

EFFECT OF DEEP-FRYING FISH ON RISK FROM MERCURY

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*The U.S. Environmental Protection Agency (EPA) and many states have issued advisories to limit or avoid the consumption of certain fish or fish from certain waters, particularly by pregnant women and young children or even women of childbearing age. Typically, risk is calculated by multiplying contaminant concentrations in fish tissue, frequency of meals, and meal size, compared to some criterion, usually the U.S. EPA reference dose (RfD). Site-specific data on mercury concentrations, meal size, and consumption frequency by fishermen were used to determine how frying fish affected risk estimates. In consumption studies fishermen typically estimate the size of portions as they appear on the plate (i.e., cooked), yet assessors calculate risk based on contaminant levels in uncooked fish. Largemouth bass (*Micropterus salmoides*, n=39) were collected from the contaminated L Lake on the Savannah River Site, South Carolina. Fillets from the opposite sides of the same fish were divided and randomly assigned to a raw or fried treatment (the commonly used local cooking method). The fried fillet was further divided in half for a breaded or nonbreaded treatment. Mercury averaged 0.44 µg/g (ppm, wet weight) in raw fish, 0.63 µg/g in fried and breaded fish, and 0.76 µg/g in fried, unbreaded fish. The maximum concentration was 1.5 µg/g in raw fish (1.9 µg/g in cooked fish). Deep-frying with and without breading resulted in weight loss of 25% and 39%, while mercury levels increased by 45% and 75%, perhaps due to the breading and absorption of oil. At the mean fish*

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consumption rate of people fishing locally, mercury intake exceeded the U.S. EPA RfD of 0.1 $\mu\text{g}/\text{kg}/\text{d}$ for all except white females. Thus consumption of fish from this lake would exceed acceptable levels. Risk assessments should be conducted with site-specific data on contaminants and consumption of cooked fish and consumption studies should specify whether portion size was pre- or postpreparation. Fishermen estimate the amounts of fish they eat based on a meal size (usually cooked), while risk assessors determine mercury levels in raw fish. A conversion factor of about 2 for mercury increase during cooking is reasonable and conservative.

In many rural and some urban regions of the United States fishing is an important aspect of recreation, culture, and tradition (Fleming et al., 1995; Toth & Brown, 1997; Burger, Stephens et al., 1999; Burger, Pflugh et al., 1999). Yet contaminants, such as mercury and polychlorinated biphenyls (PCBs), are sufficiently high in some fish and seafood to pose a risk to human consumers (ATSDR, 1996; IOM, 1996; Ratcliffe et al., 1996; Weiss & Elsner, 1996; Weihe et al., 1996; Kamrin & Fischer, 1999; Sweet & Zelikoff, 2001). Concern about such health risks has led governmental agencies to issue consumption advisories for many waterbodies (U.S. EPA, 2002). The U.S. Environmental Protection Agency (1998) reported that 16% of the nation's total lake acres and 7% of the nation's total river miles were under consumption advisories, as well as all of the Great Lakes. Mercury accounts for most of the advisories, but PCBs, chlordane, dioxins, and DDT are also important (U.S. EPA, 2002).

Fetuses, neonates, and young children are the group considered at highest risk (Bakir et al., 1973; Jacobson et al., 1990; Davidson et al., 1995; Jacobson & Jacobson, 1996; Weihe et al., 1996; Evens et al., 2001). Risk reduction for fetuses and neonates involves clear and balanced risk communication to pregnant women, taking into account the fact that fish are a wholesome source of nutrition during pregnancy (Sparks & Shepherd, 1994; Knuth, 1995; Ebert, 1996). Recognizing the preconception exposure contributes to maternal body burden, agencies often expand their warnings to all women of childbearing age. Risk from fish consumption involves contaminant levels in edible fish tissue, meal frequency, and meal size. Yet other factors may affect the risk from fish consumption, including preparation practices (Morgan et al., 1997; Wilson et al., 1998).

