



Mercury and polychlorinated biphenyls in Asian market fish: A response to results from mercury biomonitoring in New York City

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ABSTRACT

In 2004, the New York City (NYC) Health and Nutrition Examination Survey measured the highest blood mercury levels in Asian and foreign-born Chinese demographic groups. Fish consumption was the strongest predictor of exposure. The survey did not inquire about consumption of individual species, and subsequent visits to fish markets serving the Asian community suggested that many popular species lack contaminant data. Our objective was to supplement existing information on contaminants in commercial fish by collecting data on species present in markets serving the Asian community.

We measured total mercury and the sum of 101 polychlorinated biphenyl (PCB) congeners in 282 individual specimens of 19 species or products from retail fish markets in Chinese neighborhoods in NYC. Species were selected based on their volume in the market, and an absence or insufficiency of national data on mercury levels. PCBs were measured because they are also contaminants of concern. All measurements were made on a wet weight basis on whole fillets (with skin) or products (drained of liquid). Mean mercury levels ranged from below the limit of detection (0.004 µg/g) in tilapia to 0.229 µg/g in tilefish. The highest mercury level (1.150 µg/g) was measured in a tilefish specimen, and mercury levels in tilefish increased with the specimen size. Mean PCB levels ranged from 1 ng/g in red snapper to 98 ng/g in buffalo carp. The highest PCB levels were measured in a buffalo carp (469 ng/g) and a yellow croaker (495 ng/g). Species-specific differences in PCB levels accounted for only 6.3% of total variability, in contrast with 39.2% for mercury.

Although we did not measure high mean mercury levels in the species we sampled, frequent consumption of fish with low to moderate levels can also elevate blood mercury. The data we collected can be used to guide fish consumption in Asian communities. However, risk-benefit trade-offs also need to be considered.

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1. Introduction

Fish and shellfish can be an important part of a healthy diet, because they are a source of protein and other essential nutrients. Long-chain omega-3 polyunsaturated fatty acids, which may reduce risk of adverse cardiovascular events and be beneficial to early neurodevelopment (Mozaffarian and Rimm, 2006), are present in fish to varying degrees (Mahaffey et al., 2008).

However, fish may also accumulate methylmercury and polychlorinated biphenyls (PCBs). Methylmercury is a known neurotoxicant that is particularly toxic to the developing nervous

system (National Research Council, 2000). PCBs are classified by the United States (U.S.) Environmental Protection Agency (EPA) and the International Agency for Research on Cancer (IARC) as probable carcinogens, mostly based on animal studies (U.S. EPA, 1997; IARC, 1998). However, at exposure levels that may occur through frequent fish consumption in the general population, neurodevelopmental risks from prenatal exposure may be the more relevant toxicological endpoint (Stewart et al., 2008; Schantz et al., 2003).

In 2004, the New York City (NYC) Department of Health and Mental Hygiene (DOHMH) conducted a Health and Nutrition Examination Survey (HANES) that measured blood mercury concentrations in a representative sample of 1811 adult New Yorkers (McKelvey et al., 2007). The geometric mean (2.73 µg/L) was approximately three times the national HANES estimates for 1999 to 2006 (Caldwell et al., 2009). Asians registered higher blood mercury (4.11 µg/L), with the geometric mean among

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foreign-born Chinese New Yorkers (7.26 µg/L) almost three times that of NYC overall. An estimated 72% of foreign-born Chinese New Yorkers had blood mercury at or above the New York State reportable level (5 µg/L). According to NYC HANES, Asian New Yorkers consumed fish most frequently, with an estimated 19% reporting 20 or more meals of fish or shellfish in the last 30 days, compared with 5.5% of non-Hispanic whites or blacks. Fish consumption was the strongest predictor of elevated blood mercury.

The New York State (NYS) Department of Health (DOH) issues extensive advisories on recreationally caught fish (NYS DOH 2009). However, most fish consumed in the U.S. is commercial (Stern et al., 1996; Sunderland, 2007). To address this issue, the NYC DOHMH developed a brochure to guide frequent consumers in selecting commercial fish species lower in mercury (NYC DOHMH, 2007). The brochure targets pregnant and breastfeeding women and young children—those considered at highest risk (National Research Council, 2000). It lists species that have contaminant data available from the U.S. Food and Drug Administration (FDA) Center for Food Science and Applied Nutrition (CFSAN) (U.S. FDA 2006). However, subsequent visits to fish markets serving the Asian community suggested that many popular species have not been routinely tested by FDA. There was an additional concern that contaminants in commercial fish might differ regionally—as a function of varying local sources for commercial fish, or varying import patterns related to the demographic make-up of consumers (Burger et al., 2004; Burger and Gochfeld, 2006; Sunderland, 2007).

Our aim in conducting the present study was to supplement existing information on contaminants in commercial fish by measuring mercury and PCB concentrations in species present in markets serving the Asian community in NYC. We included PCBs because they are co-occurring contaminants that may also be harmful to the developing nervous system (Stewart et al., 2008; Schantz et al., 2003). The anticipated outcome was an expansion of the information base from which fish consumption guidelines may be drawn. The data we report may be used to support outreach on limiting exposure to mercury in Asian communities where fish consumption is frequent.

2. Materials and methods

2.1. Species selection

We selected fish species for this study by first observing commercial fish market displays in NYC's largest Chinese communities: Chinatown in Lower Manhattan; the vicinity of Sunset Park and Borough Park, Brooklyn; and Flushing, Queens, from March to May, 2007. We focused on the Chinese, since they are the largest Asian population in NYC, and their stores often serve Korean and other Asian subgroups that eat fish frequently. A fisheries biologist (M. Chang) visually identified the species that were present in the largest volume. From among the species identified, we used the following criteria to select species for study: (1) little or no current mercury concentration data were available from the U.S. FDA (U.S. FDA, 2008), (2) mercury levels were considered low, but there was potential for PCB contamination based on ecological considerations, or (3) there were potential changes in commercial waters of origin, due to changes in import patterns since contaminant data were last collected. For example, in 1993, 7% of tilapia imports to the U.S. were from China, compared with 71% in 2009 (National Marine Fisheries Service, 2010). We selected 19 species (fresh or previously frozen fish, or packaged items) for analysis of mercury and PCBs. We opted to collect a fixed sample size of 15 specimens per species or product to facilitate data collection.

