Contaminants in Fish of the Hackensack Meadowlands (V. 3.0)

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Preface: This is a report on part of the Aquatic Animal Inventory Assessment, funded by the Meadowlands Environmental Research Institute and conducted 2001 - 2004. This was a multi-faceted study of the fish and invertebrates that inhabit the water and sediment of the lower Hackensack River and its major tributaries within the Meadowlands. The study consisted of several parts:
1. Collection, identification and enumeration of fish, crustaceans (shrimp and crabs) and benthic macroinvertebrates.
2. Analysis of fish tissue for heavy metals (arsenic, copper, zinc, lead, chromium and mercury) and organic contaminants (PCBs and pesticides).
3. Investigation of reproductive health and food consumption habits of white perch.
4. Analysis of sediments for metals and organics

The purpose of the study was to provide recent ecological data to state and federal resource agencies, consultants, and the interested public, to compare current fish and benthic abundance and diversity with a similar study conducted by the NJMC in 1987-1988, and to collect "baseline" data on contaminants in selected fishes and sediments in relation to health of the fish and risk to consumers.

This report addresses *specifically part 2* (analysis of fish tissues). It does *not* describe other studies directed toward fish health, diet, and reproduction, or for terrapin natural history.

1.0 Introduction

The Hackensack Meadowlands Development Commission (now the New Jersey Meadowlands Commission - NJMC) Commissioners authorized a fisheries resource inventory of the lower Hackensack River and its major tributaries. This "new" study seeks to replicate the work that was done during the initial NJMC fisheries study between February 1987 and December 1988. The primary purpose of the new study was to inventory the fishery resources of the lower Hackensack River, with an emphasis on determining if the fish community has responded to perceived improvements in water quality over the past 13 years. The prospect of a new NJMC fisheries resource inventory led scientists from Rutgers University and the University of Medicine and Dentistry of New Jersey (UMDNJ), under the auspices of the (then) NJMC/Rutgers' Meadowlands Environmental Research Institute (MERI), to propose additional studies that both compliment and supplement the work to be conducted by the NJMC. That work is now complete.

2.0 Research Plan

2.1 Fish Tissue Analysis

The water quality of the Hackensack River has been improving since the 1970's. This has resulted in an increase in recreational fishing and crabbing within the Hackensack Meadowlands District (HMD). Although the water quality has improved, river sediments can act as a persistent reservoir or source of contaminants that can migrate through the food web. Our analysis of tissues from selected fish species collected during the fishery inventory should provide baseline data on the extent to which the fishes living in the River are accumulating contaminants. This data could be used to determine whether the level of contamination poses a human health and/or ecological risk concern.

2.1.1 Selection of Contaminants of Concern (COCs)

Given the historical anthropogenic impacts that the Meadowlands has been subjected to over the past three centuries and the many studies that have analyzed sediments and surface waters within the HMD in the past 20 years, the COCs in the Meadowlands are well known. COCs analyzed for in tissue samples include metals and organic chemicals that are known to bioaccumulate. Therefore, the COCs would include the metals arsenic, cadmium, chromium, copper, mercury, lead, and zinc. Organic chemicals that are known to bioaccumulate (and would therefore be considered COCs) include chlorinated pesticides, polychlorinated biphenyls (PCBs) and dioxins/furans. Of those COCs that *bioaccumulate*, only the organics and mercury *bioconcentrate*, i.e., accumulating to higher levels (typically an order of magnitude) with each trophic level, making them of special concern.

2.1.2 Selection of Target Species

Species targeted for tissue analysis include common resident non-migratory species that are consumed by humans (i.e., "game" fish) and smaller resident species with small home ranges that are consumed by fishes, birds, mammals, etc. (i.e., "forage" fish). The target game fish included white perch (*Morone americana*), carp (*Cyprinus carpio*), pumpkinseed (*Lepomis gibbosus*) and brown bullhead (*Ictalurus nebulosus*). The target forage fish included the mummichog (*Fundulus heteroclitus*), Atlantic silverside (*Menidia menidia*) and the inland silverside (*Menidia beryllina*). Of these species, the only game fish collected in sufficient numbers allowing both seasonal and site comparisons was the white perch, and the only forage fish commonly found were the mummichog and the Atlantic silverside. Other game fish collected and analyzed were the carp and bullhead; these data were analyzed only *in toto*, rather than by season and site because of inadequate data distribution.

