

# Distribution of Organochlorines and PCB Congeners In Korean Human Tissues

Mee-Jung Park, Sang-Ki Lee, Ja-Yeol Yang, Ki-Wook Kim, Su-Yeon Lee, Won-Tae Lee, Kyu-Hyuck Chung<sup>1</sup>, Yeo-Pyo Yun<sup>2</sup>, and Young-Chan Yoo<sup>2</sup>

National Institute of Scientific Investigation, Seoul 158-707, Korea, <sup>1</sup>College of Pharmacy, Sungkyunkwan University, Suwon 440-746, Korea, and <sup>2</sup>College of Pharmacy, Chungbuk National University, Cheongju 361-763, Korea

(Received April 17, 2005)

In order to investigate the residual amounts of organochlorines and polychlorinated biphenyls (PCBs) in Korean human tissues (blood, adipose tissue, liver, kidney cortex, and lung), the samples were collected from the autopsied cadavers of 40 men and 40 women (from teens to seventies of age).  $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC,  $\delta$ -BHC, p,p'-DDT, p,p'-DDD, p,p'-DDE, endrin, dieldein, aldrin, and 7 marker PCBs (28, 52, 101, 118, 138, 153, and 180) were determined in human tissues. The levels of organochlorines and PCB congeners indicated that they have been widely distributed in Korean human body. Positive correlations in terms of age were observed for the following cases: p,p'-DDE, p,p'-DDT, p-DDT, PCB 118, PCB 138, PCB 153, and p-PCB in the adipose tissue, and p-PCB in the lung. Concentration of these compounds showed a significant age-related increase. Accumulation of these compounds in aged people revealed that these compounds were more slowly eliminated in our environment and risk assessment was necessary for further proper action. Significant differences in the levels of PCBs between genders were found for PCB 118 in the adipose tissue and PCB 138 in the liver. Positive correlation coefficients between tissues were detected with p-p'-DDE and p-BHC.

Key words: Organochlorines, PCB congeners, Human tissues, Korean, Autopsy cases

#### INTRODUCTION

Organochlorines and PCB congeners are effective against a variety of insects. These chemicals were introduced in the 1940s and had widely been used because of their potent effect and relatively low cost but are rarely used today because of their environmental persistence.

Although production of organochlorines and PCB congeners has been stopped in Korea and most other parts of the world, but still they are considered as ubiquitous pollutants due to their persistence in the environment. In aquatic systems, organochlorines are adsorbed onto sediments in water that can bioconcentrate in marine mammals. Because these chemicals are soluble in fat, they are found at higher concentrations in fatty foods. Diets that contain fats that may be contaminated with organochlorines and PCB congeners (e.g., contaminated

milk and dairy products, fish) lead to increased exposure to these chemicals. Bioaccumulation of organochlorines and PCB congeners through the food chain has resulted in high concentrations of these compounds in meat, milk and fish. Because of their lipophilic properties, organochlorines and PCB congeners are not readily eliminated from the body. Estimated half-lives of individual organochlorines and PCB congeners in humans ranges from < 1 month to > 40 years (Wolff et al., 1992). An increased interest on endocrine disrupting substances has again attracted attention towards organochlorines and PCB congeners (Colborn et al., 1993, Hillman, 1993, Soto et al., 1995). The presence of organochlorines and PCB congeners in human adipose tissue has recently caused concern due to their antiandrogenic and estrogenic properties and their effects on sexual activity and development of breast cancer. Until recently, several studies have investigated the residual levels of these compounds in human milk, blood and adipose tissue collected from people of different countries (WHO, 1988, Schecter et al., 1989). For the evaluation of the risk assessment, estimation of exposure level has to be performed. In Korea, there is

Correspondence to: Sang-Ki Lee, National Institute of Scientific Investigation, 331-1, Shinwol-7-dong, Yangcheon-gu, Seoul 158-707, Korea

