調味料で味付した魚と魚を味付に使用した調味料中の多環芳香族炭化水素の定量

〔東京家政大学研究紀要　第41集　(2), p.45～56, 2001〕

調味料で味付した魚と魚を味付に使用した調味料中の多環芳香族炭化水素の定量

舘野　つや子

(平成12年10月5日受理)

Polycyclic Aromatic Hydrocarbons Resulted from Soak Fish and Soaked Seasoning

Tsuyako TATENO

(Received on October 5, 2000)

キーワード：多環芳香族炭化水素，ベンゾ(a)ピレン，魚，調味料

Key words : polycyclic aromatic hydrocarbons, benzo(a)pyrene, fish, seasoning

緒　言

魚類についての多環芳香族炭化水素(Polycyclic aromatic hydrocarbons:PAHと略す)の定量はOva, Gら[1]，尾花ら[2]，山崎ら[3]，Masudaら[4]及び舘野ら[5),6]，加熱した魚類のPAHについては，Kangsadalampai, K.ら[7]，Saint-Aubert, B.ら[8]及び舘野ら[9),10]，また，魚のくん製のPAHについては，Moret, Sabrinaら[11]及びWang, Guangdiら[12]が行っている．

しかし，調味料で味付した焼魚のPAH定量については，舘野[9),10]の報告以外には，ほとんど見られない．舘野[9),10]の今までの報告において調味料で味付した魚と対照として味付しない魚を試料とし，味付に用いた調味料(照り焼用及び味噌漬用)の違い，魚の油脂含有量の違い，加熱方法の違い及び加熱程度[9),10]の違いによるPAH生成について検討を行った．

その結果[9]48試料中42試料(88%)が調味料に漬け味付して加熱した試料の方が対照として味付しないで加熱した試料よりPAH検出量が低い傾向が見られた．

上記の結果をふまえ，今回は，網焼及び鉄板焼等の加熱によるものでなく，生魚試料と調味料間のPAH含有量について，調味料浸漬時の変化なども加え検討を行った．

食品衛生学第1研究室

分析方法

1　試料

魚：前報と同様の調味料に漬けて味付[13](照焼及び味噌漬焼)する魚試料として，ブリ(100g中油脂含有量約12g[14])及びカジキマグロ(100g中油脂含有量3.0g[14])の切身を用いた．

照焼用の調味料[13]：醤油215ml，砂糖70g，日本酒120ml，みりん215ml及び水60mlを混ぜたものを用いた．

味噌漬け用の調味料[13]：味噌300g，砂糖30g，日本酒50ml及びみりん50mlを混ぜたものを用いた．

各試料の魚及び調味料は平成9年2月～11年10月都内で市販されていたものを用いた．

2　試薬

n-ヘキサン(HPLC用)及びジエチルエーテル(残留農薬，PCB試験用)はいずれも和光純薬工業㈱製を用いた．他の試薬は前報[10]及び今までの報告[9),15),16]と同じものを用いた．

3　装置及び器具

ソックスレー抽出器及び液体抽出器その他のものは，いずれも前報[10]と同じものを用いた．

4　試料の調製

照焼用及び味噌漬用の2種類の調味料にブリ及びカジキマグロの①魚の切り身(70～100g)を1日～7日間冷蔵庫内で漬けて味付した試料，②味付に用いた使用済みの各調味料，また，③調味料で味付していない魚の切り身及び④魚を漬けて味付する前の照焼用及び味噌漬用の各

調味料を試料とした.

それぞれの魚試料及び魚を漬けて味付に使用した味噌漬用調味料及び対照として味付する前の味噌漬用調味料は，乾燥器(60～70℃)で乾燥したものを調製試料とした.また，魚を漬けて味付に使用した照焼用調味料は，そのままを試料(50ml)とした.

5　抽出及びPAH分析

1）試料の魚：試料の魚は前報[10]に従いソックスレーによる抽出，次いで液々抽出，カラムクロマトグラフィー及び蛍光測定を実施した.

2）調味料

味噌漬用調味料：ソックスレー抽出以下上記魚試料と同様に行った.

照り焼用調味料：液体抽出以下前報[10]に従って行った.

以上64試料は各1試料につき2回ずつ採取しそれぞれ分析を行った.

結果及び考察

1．ブリ試料(ブリ切身)についての実験結果

1）ブリを照焼用調味料に0～7日間浸漬した際のブリ試料中のPAH検出量をTable 1に，また，浸漬に使用した調味料中のPAH検出量をTable 2に示した.

ブリ試料中浸漬日数によりPAH検出量の増加傾向が見られたものは Benzo(a)pyrene, Pyrene, Anthracene, Fluoranthene, Phenanthrene 及び9, 10-Dimethylbenz(a)anthraceneであった。その他のPAHでは一定の傾向が認められなかった.