2.2. Sample selection

We obtained a list of stores that held permits issued by the NYS Department of Agriculture and Markets to process fish or seafood, or with the words “fish”

or “seafood” in the trade or owner name. We then identified stores located in the top 10% Chinese-populated census tracts, according to the U.S. Census 2000, by geocoding addresses and joining them with census tract boundaries for NYC. For each targeted species, we selected one specimen per store from 4 stores in Manhattan, 5 stores in Brooklyn, and 6 stores in Queens. This distribution of specimens within target species was proportional to the Chinese population size in each borough. We selected stores by taking a borough-stratified, probability proportional to store size (square footage), representative sample. Fish were purchased August–September, 2007, by NYC Health Department staff, assisted by Chinese-speaking students from Hunter College, and accompanied by a fisheries biologist (M. Chang or J. Waldman) to assist in identifying species.

2.3. Tissue preparation

We purchased whole fish, except in cases where specimens were so large that they were sold only as steaks or sections. We shipped specimens in plastic Ziploc® bags to the processing laboratory (Institute for Health and the Environment, University at Albany) on blue ice via an overnight delivery. Fish were wrapped in aluminum foil before they were inserted into plastic bags to avoid plastic-based interferences in measuring PCBs.

All specimens were stored in the processing laboratory at -80°C . When ready for processing, whole fillets, including skin and belly flap, were cut from partially thawed specimens. Edible meat (including hepatopancreas) was removed from 2 or 3 crabs per sampling location to form a crab composite. Non-fish ingredients were removed and liquid was drained from cooked, packaged items. Work was done on a cutting board covered with a fresh piece of Teflon for each specimen. Tissue from whole fish and other specimens was homogenized using a Robot Coupe R6N food processor. All knives and apparatus were rinsed with Alconox soap and tap water, followed by hexane (EMD OmniSolv High Purity) and 5% nitric acid, and allowed to dry between uses. Homogenate was stored at -80°C in glass jars or vials until analysis.

We measured total mercury and PCBs on a wet weight (raw) basis, except for specimens of canned dace and eel and frozen unagi eel, which were previously cooked and seasoned.

2.4. Measurement of total mercury

Total mercury was measured in the Arnason laboratory, Department of Earth and Atmospheric Sciences (University at Albany). Sample aliquots were digested with a mixture of concentrated nitric acid (Alpha Aesar, Environmental Grade) and hydrogen peroxide (FMC Corporation Ultrapure) using a CEM MARS V® microwave digestion oven (EPA method 3052). Samples were diluted to 50 mL with de-ionized water, and total mercury was measured by cold vapor atomic absorption spectroscopy on a Leeman Labs Hydra AA® mercury analyzer with autosampler.

Calibration curves were generated using four mercury standard solutions (0.01–20 µg/L) for each analytical batch of specimens. Calibration was verified using 0.1 and 1.0 µg/L laboratory fortified blank solutions. Recoveries were required to be between 80% and 120%.

Other quality control measures included use of laboratory reagent blanks, sample duplicates, blind duplicates, spiked samples and certified reference material (NIST 1566b Oyster Tissue, DORM 3 Dogfish muscle, and DOLT 3 Dogfish liver). Method precision was assessed by analyzing 38 sample duplicates which yielded an average relative percent difference of 6%. Blind duplicates averaged 8.2% relative percent difference. Mean recoveries of certified reference material and spiked samples were both 88%. The limit of detection (LOD) was 0.004 µg/g in sample. For statistical calculations, values below the LOD were assigned the limit of detection divided by the square root of 2.

2.5. Measurement of total PCBs

The sum of 101 PCB congeners was measured by the Institute for Health and Environmental Sciences (University at Albany), using a method based on EPA method 8082 (DeCaprio et al., 2000).

Specifically, PCBs were extracted from homogenized fish tissue that was mixed with anhydrous sodium sulfate, 1:1 hexane (EMD OmniSolv High Purity):acetone (B&J ACS/HPLC grade), and an internal standard (PCB IUPAC #104 in hexane) and a surrogate standard (PCB IUPAC# 125 and # 192) obtained from AccuStandard®, Inc. The mixture was blended to a uniform consistency using a tissue miser. After extraction, samples were subjected to a Florisil® column cleanup procedure to remove polar lipids and other interferences.

The concentrations of 101 PCB congeners or congener groups (Table 1) were quantified using a Hewlett-Packard Model 6890 gas chromatograph equipped with dual Model 7638 autosamplers, capillary columns (configured in parallel), and Hewlett-Packard ^{63}Ni micro-EC detectors. The method described here allows simultaneous quantification of concentrations of three organochlorine pesticide contaminants (hexachlorobenzene (HCB), 1,1-dichloro-2,2-(p-chlorophenyl)ethylene (p,p'-DDE), and Mirex).

Table 1

Congeners and practical quantitation limits (PQL) used to measure total PCB concentrations in the NYC Asian market fish study.