This article examines the effect of deep-frying on mercury levels in large-mouth bass (*Micropterus salmoides*), a preferred fish of people in South Carolina and elsewhere in the south (Fleming et al., 1995; Toth & Brown, 1997). Over 80% of the people interviewed along the Savannah River deep-fry their fish regularly (Burger, Stephens et al., 1999), making it critical to understand whether this cooking method affects mercury levels in fish. Most of the fish-frying is done with batter and breading, and the effect of breading on mercury levels was also examined. Thus, the hypothesis was tested that the levels of mercury in raw and deep-fried fish (with and without breading) do not differ. If the levels of mercury differ in cooked and raw fish, this can have major implications for risk assessment because most consumption studies report consumption

on the basis of cooked fish (Fleming et al., 1995; Ratcliffe et al., 1996; Burger, Stephens et al., 1999; Kamrin & Fischer, 1999), yet contaminant data are reported for raw fish (Knuth, 1995; Morgan et al., 1997; Burger, Gaines & Gochfield, 2001; Burger, Gaines, Peles et al., 2001).

The overall objective was to determine whether deep-frying affects the resultant risk assessment for fish consumption, including pregnant women. Site-specific information on consumption patterns (meal frequency, meal size), cooking methods (deep-frying), and contaminant levels in largemouth bass, a popular, predatory fish specifically mentioned in consumption advisories for the Savannah River, was used (SCDHEC, 1996, 2001; GDNR, 2001). Second, the potential risk to consumers from eating these fish was examined, considering the possibility that recreational fishing may someday be allowed in a currently contaminated lake.

Several studies have examined the effect of fat trimming, skin removal, and cooking on PCBs and other fat-soluble contaminants in fish (Morgan et al., 1997), but much less attention has been devoted to mercury (Wilson et al., 1998). Trimming and cooking fish by various means reduces the levels of fat-soluble contaminants, including dioxins, PCBs, and organochlorine pesticides (Reinert et al., 1972; Sanders & Haynes, 1988; Morgan et al., 1997). This has led some state agencies to use a reduction factor in calculating the dose of fat-soluble contaminants from raw fish in developing fish advisories (Wilson et al., 1998).

Information about the effects of cooking on mercury is less clear. Two studies showed no statistical difference in mercury in cooked and raw fish (Armbruster et al., 1987; Gutenmann & Lisk, 1991), while another showed higher levels in cooked fish on a wet weight basis (Morgan et al., 1997). Morgan et al. (1997) examined the effect of cooking practices on mercury for two commonly caught fish from Lake Wisconsin (walleye, lake trout). They found that mercury concentrations in pan-fried, baked, and broiled fish fillets were from 1.1 to 1.6 times higher than in corresponding raw portions. They found that total mercury amounts were constant before and after cooking (i.e., mercury was not driven off during cooking). However, they did not deep-fry fillets, and data on walleye and trout may not be applicable to the fish commonly caught in much of the eastern and southeastern United States. Further, without site-specific consumption data, they did not examine the effect on risk assessment and risk management.

METHODS

Study Area

Fish were collected from L Lake on the Department of Energy's Savannah River Site (SRS) (33.1°N, 81.3°W). SRS is a 780-km² former nuclear weapons production and research facility operated by the U.S. government since the early 1950s.

Fish Collection and Cooking

Thirty-nine largemouth bass were collected under appropriate state permits, and with protocol approvals from the University of Georgia Institutional Animal Care and Use Committee (A960205) and Rutgers University Institutional Review Board (07-017). Fish were collected using rod and reel, and taken to the Savannah River Laboratory for dissection into fillets. In the laboratory, after skinning, a 2-cm-thick fillet (200 g) was removed from each side of the fish. One fillet was randomly assigned to raw (only half was used in the analysis) and one was assigned to the fry treatment. The 2-cm-thick fillet assigned to fry was further divided into half; one to receive breading and one to be without breading. The 100-g fillets were immediately frozen (-4°C) and labeled by fish number, date, collection location, and treatment.

For breading, the wet fillet was dipped in a commercial breading mix (Zatarain's Seasons Fish Fry) until the surface was lightly covered. Cooking was done according to local customs using local deep frying vats (uncovered), and with a commonly used cooking oil (Dukes peanut oil) as is the local custom. No mercury was detectable in the cooking oil when analyzed alone. Fillets were weighed before and after cooking. Fillets were submerged in hot oil (approximately 190°C) until the fillet began to float (typically 5–7 min), indicating that cooking was complete. Fillets were then patted dry of oil, weighed, and frozen for transportation to the Environmental and Occupational Health Sciences Institute.