照焼用調味料中のPAH検出量では，Pyreneが調味料に含まれているが，試料の浸漬により減少傾向が見られ，調味料から試料の方へ浸漬により移行したものと推測される．また，5,12-Dihydronaphthaceneでは，調味料から試料への移行が推測される.

2）ブリを味噌調味料に0～7日間浸漬した際のブリ試料中のPAH検出量をTable 3に，また，浸漬に使用した調味料中のPAH検出量をTable 4に示した.

ブリ試料中高い検出量の PAH は Benzo (e)-pyrene, 9, 10-Dimethylbenz (a) anthracene 及び Phenanthrene であるが，前二者は，味噌調味料への浸漬により PAH 含有量に減少傾向が見られるが，Phenanthrene では減少傾向がほとんど認められない.

味噌調味料中のPAHでは，Benzo (e) pyrene, Fluoranthene, Anthracene, Phenanthrene 及び 9,10-Dimethylbenz (a) anthracene の検出量が比較的多かったが，Benzo (e) pyrene は調味料への浸漬によって減少したが，他は調味料浸漬によっても量的変化はほとんど認められなかった.

2　カジキマグロ試料(カジキマグロ切味)についての実験結果

1）カジキマグロを照焼用調味料に0～7日間浸漬した際のカジキマグロ試料中のPAH検出量をTable 5に，また，浸漬に使用した調味料中のPAH検出量をTable 6に示した.

カジキマグロ試料中高い検出量の PAH は，Benz(a) anthracene 及び Phenanthrene であるが，調味料浸漬によりPAH含有量は減少した.

調味料中のPAH検出量はTable 1と同様Pyreneの含有量が高く試料の浸漬によって減少し，試料への移行が認められた.

2）カジキマグロを味噌調味料に0～7日間浸漬した際のカジキマグロ試料中のPAH検出量をTable 7に，また，浸漬に使用した調味料中のPAH検出量をTable 8に示した.

カジキマグロ試料中高い検出量の PAH は Table 5と同様 Benz (a) anthracene 及び Phenanthrene であるが，前者は調味料浸漬によって減少しているが，後者はほとんど変化していない.

調味料中の PAH 検出量は Table 4 と同じ数値を用いたものであるが，浸漬により Phenanthrene が増加し，他のPAHについては，ほとんど変化は認められない.

まとめ

1．魚試料では，カジキマグロに Benz (a) anthracene 及び Phenanthrene の検出量が高く，その他のPAHは微量ないし検出しなかった.

2．各調味料中の PAH 検出量は微量または，検出しなかった.

照焼調味料を使用した際の魚試料中のPAH検出量が高い際には調味料中に移行して，試料中のPAHが減少する傾向があり，調味料中PAH検出量が比較的高く，魚試料中に少ない場合には，逆の移動傾向があった.

味噌調味料の場合には，上記のような傾向はほとんど認められなかった.