Tel: 82-2-2600-4925, Fax: 82-2-2600-4919

E-mail: skleedoc@nisi.go.kr

a lack of data on the concentration of organochlorines and PCB congeners in human tissues. Therefore, we performed the present work for the purpose of determination of the levels of organochlorines and PCB congeners in human tissues such as blood, adipose tissue, liver, kidney cortex, and lung obtained from autopsy material. We have also correlated its relation with age, sex, and variation between the tissue samples. The halogenated compounds studied in this article include the organochlorines such as  $\alpha$ -,  $\beta$ -,  $\gamma$ -,  $\delta$ -benzenehexachloride (BHC), the DDT group (p,p'-DDE, p,p'-DDT, p,p'-DDE), aldrine, dieldrine, endrine, and 7 marker of PCB congeners (PCB 28, 52, 101, 118, 138, 153, and 180).

#### MATERIALS AND METHODS

#### Chemicals

α-BHC, β-BHC, γ-BHC, δ-BHC, p,p'-DDE, p,p'-DDT, and p,p'-DDE, aldrine, dieldrine, endrine, and mirex were purchased from Dr. Ehrenstorfer GmbH Co. (Atlanta, GA). PCB congeners (28, 52, 101, 118, 138, 153, 180, and 204) were purchased from ChemService Co. (West Chester, PA). All other chemicals and solvents were of organic residue analytical grade. The standard stock solutions of the organochlorines and PCB congeners were 1 mg/mL in methanol. Working standards were prepared by dilution with methanol. Mirex and PCB congener 204 were used as internal standard for organochlorines and PCB congeners, respectively.

# Sample collection

The analyzed postmortem tissues were obtained from autopsied cadavers subjected to undergoing forensic medical examinations at the National Institute of Scientific Investigation of Korea, from 2000 to 2003. The number of male and female cadavers were 20~40 (Table I). We obtained samples from persons who died due to trauma, traffic accident, hanging, stangulation *etc.*, (people who died without any specific disease). Blood, adipose tissue, liver, kidney cortex, and lung were taken and stored at -30 °C until the analysis.

Table I. Distribution of age and gender of individual subjects

Age group	male	Female	Total
Teens	2	-	2
Twenties	7	13	20
Thirties	12	11	23
Forties	6	5	11
Fifties	3	5	8
Over Sixties	10	6	16
Total	40	40	80

# Sample preparation and purification

Sample homogenization, extraction and column chromatographic cleanup were carried out according to the previously described procedure (Yoo *et al.*, 2001). Briefly, liver, kidney cortex, and lung were weighed to be 5 g and homogenized with sodium sulfate solution in a tissue grinder. Homogenized samples and adipose tissues were liquid phase extracted with *n*-hexane and exact weight of lipid component was determined. Thereafter, organochlorines and PCB congeners residues were extracted thrice with acetonitrile. The acetonitrile layer was pooled, and 2% sodium chloride was added and extracted thrice with *n*-hexane and evaporated to approximately 5 mL. The residual samples were subjected to chromatographic purification on deactivated Florisil column.

#### Instrumental condition

Organochlorines and PCB congeners were identified with GC coupled with electron capture detector (GC/ECD,  $^{63}$ Ni) in a fused silica capillary DB-5 and DB-1 column (30 m×0.25 mm, 0.25 µm film thickness, respectively). Quantification was performed with DB-5 column. Mirex and PCB congener 204 solution (0.1 µg/mL) were used as internal standards for organochlorines and PCB congeners, respectively. The ratio of the peak area of target analytes to that of internal standard was utilized to calculate the concentration of the analytes in the specimen. Statistical

**Table II.** LOD (limit of detection) and recoveries of organochlorines and PCB congeners added to liver, adipose tissue, and blood (added amount :  $0.1~\mu g$ )

