

COMPARING PCDDs, PCDFs, AND DIOXIN-LIKE PCBs IN FARM-RAISED AND WILD-CAUGHT CATFISH FROM SOUTHERN MISSISSIPPI

Ferriby, LL¹; Williams, ES¹; Luksemburg, WJ²; Paustenbach, DJ³; Haws LC⁴; Birnbaum, LS⁵; and Harris, MA¹

¹ChemRisk, Houston, TX; ²Vista Analytical Laboratory, El Dorado Hills, CA; ³ChemRisk, San Francisco, CA; ⁴ChemRisk, Austin, TX; ⁵U.S. Environmental Protection Agency, Research Triangle Park, NC

Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) [hereafter referred to as “dioxin-like compounds”] are persistent environmental contaminants that have been found to be ubiquitous in environmental media and biota. Because of their persistence and widespread distribution in the environment, humans are exposed via a number of different exposure pathways. While non-occupational exposure to dioxin-like compounds can occur through the inhalation of ambient air and incidental ingestion of water and soil, the most important exposure pathway for the general population is the consumption of food and food products, primarily meat, fish and dairy products.¹ Today, well over 90% of the daily intake of PCDDs, PCDFs and PCBs for the majority of the population has been estimated to come from food products.^{2,3}

In order to better understand potential exposures to dioxin-like compounds, both the U.S. Food and Drug Administration (USFDA) and the U.S. Environmental Protection Agency (USEPA) have conducted large, nationwide studies to assess the levels of PCDD/Fs and PCBs in fish.^{4,5,6} Independent studies by Schechter and coworkers have also characterized the levels of these compounds in fish,^{7,8,9} while other researchers have conducted more species specific studies, especially for farm-raised catfish.¹⁰⁻¹³ Unfortunately, however, very little recent data are available regarding the levels of dioxin-like compounds in wild catfish in the U.S. Furthermore, much of the data in the published literature was collected more than ten years ago limiting the ability to establish current background exposures through consumption of this particular food source for the general U.S. population.

Given the relatively high consumption rates of catfish in the U.S., especially in states in the southeast, it is believed that catfish could contribute appreciably to the dietary intake of PCDD/Fs and dioxin-like PCBs. For example, research conducted at Mississippi State University estimated the total consumption of catfish to be 281 million pounds and 136 million pounds per year for the U.S. and south central populations, respectively.¹⁴ With states such as Arkansas and Mississippi leading the U.S. in average per capita consumption of catfish at 5.95 and 4.61 pounds per person per year, respectively, there is a clear need to characterize the concentration of PCDD/Fs and dioxin-like PCBs in catfish, particularly in the south central states.¹⁴ In this study, we quantified the current PCDD, PCDF, and dioxin-like PCB levels in tissue from a number of wild-caught and farm-raised catfish collected throughout Southern Mississippi which appears to represent the most substantial assessment conducted to date.

Materials and Methods

Sixty-one wild-caught and farm-raised catfish samples from Southern Mississippi were collected in March 2006 and three farm-raised catfish samples from the same region were collected in May 2005. Wild catfish (n=33) were caught by local fisherman along the Mississippi, Pearl, and Leaf Rivers. Samples were collected in one location along the Mississippi River (MR), two locations along the Pearl River (PR and the Ross Barnett Spillway or RBS), and two locations along the Leaf River (LR1 and LR2). Immediately after the fish were caught they were measured for length, weighed, and then filleted using clean knives and glass plates. Final filet weights were also recorded prior to packaging. Farm-raised catfish samples (n=31) were purchased either directly from a Mississippi farm or from local seafood markets and/or grocery stores (ten stores visited) that obtain catfish from farms in Mississippi. All samples were wrapped individually in aluminum foil (shiny side out), placed in labeled plastic bags and frozen on dry ice in uncontaminated coolers. Samples were kept frozen until analysis.

