Polychlorinated Biphenyl Concentration in Raw and Cooked North Atlantic Bluefish (*Pomatomus saltatrix*) Fillets

A. A. Salama,[‡] M. A. M. Mohamed,[§] B. Duval,[†] T. L. Potter,*[,†] and R. E. Levin[†]

Department of Food Science, Chenoweth Laboratory, University of Massachusetts, Amherst, Massachusetts 01003, Food Science and Technology Department, Faculty of Agriculture, Kafr El-Sheikh, Tanta University, Egypt, and Biochemistry Department, Faculty of Agriculture, Cairo University, Egypt

Polychlorinated biphenyls (PCBs) were measured before and after fillets of six bluefish (*Pomatomus saltatrix*) were cooked. Six cooking treatments were used with one fish per treatment. PCB concentrations ranged between 0.114 and 0.748 mg/kg in raw fish and between 0.102 and 0.315 mg/kg in cooked fish. The average for the raw fish was 0.31 and for the cooked fish was 0.19 mg/kg. PCB concentrations in both raw and cooked fillets were well below levels reported for raw fillets from fish tested in the mid-to-late 1980s and FDA health advisories. A decrease in PCB concentration in four of the six cooking treatments was observed. They included, in order of greatest decrease, smoking > microwave > chrabroiling (skin off) > charbroiling (skin on). There was no change with convection oven baking and pan frying. When adjusted for weight loss during cooking, all cooking methods showed PCB loss (average = 46%). The most effective cooking methods for removing PCBs from bluefish included smoking and microwave baking. The percentages of PCBs lost were 65 and 60, respectively. Losses for the other treatments were 46% (charbroiling with skin off), 37% (charbroiling with skin on), 27% (pan frying), and 39% (convection oven baking). The data suggest a generally decreasing trend in PCB concentration in bluefish and point to the need to account for cooking losses when estimating PCB exposures from bluefish consumption.

Keywords: *Polychlorinated biphenyls (PCBs); bluefish; cooking methods*

INTRODUCTION

The bluefish (*Pomatomus saltatrix*) is one of the most common species caught by sport fishermen along the North and Mid-Atlantic coast of the United States. Commercial catches also rose steadily through the 1980s with the exception of a sharp decline in 1988 (Eldrige and Meaburn, 1992). Their relatively high lipid content, feeding habits, and moderately long life span make bluefish susceptible to accumulation of lipophilic contaminants such as polychlorinated biphenyls (PCBs) and chlorinated hydrocarbon pesticides (Sanders and Haynes, 1988).

Surveys conducted in the mid-1980s showed that fish of fork length >500 mm often had PCB concentration in fillets in excess of the FDA tolerance level of 2 mg/kg in the edible portion of raw fish (Eldridge and Meaburn, 1992; Sanders and Haynes, 1988; Trotter et al., 1989; NOAA/FDA/EPA, 1987). These data have indicated that, depending on consumption rates, bluefish may present a significant dietary source of PCBs. However, current dietary exposure estimates are limited by the lack of more recent data. Nearly 10 years have elapsed since these surveys of PCB concentration in bluefish tissue were completed. Experience with Great Lakes salmonids (Stow, 1995) has suggested a downward trend in PCB concentration. The magnitude of the decrease for bluefish is unknown.

Another uncertainty in the assessment of dietary PCB exposure by bluefish consumption is the impact of cooking. Only a single published study was identified in which PCB concentrations were reported in cooked bluefish tissue. In this case, Trotter et al. (1989) found that baking fillets for 1 h at 325 °F in a pan which allowed liquids to drain freely from the tissue, followed by skin removal, resulted in a 27% (average) decrease in PCB concentration. Studies using other fish species and other cooking techniques have given mixed results. Armbruster et al. (1987) reported that baking, broiling, frying, microwaving and poaching reduced PCB levels in striped bass fillets by 12−23%, while steaming gave a 5% increase in PCB concentration. With five species of Great Lakes fish, baking, charbroiling, pan frying, deep fat frying, salt boiling, canning, and smoking reduced PCB levels by 20−47% (Zabik and Zabik, 1996). The greatest reduction was with smoking. Earlier studies with Chinook and Coho salmon and carp showed inconsistent or minimal losses with various cooking techniques (Smith et al., 1973; Zabik et al., 1982, 1995).

On the basis of these variable and limited results, it was concluded that additional studies were needed to guide consumers in cooking techniques which can reduce dietary PCB exposure from bluefish consumption and to provide toxicologists with more accurate PCB exposure estimates. In the study reported here, PCB levels were measured in bluefish fillets before and after cooking. Six common cooking techniques were used.