In addition to these fish species mentioned above, a common invertebrate crustacean, the blue crab, was also a target species for tissue analysis. Although eating, selling or harvesting blue crabs from the Hackensack River (or anywhere in the Newark Bay complex) is prohibited, there is nevertheless a recreational fishery for this delectable crustacean on the Hackensack River. Although we expected to capture the blue crab throughout the Meadowlands District (and in the first year we did find many), actually collecting them for analysis was planned as a second year activity. At this later time, crabs were relatively scarce; nevertheless, we present the small amount of data derived from these few crabs.

2.1.3 Processing and Analysis of Tissue Samples

The NJMC fishery team provided the specimens for analysis. For each collection, they filled out a chain-of-custody form listing the species, collection location, gear type, date, time, length, weight and any abnormalities observed for each specimen. Specimens (in labeled Ziploc® bags) were placed in ice and brought back to the NJMC lab at the end of the day. The specimens were kept on ice and dissected at the NJMC lab, providing the tissue necessary for UMDNJ, NJMC, and the contract laboratories analysis. Standard edible fillets ("skin off") were cut from the game fish specimens and the remainder of the carcasses were archived at -80 C. These species included the white perch, brown bullhead, and common carp. For purposes of either elemental or organic analysis, individual fish were used; it was not necessary to combine or pool specimens. A total of 168 white perch were analyzed for metals and 30 of these were also analyzed for organics. In addition, 29 brown bullheads, 9 carp and four blue crabs were analyzed for metals.

Forage fish were analyzed whole (including stomach contents). In the lab, composite samples were sliced vertically and 7-10 pieces (utilizing sections that included all body parts and organs) from 3-4 fish were combined for analysis. The lengths and weights of the individual fish used to make up each composite sample were not recorded, but the number of specimens per composite was noted. A total of 30 mummichog and 8 silverside composites were analyzed for metals, and 9 mummichog and 6 silverside composites were analyzed for organics.

2.1.3.1 Metals Analysis.

For metals analysis, a sufficient amount of tissue (2.0 ± 0.2 g wet weight, yielding ~0.5 g dry weight) was excised (or in the case of homogenized forage fish, combined as described above), oven-dried, weighed on a calibrated analytical balance to the nearest milligram, the dry weight recorded, and mineralized in 10 ml Trace Metal Grade HNO_3 (Fisher Scientific) in Teflon bombs in a MARS-5 programmed microwave digester (CEM Corp., Mathews, NC). The resultant mineralized solution was boiled off to near dryness, restored to 10 ml volume with 1% HNO_3, and divided in half. One half was used by the NJMC laboratory for analysis of Cd, Cr, Cu, Pb and Zn by graphite furnace atomic absorption spectrophotometry (AAS). Eventually Zn was replaced by Ni in the NJMC analyses for reasons not transmitted to this writer. The other half was used by UMDNJ for Hg analysis by cold-vapor AAS in a Bacharach MAS-50D mercury analyzer and for As analysis in a Perkin-Elmer 3100Z spectrophotometer by graphite furnace AAS with Zeeman effect. Wet weight metal levels (from which government agencies derive their risk analyses) were back calculated by dividing our dry weight values by 4, since our moisture contents were 74 - 78%.

2.1.3.2 Lipid Analysis.

Lipid content: additional aliquots of muscle and of forage fish homogenates were extracted for total lipid analysis. Such data can be used to indicate fish health, as well as for lipid normalization of organic contaminants. The traditional method was used. This involves a 50/50 mix of acetone and diethyl ether, 5 ml per gram of wet sample, in which it was ultrasonicated. The mix was centrifuged and repeated two more times. The three solvent eluates were combined and the liquid layer evaporated under N_2 (to avoid adding oxygen to unsaturated lipids). Then, the residue was weighed on a calibrated analytical balance to the nearest 0.1 milligram. Data are expressed as percent of wet weight.

2.1.3.3 Organic Analysis.

A subsample of tissue (prepared by UMDNJ)was sent to a contract laboratory (Technion Labs) for solvent extraction, extract concentration and cleanup, and initial phase confirmation of organics (pesticide/PCB's) by GC/MS. Technion sent the resulting extract to the NJMC lab for final pesticide/PCB analysis, using EPA Method 608. The final analysis was done using a Gas Chromatograph with an Electron Capture Dectector. This process proved to be inadequate because of the NJMC facilities being inadequately set up for such work. Subsequently, separate homogenates were sent to Dr. Jeff Ashley at the Academy of Natural Sciences, Philadelphia. Dr. Ashley's report has been provided separately, although his results are discussed in this document.