			·	<del></del>
	LOD	Rec	overy (Mean ± SD	0, %)
Analytes	(pg)	Blood (n=3)	Adipose tissue (n=3)	Liver (n=3)
α-BHC	1.25	84.5 ± 10.76	114.0 ± 10.16	106.0 ± 10.74
β-ВНС	2.5	77.7 ± 3.64	85.2 ± 4.41	88.4 ± 6.17
γ-BHC	1.25	83.8 ± 5.41	75.8 ± 2.89	81.8 ± 9.31
δ-BHC	1.25	90.8 ± 9.55	96.3 ± 3.95	97.8 ± 6.25
<i>p,p'</i> -DDT	5	89.0 ± 3.63	98.4 ± 10.78	102.4 ± 11.15
<i>p,p'</i> -DDD	2.5	$92.3 \pm 4.36$	86.7 ± 6.70	94.5 ± 12.53
p,p'-DDE	2.5	92.0 ± 5.74	83.4 ± 8.34	81.8 ± 4.07
endrin	1.25	78.4 ± 4.91	$72.5 \pm 6.39$	71.0 ± 5.80
dieldein	1.25	91.6 ± 13.06	78.2 ± 13.84	$70.7 \pm 4.59$
aldrin	0.625	65.4 ± 3.40	71.2 ± 6.91	65.5 ± 2.07
PCB 28	1.6	77.3 ± 9.81	$80.9 \pm 7.39$	86.7 ± 5.49
PCB 52	1.6	81.3 ± 4.62	89.7 ± 2.92	$80.5 \pm 5.39$
PCB 101	3.2	67.3 ± 0.94	75.3 ± 1.09	71.6 ± 5.56
PCB 118	3.2	107.2 ± 15.80	96.0 ± 15.89	101.8 ± 17.78
PCB 138	1.6	73.1 ± 0.19	91.7 ± 11.64	82.2 ± 6.31
PCB 153	1.6	77.8 ± 8.11	79.6 ± 9.03	85.6 ± 10.44
PCB 180	1.6	69.0 ± 0.83	72.8 ± 2.72	78.1 ± 3.30

Table III. Concentration of organochlorine pesticides and PCB congeners (µg/g extracted fat basis) in blood, adipose tissue, liver, kidney cortex, and lung

								Blood								
Sex Fre.	a:	Mean±SD	Range	z	OCs& PCBs	Sex	Fie	Mean±SD	Range	z	PCBs	Sex	F. G.	Mean±SD	Range	z
M 3%	i	0.0009±0.0054	ND~0.0344	9		Σ	%0	QN	QV	40		Σ	%0	9	Ð	30.
10%	%	10% 0.0269±0.0918	ND-0.4720	40	Aldrin	ட	3%	$0.0267\pm0.1687$	ND~1.0670	40	<u>유</u>	ш	%0	9	<u>N</u>	30
<b>%9</b>	_0	$0.0139\pm0.0659$	ND~0.4720	8		<b>-</b>	1%	$0.0133\pm0.1193$	ND~1.0670	8	2	<b>-</b>	%0	Q	QN	09
M 5%		0.0350±0.1751	ND~1.0639	9		Σ	%02	0.4816±0.7713	ND~3.7810	49		Σ	33% 0.3	0.3570±0.8023	ND~3.1417	39