Congener	IUPAC #	PQL (ng/g)	Congener	IUPAC #	PQL (ng/g)	Congener	IUPAC #	PQL (ng/g)
2	1	1.97	236/4	64	0.12	2356/236	179	0.16
4	3	1.03	23/23	40	0.06	2345/24	137	0.03
2/2+3	4+2	1.68	245/3	67	0.01	2346/236	176	0.02
2/6	10	0.04	235/4	63	< 0.01	234/235	130	0.05
24	7	0.15	245/4	74	0.08	236/345+234/245+2356/34	164+163+138	0.95
25	9	0.11	25/34	70	0.25	2346/34	158	0.09
2/3	6	0.35	24/34	66	0.09	2345/23	129	0.03
2/4	8	1.04	236/25	95	0.5	2356/245	187	0.35
26/2	19	0.08	236/24	91	0.06	2346/245	183	0.1
3/4	13	0.21	23/34	56	0.03	234/234	128	0.11
25/2	18	0.66	235/25	92	0.14	23456/25	185	0.04
4/4	15	0.27	236/23	84	0.16	2345/236	174	0.33
24/2	17	0.26	235/24+245/25	90+101	0.85	2356/234	177	0.18
236+26/3	24+27	0.05	245/24	99	0.25	2346/234	171	0.11
26/4+23/2	32+16	0.43	235/23	83	0.03	2345/34	156	0.09
245	29	0.01	245/23	97	0.17	2346/2356	201	0.06
25/3	26	0.11	234/25	87	0.33	2345/235	172	0.06
24/3	25	0.06	236/236	136	0.4	2345/245	180	0.61
25/4	31	0.6	236/34	110	0.68	23456/236	200	0.03
24/4	28	0.62	34/34	77	0.58	2345/234	170	0.23
34/2	33	0.22	2356/25	151	0.29	23456/34	190	0.05
25/26	53	0.1	2346/25	144	0.05	2345/2356	199	0.14
24/26	51	0.03	2356/24+235/34	147+109	0.04	23,456/245	203	0.07
23/4	22	0.24	345/24+236/245	123+149	0.71	2345/2346	196	0.06
236/2	45	0.09	245/34	118	0.48	23,456/234	195	0.06
23/26	46	0.04	2356/23	134	0.06	2345/2345	194	0.13
25/25	52	0.63	2345/4	114	0.02	23456/2345	206	0.04
24/25	49	0.24	235/245	146	0.13			
24/24+236/3	47+59	0.1	245/245	153	0.79			
23/25	44	0.43	234/236	132	0.38			
23/24	42	0.09	234/34	105	0.16			
26/34	71	0.07	2345/25	141	0.25			

A 5-point calibration curve was generated every two to three weeks, or 70–100 samples, as needed, using a stock 1:1:1:1 mixture of Aroclors 1016, 1221, 1254, and 1260 (20 µg/mL of each in hexane), fortified with PCB IUPAC #77 (3,4,3',4'-tetrachlorobiphenyl), HCB, *p,p'*-DDE, and Mirex (2, 1, 1, and 1 µg/mL, respectively), obtained from Accustandard[®] Inc. The stock mixture was diluted to form working solutions that ranged from 5–200 ng/mL. Response factors for each peak were required to be no more than 20% relative percent difference from expected values for the calibration to pass. Calibration was verified using the same stock mixture at a concentration of 20 ng/mL. At least 80% of congeners were required to fall within 15% of the expected value for the batch to run.

Certified reference material (CRM) consisted of Hudson River white perch, obtained from the National Institute for Standards and Technology. CRMs were run in a blind fashion for every 20 unknown determinations. Recoveries were between 94% and 108% for the sum of the 38 congeners that were measured by both laboratories.

Other quality assurance and control measures included use of a surrogate standard, method blanks, and duplicate analysis. Surrogate standard recoveries were required to be within 60–130%. Hexane method blanks were treated identically as unknown samples and carried through from extraction to end. If any congener concentrations in the blanks were above detection limits, the extraction batch was rejected and re-extracted. Method precision was assessed by analyzing 9 duplicate samples, which yielded an average relative percent difference of 6%.

Limits of detection were based on the practical quantitation limits (PQL). PQLs were determined by preparing eight identical samples containing 5 g homogenized fish tissue, spiked with 1 mL of the same stock mixture used for calibration, at a concentration of 0.2 ng/mL. The standard deviations for each congener (or congener group) were calculated and multiplied by seven. The PQLs for each congener (or congener group) ranged < 0.01–1.97 ng/g (Table 1). We assigned a value of zero wherever a congener fell below the PQL. We conducted a sensitivity analysis, in which we also used the instrument detection limits (IDL) (calculated by taking three times the standard deviations from stock mixture samples that have not gone through a tissue preparation process) to sum total congeners. The IDLs ranged < 0.01–0.017 ng/g.

2.6. Statistical analysis

We calculated species-specific arithmetic means, which are typically used to calculate allowable meal frequencies. We rounded the sum of PCB congener

concentrations to the nearest ng/g, which in some cases resulted in concentrations of “0” ng/g. We provide the 95% confidence intervals around the geometric means as an indication of the precision of our estimates of central tendency, since contaminant distributions were more log-normal than normal. We assessed correlations between mercury and total PCBs, and between total fish length and contaminant concentration, using Spearman rank correlation coefficients, to avoid the need to assume a normal distribution. Intraclass correlation is defined as the percent of total variability that is “between-species.”

2.7. Calculating meal frequencies

We used the U.S. EPA reference doses (RfD) for methylmercury (0.1 µg/kg bodyweight/day) (National Research Council, 2000) and PCBs (non-cancer outcomes) (0.02 µg/kg bodyweight/day) (U.S. EPA, 2000) to calculate allowable meals per week. We based our calculations on a 60 kg woman who eats 6-oz (170 g) serving sizes. We used total mercury as a proxy for methylmercury concentration. Using these inputs, we created consumption categories of up to five meals per week (mercury concentration up to 0.049 µg/g); up to two meals per week (mercury concentrations up to 0.126 µg/g); and up to one meal per week (mercury concentrations up to 0.260 µg/g). Similar categories for total PCB concentrations used thresholds of 10, 25 and 52 ng/g, respectively. We note that different bodyweight and serving size inputs are likely to produce different category thresholds.

3. Results

3.1. Characteristics of selected specimens

We measured total mercury on 282 specimens, and we measured 101 PCB congeners on 196 specimens, representing 19 different species/species groups or fish products (Table 2). Sleeper, blackfish, and most tilapia and hybrid striped bass specimens were purchased live from a tank. Dace (mud carp) was purchased cooked and seasoned in canned form; eel

Table 2

Scientific names of species or species groups selected for the NYC Asian market fish study.