2.1.4 Quality Assurance/Quality Control

Quality control for the analysis of fish and crustacean tissue included: chain-of-custody documentation of all materials selected for analysis and archiving; the use of carbon-steel dissection instruments to avoid chromium contamination from stainless steel; the use of deionized/distilled water; acid-washing and triple rinsing of glassware; use of an analytical balance calibrated with both internal and external standards; inclusion of the NRC-Canada certified reference material (CRM) dogfish liver tissue (DOLT-2) and method blanks (one each with each 12 unknowns). An acceptable run was one in which the CRM data were within the published 95% CI. (An exception to this was arsenic analysis, for which we were consistently at 75% of the published value). Minimum detection levels were defined as three times the standard deviation of the blanks.

2.1.5 Methylmercury analysis

In addition to the above analytical activities, we sent 12 subsamples of fish and crab tissues for methylmercury analysis to the University of Georgia's Skidaway Institute (second year only). There, our samples were analyzed by atomic fluorescence spectrophotometry. They used DORM-2 (dogfish muscle tissue) for a CRM as part of their QA/QC.

3. Results and Discussion

3.1 Metals (generalities).

A total of 168 white perch were caught, representing virtually all locations and all seasons. This is significant because this species is the one most often sought and caught by recreational fishers, thus our data are particularly relevant. In addition, 30 pooled mummichog samples were found year-round at 5 locations, and 8 pooled Atlantic silverside samples were found at several seasons at the three seining locations. While not relevant to human health, these two species are forage fish and thus important in the ecology of the HMD.

The results for overall metal burdens are summarized in the following table:

Table 1. Metal burdens in fish, µg/g dry weight - means and stand. dev.

Species	n	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
White Perch	168	0.19 (0.24)	0.13 (0.15)	0.24 (0.19)	2.49 (1.58)	1.75 (0.93)	1.94 (1.60)	0.70 (0.98)	9.95 (11.31)
Brown Bullhead	29	0.50 (1.27)	0.11 (0.08)	0.24 (0.17)	2.42 (0.70)	0.67 (0.59)	0.94 (0.74)	0.69 (0.62)	24.7 (3.36)
Carp	9	0.06 (0.07)	0.08 (0.07)	0.22 (0.08	2.57 (1.38)	1.20 (0.71)	1.40 (1.05)	0.24 (0.44)	n.a.
Mummi-chog	30*	0.14 (0.17)	0.07 (0.05)	1.12 (1.24)	11.76 (7.49)	0.25 (0.16)	4.94 (6.28)	0.93 (1.21)	11.99 (6.23)
Atlantic Silversides	8*	0.53 (0.36)	0.09 (0.07)	0.83 (1.05)	3.79 (1.45)	0.48 (0.21)	2.13 (2.05)	0.77 (0.74)	3.73 (2.51)

*Composites of 2-5 fish each.

As stated above, the white perch are of special interest because of their frequency and desirability on the part of fishers. The amounts of metals in the white perch fillets are as follows:

1. Arsenic- ranges from undetectable to 1.01 µg/g dry weight, equivalent to 0 - 0.25 µg/g wet weight.
2. Cadmium - ranges from undetectable to 0.94 µg/g dry weight, equivalent to 0 - 0.23 µg/g wet weight
3. Chromium -ranges from 0.03 to 1.28 µg/g dry weight, equivalent to 0.01 - 0.32 µg/g wet weight.
4. Copper - ranges from 0.86 to 6.48 µg/g dry weight, equivalent to 0.22 - 1.62 µg/g wet weight (two outliers were omitted because they were one and two orders of magnitude higher than the other 166 data points; white perch are notorious for accumulating Cu in liver (mg/g, rather than µg/g (Bunton et al, 1987), and liver tissue may have contaminated these two muscle samples).
5. Mercury - ranges from 0.07 to 4.66 µg/g dry weight, equivalent to 0.02 - 1.16 µg/g wet weight.
6. Nickel – ranges from 0.24 to 7.27 µg/g dry weight, equivalent to 0.06 - 1.82 µg/g wet weight.
7. Lead - ranges from undetectable to 4.25 µg/g dry weight, equivalent to 0 - 1.06 µg/g wet weight.
8. Zinc - ranges from undetectable to 23.1 µg/g dry weight, equivalent to 0 - 5.75 µg/g wet weight.