Table 1. Concentration of Polycyclic Aromatic Hydrocarbons in Row Yellowtail Samples
(not Soaked in Teriyaki-Seasoning and Soaked Row Yellowtail in Teriyaki-Seasoning of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked																				
				1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	ND*	ND		0.01	ND	(0.01)	0.02	ND	(0.01)	ND	ND		ND	ND		0.02	ND	(0.01)	ND	ND		ND	ND	
Benzo(a)pyrene	ND	0.03	(0.02)	0.01	0.06	(0.04)	0.03	0.06	(0.05)	0.02	ND	(0.01)	0.01	ND	(0.01)	0.03	ND	(0.02)	0.12	0.11	(0.12)	0.15	0.17	(0.16)
Dibenz(ah)anthracene	ND	ND		0.05	ND	(0.03)	0.01	ND	(0.01)	ND	ND		ND	ND		0.03	ND	(0.02)	ND	ND		ND	ND	
3-Methylcholanthrene	0.07	0.42	(0.25)	0.01	ND	(0.01)	ND	ND		0.01	0.05	(0.03)	ND	ND		0.02	ND	(0.01)	ND	ND		ND	ND	
Benzo(e)pyrene	ND	ND		1.03	1.05	(1.04)	ND	ND		ND	ND		0.36	ND	(0.18)	0.39	ND	(0.20)	ND	ND		ND	ND	
Pyrene	ND	ND		ND	ND		ND	5.01	(2.51)	3.17	ND	(1.59)	0.72	ND	(0.36)	3.80	1.60	(2.70)	3.86	1.20	(2.53)	0.69	0.50	(0.60)
Fluoranthene	ND	ND		0.37	ND	(0.19)	0.46	ND	(0.23)	0.38	ND	(0.19)	ND	0.64	(0.32)	1.23	0.53	(0.88)	0.09	ND	(0.05)	0.55	0.62	(0.59)
Anthracene	0.05	ND	(0.03)	0.02	ND	(0.01)	0.02	0.11	(0.07)	0.05	ND	(0.03)	0.02	ND	(0.01)	0.08	0.04	(0.06)	0.11	0.05	(0.08)	0.18	0.20	(0.19)
Phenanthrene	ND	ND		0.72	0.63	(0.68)	ND	ND		1.14	ND	(0.57)	ND	0.42	(0.21)	0.39	ND	(0.20)	1.95	1.05	(1.50)	0.16	0.25	(0.21)
Coronene	2.94	ND	(1.47)	ND	ND		0.01	2.61	(1.31)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		1.66	ND	(0.83)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	0.05	ND	(0.03)	ND	ND		ND	0.17	(0.09)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Perylene	0.01	0.01	(0.01)	0.01	ND	(0.01)	0.33	0.01	(0.17)	0.01	ND	(0.01)	ND	ND		0.01	ND	(0.01)	0.01	ND	(0.01)	0.02	ND	(0.01)
Dibenz(ac)anthracene	ND	ND		0.01	ND	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	ND	0.29	(0.15)	1.42	ND	(0.71)	0.12	ND	(0.06)	0.02	0.93	(0.48)	0.50	0.06	(0.28)	ND	0.13	(0.07)	0.61	0.05	(0.33)	ND	ND	
9-Methylanthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
5,12-Dihydronaphthacene	5.56	ND	(2.78)	ND	ND		ND	ND		ND	ND		ND	ND	(0.01)	ND	ND		ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	ND		0.01	0.02	(0.02)	ND	0.01	(0.01)	ND	0.01	(0.01)	0.01	ND		ND	0.01	(0.01)	0.01	ND	(0.01)	ND	ND	
Acenaphthene	ND	ND		ND	ND		ND	ND		0.17	ND	(0.09)	ND	ND		ND	ND		ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(4.74)			(2.76)			(5.35)			(3.01)			(1.38)			(4.19)			(4.63)			(1.76)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 2. Concentration of Polycyclic Aromatic Hydrocarbons in Teriyaki-Seasoning Samples
(not Soaked in Teriyaki-Seasoning and Teriyaki-Seasoning after Soaked Row Yellowtail of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	ND*	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(a)pyrene	0.01	ND	(0.01)	0.01	0.01	(0.01)	0.03	0.01	(0.02)	0.01	0.02	(0.03)	0.01	0.01	(0.01)	ND	0.01	(0.01)	ND	ND		0.01	0.01	(0.01)
Dibenz(ah)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
3-Methylcholanthrene	0.03	0.01	(0.02)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(e)pyrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	0.06	(0.03)	ND	ND		ND	ND		ND	ND	
Pyrene	0.56	0.10	(0.33)	ND	0.05	(0.03)	ND	0.02	(0.01)	0.03	0.02	(0.03)	0.05	0.06	(0.06)	ND	0.09	(0.05)	ND	ND		ND	ND	
Fluoranthene	0.11	0.19	(0.15)	0.19	0.16	(0.18)	0.18	0.24	(0.21)	0.15	0.27	(0.21)	0.13	0.14	(0.14)	0.17	0.14	(0.16)	0.03	0.02	(0.03)	0.06	0.05	(0.06)
Anthracene	ND	ND		ND	ND		0.03	0.01	(0.02)	ND	ND		ND	ND		0.03	0.01	(0.02)	0.01	0.01	(0.01)	ND	0.01	(0.01)
Phenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Coronene	0.01	0.02	(0.02)	0.43	ND	(0.22)	ND	0.03	(0.02)	0.01	0.03	(0.02)	ND	0.02	(0.01)	ND	0.01	(0.01)	ND	ND		ND	ND	
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Perylene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	0.02	(0.01)	ND	ND		ND	ND	
9-Methylanthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
5,12-Dihydronaphthacene	ND	ND		1.40	ND	(0.70)	ND	ND		2.03	ND	(1.02)	1.80	ND	(0.90)	ND	1.79	(0.90)	ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	ND		ND	ND		ND	ND		ND	0.01	(0.01)	ND	ND		ND	ND		ND	ND		0.01	ND	(0.01)
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(0.53)			(1.14)			(0.28)			(1.32)			(1.15)			(1.16)			(0.04)			(0.09)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 3. Concentration of Polycyclic Aromatic Hydrocarbons in Row Yellowtail Samples
(not Soaked in Miso-Seasoning and Soaked Row Yellowtail in Miso-Seasoning of 7 days from 1 day)

(ppb)