Common name	Species	Habitat
Bass, Black Sea	<i>Centropomus striata</i>	Marine
Bass, Hybrid Striped	<i>Morone saxatilis x chrysops</i>	Freshwater (farmed)
Blackfish (Tautog)	<i>Tautoga onitis</i>	Marine
Carp, Bighead	<i>Aristichthys nobilis</i>	Freshwater
Carp, Buffalo (Buffalofish)	<i>Ictiobus spp.</i>	Freshwater
Crab, Blue	<i>Callinectes sapidus</i>	Estuarine
Croaker, Yellow	<i>Pseudosciaena polyactis</i>	Marine (farmed)
Cutlass (Beltfish)	<i>Tentoriceps sp.</i> <i>Trichiurus sp.</i> <i>Lepturacanthus sp.</i>	Marine
Dace (Mud Carp), Canned	<i>Cirrhinus molitorella</i>	Freshwater
Eel, Canned (or Frozen “Unagi”)	<i>Anguilla japonica</i>	Estuarine (farmed)
Flatfish (Rex, Dover and Gray Sole; Summer, Winter and Windowpane Flounder; American Plaice)	<i>Errex zachirus</i> <i>Microstomus pacificus</i> <i>Glyptocephalus cynoglossus</i> <i>Paralichthys dentatus</i> <i>Pseudopleuronectes americanus</i> <i>Scophthalmus aquosus</i> <i>Hippoglossoides platessoides</i>	Marine
Mackerel, Spanish	<i>Scomberomorus maculatus</i>	Marine
Pompano, Golden	<i>Trachinotus blochii</i>	Marine (farmed)
Pompano, White (or Butterfish)	<i>Pampus spp.</i>	Marine
Porgy	<i>Peprilus alepidotus</i> <i>Stenotomus chrysops</i> <i>Pagrus pagrus</i>	Marine
Sleeper	<i>Dormitator maculatus</i>	Freshwater (farmed)
Snapper, Red or Vermillion	<i>Lutjanus campechanus</i> <i>Rhomboplites aurorubens</i>	Marine
Tilapia	<i>Oreochromis spp.</i>	Freshwater (farmed)
Tilefish	<i>Lopholatilus chamaeleonticeps</i>	Marine

specimens were purchased cooked and seasoned in both canned and frozen form. The remaining species were purchased mostly as whole fish, on ice, some having been previously frozen. Some carp and tilefish specimens were too large to purchase whole, and in these instances, we purchased steaks or sections, instead of whole fish.

In some cases, specimens purchased under a single common name represented several genera. For example, cutlassfish comprised three; “flatfish” comprised seven; and it was not possible to distinguish the genera that include red and vermillion snapper with a high degree of certainty.

We were unable to collect reliable data on country of origin for most specimens, with the exception of prepared eel and dace products, which were all labeled imported from China or Taiwan. All yellow croaker specimens that were previously frozen and wrapped with loose packaging were labeled from China.

3.2. Contaminant levels in selected species

Mean mercury concentrations ranged from below the LOD (0.004 µg/g) in tilapia to 0.229 µg/g in tilefish (Table 3). The geometric mean concentrations were similar to medians (data not shown). Several species contained outlier mercury levels: hybrid striped bass specimens were below 0.045 µg/g, except for one

specimen that contained 0.167 µg/g; tilefish specimens were below 0.38 µg/g, except for one specimen that contained 1.15 µg/g, which is over the U.S. FDA action level of 1.0 µg/g (1 ppm).

Mean PCB concentrations ranged 1–98 ng/g, and the within species variability was high. Based on intraclass correlation, only 6.3% of total variability in PCB concentration is explained by species differences, in contrast with 39.2% of mercury variability. Arithmetic mean PCB concentration across all specimens was 32 ng/g. The highest concentrations were measured in a buffalo carp (469 ng/g) and a yellow croaker (495 ng/g). Several species contained notable outlier PCB levels. Blue crab PCB levels were under 10 ng/g in all but two specimens, which measured 257 and 39 ng/g. These specimens were very low in mercury (< 5 ng/g), and they had no distinguishing characteristics. Flatfish PCB levels were less than 22 ng/g in all but two specimens, which were 90 and 260 ng/g. In general, the geometric means (which minimize the contribution of high, outlier values) were considerably lower than arithmetic means. We combined canned and frozen eel results in PCB analyses because: they are presumed to represent the same species, and combining them resulted in a more robust sample size. Use of the IDL rather than PQL as the limit of detection resulted in PCB concentrations ranging 0–8 ng/g higher in individual specimens and up to 4 ng/g higher for species-specific averages (data not shown).

Correlation between mercury and PCBs was mostly low within species. Only buffalo carp generated a Spearman R^2 higher in magnitude than 50% (57%), and with a p -value less than 0.05. Buffalo carp is low in mercury but higher in PCBs.

Fish length and weight showed little variability within most species for sale at markets, so we were unable to assess correlations between these characteristics and contaminant levels. Tilefish length, however, varied from 16 to over 34 in. We were able to reliably measure total length in seven specimens (other specimens were purchased as steaks or cut before we had an opportunity to measure them accurately), and length explained almost all the variability in mercury concentration among these (Spearman $R^2=90\%$) (Fig. 1). We were unable to measure the length of a tilefish specimen that was without a doubt the largest of all the fish we sampled. We made qualitative note of its size before it was cut for sale. This specimen turned out to have the highest mercury concentration of all the specimens measured (1.150 µg/g). In contrast, length explained very little of the variability in PCB concentrations (Spearman $R^2=4\%$), which ranged 2–132 ng/g.

We have not presented organochlorine pesticide concentrations, because almost all were very low. However, it may be noteworthy that outlier concentrations of each of the measured contaminants were in two yellow croaker specimens (highest levels of p,p'-DDE, HCB, and Mirex were 69, 4.7, and 6.1 ng/g, respectively).

3.3. Using contaminant data to calculate allowable meal frequencies

Using the U.S. EPA RfDs for methylmercury (0.1 µg/kg body-weight/day), and PCBs (non-cancer outcomes: 0.02 µg/kg body-weight/day), we calculated the number of 6-ounce (170 g) servings that a 60 kg woman could theoretically eat per week (Tables 3 and 4). Only four species had average mercury levels high enough to be limited to one meal per week, compared with 11 species that were limited to one meal per week based on PCB concentration. PCBs were more restrictive than mercury in all but two species (sleeper and Spanish mackerel). A cross-tabulation of allowable meals per week based on restrictions for both methylmercury and PCBs allows only dace,

Table 3
Mercury concentrations in whole fillets of fish (skin on) and fish products ($\mu\text{g/g}$ wet weight) in ascending order of arithmetic means, NYC Asian Market Fish Study, 2007.