Chemical and Statistical Analysis

A 2-g sample was cut from the center of each 100-g fillet and completely digested in 3 ml Ultrex ultrapure nitric acid in a microwave (MD 2000 CEM), using a digestion protocol of 3 stages of 10 min each under 50, 100, and 150 pounds per square inch (3.5, 7.0, and 10.6 kg/cm^2) at $70 \times$ power (Burger, Gaines & Gochfeld, 2001). Digested samples were diluted in 20 ml deionized water. Mercury was analyzed by cold vapor technique. All concentrations are expressed in parts per million ($\mu\text{g/g}$ on wet weight).

Mercury was analyzed with a HGA4 mercury analyzer in the Environmental and Occupational Health Sciences Institute. Detection limits were 0.2 ppb for mercury (instrument detection limits). All specimens were run in batches that included blanks, a standard calibration curve, and spiked specimens. The accepted recovery levels for spikes was 85%. All spikes exceeded this value. The coefficient of variation on replicate samples ranged from 4 to 10%. Further quality control included blind runs of duplicate samples during the analysis for each metal (acceptable criterion $\pm 15\%$). Twelve replicates were also analyzed using the pyrolysis feature of the Lumex mercury analyzer and yielded a high correlation ($r=.94$) between the two methods. Like most analytic reports, these procedures analyze the total mercury content, while risk assessments are based on methylmercury. Other studies have shown that in most cases at least 90% of the mercury in fish is in the methyl form. Thus this adds a slight additional protective value of up to 10% to the risk assessment, since not all the measured mercury is methyl.

Mercury concentrations were compared among treatments using the Kruskal-Wallis nonparametric analysis of variance (SAS PROC NPAR1WAY with Wilcoxon Option), which generated a χ^2 value to examine differences among treatments (SAS Institute, Inc., 1995). Before and after mercury concentrations for the 39 fish were compared with a Kendall tau. An a priori significance level of $<.05$ was designated.

Risk Analysis Methods

Development of the nervous system in the fetus is the most sensitive end point for organic mercury (Stern, 1993; Young et al., 1997) and is now used in risk assessments to develop RfDs or their equivalent. The RfD is based on chronic exposure, no observed effect levels, and various uncertainty factors, including one to protect sensitive subgroups, hence it can apply to either adults or children. The development of the U.S. Environmental Protection Agency RfDs was originally based on the Iraq organomercury epidemic in the early 1970s (Bakir et al., 1973), although Stern (1993) computed a lower RfD of $0.07 \mu\text{g}/\text{kg}/\text{body weight}/\text{d}$ based on data from Iraq. Other suggested RfDs have been based on recent prospective longitudinal studies in the Seychelles (Bakir et al., 1973) and the Faroe Islands (Weihe et al., 1996). The National Research Council recently reviewed the studies and suggested that the Faroe Island results were a reasonable basis for a revised RfD, and the U.S. EPA recalculated the RfD, arriving again at a value of $0.1 \mu\text{g}/\text{kg}/\text{d}$, which is documented in the U.S. EPA IRIS database (U.S. EPA, 1997, 1998). However, the U.S. EPA Division of Water has based its fish advisories on an oral RfD of $0.06 \mu\text{g}/\text{kg}/\text{d}$. Recently, the Agency for Toxic Substances and Disease Registry has proposed a minimum risk level of $0.3 \mu\text{g}/\text{kg}/\text{d}$ (ATSDR, 1999) based on the Seychelles neurodevelopmental study (Davidson et al., 1995).

The risk assessment in this study is based on the U.S. EPA oral RfD for methylmercury of $0.1 \mu\text{g}/\text{kg}/\text{d}$ and on the minimum risk level (MRL) of $0.3 \mu\text{g}/\text{kg}/\text{d}$, which is sometimes used as an unofficial RfD for adults who are not pregnant. The estimated daily intake of mercury, derived from consumption studies (Burger, Stephens et al., 1999), was compared with the RfD values, using the median, mean, 75%, and 95% of the consumption distributions reported for males and females. Since there was a clear ethnic differences in consumption rates among fishermen in the area (Burger, Gaines & Gochfeld, 2001), black and white fishermen are reported separately (Table 1).