MATERIALS AND METHODS

Fish Source. Bluefish caught commercially in Massachusetts waters were donated by the University of Massachusetts

* Author to whom correspondence should be addressed [e-mail tpotter@fnr.umass.edu; fax (413) 545-1262].

[†] University of Massachusetts.

[‡] Tanta University.

[§] Cairo University.

S0021-8561(97)00714-0 CCC: $15.00
Published on Web 02/28/1998

Marine Station (Gloucester, MA). The fish were transported directly to the laboratory on ice, rinsed with distilled water, wrapped in foil, and stored in a −20 °C freezer. The seven specimens used in the study had an average weight of 2.0 kg and a length of 60 cm. Bluefish of this fork length are generally between 3 and 5 years in age, with the majority of 60-cm fish belonging to the 3−4-year-old class (Terceiro and Ross, 1993). A single fish was used to evaluate the effects of each of the six cooking treatments. Prior to cooking, all fish were filleted along the backbone. Two whole fillets were obtained from each fish. One was analyzed raw. The second was cooked and then analyzed. In addition, both fillets of a single fish were analyzed raw. Triplicate analyses of subsamples from each of these fillets revealed that there was no statistically significant difference ($P < 0.01$) in PCB concentration between the fillets. This observation provided the basis of the experimental approach used in the evaluation of cooking methods in which the PCB concentrations in the two fillets obtained from individual fish were compared.

Cooking Methods. After filleting, the fish tissue was rinsed with tap water and then distilled water and patted dry with paper towels.

(1) Smoking. Whole fillets were smoked using a hot smoked fish process involving soaking the fish in brine solution (98 g of NaCl and 1.5 g of $NaNO_2$ per liter of distilled water), drying the fish at room temperature for 3 h, and smoking for 4 h at 54−66 °C.

(2) Microwave. The fillets were cut into 2.5-cm-thick steaks with cuts made perpendicular to the "head to tail" axis. Steaks were cooked uncovered in a round Pyrex glass dish in a standard household microwave oven (2450 MHz) operated on the "high" setting for 3−5 min. Cooking times were varied to compensate for differences in steak size and to achieve similar degrees of doneness in all steaks. The cooking endpoint was based on the visual appearance of the tissue at the center of the steaks changing, from translucent to white.

(3) Charbroiling (without Skin). Whole fillets were cooked over charcoal for 18 min to an internal temperature of 65−70 °C.

(4) Charbroiling (with Skin). Whole fillets were cooked over charcoal for 18 min to an internal temperature of 65−70 °C.

(5) Pan Frying. The fillets were cut into 1.25-cm-thick steaks with cuts made perpendicular to the "head to tail" axis. Steaks were pan-fried in soybean oil at 190 °C for 3−5 min until golden brown on each side. Cooking times were varied to compensate for differences in steak size and to achieve similar degrees of doneness in all steaks. The cooking endpoint was based on the visual appearance of the tissue at the center of the steaks, changing from translucent to white.

(6) Baking. Whole fillets were baked in an oven at 204 °C for 15 min. The internal temperature of the fish after cooking was 80 °C. Liquids were not allow to drain from the fish after cooking.

Sample Preparation. Whole fillets were homogenized in a stainless steel blender with sodium sulfate. With the microwave and pan-frying techniques, all steaks from a given fillet were blended together to yield single samples for these cooking techniques. This approach, that is, using whole fillets, was taken to reduce subsampling variation when cooked and control samples were compared. PCBs are not uniformly distributed in bluefish muscle tissue (Sanders and Haynes, 1988).

Extraction and Cleanup. Duplicate 10-g subsamples of the homogenized and desiccated tissue were sequentially extracted with three 50-mL aliquots of *n*-pentane/methylene chloride (1:1) in glass centrifuge tubes. The extracts were combined with 10 mL of isooctane added as a keeper solvent, followed by Rotoevaporation of the extracts to ≈50 mL. Erney (1974) used a similar approach. All chemicals and solvents were of "pesticide grade" (Fisher Scientific, Medford, MA). Lipid removal was accomplished by adding 25 mL of concentrated sulfuric acid to solvent extracts in 250-mL round-bottom flasks, followed by gentle shaking. After 12−16 h, the clear solvent layers were transferred quantitatively to round-bottom flasks and evaporated on a rotary evaporator to 20 mL. The samples were further concentrated to 5 mL under the flow of nitrogen. Extracts were then treated on Florisil columns (U.S. Environmental Protection Agency, 1992). Decafluorobiphenyl (Ultra Scientific, Hope, RI), which served as a surrogate compound and retention time marker, was added (2 μg) to all samples prior to solvent extraction. The recovery was 84−93%.