Fish tissue samples were analyzed by Vista Analytical Laboratory (El Dorado Hills, CA) for the 17 laterally-substituted PCDD/Fs and 12 dioxin-like PCBs using EPA Methods 1613 and 1668, respectively. Samples with concentrations below the limit of detection (LOD) were assumed to have a concentration equal to the LOD divided by the square root of two. PCDD/F, PCB, and total TCDD TEQs for individual samples were calculated by summing the product of each congener's concentration and the associated 1998 World Health Organization toxic equivalency factor.¹⁵ All data analyses were conducted using Microsoft Excel. The mean, median, and 25th and 75th percentile TEQ concentrations were characterized by fish type (wild-caught and farm-raised), sample type (fillet, whole, dressed, and nugget) and

Levels in feed and food (fish)

collection site. Ninety-five percent confidence intervals for the mean TEQ levels were also calculated using Microsoft Excel. All TEQ concentrations are presented as wet weight unless otherwise noted.

Results and Discussion

Table 1 presents descriptive data for the various types of fish and types of samples collected. A total of 17 blue, 3 appaloosa, and 1 willow catfish were filleted for tissue analysis. As expected, farm-raised catfish fillets were larger than wild-caught fillets and had a higher average lipid fraction.

Table 1: Descriptive data for wild-caught and farm-raised catfish collected in Southern Mississippi

Species Collected		Avg. % Lipid	Avg. Tissue Weight ^a	Avg. Fish Weight ^a	Avg. Fish Length ^b
Wild-Caught	Channel, Blue, Appaloosa, Willow	1.53	6.92	18.63	14.10
Fillet	Blue, Appaloosa, Willow	1.78	9.51	24.65	16.04
Whole	Channel	0.87	2.56	2.56	8.94
Farm-Raised	Channel ^c	7.97	13.07	---	---
Fillet	Channel ^c	8.20	11.67	---	---
Dressed	Channel ^c	4.92	17.44	---	---
Nuggets	Channel ^c	8.51	20.88	---	---

^aMeasured in ounces

^bMeasured in inches

^cAssumed – catfish farmers primarily raise channel cats due to their hardiness and growth rate.

Table 2 shows the mean, range and various percentiles of total measured concentrations of PCDDs, PCDFs, and dioxin-like PCBs. As seen in Table 2, concentrations ranged from 0.13 to 4.96 pg TEQ/g wet wt for wild-caught fish and from 0.15 to 2.56 pg TEQ/g wet wt for farm-raised fish, with average values of 1.50 and 0.98 pg TEQ/g wet wt and median values of 1.41 and 0.97 pg TEQ/g wet wt, respectively. Surprisingly, there was no statistically significant difference between the mean TEQ for wild-caught and farm-raised catfish fillets ($p=0.11$). Although not significantly different ($p=0.45$), whole wild-caught fish had a slightly higher mean TEQ than wild-caught fillets even though the whole fish were appreciably smaller in size than the fish from which the fillets were taken (Table 1). Further examination of the relationships between total TEQ concentration and the length and weight of the individual fish indicated that fish weight is a moderate predictor of TEQ concentration ($p=0.07$); however, in this study fish length was not predictive of total TEQ fish concentrations ($p=0.30$). Regardless, more comprehensive analyses will be needed to determine specifically what characteristics impact TEQ concentrations in catfish from this region of the country.

Table 2: TEQ summary statistics for the 29 dioxin-like congeners by fish type and sample type.

	N	Mean	95% CI	TEQ (pg/g)			Range
				25 th Percentile	Median	75 th Percentile	
All Samples	64	1.25	1.01 - 1.49	0.43	1.17	1.71	0.13 - 4.96
Wild-Caught	33	1.50	1.13 - 1.88	0.68	1.41	1.73	0.13 - 4.96
Fillet	24	1.39	1.02 - 1.76	0.63	1.54	1.74	0.13 - 3.43
Whole	9	1.80	0.85 - 2.76	0.91	1.28	1.68	0.64 - 4.96
Farm-Raised	31	0.98	0.72 - 1.24	0.22	0.97	1.56	0.15 - 2.56
Fillet	25	0.99	0.69 - 1.29	0.21	0.98	1.51	0.15 - 2.56
Dressed	2	1.42	0.54 - 2.30	1.19	1.42	1.64	0.97 - 1.87
Nuggets	4	0.64	0.00 - 1.36	0.27	0.33	0.69	0.16 - 1.74