Sample / Day	not Soked			Days of Soaked 1			2			3			4			5			6			7		
PAH	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	ND*	0.65	(0.33)	0.01	ND	(0.01)	0.04	0.12	(0.08)	ND	ND		0.05	ND	(0.03)	ND	ND		ND	ND		ND	ND	
Benzo(a)pyrene	ND	0.09	(0.05)	ND	0.07	(0.04)	ND	ND		0.01	0.02	(0.02)	ND	ND		0.08	0.01	(0.05)	0.15	0.02	(0.09)	0.04	0.04	(0.04)
Dibenz(ah)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		0.04	ND	(0.02)	ND	ND		ND	ND		ND	ND	
3-Methylcholanthrene	ND	ND		ND	ND		ND	ND		ND	ND		0.02	0.03	(0.03)	ND	ND		0.04	ND	(0.02)	ND	ND	
Benzo(e)pyrene	3.40	0.10	(1.75)	5.13	ND	(2.57)	8.08	5.92	(7.00)	1.56	0.51	(1.04)	0.54	ND	(0.27)	0.15	ND	(0.08)	0.11	ND	(0.06)	ND	ND	
Pyrene	ND	0.05	(0.03)	ND	0.17	(0.59)	ND	ND		ND	ND		ND	3.21	(1.61)	ND	0.87	(0.44)	ND	0.03	(0.02)	ND	ND	
Fluoranthene	ND	0.70	(0.35)	ND	0.68	(0.34)	ND	ND		ND	0.65	(0.33)	ND	ND		ND	ND		1.81	1.06	(1.44)	1.13	ND	(0.57)
Anthracene	ND	0.60	(0.30)	ND	ND		ND	ND		ND	0.03	(0.02)	ND	ND		0.01	0.08	(0.05)	0.01	0.16	(0.09)	0.13	0.02	(0.08)
Phenanthrene	ND	12.93	(6.47)	5.15	ND	(2.58)	18.05	4.60	(11.33)	7.34	1.97	(4.66)	4.83	3.57	(4.20)	ND	4.61	(2.31)	6.31	ND	(3.16)	26.80	16.03	(21.42)
Coronene	ND	0.23	(0.12)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	0.09	(0.05)	ND	ND	
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.02	ND	(0.01)	ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	0.11	(0.06)	ND	ND		0.28	ND	(0.14)	ND	ND		0.88	0.17	(0.53)	ND	0.18	(0.09)
Perylene	ND	0.01	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		0.02	ND	(0.01)	0.03	0.01	(0.02)	ND	ND	
Dibenz(ac)anthracene	ND	ND		ND	ND		0.13	ND	(0.07)	ND	ND		0.02	ND	(0.01)	ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	12.14	2.92	(7.53)	ND	ND		ND	0.51	(0.26)	ND	ND		0.34	2.29	(1.32)	ND	ND		ND	0.04	(0.02)	ND	ND	
9-Methylanthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.01	ND	(0.01)	ND	ND	
5,12-Dihydronaphth acene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	ND		ND	0.02	(0.01)	ND	ND		ND	0.01	(0.01)	ND	ND		0.01	0.01	(0.01)	0.01	0.61	(0.31)	ND	ND	
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		0.83	0.17	(0.50)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(16.94)			(6.64)			(18.80)			(6.08)			(7.63)			(2.95)			(5.83)			(22.20)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 4. Concentration of Polycyclic Aromatic Hydrocarbons in Miso-Seasoning Samples
(not Soaked in Miso-Seasoning and Miso-Seasoning after Soaked Row Yellowtail of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	0.06	0.90	(0.48)	0.06	0.06	(0.06)	0.03	0.03	(0.03)	ND	ND		ND	ND		ND	ND		ND	0.03	(0.02)	0.03	0.03	(0.03)
Benzo(a)pyrene	0.06	0.10	(0.08)	0.02	0.02	(0.02)	0.01	0.03	(0.02)	ND	0.04	(0.02)	ND	0.01	(0.01)	ND	ND		0.03	0.01	(0.02)	0.04	0.01	(0.03)
Dibenz(ah)anthracene	0.08	0.11	(0.10)	ND	ND		ND	ND		ND	1.73	(0.87)	ND	ND		ND	ND		ND	ND		ND	ND	
3-Methylcholanthrene	0.01	0.01	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(e)pyrene	0.19	0.26	(0.23)	ND	0.04	(0.02)	ND	ND		0.56	ND	(0.28)	ND	ND		ND	ND		ND	ND		ND	ND	
Pyrene	0.02	0.03	(0.03)	ND	0.16	(0.08)	0.19	ND	(0.10)	0.01	ND	(0.01)	0.13	0.04	(0.09)	0.06	0.01	(0.04)	0.13	ND	(0.07)	0.03	ND	(0.02)
Fluoranthene	0.16	ND	(0.08)	0.11	0.28	(0.20)	0.58	1.27	(0.93)	0.25	0.27	(0.26)	ND	0.20	(0.10)	0.52	0.23	(0.38)	0.57	0.23	(0.40)	0.26	0.23	(0.25)
Anthracene	0.11	0.16	(0.14)	ND	ND		0.03	0.03	(0.03)	0.01	0.05	(0.03)	0.03	0.04	(0.04)	0.10	ND	(0.05)	0.10	0.02	(0.06)	0.11	0.04	(0.08)
Phenanthrene	ND*	2.38	(1.19)	ND	0.46	(0.23)	ND	1.64	(0.82)	ND	ND		ND	ND		ND	1.70	(0.85)	2.13	ND	(1.07)	2.88	1.79	(2.34)
Coronene	0.01	0.04	(0.03)	0.09	0.16	(0.13)	0.05	0.09	(0.07)	0.05	0.07	(0.06)	0.09	0.05	(0.07)	0.20	0.08	(0.14)	ND	0.10	(0.05)	0.01	0.10	(0.06)