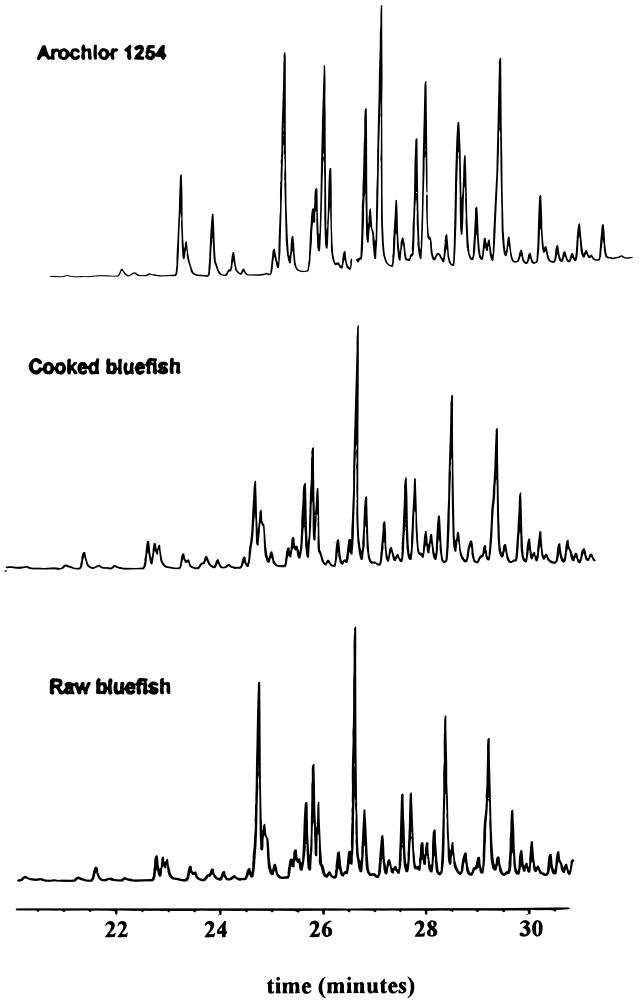

Figure 1. GC-ECD chromatograms of Arochlor 1254 and PCBs extracted from raw and cooked bluefish.

Instrumental Analysis. Extracts were analyzed using a Hewlett-Packard model 5890 series II gas chromatograph equipped with an electron capture detector (ECD). The oven was fitted with a 30 m × 0.25 mm i.d., DB-5 (J&W Scientific, Folsom, CA) fused silica capillary column. Helium carrier head pressure was 100 kPa with nitrogen gas makeup at 30 mL/min. The injector and detector temperatures were 270 and 280 °C, respectively.

The initial oven temperature was 60 °C. This temperature was held for 1 min followed by an increase to 290 °C at 8 °C/min. The final temperature was held for 8 min. Manual injections were in the splitless mode. PCBs were quantified using a set of Arochlor 1254 standards prepared from 1000 μg/mL stock in hexane (Chemical Service Co., West Chester, PA). Aroclor 1254 was selected for quantitation on the basis of the relatively close match between the chromatograms of the standard and extracts obtained from raw and cooked fish. This similarity in chromatographic profiles of the Aroclor 1254 standard and extracts obtained from raw and smoked fish is shown in Figure 1.

RESULTS AND DISCUSSION

Results of all analyses are summarized in Table 1. In general, a close match was observed between repli-

Table 1. PCB Concentration (Milligrams per Kilogram) in Cooked and Raw Bluefish Fillets[a]

cooking method	raw	cooked
smoking	0.69	0.30
microwave baking	0.44	0.23
charbroiling (skin off)	0.15	0.10
charbroiling (skin on)	0.32	0.26
pan frying	0.12	0.12
baking	0.14	0.14

[a] Averages of duplicate analyses.

Table 2. PCB Concentration Change and Percent Loss[a]

cooking method	% change	% loss
smoking	55	65
microwave baking	48	60
charbroiling (skin off)	29	47
charbroiling (skin on)	18	37
pan frying	no change	27
baking	no change	39

[a] Percent loss was computed from the observed concentration change by adjustment for the weight loss during cooking.

cate analyses. The coefficient of variation (CV) of these data pairs was 3.4−17% (average = 9.1%).