Species/Product	Arithmetic mean	Geometric mean (95% CL)	Minimum	Maximum	No. of samples	Allowable servings/wk ^a
Tilapia	< LOD ^{**}	< LOD ^{**}	< LOD ^{**}	< LOD ^{**}	15	Up to 5/wk
Pompano, White	0.010	0.006 (0.003, 0.010)	< LOD ^{**}	0.039	15	Up to 5/wk
Crab, Blue	0.017	0.010 (0.005, 0.019)	< LOD ^{**}	0.052	14	Up to 5/wk
Dace, Canned ^b	0.022	0.018 (0.012, 0.027)	< LOD ^{**}	0.052	15	Up to 5/wk
Carp, Bighead	0.023	0.014 (0.008, 0.027)	< LOD ^{**}	0.092	15	Up to 5/wk
Bass, Hybrid Striped	0.028	0.018 (0.011, 0.028)	0.005	0.167	15	Up to 5/wk
Pompano, Golden	0.031	0.026 (0.017, 0.038)	< LOD ^{**}	0.068	15	Up to 5/wk
Croaker, Yellow	0.033	0.032 (0.027, 0.037)	0.021	0.068	15	Up to 5/wk
Snapper, Red or Vermillion	0.035	0.030 (0.021, 0.041)	0.008	0.082	15	Up to 5/wk
Carp, Buffalo	0.042	0.032 (0.020, 0.050)	0.007	0.079	15	Up to 5/wk
Cutlass	0.056	0.043 (0.028, 0.066)	0.013	0.149	15	Up to 2/wk
Sleeper	0.060	0.051 (0.033, 0.081)	< LOD ^{**}	0.091	15	Up to 2/wk
Porgy	0.094	0.087 (0.070, 0.108)	0.048	0.181	15	Up to 2/wk
Flatfish (Flounder/Sole)	0.095	0.071 (0.046, 0.109)	0.024	0.308	15	Up to 2/wk
Eel, Frozen Unagi ^b	0.097	0.091 (0.069, 0.121)	0.059	0.181	8	Up to 2/wk
Bass, Black Sea	0.105	0.097 (0.076, 0.123)	0.044	0.200	15	Up to 2/wk
Mackerel, Spanish	0.147	0.133 (0.097, 0.182)	0.039	0.244	13	Up to 1/wk
Blackfish	0.158	0.145 (0.114, 0.185)	0.089	0.298	14	Up to 1/wk
Eel, Canned ^b	0.181	0.144 (0.074, 0.282)	0.029	0.391	8	Up to 1/wk
Tilefish	0.229	0.163 (0.107, 0.247)	0.063	1.150	15	Up to 1/wk

^a Based on a 60 kg person ingesting the U.S. EPA RfD (0.1 $\mu\text{g/kg/day}$); meal size=6 oz (170 g).

^b Measurements made on cooked and seasoned items, drained.

^{**} Limit of detection (LOD)=0.004 $\mu\text{g/g}$.

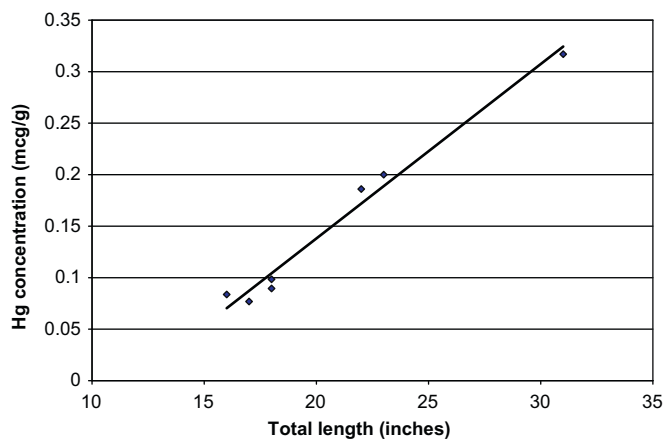


Fig. 1. Mercury concentration ($\mu\text{g/g}$) by whole body length (inches) in whole tilefish specimens.

red snapper and white pompano to be eaten up to five times per week (Table 5).

4. Discussion

4.1. Mercury and PCB levels in selected species

In response to a NYC-wide survey that found that frequent fish consumption in Asian communities was associated with elevated blood mercury levels (McKelvey et al., 2007), we measured mercury and PCBs in 19 fish species or products that are commonly sold in Asian retail markets, and for which contaminant data are lacking. Although we did not measure high mean mercury levels in the species we sampled, frequent consumption of fish with low to moderate levels can also elevate blood mercury.

The total mercury and PCB levels that we measured were comparable to levels that have been reported by the U.S. FDA CFSAN (U.S. FDA, 2008) and published studies (Domingo and

Bocio, 2007; Bocio et al., 2007; Burger and Gochfeld, 2005). Nonetheless, a 60 kg woman who consumed five 6-oz servings per week of a species in the moderate mercury concentration range (0.100 $\mu\text{g/g}$) would be ingesting about twice the RfD. PCB levels in most of the species we tested were similar in range to levels measured in wild and farmed salmon (Shaw et al., 2006; Hites et al., 2004; Easton et al., 2002). However, several of our species had average PCB levels that were substantially higher than those that have been reported in farmed salmon (Fig. 2). Farmed salmon has been singled out in recent years as a potentially important source of PCB exposure (Hites et al., 2004), perhaps because it is commonly consumed by the general population. However, in populations that eat fish as often as daily, other species may be equally if not more important sources.

We measured mercury in several species (tilapia, Spanish mackerel, flatfish, and tilefish), for which data are provided by U.S. FDA (U.S. FDA, 2008). The mean mercury level in NYC flatfish specimens was twice as high, which may be a result of sampling different mixes of distinct species, since both sources of data combine various species into a single estimate. It may also be the result of regional differences in locally caught fish, since our results are within the range measured in flatfish specimens caught off the coast of New Jersey (Burger et al., 2009). Average mercury measured in Spanish mackerel and tilefish for the NYC study were similar to FDA estimates for specimens from the Atlantic Ocean, which are substantially lower than FDA specimens from the Gulf of Mexico. The strong correlation we measured between total length and mercury level in tilefish suggests that fish size may be influencing the differences. Greater contamination in the Gulf of Mexico is another potential explanation that we did not explore (Interagency Working Group on Methylmercury, 2004). The NYC study corroborated the very low mercury levels in tilapia, also reported by FDA.