The levels of arsenic reported here are typical of seafood in general, and are not considered of significance. Most arsenic in seafood is in the form of organoarsenicals, most of which are poorly absorbed and metabolized by consumers. Thus, they are not considered to be toxic (Irgolic, 1992). Neither the EPA nor the FDA has published guidelines for arsenic in food.

The cadmium, chromium, copper, lead and zinc levels are not particularly high, either, nor have our government agencies published guidelines for them other than chromium. They do not bioconcentrate. There are, however, European guidelines for maximum permissible levels: Cd – 0.06, Cu – 2-3, and Zn – 15 mg/d/person (European Community, 2001). These translate to Cd - 0.3 µg/g, Cu – 10-15 µg/g; and Zn – 75 µg/g for the 200 g meal, *if eaten daily*. By these European guidelines, the HMD white perch are not problematical.

The "Chromium level of concern for crustacean shellfish" is 22 μg/g (USFDA, 1993), two orders of magnitude higher than the means found in our Hackensack game fish species.

3.1.1. Mercury.

The one metal that we analyzed that is of concern here is mercury. It is the only metal known with certainty to bioconcentrate, becoming an order of magnitude higher with each trophic level. The reason for this is that the most likely form to be found in fish is methylmercury, and this especially toxic organic form is taken up by organisms in a manner similar to organic compounds. The FDA action level for mercury in fish is 1 ppm (μg/g) wet weight. This was exceeded by 4 of the 168 white perch, and these were all at the longer end of the length continuum. However, there are additional EPA and FDA joint guidelines for fish consumption vis-á-vis Hg. These agencies have recommended no more than one meal per month of 0.47 - 0.94 ppm Hg in fish. Thus, there is a good probability of catching a "risky" white perch, since 53 of 168 (32%) exceeded the 0.47 ppm level.

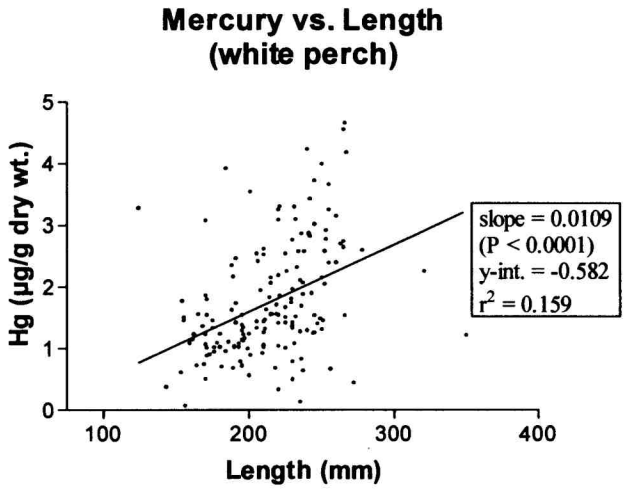
Of the other game species, only one of 29 brown bullheads and one of the nine carp exceeded that 0.47 ppm one-meal-per-month risk level.

Crab claw meat from four crabs was analyzed for the same metals. The one that could be considered "eating size" met the risk level of 0.47 μg/g Hg wet weight. It is, obviously, inappropriate to draw conclusions from these few data points.

3.1.1.1. Mercury uptake vs. fish size.

A mercury/size correlation typically exists in fish. This is demonstrated in our data (Figure 1).

1A.



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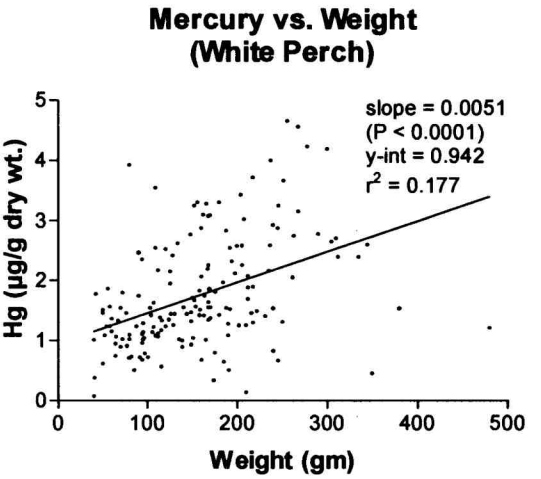


Figure 1. Size relationships for mercury uptake in white perch, by both length (A) and weight (B). A slight but significant relationship is demonstrated, using data from all sites and all seasons (P = 0.05 for both correlations).

