PCDDs, PCDFs AND PCBs IN FARM RAISED CATFISH FROM SOUTHEAST UNITED STATES

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Abstract

Nine catfish fillets, three catfish nuggets, two feed samples and one pond sediment were analyzed for PCDDs, PCDFs and PCBs. Farm raised catfish from Mississippi, Alabama, and Arkansas contained significant levels of 2,3,7,8-substituted PCDDs and PCDFs. In addition, a large number of non-2,3,7,8-substituted congeners were present in all samples. The catfish fillets and catfish nuggets also contained high concentrations of dioxin-like PCBs, as well as a number of non-dioxin-like PCBs. The toxic equivalent due to PCDDs and PCDFs ranged from 9.5 to 43.0 pg/g lipid whereas the TEQ from the dioxin-like PCBs was from 0.45 to 4.9 pg/g lipid. The major source for the PCDDs and PCBs appears to be from feed and not from the pond sediment.

Key Words Catfish, polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, polychlorinated biphenyls, non-2,3,7,8-substituted congeners, catfish feed.

1 INTRODUCTION

For the general population, food consumption is the major route of exposure (> 90% of daily intake) for polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/PCDFs).[1)] In a previous study examining PCDDs/PCDFs levels in food items from local supermarkets in southern Mississippi, USA, catfish nuggets were found to contain the highest concentration of 2,3,7,8-TCDD (0.30 to 0.83 pg/g wet wt.) of all the food items analyzed.[2,3)] In this prior study, we also determined that the catfish contained many non-2,3,7,8-substituted congeners.[2,3)] In 1995, we followed up on our initial catfish study in order further to determine: (a) if our earlier results could be reproduced; (b) if whole catfish fillets had similar PCDDs/PCDFs levels and congener profiles as the catfish nuggets of the same brand; (c) if whole catfish fillets from different sources

had similar PCDDs/PCDFs levels and congener profiles; (d) the PCDDs/PCDFs source by analyzing catfish feed and pond sediment; and (e) the concentrations of PCBs.

2 EXPERIMENTAL

We collected 15 samples for this study in 1995 (see Table 1). Three catfish nuggets (brand A) and three catfish fillets (brand A) were purchased from the same store and were distributed by the same supplier as those collected and analyzed in our earlier study. [2,3] In addition, one catfish fillet (brand B) was obtained from an Alabama supplier, and three catfish fillets were obtained from an aquaculture facility in Stoneville, MS. Of the three catfish from the Stoneville facility, one sample was pond-raised on a 4-8% fish meal diet (i.e., ground up fish); the second was from pond-raised fish fed a 4% fish meal diet; and the third was from pond-raised fish fed a 0% fish meal diet. One sample of catfish feed was collected from the Stoneville facility and contained approximately 4% fish meal. One sample of catfish pond sediment was collected from the Stoneville facility pond in which the 4-8% fish meal fed catfish were raised. Two catfish fillets were obtained from an aquaculture facility in Stuttgart, AK. One of the Stuttgart fish was raised in a pond and fed an 8% fish meal diet; the second was raised in a glass aquarium and fed an 8% fish meal diet. One sample of catfish feed was collected from Stuttgart and contained approximately 8% fish meal.

Table 1: Sample identification

Type of Sample	Sample No.	Description
Brand A - Nuggets	1	Catfish nuggets *, MS
	2	Catfish nuggets *, MS
	3	Catfish nuggets *, MS
Brand A - Fillet	1	Catfish fillet, MS
	2	Catfish fillet, MS
	3	Catfish fillet, MS
Brand B - Fillet		Catfish fillet, AL
MS Commercial	1	Catfish fillet, raised on 4-8 % fish meal
	2	Catfish fillet, raised on 4 % fish meal
	3	Catfish fillet, raised on 0 % fish meal
AK Commercial	1	Catfish fillet, raised on 8 % fish meal
	2	Catfish fillet; raised in glass aquarium
Feed MS		Fish feed, MS
Feed AK		Fish feed, AK
MS pond sediment		Sediment from pond of MS Commercial 1