F 8%	.0	0.0247±0.0914	ND-0.4503	4	p,p'-DDE	ட	%8/	$0.5221\pm0.5716$	ND~2.1640	40	2 2 2 3 4 4 7 8	ட	23% 0.4	$0.4608\pm1.0682$	ND~4.4890	30
%9	.0	$0.0298\pm0.1388$	ND~1.0639	8		<b>—</b>	74%	$0.5018\pm0.6748$	ND~3.7810	8	) :	<b>-</b>	28% 0.4	$0.4089\pm0.9381$	ND~4.4890	99
M 3%	1 .0	0.0306±0.1937	ND~1.2253	40		Σ	%0	QN	QN	9		Σ	%0	9	S	99
%8	\ o	$0.1431\pm0.7089$	ND~4.4090	4	DDT-d'd	ட	%0	N N	S	40	PCB	щ	%0	QN	Q	8
. 5%	.0	$0.0869\pm0.5195$	ND~4.4090	8		<b>—</b>	%0	ND	Q	8	3	<b>—</b>	%0	Q	Q	90
%0 W		Q.	9	6		Σ	%0	QN	2	4		Σ	%0	9	N Q	8
F 3%	~0	$0.0045\pm0.0283$	ND-0.1792	4	ddd-'q,q	щ	3%	$0.0249\pm0.1575$	ND~0.9960	40	PCB	щ	%0	QN	N	99
1%	_0	$0.0022\pm0.0200$	ND~0.1792	8		<b>-</b>	1%	0.0124±0.1114	ND~0.9960	8	<u>}</u>	<b>-</b>	%0	9	N	90
Σ		0.0665±0.2567	ND~1.2253	9		Σ		0.4816±0.7713	ND~3.7810	9		Σ	3% 0.0	0.0695±0.3807	ND~2.0850	30
		$0.1991\pm0.7382$	ND~4.4090	4	∑-DDT	ட		0.5470±0.6477	ND~2.8500	40	원 동	ட	%0	Q	2	30
		$0.1328\pm0.5531$	ND~4.4090	8		<b>—</b>		0.5143±0.7084	ND~3.7810	8	<u> </u>	<b>-</b>	2% 0.0	$0.0347\pm0.2692$	ND~2.0850	99
M 3%		0.0078±0.0490	ND~0.3100	9		Σ	3%	0.1216±0.6660	ND~3.6480	99		Σ	0.5	0.5481±1.1406	ND~3.6658	30
F 5%		0.0073±0.0325	ND~0.1675	4	% 20 80 80 80 80 80 80 80 80 80 80 80 80 80	щ	7%	$0.0726\pm0.3298$	ND~1.7752	30	Σ-PCB	щ	0.6	0.5334±1.0865	ND~4.4890	30
T 4%		0.0075±0.0413	ND-0.3100	8	}	<b>-</b>	2%	0.0971±0.5217	ND~3.6480	99		<b>-</b>	0.5	0.5408±1.1044	ND~4.4890	99
%0 W		8	9	49		Σ	%0	N O	QN	93						}
%0	~~	Q	9	4	PCB 52	щ	%0	Q	8	30						
%0	_0	QN	Q	8	ļ	<b>-</b>	%0	QN	Q	99						
	1															