We can conclude from Figure 1A that a white perch greater than 10 inches (254 mm) long will

have a 50% probability of being a risky meal, and this approximates the minimum-sized "keeper" for a recreational fisher. Considering the amount of Hg circulating in the HMD, the amount in white perch was surprisingly low. The answer can be found in the white perch's dietary habits. The stomach content analysis done for a companion study (see final report from J.S. Weis, Rutgers University) shows that they rarely eat fish, but instead eat small crustaceans, as do the mummichogs.

3.1.1.2. Mercury uptake – seasonal relationships.

The data have been analyzed for seasonal and site trends, but only for white perch, since this is the one species of game fish for which sufficient data are available. The higher Hg levels tend to occur in warmer weather, when more people are fishing. Thus, the probability of catching white perch would be increased in the summer (see Figure 2, below). [To aid in interpreting these Hg uptake graphs, keep in mind that these are data for dry weight. To convert to wet weight, divide by 4. Thus, the 0.47 µg/g consumption advisory level would be about 2 on these graphs.]

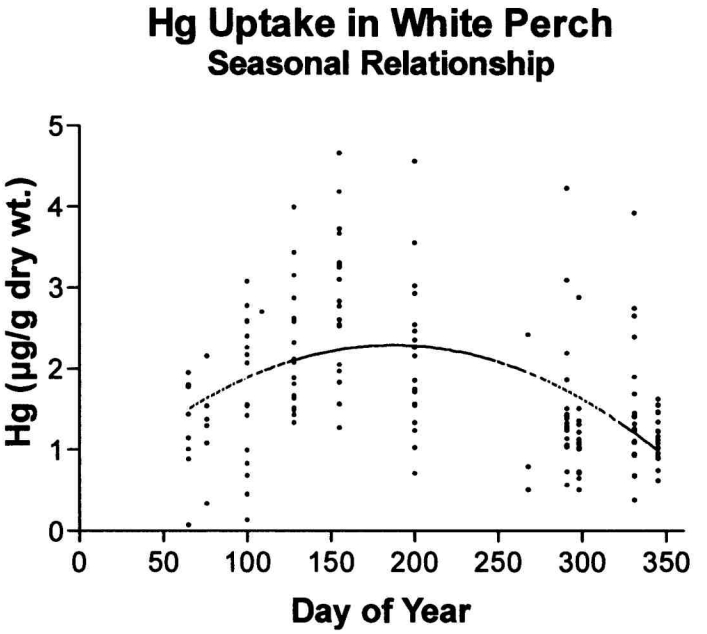


Figure 2. The relationship of season to mercury uptake - it is seen to be higher in the summer than in the cooler months. Days 130 – 240 are mid-May – September. The curve represents the predictable level, and is calculated as $y = a + bx + cx^2$. For this curve, $r^2 = 0.211$, $P < 0.01$.

The reasons for higher mercury in fish during the warmer months may represent the higher food intake at this time. Depuration occurs at a rate that, like any physiological activity or chemical reaction, is temperature-dependent. Nevertheless, it still continues during the winter, a time when food is scarce to non-existent, so that a fish will show a net loss of Hg during that time.

3.1.1.3. Mercury uptake – site relationships.

The sites from which the fish were collected have also been analyzed for mercury uptake,

averaging all sizes and seasons for each site. This is demonstrated in Figure 3.