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	ND		ND	0.04	(0.02)	ND	ND		0.07	ND	(0.04)	ND	ND		ND	ND	
Perylene	0.02	0.05	(0.04)	ND	ND		0.01	ND	(0.01)	ND	ND		ND	ND		0.01	0.01	(0.01)	ND	0.01	(0.01)	ND	ND	
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	0.56	1.79	(1.18)	2.72	0.59	(1.66)	ND	0.48	(0.24)	ND	0.52	(0.26)	ND	ND		ND	ND		ND	0.55	(0.28)	0.26	0.34	(0.30)
9-Methylanthracene	0.01	0.03	(0.02)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
5,12-Dihydronaphth:acene	ND	0.66	(0.33)	ND	0.21	(0.11)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	ND		ND	ND		0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.02	0.01	(0.02)	ND	0.01	(0.01)
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.44	ND	(0.22)	0.06	ND	(0.03)	ND	ND	
1,12-Benzoperylen	ND	ND		0.83	0.17	(0.50)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(3.94)			(3.01)			(2.26)			(1.82)			(0.32)			(1.74)			(2.03)			(3.12)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 5. Concentration of Polycyclic Aromatic Hydrocarbons in Row Swordfish Samples
(not Soaked in Teriyaki-Seasoning and Soaked Row Swordfish in Teriyaki-Seasoning of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	6.21	7.30	(6.76)	0.17	0.03	(0.10)	0.28	ND	(0.14)	ND	ND	0.28	ND	(0.14)	0.01	ND	(0.01)	0.02	0.01	(0.02)	0.09	0.07	(0.08)
Benzo(a)pyrene	0.03	ND*	(0.02)	0.16	0.41	(0.29)	1.52	0.05	(0.79)	0.01	0.08	(0.05)	0.05	ND	(0.03)	0.01	ND	(0.01)	0.02	0.02	(0.02)	0.03	0.05	(0.04)
Dibenz(ah)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.02	ND	(0.01)	ND	ND		ND	ND	
3-Methylcholanthrene	ND	ND		ND	ND		0.19	ND	(0.10)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(e)pyrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Pyrene	0.08	0.05	(0.07)	0.13	ND	(0.07)	ND	ND		1.84	ND	(0.92)	ND	ND		ND	ND		ND	ND		6.59	ND	(3.30)
Fluoranthene	0.24	0.39	(0.32)	1.85	0.53	(1.19)	2.70	5.49	(4.10)	0.05	3.02	(1.54)	3.40	0.31	(1.86)	0.40	0.93	(0.67)	0.22	0.52	(0.37)	ND	ND	
Anthracene	0.07	0.09	(0.08)	0.10	ND	(0.05)	0.52	ND	(0.26)	0.12	0.01	(0.07)	0.06	ND	(0.03)	0.50	ND	(0.03)	ND	ND		ND	ND	
Phenanthrene	3.00	5.92	(4.46)	ND	2.96	(1.48)	ND	ND		ND	ND		ND	ND		ND	ND		0.48	0.13	(0.31)	0.45	0.70	(0.58)
Coronene	0.17	0.21	(0.19)	0.04	0.44	(0.24)	2.63	0.67	(1.65)	ND	ND		0.12	ND	(0.06)	0.12	ND	(0.06)	ND	ND		ND	ND	
Fluorene	ND	ND		0.07	ND	(0.04)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Perylene	ND	0.06	(0.03)	0.02	ND	(0.01)	0.29	0.01	(0.15)	ND	ND		0.02	ND	(0.01)	ND	ND		0.02	ND	(0.01)	0.01	0.02	(0.02)
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	0.20	ND	(0.10)	1.26	ND	(0.63)	0.20	0.10	(0.15)	ND	ND		ND	ND		ND	ND		0.32	ND	(0.16)	ND	ND	
9-Methylanthracene	ND	ND		0.01	ND	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.21	ND	(0.11)
5,12-Dihydronaphth acene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	0.01	(0.01)	ND	ND		0.02	ND	(0.01)	ND	ND		ND	ND		ND	ND		0.01	ND	(0.01)	0.01	ND	(0.01)
Acenaphthene	ND	ND		ND	0.17	(0.09)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(12.04)			(4.20)			(7.35)			(2.58)			(2.13)			(0.79)			(0.90)			(4.14)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 6. Concentration of Polycyclic Aromatic Hydrocarbons in Teriyaki-Seasoning Samples
(not Soaked in Teriyaki-Seasoning and Teriyaki-Seasoning after Soaked Row Swordfish of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	ND*	ND		ND	0.01	(0.01)	ND	0.01	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(a)pyrene	ND	0.01	(0.01)	0.01	0.04	(0.03)	0.03	0.04	(0.04)	0.01	0.02	(0.02)	ND	0.04	(0.02)	0.01	0.01	(0.01)	ND	0.02	(0.01)	0.01	ND	(0.01)
Dibenz(ah)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