Table III. Continued

								β	Adipose tissue								
SOO	Sex	F G	Mean±SD	Range	z	OCs& PCBs	Sex	Fre.	Mean±SD	Range	z	PCBs	Sex	F. e.	Mean±SD	Range	z
	Σ	2%	0.0005±0.0022	ND~0.0108	40		Σ	2%	0.0002±0.0010	ND~0.0049	40		Σ	%0	N	Q	30
$\alpha$ -BHC	π	10%	$0.0008\pm0.0028$	ND~0.0159	40	Aldrin	щ	10%	$0.0025\pm0.0089$	ND~0.0428	40	PCB 5	щ	3%	$0.0010\pm0.0053$	ND~0.0293	30
	<u>-</u>	8%	0.0006±0.0025	ND~0.0159	8		<b>-</b>	%8	0.0014±0.0064	ND~0.0428	80		-	2%	0.0005±0.0038	ND~0.0293	09
	9 W	28%	0.0428±0.0712	ND~0.3995	9		Σ	100%	0.1443±0.1238	0.0015~0.6096	9		Σ	%02	0.0220±0.0346	ND~0.1486	30
β-ВНС	<u>ш</u>	%08	$0.0403\pm0.0441$	ND~0.2160	4	p,p'-DDE	ட	100%	$0.2030\pm0.1915$	0.0217~0.8987	40	3 2 3 3	ᄔ	%//	0.0167±0.0158	ND~0.0585	30
	) T	%69	$0.0415\pm0.0588$	ND~0.3995	80		⊢	100%	0.1737±0.1629	0.0015~0.8987	80	2	$\vdash$	73%	$0.0193\pm0.0268$	ND~0.1486	09
	M	2%	0.0014±0.0069	ND~0.0410	9		Σ	83%	0.0339±0.0350	ND~0.1748	40		Σ	93%	0.0312±0.0376	ND~0.1677	30
γ-BHC	ıL	2%	$0.0024\pm0.0119$	ND~0.0720	40	p,p'-DDT	ட	%9/	$0.0243\pm0.0220$	ND~0.0746	40	7 23 38 28	щ	%/8	$0.0199\pm0.0144$	ND~0.0612	30
	<b>—</b>	2%	0.0019±0.0097	ND~0.0720	8		<b>-</b>	%62	$0.0291\pm0.0294$	ND~0.1748	88	2	<b>-</b>	%06	$0.0255\pm0.0288$	ND~0.1677	09
	M	%0	ND	Ð	40		Σ	28%	0.0067±0.0127	ND~0.0709	40		Σ	73%	0.0165±0.0206	ND~0.0699	99
S-BHC	ш	%0	N	2	4	$d^{\prime}d^{\prime}d$	ш	%09	$0.0054\pm0.0075$	ND~0.0288	40	PCB 153	ш	%/5	$0.0105\pm0.0144$	ND~0.0487	30
	_	%0	ND	ND	80		-	%69	0.0060±0.0104	ND~0.0709	80	3	<b>-</b>	%59	0.0135±0.0179	ND~0.0699	09
	Σ		0.0448±0.0721	ND~0.3995	4		Σ		0.1848±0.1479	0.0015~0.6845	9		Σ	87%	0.0226±0.0260	ND~0.1126	30
Σ-BHC	ட		$0.0434\pm0.0458$	ND~0.2160	40	Σ-DDT	ட		$0.2327\pm0.1888$	0.0217~0.8987	4	8 8 8	щ	87%	$0.0087\pm0.0067$	ND~0.0265	30
	_		0.0441±0.0600	ND~0.3995	80		<b>—</b>		0.2088±0.1702	0.0015~0.8987	80	2	<b>—</b>	87%	0.0157±0.0201	ND~0.1126	09
	S ∑	33%	$0.0025\pm0.0051$	ND~0.0178	40	1	Σ	%/	0.0015±0.0071	ND~0.0385	99		Σ		0.0950±0.1028	ND~0.4989	30
Endrin	F 2	28%	$0.0035\pm0.0104$	ND~0.0583	4	% 88	ш	10%	$0.0027\pm0.0094$	ND~0.0374	30	Σ-PCB	ட		$0.0595\pm0.0370$	ND~0.1462	30
	<u>⊢</u>	30%	0.0030±0.0081	ND~0.0583	8	<b> </b>	<b>-</b>	%8	$0.0021\pm0.0083$	ND~0.0385	99		<b>—</b>		0.0772±0.0787	ND~0.4989	09
	Σ	3%	0.0001±0.0008	ND~0.0052	40		Σ	3%	0.0012±0.0066	ND~0.0360	8						
Dieldrin	ш	%0	9	Q	9	22 22 23	ш	%0	N	Q	30						
	· -	1%	0.0001±0.0006	ND~0.0052	8	}	<b>—</b>	2%	$0.0006\pm0.0047$	ND~0.0360	99						