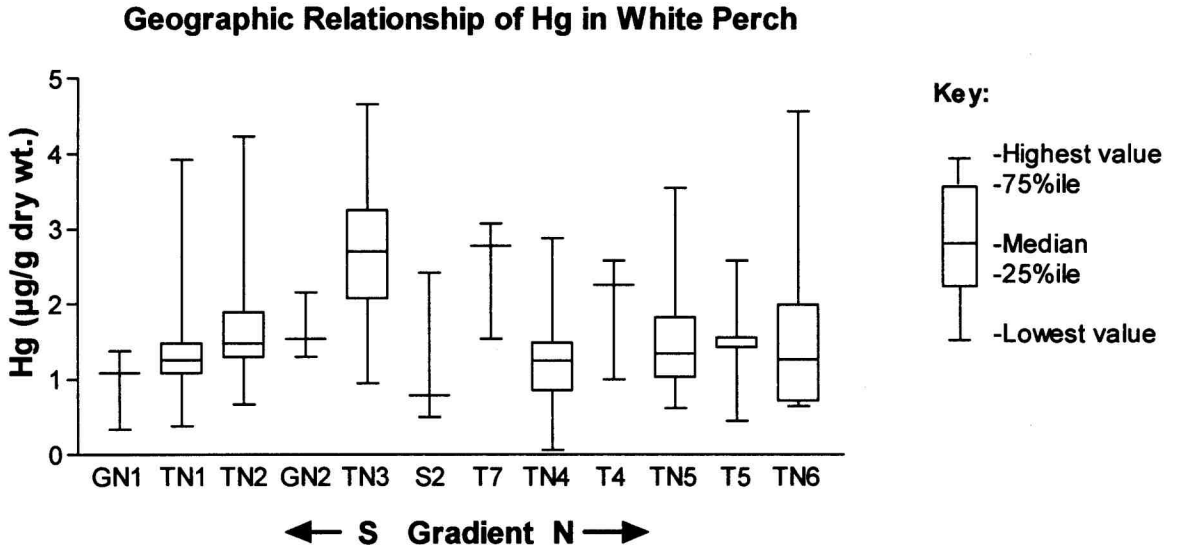


Figure 3. The north-south distribution of Hg burdens is demonstrated in this 'box and whiskers plot'. The total area covered by this graph is from Gill Net 1 at River Mile (RM) 2.7 to Trap Net 6 at RM 12.5. Data without boxes are those for which only a few fish were found at each of those sites (thus, not enough data was available to generate all the predictions available in this type of graph).

The geographic distribution of Hg shows that the highest amounts were in the area opposite Harmon Cove (TN3), but this interpretation may be spurious since most of these fish were caught in June, near the height of the curve shown in Figure 2. The site expected to be highest was the Berry's Creek Canal, since this drains an area with three Superfund areas, one of which is infamous for Hg. Unfortunately, only three fish were caught there, so that result is inconclusive. Conversely, the relatively low levels at Trap Net 4 (TN4), near Mill's Creek, may be similarly biased because these were all caught in the colder part of the year (October 25, December 11 and March 6). Changes in Hg burden in relation to season were previously reported in mummichogs (Weis et al, 1986). It would have been appropriate to have fish from each sampling site year-round, but the fish were not always available at each site. It is not known how much 'site fidelity', i.e., staying in one area, white perch have, or to what extent they school and migrate up and down stream, other than their spawning runs into fresher water in the springtime. What is conclusive from our data is that a fish with an unacceptable Hg level can be caught anywhere on the river.

3.1.1.4. Methylmercury (MeHg).

Twelve samples (8 white perch, 4 crab claws) were sent to the Univ. of Georgia Skidaway Institute for analysis. Typically, fish have virtually all of their Hg in methylated form, but only small fractions of our Hg levels were found to be methylated. The meHg levels were 0.048 ± 0.027 µg/g for the fish and 0.079 ± 0.083 µg/g for the crab claw meat. These were 11% and 6.7% of the total Hg encountered in these species, respectively. Typically, meHg as a percentage of total Hg, is close to 100% in fish (Bloom, 1992), including HMD mummichogs (Weis et al,

1986), so this was surprising. Our quality control and theirs were both 'right on' for analysis of Hg in the CRMs, so the data are reliable. However, this issue certainly needs to be revisited.

3.2 Organic contaminants

The target organic analytes were PCBs, DDT and its metabolites (collectively = DDXs), and chlordanes. The specific congeners for these three groups were all quantified, and these are in Dr. Ashley's report. The FDA action levels for fish consumption are based on the total congeners of each group, and that will be discussed here. Thirty white perch, 6 silversides composites and 9 mummichog composites were analyzed. The size and seasonal relationships for PCB levels, and the relationship to Hg levels in the same fish are illustrated in Figure 4.