In the raw fish, PCB concentrations ranged between 0.114 and 0.748 mg/kg with an average and standard deviation of 0.31 and 0.21 mg/kg, respectively. The average was 6−8 times lower than mean PCB concentration values reported in earlier studies on bluefish. Trotter et al. (1989) reported a mean of 2.5 mg/kg for fillets obtained from 20 specimens collected in Buzzards Bay, New Bedford, and Plymouth, MA, in 1986. The mean PCB concentration in the fillets of bluefish caught off the North Carolina coast in 1988 was 2.04 mg/kg (Sanders and Haynes, 1988). These fish were in the same size range as those analyzed in the current study. In another study conducted with fish captured along the Atlantic Coast in 1984−1986, it was reported that 16% of all fillets taken from bluefish of >60-cm forklength had PCB concentrations >2 mg/kg in the edible portion of raw fish (NOAA, FDA, EPA, 1987). Comparison of data obtained in the current study with previously reported results suggests a decreasing trend in PCB concentration in bluefish fillets.

The average and standard deviation of the PCB concentration in the cooked fish were 0.19 and 0.08 mg/kg, with a range of 0.102−0.315 mg/kg. These data indicate a potentially significant decrease in PCB concentration with cooking. The apparent extent of decrease was cooking method dependent. As indicated in Table 2, four of the six cooking methods resulted in a decrease in PCB concentration ranging from 55 to 18%. They included, in order of the greatest percent PCB loss, smoking > microwave baking > charbroiling (skin off) > charbroiling (skin on). Pan frying and baking did not change the concentration of PCBs in the cooked fillets. However, when the data were adjusted for weight loss during cooking, all treatments indicated a loss of PCBs. Loss ranged from 27 to 65%.

These results are in agreement with a previously reported study concerning the effect of cooking on PCB concentrations in bluefish (Trotter et al., 1989) and studies examining this tissue in other fish species (Armbruster et al., 1987; Zabik et al., 1979; Zabik and Zabik, 1996). Smoking, microwave baking, and charbroiling all resulted in a net loss of PCBs from the tissue. The greatest loss of PCBs, 65%, was with the smoking treatment. Loss due to smoking from five Great Lakes species was 48% (Zabik and Zabik, 1996).

Skin removal in the case of charbroiling, one of the most common cooking methods used by sport fishermen, appeared to increase the portion of PCBs lost. Cooking with the skin off resulted in a 46% decrease, while skin-on cooking gave a 37% decrease. The fillet was cooked skin surface down and not turned during cooking. This presumably reduced lipid loss during cooking. Studies have shown that high concentrations of PCBs are found in the oil drippings after cooking and in fish cooked with the skin (Trotter et al., 1989).

The microwave results are anomalous in that PCB loss was relatively high. Zabik et al. (1979) found that microwaving gave a 34% reduction of PCBs from fat lake trout. Armbruster et al. (1987) reported a 19% PCB loss after microwave cooking of striped bass.

That no net change in the PCB concentration was observed with pan frying and convection oven baking was presumably due to the weight loss during cooking. When this was taken into account, PCB reduction was 39% for baking and 27% for pan frying.

CONCLUSIONS

In this study, PCB concentrations in the fillets of all fish were well below the FDA tolerance level of 2 mg/kg. This is in contrast to prior studies (NOAA/FDA/EPA, 1986; Trotter et al., 1988; Sanders and Haynes, 1989). During the 10 years between the current and prior studies, there has been a major regulatory effort to control and contain PCBs throughout the United States and Canada. This may account for the apparent decrease.

A time-dependent decrease in PCB concentration in bluefish was expected on the basis of data summarized by Stow (1995) for PCB concentration in Lake Michigan salmonoids over the period 1972−1992. However, species aggregated plots of PCB concentration have remained fairly constant since the early 1980s (Stow, 1995). PCB levels in bluefish may be behaving in a similar fashion, although time scales and the concentration level which is being approached asymptotically remain uncertain. More study is needed to predict bluefish PCB concentration. It is essential to derive accurate exposure estimates of the general population from bluefish consumption. Without such data, efforts to identify relationships between fish consumption and public health impacts will remain speculative.

The data obtained in the current study also emphasize the importance of evaluating cooking when dietary PCB exposure is estimated. Cooking has the potential to significantly decrease PCB concentration in fillets. The decrease was observed to be cooking method dependent, ranging from >50% to no change.