* Nuggets are small pieces of fillet

Collection and analysis for PCDDs/PCDFs for the catfish nuggets, fillets and feed were performed as previously described.[2,3] Collection and analysis for PCDDs/PCDFs of the pond sediment was carried out as previously described.[4] In addition, all samples were analyzed for the following twelve PCB congeners: PCB

28, PCB 52, PCB 77, PCB 101, PCB 105, PCB 118, PCB 126, PCB 138, PCB 153, PCB 156, PCB 169, and PCB 180. The methods used for the fractionation and analysis of PCBs was a modification of the procedures described by van Bavel [5] and will be reported elsewhere. Separation and quantification were performed using HRGC/HRMS and $^{13}C_{12}$-labelled internal standards.

3 RESULTS

The results for the PCDDs, PCDFs, PCBs, TEQ (PCDDs/PCDFs), TEQ (PCB), and TEQ (PCDDs/PCDFs/PCBs) for all catfish nugget and fillet samples (pg/g lipid), feed (pg/g lipid) and pond sediment (pg/g dry matter) are reported in Table 2. To calculate the TEQ for dioxin-like PCBs (designated * in Table 2), the method established by WHO/IPCS was used. [6] Non-quantifiable levels (ND) with a limit of quantification (LOQ) for the individual congeners are reported in parentheses. One half of LOQ was used to calculate the I-TEQ.

All 2,3,7,8-substituted PCDDs were quantified in all samples. The Mississippi catfish nuggets and fillets had a range of 2.1-4.7 pg/g lipid of 2,3,7,8-TCDD. The two Arkansas catfish fillets, however, had 2,3,7,8-TCDD concentrations of 32 and 27 pg/g lipid. In general, the 2,3,7,8-substituted PCDFs were lower than the 2,3,7,8-substituted PCDDs in all catfish samples. The 2,3,7,8-substituted PCDFs were lower in all commercial catfish fillets collected at the Stoneville aquaculture facility (the point of origin prior to processing and shipment) compared to nuggets and fillets.

PCB 77 ranged from 22-49 pg/g lipid for all Mississippi catfish samples and one Arkansas catfish sample. The second Arkansas catfish sample had a PCB 77 concentration of 240 pg/g lipid. PCB 126 ranged from 3.5-36 pg/g lipid for all catfish in the study. The second Arkansas catfish sample had a PCB 126 concentration of 36 pg/g lipid. As expected, all catfish in this study, except the second Arkansas sample, contained the highest concentrations of PCBs for congeners 153 and 138, respectively. The second Arkansas catfish sample contained an unusually high concentration of PCB 28 (29,000 pg/g lipid).

The TEQs (PCDDs/PCDFs) for all catfish samples ranged from 5.5-42 pg/g lipid. The TEQs (PCBs) were in the range 0.45-4.9 pg/g lipid. Finally, the total TEQs (PCDDs/PCDFs/PCBs) ranged from 6.55-44.94 pg/g lipid.

The Mississippi feed was consistently lower for all 2,3,7,8-substituted PCDDs than the Arkansas feed. For example, the Mississippi feed contained 2.7 pg/g lipid of 2,3,7,8-TCDD and the Arkansas feed contained 44 pg/g lipid of this congener. The opposite was observed for the PCBs, where the levels in the Arkansas feed were consistantly much lower than the Mississippi feed. The total TEQs (PCDDs/PCDFs/PCBs) for Mississippi and Arkansas feed were 10.51 and 61.19 pg/g lipid, respectively.