3-Methylcholanthrene	0.04	ND	(0.02)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(e)pyrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Pyrene	0.64	0.45	(0.55)	ND	0.04	(0.02)	ND	ND		1.30	ND	(0.65)	0.19	ND	(0.10)	0.04	ND	(0.02)	ND	ND		ND	ND	
Fluoranthene	0.01	0.24	(0.13)	0.14	0.32	(0.23)	0.38	0.19	(0.29)	0.35	0.43	(0.39)	0.17	0.23	(0.20)	ND	0.31	(0.16)	0.23	0.15	(0.19)	0.28	0.14	(0.21)
Anthracene	ND	ND		ND	ND		ND	ND		0.03	0.03	(0.03)	ND	0.04	(0.02)	ND	ND		0.01	ND	(0.01)	0.01	ND	(0.01)
Phenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	0.17	(0.09)	ND	ND		ND	ND		ND	ND	
Coronene	ND	0.04	(0.02)	ND	0.05	(0.03)	0.01	ND	(0.01)	ND	0.03	(0.02)	ND	0.03	(0.02)	ND	0.03	(0.02)	ND	ND		ND	ND	
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Perylene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9-Methylanthracene	ND	ND		ND	ND		ND	0.01	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
5,12-Dihydronaphthacene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(k)fluoranthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(0.73)			(0.32)			(0.36)			(1.11)			(0.45)			(0.21)			(0.21)			(0.23)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 7. Concentration of Polycyclic Aromatic Hydrocarbons in Row Swordfish Samples
(not Soaked in Miso-Seasoning and Soaked Row Swordfish in Miso-Seasoning of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	8.18	9.95	(9.07)	0.01	ND	(0.01)	0.05	0.02	(0.04)	ND	0.13	(0.07)	ND	0.09	(0.05)	0.03	ND	(0.02)	0.10	0.08	(0.09)	0.03	0.02	(0.03)
Benzo(a)pyrene	0.04	0.04	(0.04)	0.04	0.02	(0.03)	ND	0.02	(0.01)	0.03	0.12	(0.08)	0.02	0.02	(0.02)	0.03	0.03	(0.03)	0.06	0.07	(0.07)	0.04	0.03	(0.04)
Dibenz(ah)anthracene	ND*	ND		0.02	ND	(0.01)	ND	ND		ND	0.01	(0.01)	ND	0.01	(0.01)	ND	0.04	(0.02)	0.01	0.16	(0.09)	ND	0.02	(0.01)
3-Methylcholanthrene	ND	ND		ND	ND		ND	ND		ND	0.01	(0.01)	ND	0.01	(0.01)	ND	0.01	(0.01)	0.01	0.01	(0.01)	ND	ND	
Benzo(e)pyrene	ND	0.06	(0.03)	ND	ND		ND	ND		ND	0.37	(0.19)	ND	ND		ND	0.04	(0.02)	0.45	0.13	(0.29)	ND	ND	
Pyrene	0.10	0.02	(0.06)	0.01	0.14	(0.08)	8.92	0.11	(4.52)	0.13	ND	(0.07)	ND	0.03	(0.02)	ND	ND					0.03	0.33	(0.18)
Fluoranthene	0.26	0.36	(0.31)	0.34	0.38	(0.36)	0.33	0.39	(0.36)	0.47	0.77	(0.62)	0.43	0.72	(0.58)	0.34	0.29	(0.32)	0.69	0.51	(0.60)	0.52	0.47	(0.50)
Anthracene	0.06	0.06	(0.06)	0.01	0.03	(0.02)	0.02	0.04	(0.03)	0.02	0.13	(0.08)	0.06	0.08	(0.07)	0.03	0.14	(0.09)	0.04	0.60	(0.32)	0.04	0.07	(0.06)
Phenanthrene	2.99	8.92	(5.96)	ND	3.81	(1.91)	0.90	7.16	(4.03)	0.31	3.86	(2.09)	ND	10.54	(5.27)	1.78	2.69	(2.24)	0.46	2.23	(1.35)	4.17	3.64	(3.91)
Coronene	0.25	0.01	(0.13)	0.23	0.07	(0.15)	0.23	0.01	(0.12)	ND	ND		0.10	0.36	(0.23)	0.05	0.04	(0.05)	0.01	0.23	(0.12)	0.01	0.01	(0.01)
Fluorene	ND	ND		ND	ND		0.01	ND	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		0.06	ND	(0.03)	ND	ND		ND	0.05	(0.03)	ND	0.18	(0.09)	ND	ND		ND	ND	
Perylene	0.04	ND	(0.02)	0.06	0.01	(0.04)	ND	ND		ND	0.05	(0.03)	ND	0.01	(0.01)	ND	ND		0.02	ND	(0.01)	0.01	0.01	(0.01)
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	0.16	0.18	(0.17)	0.34	ND	(0.17)	0.31	ND	(0.16)	1.44	2.12	(1.78)	ND	0.20	(0.10)	0.19	0.24	(0.22)	ND	1.56	(0.78)	0.25	0.49	(0.37)
9-Methylanthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.01	ND	(0.01)	ND	ND	
5,12-Dihydronaphthacene	1.30	ND	(0.65)	ND	ND		1.26	0.55	(0.91)	ND	ND		0.35	0.19	(0.27)	1.53	0.85	(1.19)	ND	0.60	(0.30)	ND	ND	
Benzo(k)fluoranthene	0.01	ND	(0.01)	0.01	0.01	(0.01)	0.01	ND	(0.01)	ND	ND		0.01	ND	(0.01)	0.01	ND	(0.01)	ND	ND		ND	ND	
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	0.07	(0.04)	0.09	0.02	(0.06)	0.04	ND	(0.02)	ND	ND		ND	0.07	(0.04)
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(16.51)			(2.79)			(10.23)			(5.07)			(6.74)			(4.33)			(4.04)			(5.16)