RESULTS

Mercury in Raw and Cooked Fish

Mercury levels were significantly higher in cooked fish (breaded and non-breaded) compared to raw fish (Kruskal-Wallis $\chi^2=13.8$, Table 2). For a given fish, there was a significant correlation between the levels of mercury in the cooked and uncooked fish ($\text{tau}=0.85$, $p<.001$). On average, an uncooked

TABLE 1. Fish Consumption

	Median	Mean	75th Percentile	95th Percentile
Black males (g/d)	51.8	70.1	131.5	187.9
Black females (g/d)	35.2	47.7	89.4	127.8
White males (g/d)	18.8	38.4	53.4	135.3
White females (g/d)	12.8	26.1	36.3	90.0
Black males (kg/yr)	18.9	25.6	48.0	68.6
Black females (kg/yr)	12.8	17.4	32.6	46.6
White males (kg/yr)	6.9	14.0	19.5	49.4
White females (kg/yr)	4.7	9.5	13.2	32.9

Note: Data are based on interviews with fishermen along the Savannah River (Burger, Stephens et al., 1999).

TABLE 2. Concentrations of Total Mercury (in $\mu\text{g/g}$ [ppm], wet weight) in Raw and Fried Fillets from Largemouth Bass from L Lake

	Mean mass of sample (g)	Arithmetic mean \pm SE	Range of Hg
Raw fish	100	0.446 \pm 0.04	0.16–1.56
Deep-fried fish			
With breeding	80	0.63 \pm 0.065	0.16–1.97
Without breeding	72	0.76 \pm 0.069	0.16–1.56

Note: Data are based on $n = 39$.

fillet of 100 g weighed 80 g after cooking with breading, and 72 g after cooking without breading. While deep-frying with and without breading resulted in weight loss of 25% and 39%, mercury levels increased by 45% (breaded) and 75% (unbreaded).

Risk Analysis

Based on site-specific data on fish consumption and mercury concentrations, risk was calculated for four subgroups (black males, black females, white males, white females) at four levels of intake (median, mean, 75th and 95th percentile) for raw and cooked fish, using the mean mercury concentrations under each condition (Table 3). Mercury intake for those with mean consumption rates eating nonbreaded fish at the mean mercury concentration of 0.76 ppm ranged from 0.19 $\mu\text{g/kg/d}$ for white females to 0.44 $\mu\text{g/kg/d}$ for black males (Table 3). At the median consumption rate, only white females (mean intake of 0.09 $\mu\text{g/kg/d}$) would not exceed the U.S. EPA reference dose.

Another method of examining risk is to calculate how much fish a pregnant women could consume without increasing her risk. Translating this into consumption patterns, a pregnant women could not eat an 8-ounce (227 g) portion of cooked fish more often than about once a month without exceeding the U.S. EPA RfD (Table 4).

TABLE 3. Mercury Intake for Four Subgroups at Four Levels of Fish Consumption

	Median	Mean	75th Percentile	95th Percentile
Raw-fish				
Black males	0.32	0.44	0.82	1.17
Black females	0.26	0.35	0.66	0.93
White males	0.12	0.24	0.33	0.85
White females	0.09	0.19	0.26	0.65
Deep-fried and breaded				
Black males	0.47	0.63	1.19	1.70
Black females	0.37	0.50	0.94	1.35
White males	0.17	0.35	0.48	1.22
White females	0.14	0.28	0.38	0.95
Deep-fried, not breaded				
Black males	0.56	0.76	1.43	2.04
Black females	0.45	0.60	1.13	1.62
White males	0.20	0.42	0.58	1.47
White females	0.16	0.33	0.46	1.14

Note: Data are based on four demographic groups, for the mean mercury concentration in each of the cooking categories (Burger, Stephens et al., 1999). Body weight of 70 kg for males and 60 kg for females was assumed. All values are in micrograms of mercury per kilogram body weight per day ($\mu\text{g}/\text{kg}/\text{d}$).