7	ď
=	<u>.</u>
1	90

									Liver								
OCs	Sex	x Fre.	Mean±SD	Range	z	OCs& PCBs	Šex	Fre	Mean±SD	Range	z	PCBs	Sex	Fre.	Mean±SD	Range	z
	Σ	%5 1	0.0059±0.0298	ND~0.1819	40		≥	3%	0.0004±0.0026	ND~0.0165	40		Σ	3%	$0.0349\pm0.1909$	ND~1.0457	30
$\alpha$ -BHC	ш	. 10%	0.0076±0.0238	ND~0.0946	40	Aldrin	ட	%0	Q	ON	40	9C 13C	ட	3%	$0.0215\pm0.1179$	ND~0.6456	30
	_	%8	$0.0068\pm0.0268$	ND~0.1819	8		<b>—</b>	%	0.0002±0.0018	ND~0.0165	80	2	_	3%	0.0282±0.1574	ND~1.0457	09
	Σ	1 58%	0.3581±0.7714	ND~4.5591	40		Σ	%86	0.3223±0.4765	ND~2.6883	40		Σ	43%	0.0568±0.1080	ND~0.4803	30
β-BHC	ш	%89	0.3734±0.4759	ND~2.0620	40	p,p'-DDE	ட	95%	0.3727±0.4159	ND~2.0759	40	38 138 138	ட	53%	$0.1277 \pm 0.2958$	ND~1.5750	30
	_	. 63%	0.3658±0.6369	ND~4.5591	8		<b>—</b>	%96	0.3475±0.4451	ND~2.6883	8	)	_	48%	0.0922±0.2237	ND~1.5750	09
	Σ	1 3%	0.0005±0.0029	ND~0.0186	40		Σ	30%	0.0438±0.1114	ND~0.5169	40		Σ	27%	0.0241±0.0531*	ND~0.2346	30
γ-BHC	ш	2%	$0.0075\pm0.0344$	ND~0.1901	40	TOO-'q,q	ட	10%	$0.0140\pm0.0577$	ND~0.3461	40	92 138 138	ட	10%	0.0022±0.0092	ND~0.0491	30
	_	. 4%	$0.0040\pm0.0245$	ND~0.1901	8		<b>—</b>	20%	$0.0289\pm0.0894$	ND~0.5169	8	3	<b>-</b>	18%	$0.0132 \pm 0.0393$	ND~0.2346	09
	Σ	3%	0.0011±0.0066	ND~0.0419	40		Σ	33%	0.0442±0.1315	ND~0.6676	9		Σ	20%	0.0095±0.0253	ND~0.1102	30
S-BHC	ш	%0	Q	Q	40	OOO-,d'd	u_	23%	$0.0514\pm0.1978$	ND~1.1643	40	PCB 153	ட	3%	$0.0008\pm0.0045$	ND~0.0248	30
	_	1%	0.0005±0.0047	ND~0.0419	8		<b>-</b>	28%	$0.0478\pm0.1669$	ND~1.1643	8	3	<b>—</b>	12%	0.0052±0.0186	ND~0.1102	09
	Σ	_	0.3655±0.7734	ND~4.5591	40		Σ		0.4103±0.6432	ND~3.4972	9		Σ	20%	0.0115±0.0383	ND~0.2042	30
Σ-BHC	ш.		0.3887±0.4783	ND~2.0620	40	Σ-DDT	ட		$0.4381\pm0.5279$	ND~2.6390	4	8 8 8	ட	3%	0.0033±0.0179	ND~0.0978	30
	_		0.3771±0.6390	ND~4.5591	8		<b>—</b>		0.4242±0.5849	ND~3.4972	80		<b>—</b>	12%	0.0074±0.0299	ND~0.2042	09
	Σ		18% 0.0176±0.0579	ND~0.3225	9		Σ	%/	0.0219±0.1163	ND~0.6373	30		Σ		$0.1586 \pm 0.2721$	ND~1.0457	30
Endrin	ш	-	15% 0.0214±0.0823	ND~0.4300	40	8 8 8	ш	20%	$0.1755\pm0.6241$	ND~3.2790	30	Σ-PCB	ட		$0.3310\pm0.9007$	ND~4.8540	30
	-	. 16%	0.0195±0.0707	ND~0.4300	80	ł	_	13%	0.0987±0.4518	ND~3.2790	09		⊢		0.2448±0.6654	ND~4.8540	09
	Σ	%0 V	Q.	9	40		Σ	%0	QN	Q.	8						
Dieldrin	ш.	3%	$0.0005\pm0.0030$	ND~0.0192	40	PCB 52	ட	%0	QN	2	30						
	-	. 1%	$0.0002\pm0.0022$	ND~0.0192	8	}	-	%0	2	9	99						