4.2. Application of RfDs for Hg and PCBs

We used U.S. EPA RfDs for methylmercury and PCBs to interpret contaminant levels in terms of allowable 6-ounce fish servings per week for a 60 kg woman. We note that other serving

Table 4

Concentrations of the sum of 101 PCB congeners in whole filets of fish (skin on) and fish products (ng/g wet weight) in ascending order of arithmetic means, NYC Asian Market Fish Study, 2007.

Species/product	Arithmetic mean	Geometric mean (95% CI)	Minimum	Maximum	No. of samples	Allowable servings/wk ^a
Snapper, Red or Vermillion	1	0 (0, 1)	0	6	11	Up to 5/wk
Dace, Canned ^b	7	3 (1, 11)	0	17	11	Up to 5/wk
Pompano, White	8	3 (1, 10)	0	44	10	Up to 5/wk
Sleeper	10	0 (0, 8)	0	78	10	Up to 5/wk
Cutlass	12	4 (1, 12)	0	62	11	Up to 2/wk
Tilapia	13	7 (3, 17)	1	38	10	Up to 2/wk
Bass, Black Sea	18	14 (9, 23)	5	43	11	Up to 2/wk
Mackerel, Spanish	23	16 (7, 37)	1	50	9	Up to 2/wk
Crab, Blue	24	1 (0, 6)	0	257	13	Up to 2/wk
Pompano, Golden	30	18 (6, 53)	2	89	8	Up to 1/wk
Tilefish	31	21 (13, 36)	8	131	12	Up to 1/wk
Eel, Canned or Frozen ^b	32	2 (0, 93)	0	150	9	Up to 1/wk
Flatfish (Flounder/Sole)	32	8 (3, 20)	2	260	13	Up to 1/wk
Blackfish	35	17 (6, 51)	1	118	9	Up to 1/wk
Bass, Hybrid Striped	50	44 (30, 65)	20	88	10	Up to 1/wk
Carp, Bighead	50	2 (0, 20)	0	279	9	Up to 1/wk
Porgy	56	28 (9, 87)	1	124	10	Less than 1/wk
Croaker, Yellow	86	27 (9, 76)	2	495	11	Less than 1/wk
Carp, Buffalo	98	48 (19, 124)	8	469	9	Less than 1/wk

^a Based on a 60 kg person ingesting the U.S. EPA RfD (0.02 µg/kg/day); meal size = 6 oz (170 g).

^b Measurements made on packaged cooked and seasoned items, drained.

Table 5

Cross tabulations of allowable 6-oz (170 g) servings per week taking into account U.S. EPA RfD values for methylmercury (0.1 µg/kg/d) and PCBs (0.02 µg/kg/d).

		Allowable meals ^a based on average mercury levels			
		Up to 5/week	Up to 2/week	Up to 1/week	Less than 1/week
Allowable meals ^a based on average PCB levels	Up to 5/week	Dace, Canned Snapper, Red Pompano, White	Sleeper		
	Up to 2/week	Tilapia Crab, Blue	Bass, Black sea Cutlass	Mackerel, Spanish	
	Up to 1/week	Pompano, Golden Carp, Bighead Bass, Hybrid Striped	Unagi, Frozen Flatfish	Blackfish Tilefish Eel, Canned	
	Less than 1/week	Croaker, Yellow Carp, Buffalo	Porgy		

^a Assuming a bodyweight of 60 kg and 6-oz (170 g) serving sizes.

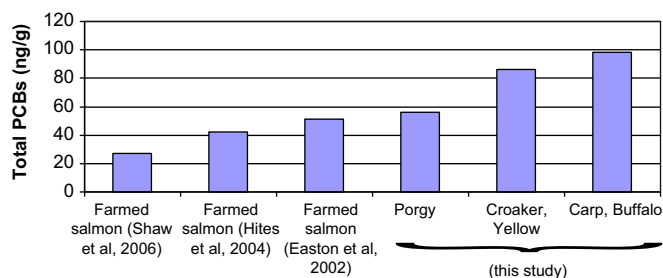


Fig. 2. Comparison of NYC Asian market fish species with the highest mean total PCB levels with studies of farmed salmon.

sizes or bodyweights may be assumed when local data suggest more relevant inputs. A 6-ounce serving size is used by the U.S. FDA/EPA fish consumption guidelines (U.S. FDA/EPA 2004), and it is in agreement with portion sizes measured among fish consumers in neighboring New Jersey (Stern, Korn et al., 1996). We assumed a 60 kg bodyweight since it was the approximate median among Asian women 20–50 years old residing in NYC, as estimated through the NYC HANES (McKelvey W, personal communication). We did not categorize species for consumption

frequency greater than five servings per week, because it would have required ranking species based on differences of several parts per billion in average contaminant levels. Our laboratory methods are not likely to be able to make such small distinctions with an adequate precision or certainty. We recognize, however, that there are population subgroups that eat fish more frequently than five times per week, and these groups also need guidance on how to limit exposure to contaminants.

Using data on methylmercury and PCB levels simultaneously for the purpose of calculating fish consumption guidelines may not be feasible in practice because of the lack of PCB data on most commercial species. Steering consumers away from species simply because data are available gives the potentially false impression that unmentioned species are low in PCBs. Furthermore, the variability we observed in PCB concentrations within species supports the notion that water of origin may be more important in predicting PCB levels than species in a commercial market setting, and it argues for stricter enforcement of the country of origin labeling law, in conjunction with monitoring for PCBs in commercial bodies of water. Agricultural or industrial run-off impacting coastal aquaculture in China may be a source of higher organochlorine levels measured in Chinese-farmed yellow croaker (Barboza, 2007).

We opted to use the PCB RfD for non-cancer outcomes, because this is the dose that the U.S. EPA recommends for calculating an allowable number of fish meals per month or week (U.S. EPA, 2000). We also took into account the fact that the primary impetus for this study was to collect information that could be targeted to pregnant woman and young children, with an exposure limited to the prenatal and early childhood periods. Using a cancer slope factor typically results in more restrictive meal frequencies (Huang et al., 2006). A limitation of both non-cancer and cancer-related reference doses is that they are based on toxicology studies of congener mixes that may not be generalizable to the patterns of exposure that occur today (U.S. EPA, 2000).