* ND : Not detected < 0.01ppb ; ( ) : Average

Table 8. Concentration of Polycyclic Aromatic Hydrocarbons in Miso-Seasoning Samples
(not Soaked in Miso-Seasoning and Miso-Seasoning after Soaked Row Swordfish of 7 days from 1 day)

(ppb)

Sample / Day / PAH	not Soked			Days of Soaked: 1			2			3			4			5			6			7		
	1	2		1	2		1	2		1	2		1	2		1	2		1	2		1	2	
Benz(a)anthracene	0.06	0.90	(0.48)	0.03	0.02	(0.03)	ND	0.05	(0.03)	ND	ND		0.02	ND	(0.01)	ND	0.05	(0.03)	0.01	0.07	(0.04)	0.10	0.05	(0.08)
Benzo(a)pyrene	0.06	0.10	(0.08)	0.01	0.02	(0.02)	0.01	0.03	(0.02)	0.02	0.01	(0.02)	0.03	0.02	(0.03)	0.02	0.04	(0.03)	0.04	0.07	(0.06)	0.06	0.03	(0.05)
Dibenz(ah)anthracene	0.08	0.11	(0.10)	ND*	ND		0.01	0.01	(0.01)	ND	ND		0.01	0.07	(0.04)	ND	ND		0.01	0.09	(0.05)	0.01	0.04	(0.03)
3-Methylcholanthrene	0.01	0.01	(0.01)	ND	ND		ND	0.01	(0.01)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Benzo(e)pyrene	0.19	0.26	(0.23)	ND	0.04	(0.02)	ND	ND		ND	ND		0.42	ND	(0.21)	ND	ND		0.01	0.15	(0.08)	0.13	0.25	(0.19)
Pyrene	0.02	0.03	(0.03)	0.11	ND	(0.06)	ND	0.01	(0.01)	0.02	0.04	(0.03)	0.05	ND	(0.03)	0.04	ND	(0.02)	0.09	0.06	(0.08)	0.08	0.05	(0.07)
Fluoranthene	0.16	ND	(0.08)	0.02	0.25	(0.14)	0.03	0.27	(0.15)	0.18	0.12	(0.15)	0.22	0.20	(0.21)	0.11	0.18	(0.15)	0.76	0.18	(0.47)	0.25	0.18	(0.22)
Anthracene	0.11	0.16	(0.14)	0.02	0.03	(0.03)	ND	0.05	(0.03)	0.05	ND	(0.03)	0.05	0.01	(0.03)	0.05	0.07	(0.06)	0.05	0.50	(0.28)	0.32	0.04	(0.18)
Phenanthrene	ND	2.38	(1.19)	0.84	0.78	(0.81)	2.22	0.84	(1.53)	ND	ND		0.10	ND	(0.05)	0.25	ND	(0.13)	1.56	9.38	(5.47)	1.21	14.10	(7.66)
Coronene	0.01	0.04	(0.03)	0.12	0.01	(0.07)	0.14	0.05	(0.10)	0.59	0.06	(0.33)	0.07	0.26	(0.17)	0.09	0.04	(0.07)	0.05	0.10	(0.08)	0.03	0.07	(0.05)
Fluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
2,3-Benzofluorene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
1-Methylphenanthrene	ND	ND		ND	ND		ND	ND		ND	ND		ND	0.26	(0.13)	0.39	ND	(0.20)	ND	ND		0.24	0.15	(0.20)
Perylene	0.02	0.05	(0.04)	ND	ND		ND	0.01	(0.01)	0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.04	0.02	(0.03)	0.12	0.03	(0.08)
Dibenz(ac)anthracene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
9,10-Dimethylbenz(a)anthracene	0.56	1.79	(1.18)	1.21	0.21	(0.71)	0.62	0.27	(0.45)	0.52	0.72	(0.62)	0.49	0.13	(0.31)	ND	ND		0.51	1.56	(1.04)	1.39	0.49	(0.94)
9-Methylanthracene	0.01	0.03	(0.02)	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		0.01	0.01	(0.01)	0.01	ND	(0.01)
5,12-Dihydronaphthacene	ND	0.66	(0.33)	ND	ND		ND	1.15	(0.58)	ND	ND		ND	ND		ND	ND		0.36	ND	(0.18)	ND	ND	
Benzo(k)fluoranthene	ND	ND		0.01	ND	(0.01)	0.01	0.01	(0.01)	0.01	0.01	(0.01)	0.01	ND	(0.01)	0.01	ND	(0.01)	ND	ND		ND	ND	
Acenaphthene	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	0.05	(0.03)	ND	ND		ND	ND	
1,12-Benzoperylen	ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND		ND	ND	
Total PAH			(3.94)			(1.90)			(2.94)			(1.20)			(1.24)			(0.74)			(7.87)			(9.76)