TABLE 4. Frequency with Which a 60-kg Woman Could Consume an 8-Ounce Meal of Each Type of Fish (Raw, Fried) and Corresponding Advisory Text

	Hg ($\mu\text{g}/\text{g}$ wet) ^a	μg Hg in 8-ounce (227-g) meal	Dose/meal or intake/60 kg ^b	Dose/RfD (HQ) ^c	Dose/ MRL	Advice for pregnant women ^d	Advice for other adults ^e
Raw fish	0.44	99.1	1.65	16.5	5.5	3 wk	1 wk
Fried and breaded	0.63	143.8	2.39	23.9	8.0	1 mo	1 wk
Fried and unbreaded	0.76	172.9	2.88	28.8	9.6	1 mo	2 wk

Note: Doses are divided by the U.S. EPA RfD (0.1 $\mu\text{g}/\text{kg}/\text{d}$) and by the CDC/ATSDR MRL (0.3 $\mu\text{g}/\text{kg}/\text{d}$).

^aConcentration of mercury in muscle in ppm (wet weight basis).

^bMercury in 8-ounce (227-g) meal divided by 60 kg body weight.

^cHazard quotient (HQ) is obtained by dividing daily dose by RfD.

^dText for fish advisory: Pregnant women should not eat an 8-ounce meal more often than every (week, month).

^eText for other adults: Do not eat this more often than once every week

DISCUSSION

The source of the mercury in the fish was primarily industrial pollution. L Lake was used as a source of cooling water for one of the nuclear reactors when it was functioning (Kennamer et al., 1998). Prior to the construction of

the cooling ponds, there was some ecosystem contamination of streams and the floodplain, and small quantities of radionuclides were released subsequently (Ashley & Zeigler, 1980; Whicker et al., 1990; Kennamer et al., 1998). There is some controversy about the source of mercury contamination in L Lake, which appears to come both from on-site releases and from mercury released upriver, which was then pumped into the lake during cooling activities and re-released to contribute to the mercury load in the Savannah River (Kvartek et al., 1994; Sugg et al., 1995). Atmospheric deposition also contributes contaminants to SRS. The U.S. EPA has determined that the Savannah River is in a zone of above average atmospheric mercury deposition ($>10\mu\text{g}/\text{m}^2/\text{yr}$; U.S. EPA, 1980).

Mercury in Raw and Cooked Fish

Heavy metal levels in fish are usually calculated on a wet weight basis, rather than a dry weight basis. For 11 species of Savannah River fish the dry weight ranged from 23 to 33% of the corresponding wet weight (i.e., water content of 67–77%). Thus for the same samples, concentrations expressed on a wet weight basis are one-fourth to one-third of the same mercury content expressed on a dry weight basis, although in some fish the ratio may be as high as one-fifth (Burger, Gaines, Boring et al., 2001). While drying fish for dry weight analysis results in complete moisture loss, cooking for human consumption removes only some of the moisture.

Deep-frying of raw fish resulted in moisture losses. However, the fillets deep-fried with breading weighed more than those without breading, no doubt due to the weight of the breading itself. When fish is deep-fried, it loses moisture, but gains weight from the breading and from the uptake of oil.

There was an apparent increase in mercury concentration in the deep-fried fish (largely because when calculated on the basis of the weight of the sample, the fried fish had lost moisture but retained the same amount of mercury). The end result of moisture loss in fish on contaminant levels is that levels are higher in cooked fish than fresh fish (on a wet weight basis of the portion itself). If drying takes a 100-g fillet to 20 g, but retains the same level of mercury, then the concentration of mercury will be five times as high in the dried fish as the wet weight fish, even though the actual amount has not changed. For example, if deep-frying results in a water loss of 50% (the weight of a fillet goes from 100 to 50 g), the concentration of mercury in the average fillet would increase from $0.45\mu\text{g}/\text{g}$ to $0.9\mu\text{g}/\text{g}$ while the mass would remain the same (45 μg). If, during the process of deep frying, the fillet absorbed 9 g cooking oil, the fillet would weigh 59 g, and the concentration of mercury would be $0.76\mu\text{g}/\text{g}$ —the average for a fried fillet in this study.

Ecologists interested in mercury levels in fish regularly use dry weight/wet weight conversion factors of 4 to 5 when determining the toxic dose for the fish themselves. While the conversion factor between raw and cooked fish is not as high as between laboratory dried and raw fish, it still is a factor that should be considered.