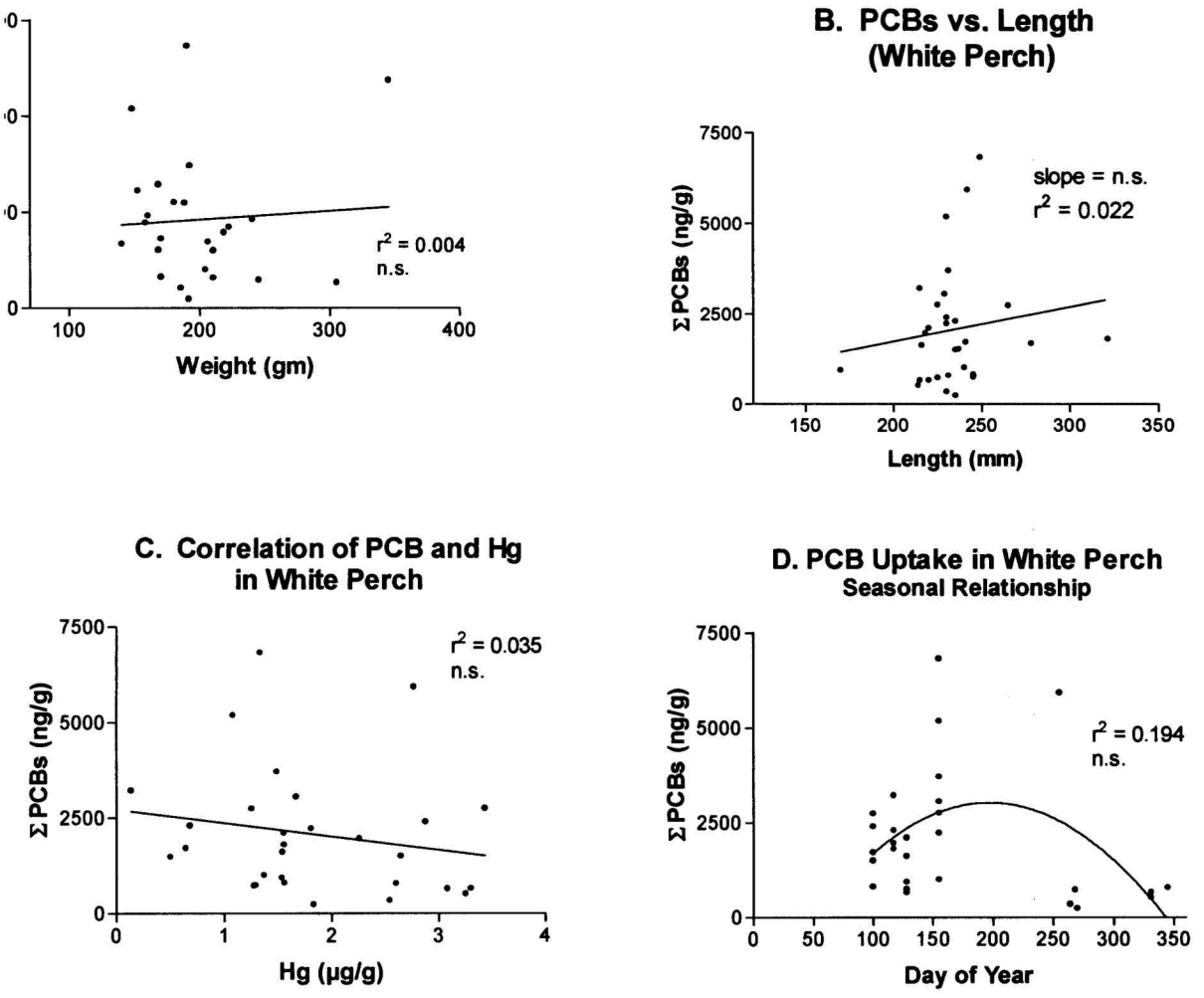


Figure 4. The correlations of total PCBs (ΓPCBs) to length, weight, mercury burdens and to season are shown here. None of the correlations are significant.

None of these correlations are significant, although there is a trend toward seasonal correlation, as with Hg. Many more specimens would have to be analyzed to demonstrate a seasonal correlation.

The FDA action level for PCBs in food is 2000 ng/g wet weight. This was met or exceeded in 12 of the 30 white perch (40%). The distribution of these 12 white perch covers the entire length of that part of the Hackensack River that was surveyed. None of the forage fish exceeded the action level for PCBs.

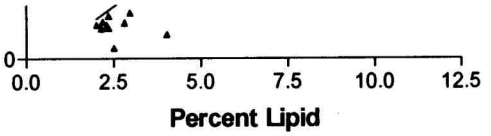
King et al (2004) measured PCB levels in fish from 14 tributaries within the Chesapeake Bay watershed and correlated their findings with the amount, type and distribution of developed land in each location. Total PCBs (ΓPCBs) in white perch were greatest in relation to the most intense development. In areas with high levels of development, the researchers found that the farther the development was from the water, the lower the contaminant levels were in the fish they sampled. An index of development inversely proportional to distance from the shoreline gave the highest correlation to ΓPCBs in white perch: $r^2 = 0.99$. King et al concluded that more than 4% total residential/commercial development in the watershed would predict exceeding the USEPA (1999) guideline of 52 ng/g ΓPCBs for more than one meal per month. It is not surprising, therefore, to find high PCBs in the HMD, considering the high proportion and history of development in the area. All of our white perch exceeded this USEPA (1999) one-meal-per-month guideline, the lowest being 242 ng/g - nearly five times the guideline.