Figure 1 illustrates the total TEQ concentration distribution for the wild-caught catfish by collection site. These box plots identify the median, average and range of concentrations (not the 5-95% distribution). Interestingly, the Ross Barnett Spillway, which is part of the Pearl River just to the northeast of Jackson, had fish with some of the lowest tissue concentrations while fish collected from the Pearl River near Picayune were relatively higher. This may be due to downstream effects, the possibility of point sources near to where the fish were collected, and even the age of the fish collected. With regard to the Leaf River, the second location not only had the highest concentrations of PCDD/Fs and

Levels in feed and food (fish)

dioxin-like PCBs, but also had the widest range of tissue concentrations. Even more importantly, these fish were some of the smallest collected indicating a potential source between the first collection site along the Leaf River (which was upstream of LR2) and the second collection site. For the Mississippi River, tissue levels were on average, lower than expected. This may be due to several factors including the larger volume of water in this river and the rapid currents compared to those of smaller and shallower rivers such as the Leaf and Pearl.

As shown in Figure 2, with the exception of the Mississippi River, PCDDs were the main contributors to the mean total TEQ for each collection site. For the Mississippi River, PCBs contributed most to the overall TEQ followed by PCDDs and PCDFs. More noteworthy are the differences in the relative amounts of PCDDs, PCDFs and PCBs that contributed to the mean total TEQ fish concentrations for the different sites on the Leaf River. While the relative contributions of PCDD/Fs decreased from LR1 to LR2, the contribution of PCBs significantly increased. A similar trend is seen with the Ross Barnett Spillway and the Pearl River; however, the differences in relative contributions are smaller than those seen with the Leaf River sites. These results indicate that TEQ concentrations in wild-caught and farm-raised fish are generally similar, but TEQ concentrations and the contribution of PCDDs, PCDFs and PCBs to the mean TEQ concentration vary by collection site for wild-caught catfish.

Figure 1: Boxplots of total TEQ concentrations in wild-caught catfish from the different collection sites.