Even though the non-cancer PCB RfD results in less restrictive meal advice than cancer-related reference doses, it is still more restrictive than the methylmercury RfD for all but two species (Table 5). However, we did not make any assumptions about PCB loss due to cooking or removal of fat and skin, which would probably have resulted in greater permitted consumption frequency (Wilson et al., 1998; Sherer and Price, 1993), although not all studies show a consistent, subsequent reduction in contaminant (Shaw et al., 2006). We measured specimens with skin and the fatty belly flap, because that is how Chinese and other Asians (our target groups) consume fish.

The allowable weekly consumption categories we have presented in Table 5 are for illustration purposes only. Their value may be limited in communities that eat fish almost daily, since they have not taken into account the trade-offs between risks and benefits of fish in the diet (Ginsberg and Toal, 2009). Fish contain long-chain omega-3 fatty acids and other nutrients, which may have beneficial effects on a developing child (Nesheim and Yaktine, 2007). Without considering benefits in conjunction with risks, avoiding or limiting fish could result in a net detrimental impact on public health by virtue of the less healthy food substitutions that may occur when traditional diets are given up in favor of “Westernized” diets.

For frequent fish consumers, Table 5 is also limited in that it does not provide information on how to select combinations of fish species that fall within different contaminant categories. For example, if a consumer eats one fish meal from the ‘Up to 2/week’ category, the tabular format does not convey information about how many meals a consumer can eat from the ‘Up to 5/week’ category. Online mercury calculators are better suited to provide this kind of information than static tables (see, for example, www.gotmercury.org—advanced mode).

4.3. Limitations in laboratory measurements

It is generally thought that almost all mercury in fish is of the more toxic organic methylmercury form, but some studies have reported lower amounts. A wide range of percentages (30–79%) were found in 37 samples of canned tuna (Forsyth et al., 2004). The U.S. EPA RfD applies specifically to methylmercury and neurodevelopmental outcomes. Inorganic forms of mercury are not well absorbed by the gastrointestinal tract and – when ingested – are likely to be considerably less toxic to the developing nervous system (National Research Council, 2000). For the purpose of calculating consumption guidelines, we have made the conservative assumption that all measured mercury is in the form of methylmercury.

There are several sources of error that may arise when measuring PCB concentrations in fish tissue. Co-occurring lipid compounds are a frequent source of interferences. It may be difficult to compare PCB measurements across laboratories because of differences in analytic methods and number of

congeners quantified. The congeners we quantified probably capture more than 95% of the PCBs routinely or sporadically present in biota, so our results should be comparable to other studies that have also quantified the large majority of congeners (A.P. DiCaprio, personal communication, November 30, 2009). In contrast, studies that have measured only 20–30 of the most frequently occurring congeners may be biased toward reporting lower levels of PCBs. Treatment of values at the limits of detection may be a source of discrepancies across studies as well. We noted up to 4 ng/g higher average PCB concentration by species when we used an IDL, which was sufficient to shift several species into an adjacent, less frequent consumption category.

4.4. Public health considerations

Reducing exposure to mercury and PCBs from fish consumption is a complex problem that requires simultaneous consideration of health benefits of fish as well as the added risks of substituting foods that may be even more harmful to health. In communities where fish consumption is very frequent, it is particularly important to steer the highest risk groups—such as reproductive age women and young children—in the direction of selecting fish that are lower in contaminants, since total exposures can mount quickly. The data we collected can be used for this purpose in Chinese and other Asian communities that may be at risk for mercury or PCB exposure, due to frequent fish consumption. Detailed data from this study can be downloaded from our website: www.nyc.gov/health/asianmarketfish

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References

- Barboza, David, 2007. In China, Farming Fish in Toxic Waters. *New York Times* December 15, 2007. Available at: http://www.nytimes.com/2007/12/15/world/asia/15fish.html?_r=1&ref=david_barboza. (accessed 14 June 2010).
- Bocio, A., Domingo, J.L., Falco, G., Llobet, J.M., 2007. Concentrations of PCDD/PCDFs and PCBs in fish and seafood from the Catalan (Spain) market: estimated human intake. *Environ. Int.* 33 (2), 170–175.
- Burger, J., Gochfeld, M., 2005. Heavy metals in commercial fish in New Jersey. *Environ. Res.* 99 (3), 403–412.
- Burger, J., Gochfeld, M., 2006. Mercury in fish available in supermarkets in Illinois: are there regional differences. *Sci. Total Environ.* 367 (2–3), 1010–1016.
- Burger, J., Jeitner, C., et al., 2009. Factors affecting mercury and selenium levels in New Jersey flatfish: low risk to human consumers. *J. Toxicol. Environ. Health A* 72 (14), 853–860.
- Burger, J., Stern, A.H., et al., 2004. Fish availability in supermarkets and fish markets in New Jersey. *Sci. Total Environ.* 333 (1–3), 89–97.
- Caldwell, K.L., Mortensen, M.E., Jones, R.L., Caudill, S.P., Osterloh, J.D., 2009. Total blood mercury concentrations in the U.S. population: 1999–2006. *Int. J. Hyg. Environ. Health* 212 (6), 588–598.
- DeCaprio, A.P., Tarbell, A.M., Bott, A., Wagemaker, D.L., Williams, R.L., O'Hehir, C.M., 2000. Routine analysis of 101 polychlorinated biphenyl congeners in human serum by parallel dual-column gas chromatography with electron capture detection. *J. Anal. Toxicol.* 24 (6), 403–420.
- Domingo, J.L., Bocio, A., 2007. Levels of PCDD/PCDFs and PCBs in edible marine species and human intake: a literature review. *Environ. Int.* 33 (3), 397–405.
- Easton, M.D., Luszniak, D., Von der Geest, G.E., 2002. Preliminary examination of contaminant loadings in farmed salmon, wild salmon and commercial salmon feed. *Chemosphere* 46 (7), 1053–1074.