The action level for DDXs is 5000 ng/g wet weight. None of the white perch met or even approached that level; the highest was 2159 ng/g, and most were less than one-tenth of the action level. None of the forage fish exceeded that level, either. There is a very hot spot for DDXs in the lower Passaic River, but this problem apparently does not extend into the Hackensack (Rosman, 2004).

The action level for total chlordanes is 300 ng/g wet weight. One of the white perch exceeded that with 304 ng/g. Most were substantially lower, even into the single digits. None of the forage fish exceeded that level.

3.2. Lipid normalization.

When analyzing organic contaminants in tissues, it is sometimes useful to correlate the chemical burdens with the total lipid in the tissues. This shows the relationship of the fat-soluble contaminants with the state of health of the fish (as suggested by their fattiness). This is illustrated in Figure 5, below, for the white perch.



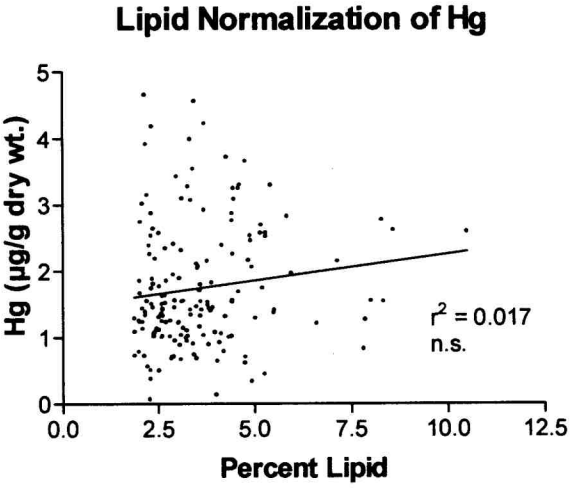


Figure 5. Correlation of PCB (left) and Hg (right) burdens in white perch with the total lipid content of the muscle tissue.

Thus, it can be seen that the uptake of PCBs, but not Hg, correlates well with the amount of lipid in the muscle tissue.

4. Conclusions and Recommendations

The safety (health risk) of Hackensack River fish and crabs has been assessed. There were too few crabs and carp to reach logical conclusions. The white perch collection, on the other hand, was large enough to enable valid conclusions.

It is suggested that white perch are not to be considered edible. The Hg level exceeded the 'one meal per month' action level of 0.47 μg/g wet weight (ppm) in 32% of our catch, and 2.5% exceeded the 'no consumption at all' level of 1 μg/g. A more refined analysis of the data shows that the larger fish represent greater risk Furthermore, the warmer months, when more recreational fishing takes place, may present the greater risk for Hg, as well. Another reason for avoiding white perch is the PCB contamination, since 40% of these fish exceeded the FDA action level for this class of compound and all exceeded the USEPA guideline of no more than one meal/month (USEPA, 1999). In fact, nearly all were 10 times that advisory level!

Brown bullheads may be safe to eat in relation to metals. They have not been analyzed for organics, however.

The relationship of sampling site to mercury cannot be demonstrated because of the inability to obtain sufficient numbers of game fish at many sites at all seasons.

Thus, it is recommended that:

1. further sampling of white perch be made in order to determine the migratory patterns of this species.
2. further analysis of *all* game fish be pursued for organics, specifically for PCBs, since only 30 white perch could be assayed at this time, and for dioxins/furans, since these are problematical elsewhere in the Newark Bay complex.
3. 'fingerprinting' of PCBs be performed from current and future data. Calculating the ratios of the specific congeners of PCBs found at the several sites from which sediments were analyzed, and comparing this to the congeners in the fish collected at the same sites, will give some insight into the fish migratory patterns.
4. a similar survey be performed for the natural piscivores resident in the HMD, i.e., birds, including cormorants and the several species of herons and egrets that are present. This will demonstrate the extent of the trophic transfer of Hg and organics in the area.
5. blue crabs be sought again in order to add to the small amount of data we have so far for this desirable edible species.

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