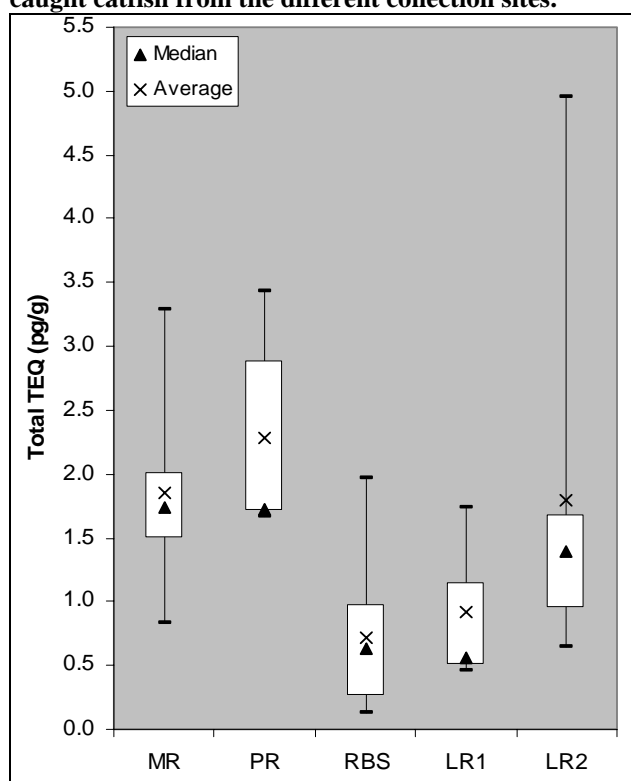
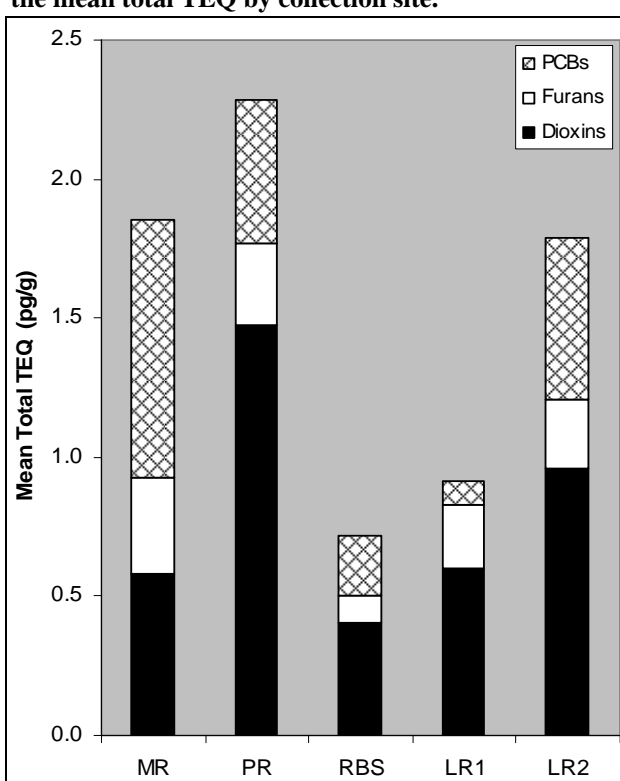


Figure 2: Contribution of dioxins, furans, and PCBs to the mean total TEQ by collection site.



When comparing these results to those for fish from a primarily urbanized area,¹⁶ the TEQ concentrations in fish tissue from rural Southern Mississippi had a much smaller range of values (0.13 – 4.96 vs. 0.4 – 41 in Suarez et al.¹⁶). However, the average and median TEQ concentrations for samples collected in the highly industrialized Houston ship channel were only 3.5 and 2.5 times greater than the concentrations found in rural Mississippi, suggesting that little difference exists between vastly urbanized areas and those that are relatively rural.

A direct comparison of catfish nugget concentrations reported in Fiedler et al.¹² with data for our farm-raised nuggets indicate a continuation of the downward trend of PCDD/F and PCB levels in food products initially reported by several researchers in the mid-1990s.^{17,18,19} The continued decrease in PCDD/PCDF concentrations in blood over the past 30 years has also been well documented.²⁰ While the mean concentration for the 1997 study was 2.85 pg TEQ/g wet wt, the mean concentration for farm-raised catfish nuggets analyzed in this study was significantly lower ($p=0.02$) at 0.64 pg TEQ/g wet wt. These results provide a reasonable data set describing the current background levels of PCDDs,

Levels in feed and food (fish)

PCDFs and dioxin-like PCBs in catfish representative of the Southeastern region of the United States and will allow for the determination of current background intake of these chemicals due to this particular food source. It should be noted that prior work has shown that cooking practices will often lower the actual quantity of PCDD/Fs and PCBs in the prepared foods which are ingested and this must be considered in any risk assessment.^{21,22}

Acknowledgements

We would like to thank the local fishermen and women of southern Mississippi for their efforts in helping us obtain catfish for this study. Special thanks are also given to Ken Unice for his statistical insight and help with data quality verification. Funding for the analyses described in this paper was provided in part by Joslyn Manufacturing, Inc. and Liberty Mutual who are currently involved in litigation associated with the dioxins/furans. Substantial additional funding was provided by ChemRisk, Inc. and Vista Analytical Laboratory. The contents of this paper reflect the opinions and view of the authors and do not represent the official views of the USEPA. The mention of trade names or commercial products does not constitute endorsement or recommendation for use.