ACKNOWLEDGMENT

We thank Herbert Hultin and Stephen Kelleher at the University of Massachusetts Marine Station, Gloucester, MA, for donating the bluefish used in this study.

LITERATURE CITED

Armbruster, G.; Gerow, K. G.; Gutenmann, W. H.; Littman, C. B.; Lisk, D. J. The effects of several methods of fish preparation on residues of polychlorinated biphenyls and sensory characteristics in striped bass. *J. Food Saf.* **1987**, *8*, 235−243.

Eldridge, P. J.; Meaburn, M. Potential impact of PCB's on bluefish, *Pomatomus saltatrix,* management. *Mar. Fish. Rev.* **1992**, *54*, 19−24.

Erney, D. R. Rapid screening method for analysis of chlorinated pesticide and polychlorinated biphenyl residues in fish. *J. Assoc. Off. Agric. Chem.* **1974**, *57*, 576−579.

Faria, S. M. A manual of seafood products, marketing, and utilization. *Mass. Div. Mar. Fish.* **1984**, 109−122.

Madenjian, C. P.; Carpenter, S. R.; Rand, P. S. Why are the PCB concentrations of salmonine individuals from the same lake so highly variable? *Can. J. Fish. Aquat. Sci.* **1994**, *51*, 800−807.

NOAA/FDA/EPA (U.S. National Oceanic and Atmospheric Administration, U.S. Food and Drug Administration, U.S. Environmental Protection Agency). Report on the 1984-1986 federal survey of PCBs in Atlantic coast bluefish. Interpretive report; PB87-214672/XAB, March 1987; 76 pp.

Sanders, M.; Haynes, B. Distribution pattern and reduction of polychlorinated biphenyls (PCB) in bluefish *Pomatomus saltatrix* (Linnaeus) fillets through adipose tissue removal. *Bull. Contam. Toxicol.* **1988**, *41*, 670−677.

Smith, W. E.; Funk, K.; Zabik, M. E. Effects of cooking on concentrations of PCB and DDT compounds in chinook (*Onchorhynchus tshawytscha*) and coho (*O. kisutch*) salmon from Lake Michigan. *J. Fish Res. Board Can.* **1973**, *30*, 702−706.

Stow, C. A.; Carpenter, S. R.; Amrhein, J. F. PCB concentration trends in Lake Michigan coho (*Oncorhynchus kisutch*) and chinook salmon (*O. tshawytscha*). *Can. J. Fish. Aquat. Sci.* **1994**, *51*, 1384−1390.

Stow, C. A.; Jackson, L. J.; Amrhein, J. F. An examination of the PCB:lipid relationship amoung individual fish. *Can. J. Fish. Aquat. Sci.* **1997**, *54*, 1031−1038.

Terceiro, M.; Ross, J. L. A comparison of alternative methods for the estimation of age from length data for Atlantic coast bluefish (Pomatomus saltatrix). *Fish. Bull.* **1993**, *91*, 534−549.

Trotter, W. J.; Corneliussen, P.; Laski, R. R.; Vannelli, J. J. Levels of polychlorinated biphenyls and pesticides in bluefish before and after cooking. *J. Assoc. Off. Anal. Chem.* **1989**, *72*, 501−503.

U.S. Environmental Protection Agency. Florisil column cleanup method 3620A, 1992; 3620A1-11.

Zabik, M. E.; Zabik, M. Influence of processing on environmental contaminants in foods. *Food Technol.* **1996**, *May*, 225−229.

Zabik, M. E.; Hoojjat, P.; Weaver, C. M. Polychlorinated biphenyls, dieldrin and DDT in Lake Trout cooked by broiling, roasting or microwave. *Bull. Environ. Contam. Toxicol.* **1979**, *21*, 136−143.

Zabik, M. E.; Merrill, C.; Zabik, M. J. PCBs and other xenobiotics in raw and cooked carp. *Bull. Environ. Contam. Toxicol.* **1982**, *28*, 710−715.

Zabik, M. E.; Zabik, M.; Booren, A.; Nettles, M.; Song, J.; Welch, R.; Humphrey, H. Pesticide and total polychlorinated biphenyls in chinook salmon and carp harvested from the Great Lakes: effects of skin-on and skin-off processing and selected cooking methods. *J. Agric. Food Chem.* **1995**, *43*, 993−1001.

Received for review August 18, 1997. Revised manuscript received January 16, 1998. Accepted January 23, 1998. We acknowledge Tanta University for providing financial support for Dr. Salama and Cairo University for supporting Mr. Mohamed.

JF9707142