Table III. Continued

								<b>x</b> .	Kidney Cortex								
S)O	Sex	F. G.	Mean±SD	Range	z	OCs& PCBs	Sex	Fre.	Mean±SD	Range	z	PCBs	Sex	Fre.	Mean±SD	Range	z
	Σ	3%	0.0111±0.0606	ND~0.3322	8		Σ	%0	QN .	2	8	!	Σ	%0	Q	Q	20
α-BHC	ш	3%	0.0070±0.0382	ND~0.2091	30	Aldrin	щ	3%	0.0049±0.0269	ND-0.1473	93	8 등	ட	%0	Q	Q	20
	-	3%	$0.0090\pm0.0503$	ND~0.3322	99		<b>-</b>	2%	0.0025±0.0190	ND~0.1473	99		<b>—</b>	%0	Q	9	40
	Σ	30%	0.2245±0.4263	ND~1.3600	8		Σ	77%	0.2727±0.3906	ND~1.9215	8		Σ	15%	0.0313±0.1024	ND~0.4513	70
д-внс	щ	30%	0.2021±0.3714	ND~1.0814	30	p,p'-DDE	щ	73%	$0.3595\pm0.4571$	ND~1.7370	8	138 138 138	ட	15%	$0.1310\pm0.4766$	ND~2.1060	20
	<b>—</b>	30%	$0.2133\pm0.3965$	ND~1.3600	99		-	75%	$0.3160\pm0.4238$	ND~1.9215	8	•	<b>-</b>	15%	0.0812±0.3440	ND~2.1060	40
	Σ	%0	2	ON.	8		Σ	%0	9	QN	8		≥	%0	2	S	70
γ-BHC	щ	1%	0.0295±0.1145	ND~0.5300	30	p,p'-DDT	ட	%0	2	9	30	90 23 28 28	ட	%0	9	Q	20
	<b>—</b>	3%	0.0147±0.0816	ND~0.5300	99		-	%0	Q	Q	8	3	<b>-</b>	%0	QN	QN	4
	Σ	%	QN	Q	30		Σ	3%	0.0010±0.0054	ND~0.0294	8		Σ	%0	9	9	70
8-BHC	ட	%0	Q	S	30	OOO-,d'd	ட	%2	$0.0045\pm0.0185$	ND~0.0940	30	PGB 153	ட	%0	9	Q	20
	<b>—</b>	%0	QN	Q	99		⊢	2%	$0.0027\pm0.0136$	ND~0.0940	8		<b>-</b>	%0	QN	Q	4
	Σ		0.2356±0.4246	ND~1.3600	8		Σ		0.2736±0.3924	ND~1.9215	99		Σ	%0	2	9	70
Σ-BHC	ш		$0.2386\pm0.4068$	ND~1.3136	30	∑-DDT	ட		$0.3640\pm0.4567$	ND~1.7370	30	2 2 2 2 3 3 3	ட	%0	9	Q	70
	<b>—</b>		0.2371±0.4122	ND~1.3600	99		<b>—</b>		0.3188±0.4246	ND~1.9215	09		<b>—</b>	%0	e Q	Q	40
	Σ	1%	0.0844±0.3407	ND~1.6980	೫		Σ	2%	0.0125±0.0559	ND~0.2500	20		Σ		0.0438±0.1131	ND~0.4513	70
Endrin	u_	10%	$0.1201\pm0.4006$	ND~1.7540	30	% 88 8	ш	20%	$0.1717\pm0.3669$	ND~1.1677	20	Σ-PCB	ட		$0.3027 \pm 0.5607$	ND~2.1060	20
	<b>—</b>	%8	$0.1023\pm0.3691$	ND~1.7540	99	i	<b>-</b>	13%	0.0921±0.2713	ND~1.1677	40		<b>—</b>		0.1733±0.4202	ND~2.1060	40
	Σ	3%	0.0009±0.0049	ND~0.0265	99		Σ	%0	9	S	20						
Dieldrin	ட	3%	$0.0044\pm0.0243$	ND~0.1330	30	25 S	ட	%0	QN	2	20						
İ	<b>—</b>	3%	0.0027±0.0175	ND~0.1330	99		_	%0	ND	ON	40						
							l										