* ND : Not detected < 0.01ppb ; ( ) : Average

文　献

1）Ova, G.：Onaran, S. Adv. Food Sci **20** (5/6), 168～172 (1998)
2）尾花裕孝，堀伸二郎，田中淳一，樫本　隆：食衛誌. **25**, 35～40 (1984)
3）山崎裕康，南純三郎，市川富夫，近藤雅臣：食衛誌. **18**, 368～374 (1977)
4）Masuda, Y., Mori, K., Kuratsune, M.：Gann **57**, 133～142 (1966)
5）舘野つや子：東京家政大紀要. **26**, 85～89 (1986)
6）舘野つや子：東京家政大紀要. **28**, 103～107 (1988)
7）Kangsadalampai, K.：Butryee, G.；Manoonphol, K.：Food Chem. Toxicol. **35**, 213～218 (1997)
8）Saimt-Aubert, B.：Coopey, J. F.；Astre, C.：Spiliotis, J.：Joyeux, H.：J. Food Compos. Anal. **5**, 257～263 (1992)
9）舘野つや子：食衛試. **39**, 159～164 (1998)
10）舘野つや子：東京家政大紀要. **38**, 117～121 (1998)
11）Moret, Sabrina；Conte, Lanfranco；Dean, Daniela：J. Agric. Food Chem. **47**, 1367～1371 (1999)
12）Wang, Guangdi：Lee, Arthur S.：Lewis, Monique；Kamath, Burde；Archer, Renita K.：J. Agric. Food Chem. **47**, 1062～1066 (1999)
13）群羊社編："味つけ百科" p.3～18 (1983) 緒方出版.
14）科学技術庁資源調査会編："日本食品成分表" 山口廸夫監修 p.64～83 (1996) 医歯薬出版
15）舘野つや子，南雲葉子，末永泉二：食衛試. **31**, 271～276 (1990)
16）舘野つや子，末永泉二：食衛誌. **35**, 206～209 (1994)
17）日本薬学会編："衛生試験法・注解" p.323 (1995) 金原出版

Summary

There are few reports about Polycyclic Aromatic Hydrocarbons (PAH) resulted from soak fish. PAH detection of soak flsh are held twice each fish from first day to 7th-day. Those fish are yellowtail and swordfish of teriyaki-seasoning or miso-seasoning. The number of cases is 64. PAH detection of soaked seasoning is held also. As a result, in case of teriyaki-seasoning, if PAH of teriyaki-seasoning fish is high, PAH tends to transfer into the seasoning. On the contrary, if PHA of the seasoning is high, PAH tends to transfer into the fish. In case of miso-seasoning, there is no tendency above.