Table 2: Summary of PCDD, PCDF, and PCB analyses. Sample identification, see Table 1. Concentrations in pg/g lipid, LOQ in parentheses

Type of Sample	Brand A Nuggets			Brand A Fillet			Brand B	MS Commercial			AK Commer.		Feed		Sediment
Sample No.	1	2	3	1	2	3	Fillet	1	2	3	1	2	MS	AK	(d.m.)
2,3,7,8-TCDD	3.2	2.7	2.5	3.0	4.1	4.7	2.4	2.2	2.6	2.1	32	27	2.7	44	0.17
1,2,3,7,8-PeCDD	5.1	4.9	5.0	12	15	16	9.5	3.6	8.4	4.9	16	14	3.1	19	0.42
1,2,3,4,7,8-HxCDD	4.9	4.5	4.5	12	16	16	9.9	1.9	6.9	4.0	1.4	1.9	2.4	2.9	0.72
1,2,3,6,7,8-HxDD	7.6	7.1	7.0	17	23	25	15	4.2	12	6.2	5.7	6.1	4.0	8.9	1.7
1,2,3,7,8,9-HxCDD	6.8	5.9	6.1	11	15	18	11	2.3	8.4	4.6	14	15	4.5	39	2.6
1,2,3,4,6,7,8-HpCDD	47	42	47	97	130	140	96	11	67	49	8.8	10	43	65	63
OCDD	370	310	370	650	990	880	480	48	900	610	49	64	580	1300	1700
2,3,7,8-TCDF	1.1	1.2	1.2	1.2	1.1	1.2	0.68	0.71	0.29	0.24	0.70	0.45	2.4	0.18	0.27
1,2,3,7,8-PeCDF	0.59	0.87	0.78	1.7	1.6	2.4	0.29	(0.40)	0.20	(0.21)	0.33	0.26	0.42	(0.249	0.13
2,3,4,7,8-PeCDF	1.9	2.2	2.1	4.2	5.4	7.3	0.62	0.58	0.32	0.22	0.76	0.74	0.99	(0.25)	0.14
1,2,3,4,7,8-HxCDF	2.3	2.9	2.7	3.4	3.9	5.6	0.47	(0.66)	(0.20)	(0.30)	(0.38)	(0.38)	(0.94)	(0.33)	0.54
1,2,3,6,7,8-HxCDF	1.2	1.4	1.5	2.2	3.0	4.4	0.31	(0.55)	(0.17)	(0.25)	(032)	(0.31)	(0.78)	(0.28)	0.28
1,2,3,7,8,9-HxCDF	(0.25)	(0.24)	(0.38)	0.26	(0.28)	0.52	(0.31)	(0.76)	(0.24)	(0.34)	(0.44)	(0.43)	1.1	(0.39)	(0.25)
2,3,4,6,7,8-HxCDF	2.3	2.7	2.5	4.2	5.4	7.9	0.47	(0.69)	(0.22)	(0.31)	(0.40)	(0.39)	(0.99)	(0.35)	0.25
1,2,3,4,6,7,8-HpCDF	2.1	2.1	2.1	3.0	3.0	4.8	1.1	0.83	0.44	0.44	0.67	1.1	2.4	1.1	5.3
1,2,3,4,7,8,9-HpCDF	(0.3)	(0.32)	(0.53)	(0.29)	(0.37)	(0.28)	(0.43)	(1.0)	(0.32)	(0.46)	(0.61)	(0.58)	(1.4)	(0.52)	(0.41)
OCDF	0.53	(0.40)	(0.68)	0.87	0.92	1.0	0.76	(1.4)	0.71	(0.55)	(0.77)	(0.70)	3.4	1.3	6.4
TEQ (PCDD/PCDF)	***10***	***9.6***	***9.5***	***18***	***23***	***27***	***13***	***5.5***	***11***	***7.3***	***43***	***37***	***7.2***	***61***	***3.5***
PCB 77 (33',44') *	41	45	44	49	49	49	36	29	22	22	42	240	180	7.5	1.9
PCB 126 (33'44'5) *	11	11	11	19	22	27	6.8	8.0	8.1	3.5	14	36	23	1.3	0.36
PCB 169 (33'44'55') *	3.3	3.4	3.7	5.9	6.7	8.2	2.8	2.4	1.5	0.68	3.4	6.3	4.3	0.52	0.33
PCB 105 (233'44') *	390	410	410	430	450	560	320	380	430	140	730	2300	1300	64	7.6
PCB 118 (23'44'5) *	1300	1400	1400	1400	1500	1900	1000	1100	1300	410	2300	5700	4700	200	18
PCB 156 (233'44'5) *	160	170	170	150	160	190	110	120	140	43	310	560	470	36	2.8
PCB 180 (22'344'55') *	1300	1400	1500	1000	1100	1400	700	750	850	280	2200	3300	4100	190	20
PCB 28 (244')	410	410	420	610	610	690	570	590	480	420	960	29000	1300	180	15
PCB 52 (22'55')	920	950	950	1000	1000	1300	900	860	1100	470	1300	13000	3600	76	16
PCB 101 (22'455')	2000	2000	2000	1800	1900	2400	1400	1400	1700	540	2900	5700	6800	99	21
PCB 138 (22'33'56)	3100	3300	3300	2600	2900	3700	1800	2100	2300	680	5400	9200	9500	380	35
PCB 153 (22'44'55')	3500	3700	3700	2900	3200	4000	1900	2200	2300	690	5700	9400	11000	410	30
TEQ (PCB)	***1.42***	***1.44***	***1.44***	***2.25***	***2.58***	***3.16***	***0.92***	***1.05***	***1.09***	***0.45***	***1.94***	***4.9***	***3.31***	***0.19***	***0.04***
TEQ (PCDD/PCDF/PCB)	**11.42**	**11.04**	**10.94**	**20.25**	**25.58**	**30.16**	**13.92**	**6.55**	**12.09**	**7.75**	**44.94**	**41.90**	**10.51**	**61.19**	**3.54**