References

1. Agency for Toxic Substances and Disease Registry (ATSDR). Toxicological profile for chlorinated dibenzo-*p*-dioxins. Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service, 1998.
2. Agency for Toxic Substances and Disease Registry (ATSDR). Toxicological profile for polychlorinated biphenyls (PCBs). Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service, 2000.
3. U.S. Environmental Protection Agency (USEPA). *Draft exposure and human health risk assessment of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) and related compounds, Parts I, II and III*. Washington, D.C.: U.S. Environmental Protection Agency, Office of Research and Development, National Center for Environmental Assessment, Exposure Assessment and Risk Characterization Group, 2003. <http://www.epa.gov/ncea/pdfs/dioxin/nas-review/>.
4. Jensen E, Bolger PM. *Food Addit Contam* 2001; 18:395-403.
5. Jensen E, Canady R, Bolger PM. *Organohalogen Comp* 2000; 47:318-321.
6. U.S. Environmental Protection Agency (USEPA). National study of chemical residues in fish. Washington, D.C.: Office of Science and Technology. EPA/823-R-02-008, 1992.
7. Schechter A, Pöpke O, Ball M, Startin JR, Wright C, Kelly M. *Organohalogen Comp* 1993; 13:97-100.
8. Schechter A, Cramer P, Boggess K, Stanley J, Olson JR. *Chemosphere* 1997; 34:1437-1447.
9. Schechter A, Cramer P, Boggess K, Stanley J, Pöpke O, Olson J, Silver A, Schmitz M. *J Toxicol Environ Health, Part A* 2001; 63:1-18.
10. Cooper K, Fiedler H, Bergek S, Andersson R, Hjelt M, Rappe C. *Organohalogen Comp* 1995; 26:51-57.
11. Cooper K, Bergek S, Fiedler H, Hjelt M, Bonner M, Howell F, Rappe C. *Organohalogen Comp* 1997; 28:197-202.
12. Fiedler H, Cooper KR, Bergek S, Hjelt M, Rappe C. *Chemosphere* 1997; 34:1411-1419.
13. Fiedler H, Cooper K, Bergek S, Hjelt M, Rappe C, Bonner M, Howell F, Willett K, Safe S. *Chemosphere* 1998; 37:1645-1656.
14. Dean S, Hanson T, Murray S. Economic Impact of the Mississippi farm-raised catfish industry at the year 2003. Publication 2317: Mississippi State University Extension Service, Starkville, MS. 2003.
15. van den Berg M, Birnbaum L, Bosveld ATC, Brunström B, Cook P, Feeley M, Giesy JP, Hanberg A, Hasegawa R, Kennedy SW, Kubiak T, Larsen JC, van Leeuwen FXR, Liern AKD, Nolt C, Peterson RE, Poellinger L, Safe S, Schrenk D, Tillitt D, Tysklind M, Younes M, Wærn F, Zacharewski T. *EHP* 1998; 106:775-792.
16. Suarez MP, Rifai HS, Palachek RM, Dean KE, Koenig L. *Environ Engin Sci* 2005; 22:891-906.
17. Furst P, Wilmers K. *Organohalogen Comp* 1995; 26:101-104.
18. Liem AKD, de Jong APJM, Theelen RMC, van Zorge JH. Occurrence of dioxins and related compounds in Dutch foodstuffs – Part I: Sampling strategy and analytical results. 11th International Symposium on Chlorinated Dioxins and Related Compounds, Research Triangle Park, NC, Abstract P-156: 1991, p. 365.
19. Mayer R. *Organohalogen Comp* 1995; 26:109-111.
20. Aylward LL, Hays SM. *J Expo Anal Environ Epidemiol* 2002; 12:319-328.
21. Schechter A, Dellarco M, Pöpke O, Olson J. *Chemosphere* 1998; 37:1723-1730.
22. Hori T, Nakagawa R, Tobiishi K, Iida T, Tsutsumi T, Sasaki K, Toyoda M. *J Agric Food Chem* 2005; 53:8820-8828.