. (a) Parameters include a fixed  $\alpha$  value. (b) parameters include an  $\alpha$  value that changes with log  $k_{ow}$ .

Overall, the Thomann model predicted the measured data surprisingly well, given the uncertainties in parameters such as  $\alpha$ ,  $k_u$  and  $K$ . This is the best evidence that diet is an important source of PCBs to YOY bluefish. The best fit of model and data is on 6/30 (Figures 9a, 9b, 9c), which is also the time when the diet was mostly dominated by striped bass (J. Buckel, personal communication). Over time, the data change from well predicted by the model (Figure 9a, 9b, 9c) to being underestimated by the model (Figure 11a, 11b, 11c). It is not surprising that the rates of accumulation of PCBs into these fish are not at steady state, given the enormous change in the estimates of growth rates and consumption rates, over the size range of the YOY fish (Table 3). In addition, the large increase in lipid weight on the September date caused a new PCB gradient between the organism and the water, which would be expected to result in increased partitioning from the water into the organism. This is consistent with the results seen in Figure 7c, where the equilibrium partitioning model closely matches the observed data.

### CONCLUSIONS

- 1) YOY bluefish began rapidly accumulating PCBs, upon entering the Hudson River estuary.
- 2) Differences over time in the patterns of accumulation of congeners with different amounts of chlorine substitution may reflect differences in the rate of uptake and excretion between these homologue groups.
- 3) Equilibrium partitioning models underestimate measured bioaccumulation factors, which may reflect the importance of diet in uptake of PCBs, or may be due to the partitioning of PCBs from the dissolved phase into a non-lipid portion of the fish.
- 4) The food chain model predicted measured BAF values well, to a first approximation. Differences between predicted and measured values could be due to inadequate

representation of prey, or to a non-steady state situation resulting from the rapidly changing growth of these fish, and for September, to an increase in lipids production.

### RECOMMENDATIONS FOR FUTURE WORK

In order to better understand these data, it would be a good idea to model the time-dependent, non-steady state uptake. This study would also be enhanced by analyzing more bluefish samples, in order to estimate inter-station and intra-station variability. The analysis of more prey species, with corresponding comparison to the results of the 1993 stomach content analysis would enable us to obtain more representative prey concentrations. Finally, a comparison of lipid extraction procedures, where the acetone extractions used in this study are compared to petroleum ether and chloroform/methanol extractions, would provide valuable insight into a question that is not resolved in the literature, as to the best way to measure lipids.

### ACKNOWLEDGMENTS

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