- Forsyth, D.S., Casey, V., Dabeka, R.W., McKenzie, A., 2004. Methylmercury levels in predatory fish species marketed in Canada. *Food Addit. Contam.* 21 (9), 849–856.
- Ginsberg, G.L., Toal, B.F., 2009. Quantitative approach for incorporating methylmercury risks and omega-3 fatty acid benefits in developing species-specific fish consumption advice. *Environ. Health Perspect.* 117 (2), 267–275.
- Hites, R.A., Foran, J.A., Carpenter, D.O., Hamilton, M.C., Knuth, B.A., Schwager, S.J., 2004. Global assessment of organic contaminants in farmed salmon. *Science* 303 (5655), 226–229.
- Hites, R.A., Foran, J.A., Schwager, S.J., Knuth, B.A., Hamilton, M.C., Carpenter, D.O., 2004. Global assessment of polybrominated diphenyl ethers in farmed and wild salmon. *Environ. Sci. Technol.* 38 (19), 4945–4949.
- Huang, X., Hites, R.A., et al., 2006. Consumption advisories for salmon based on risk of cancer and noncancer health effects. *Environ. Res.* 101 (2), 263–274.
- IARC 1998. Monographs on the Evaluation of Carcinogenic Risks to Humans. Overall Evaluations of Carcinogenicity. An Updating of IARC Monographs vol. 1–42. Supplement 7. Available at: <<http://monographs.iarc.fr/ENG/Monographs/suppl7/suppl7.pdf>>. (accessed 14 June 2010).
- Interagency Working Group on Methylmercury. 2004. Methylmercury in the Gulf of Mexico: State of Knowledge and Research Needs. Available at: <http://www.flseagrant.org/program_areas/ecosystem_health/publications/Methylmercury_In_Gulf_of_Mexico.pdf>. (accessed 14 June 2010).
- Mahaffey, K.R., Clickner, R.P., Jeffries, R.A., 2008. Methylmercury and omega-3 fatty acids: co-occurrence of dietary sources with emphasis on fish and shellfish. *Environ. Res.* 107 (1), 20–29.
- McKelvey, W., Gwynn, R.C., Jeffery, N., Kass, D., Thorpe, L.E., Garg, R.K., Palmer, C.D., Parsons, P.J., 2007. A biomonitoring study of lead, cadmium, and mercury in the blood of New York city adults. *Environ. Health Perspect.* 115 (10), 1435–1441.
- Mozaffarian, D., Rimm, E.B., 2006. Fish intake, contaminants, and human health: evaluating the risks and the benefits. *J. Am. Med. Assoc.* 296 (15), 1885–1899.
- National Marine Fisheries Service, 2010. Annual Trade Data Summarized by Country/Association. NOAA Fisheries: Office of Science and Technology. Available at: <http://www.st.nmfs.noaa.gov/st1/trade/annual_data/TradeDataAnnualProductCountrySummary.html>. (accessed 14 June 2010).
- National Research Council, 2000. Toxicological Effects of Methylmercury. The National Academies Press, Washington, DC. Available at: <http://www.nap.edu/catalog.php?record_id=9899>. (accessed 14 June 2010).
- Nesheim, M.C., Yaktine, A.L. (Eds.), 2007. *Seafood Choices: Balancing Benefits and Risks*. The National Academies Press, Washington, DC.
- NYC DOHMH, 2007. Eat Fish, Choose Wisely. Available at: <http://www.nyc.gov/html/doh/downloads/pdf/edp/mercury_brochure.pdf>. (accessed 14 June 2010).
- NYS DOH, 2009. Chemicals in Sportfish and Game: 2009–2010 Health Advisories. Available at: <<http://www.health.state.ny.us/environmental/outdoors/fish/fish.htm>>. (accessed 14 June 2010).
- Schantz, S.L., Widholm, J.J., Rice, D.C., 2003. Effects of PCB exposure on neuropsychological function in children. *Environ. Health Perspect.* 111 (3), 357–376.
- Shaw, S.D., Brenner, D., Berger, M.L., Carpenter, D.O., Hong, C.S., Kannan, K., 2006. PCBs, PCDD/Fs, and organochlorine pesticides in farmed Atlantic salmon from Maine, eastern Canada, and Norway, and wild salmon from Alaska. *Environ. Sci. Technol.* 40 (17), 5347–5354.
- Sherer, R.A., Price, P.S., 1993. The effect of cooking processes on PCB levels in edible fish tissue. *Qual. Assur.* 2 (4), 396–407.
- Stern, A.H., Korn, L.R., et al., 1996. Estimation of fish consumption and methylmercury intake in the New Jersey population. *J. Expo. Anal. Environ. Epidemiol.* 6 (4), 503–525.
- Stewart, P.W., Lonky, E., Reihman, J., Pagano, J., Gump, B.B., Darvill, T., 2008. The relationship between prenatal PCB exposure and intelligence (IQ) in 9-year-old children. *Environ. Health Perspect.* 116 (10), 1416–1422.
- Sunderland, E.M., 2007. Mercury exposure from domestic and imported estuarine and marine fish in the U.S. seafood market. *Environ. Health Perspect.* 115 (2), 235–242.
- U.S. EPA. 1997. Integrated risk information system: polychlorinated biphenyls (PCBs) (CASRN 1336-36-3). Available at: <<http://www.epa.gov/iris/subst/0294.htm>>. (accessed 14 June 2010).
- U.S. EPA. 2000. Guidance for Assessing Chemical Contaminant Data for Use in Fish Advisories, vol. 2. Risk Assessment and Fish Consumption Limits, Washington, DC. Available at: <<http://www.epa.gov/waterscience/fish/advice/volume2/>>. (accessed 14 June 2010).
- U.S. FDA 2008. Mercury Levels in Commercial Fish and Shellfish. U.S. Department of Health and Human Services. Available at: <<http://www.fda.gov/food/foodsafety/product-specificinformation/seafood/foodbornepathogenscontaminants/methylmercury/ucm115644.htm>>. (accessed 14 June 2010).
- U.S. FDA/EPA. 2004. What You Need to Know about Mercury in Fish and Shellfish. Available at: <<http://www.fda.gov/Food/ResourcesForYou/Consumers/ucm110591.htm>>. (accessed 14 June 2010).
- Wilson, N.D., Shear, N.M., Paustenbach, D.J., Price, P.S., 1998. The effect of cooking practices on the concentration of DDT and PCB compounds in the edible tissue of fish. *J. Expo. Anal. Environ. Epidemiol.* 8 (3), 423–440.