In largemouth bass, mercury levels were 45–75% higher in the cooked fillets compared to uncooked fillets (on a wet weight basis), which is slightly higher than the range reported by Morgan et al. (1997) for fish from Lake Superior. In some of the samples, mercury concentrations were twice as high in cooked as in uncooked fish.

Risk Assessment: Fish from L Lake

The mercury in the fish from L Lake, averaging 436 ± 43 ppb in raw fish, can be used to compute the risk to fishermen if they ate these fish. While at present fishing is not allowed on L Lake, the possibility exists for illegal fishing, and future land use scenarios for SRS include recreational fishing. Fishermen often catch more than a single fish and even if they fish infrequently, may take home enough fish for several meals (Burger, Stephens et al., 1999). The data from this study clearly indicate that at this time, there would be a risk to people consuming largemouth bass from L Lake more frequently than once a month. From a risk management perspective, it means that DOE officials should be diligent about preventing fishing on this site, and aware of the risk in any future land use decisions, until mercury levels in fish decline substantially.

Risk Assessment: Raw Versus Cooked Fish

The primary objective was to understand the effect of local cooking methods on mercury in largemouth bass, one of the preferred local fishes (Burger, 1998). The data from this study clearly indicate that the concentration of mercury (on a wet weight basis) in uncooked fish is less than in cooked fish (for the same portion size). The actual conversion factor will depend upon the species of fish and the cooking method. While it is clear from these data that a conversion factor should be used in risk assessment, there are several questions that need to be answered for adequate risk assessment and management, including: (1) Are estimates of meal size or amount eaten based on cooked or uncooked fish? (2) Are contaminant values based on cooked or uncooked fish? (3) What are the conversion values for a specific fish and a specific cooking method? (4) What are the relative contributions of the different fish species and cooking methods to the total fish consumption of people eating wild-caught fish?

Morgan et al. (1997) suggested using food preparation factors in risk assessment, but these have generally not been applied. Preparation factors (mercury concentration in cooked fish/mercury concentration in raw fish) in their study generally ranged from 1.3 to 1.6 for fillets from Great Lakes fish, compared to 1.5 to 1.8 for largemouth bass in this study. These two studies suggest that a preparation conversion factor of 2 would be a suitable, protective default. Thus, risk assessors who do not take into account cooking method, but use contaminants data from raw fish, may be overestimating safe consumption levels. This factor should be considered by state agencies setting consumption levels for high risk populations.

The risk assessment presented in this paper for largemouth bass from the Savannah River Site indicates that at the mean, median, and 75th and 95th

percentile consumption levels, black and white men and women exceed the U.S. EPA reference dose. Translated into consumption levels, it means that people should not eat an 8-ounce portion of raw bass more than every 17 d, or an 8-ounce portion of cooked fish every 24–29 d. The average meal size of South Carolina fishermen interviewed was actually close to 10 ounces per meal (Burger, Stephens et al., 1999). In Table 4 a suggested interval for fish consumption is provided. If risk assessors are basing their risk estimates on mercury in raw fish (the usual practice) and consumers estimate their meal size from cooked fish (the usual practice), then the risk estimates are biased downward. This is counterbalanced by the fact that the RfD is based on methylmercury, while the analytic results are based on total mercury, and the risk calculations assume 100% is methylmercury. Since methylmercury actually comprises about 90–95% of the total mercury in fish, this adds an additional 5–10% protection.

Calculations of the RfD and other criteria values incorporate one or more safety or uncertainly factors to assure protection of sensitive individuals. Thus, what appears unacceptably risky to a public health agency does not necessarily mean that an individual's health will be measurably impacted.

Finally, it is clear that the mass of mercury in the fillet itself has not changed; what has changed between raw and cooked fish is the perception of the quantity of fish consumed. The human health risk is based on the dose (mass of mercury). However, if people estimate their consumption based on cooked fish but risk assessors compute risk on raw fish, the estimates are underestimates of the actual risk. Another implication of this research is that when people are asked about fish consumption, whether their answers are based on cooked or uncooked fish should be clearly stated. Unlike fishermen, people who purchase fish in stores may have weighed fillets and may estimate their intake on the basis of precooked weights.

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