4 DISCUSSION

In this study we were able to reproduce the results we previously reported on the concentrations of PCDDs and PCDFs in farm raised catfish.[3] In our earlier study, we reported 2,3,7,8-TCDD concentrations in three Mississippi catfish nuggets ranging from 2.5-8.8 pg/g lipid. In our current study, we quantified 2,3,7,8-TCDD ranging from 2.5-3.2 pg/g lipid in three Mississippi catfish nuggets from the same supplier. In addition, three catfish fillets from the same Mississippi supplier contained 2,3,7,8-TCDD levels ranging from 3.0-4.7 pg/g lipid. Further verification was demonstrated in three catfish obtained from a Mississippi commercial supplier where we quantified 2,3,7,8-TCDD from 2.1-2.6 pg/g lipid. Similarly, the TEQ that we observed in our earlier study ranged from 10.2-27.8 pg TEQ/g lipid for the three Mississippi catfish nuggets. In the present study, the three nuggets from the same supplier had TEQs (PCDDs/PCDFs) ranging from 9.5-10 pg TEQ/g lipid. The Mississippi fillets from the same supplier ranged from 18-27 pg TEQ (PCDDs/PCDFs)/g lipid. The three catfish obtained from the commercial supplier contained TEQ (PCDDs/PCDFs) ranging from 5.5-11 pg TEQ/g lipid.

The source of the PCDDs and PCDFs in the Mississippi and Arkansas catfish appears to be from the catfish feed. For example, Mississippi catfish feed was used to feed the catfish at the Stoneville facility. Approximately five tons of catfish feed is used per production acre of water per year.[7] The MS Commercial 1 sample was raised in a pond where catfish were fed the Mississippi feed we analyzed (see Table 2). There is good agreement with all PCDDs and PCDFs congeners, with the exception of OCDD and 1,2,3,4,6,7,8-HpCDD, between the feed and the catfish. In contrast, the sediment from the pond where MS Commercial 1 was raised had significantly lower levels of most 2,3,7,8-substituted PCDDs. In addition, while MS Commercial 1 had mostly non-detectable 2,3,7,8-substituted PCDFs, the pond sediment had detectable levels of all these congeners except 1,2,3,7,8,9-HxCDF and 1,2,3,4,7,8,9-HpCDF.

The contribution of the PCBs to the total TEQs (PCDDs/PCDFs/PCBs) for all catfish samples ranged from approximately 4-16%. The Arkansas feed had a PCB contribution to the total TEQ of less than 0.5%. The Mississippi feed, on the other hand, had a PCB contribution to the total TEQ of 31.5%.

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