Table III. Continued

ocs	Sex	Fe	Mean±SD	Range	z	OCs& PCBs	Sex	F.	Mean±SD	Range	z	PCBs	Sex	Fre.	Mean±SD	Range	z
	Σ	%0	S	9	70		Σ	%0	QN	QN	20		Σ	%0	9	Q	70
$\alpha$ -BHC		%0	Q	Q	20	Aldrin	ட	2%	$0.0653\pm0.2918$	ND~1.3050	20	<u>2</u> 5	ட	2%	0.0227±0.1015	ND~0.4540	20
	_	%0	Q	2	9		-	3%	0.0326±0.2063	ND~1.3050	40		-	3%	0.0113±0.0718	ND~0.4540	40
	Σ	10%	0.3340±1.1101	ND~4.6320	20		Σ	%09	$0.5883\pm0.9450$	ND~3.7000	70	(	Σ	15%	$0.0569\pm0.1395$	ND~0.4211	70
в-внс	Т	10%	0.0597±0.1908	ND~0.7553	20	p,p'-DDE	щ	22%	$0.3437\pm0.5559$	ND~2.3617	20	£ 8€	ш.	20%	$0.1512\pm0.3645$	ND~1.2166	20
	· -	10%	$0.1968\pm0.7984$	ND~4.6320	40		-	28%	0.4660±0.7752	ND~3.7000	40		۳	18%	0.1041±0.2766	ND~1.2166	40
	Σ	%0	9	Q	70		Σ	%0	QN	QN	20	1	Σ	%0	Q	Q	20
γ-BHC	ட	2%	$0.0360\pm0.1610$	ND~0.7199	20	p,p'-DDT	ட	2%	$0.1872\pm0.8372$	ND~3.7440	20	2.5 8.5 8.5	ட	%0	9	9	20
	⊢	3%	$0.0180\pm0.1138$	ND~0.7199	40		<b>—</b>	3%	$0.0936\pm0.5921$	ND~3.7440	40		<b>-</b>	%0	QN	QN	40
	Σ	%0	9	2	70		Σ	10%	0.0247±0.0889	ND~0.3890	20		Σ	%0	9	N Q	20
8-BHC	ட	%0	9	2	20	000-,d'd	ட	10%	$0.0170\pm0.0549$	ND~0.2220	20	7 2 3 3 3 3	щ	%0	9	P	20
	-	%0	Q	Q	40		<b>—</b>	10%	$0.0209\pm0.0730$	ND~0.3890	40		-	%0	QN	QN	4
	Σ		0.3340±1.1101	ND~4.6320	20		Σ		0.6132±0.9501	ND~3.7000	70	1	Σ	%0	9	Q	20
Σ-BHC	ш		$0.0957\pm0.2404$	ND~0.7553	20	Σ-DDT	ட		$0.5480\pm0.9808$	ND~3.9948	70	58	ட	%0	Q	Q	20
	<b></b>		0.2148±0.8019	ND~4.6320	40		<b> </b>		$0.5805\pm0.9537$	ND~3.9948	40	:	r	%0	QN	9	9
	Σ	10%	0.0442±0.1542	ND~0.6660	20	!	Σ	10%	0.2599±0.9753	ND~4.3190	70		Σ		0.3167±0.9693	ND~4.3190	20
Endrin	u.	2%	$0.0201\pm0.0897$	ND~0.4010	20	8 88 8 88	ш	15%	$0.0846\pm0.2322$	ND~0.9240	20	Σ-PCB	щ		$0.2585\pm0.5139$	ND~1.8649	70
	<b>—</b>	%8	$0.0321\pm0.1251$	ND~0.6660	40		<b>⊢</b>	13%	0.1722±0.7054	ND~4.3190	40		-		0.2876±0.7664	ND~4.3190	40
	Σ	%0	QN	ON	20		Σ	%0	Q	Q	20						
Dieldrin	щ	%0	2	Q	70	25 25 25 25 25 25 25 25 25 25 25 25 25 2	щ	%0	Q	N	20						
	<b>-</b>	%0	9	QN	4		<b>—</b>	%0	Q.	Q	40						

M.-J. Park et al.

analysis was performed with SPSS program and student's *t*-test.

#### RESULTS AND DISCUSSION

Calibration curves for organochlorines such as  $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC,  $\delta$ -BHC, p,p'-DDE, p,p'-DDT, and p,p'-DDE, aldrine, dieldrine, endrine, and 7 markers PCB congeners were linear in the range of 0.00625~0.25 µg/mL (r >0.99, respectively). LOD (limit of detection) of all analytes ranged from 0.625 to 5 pg (Table II). Recovery test was done in triplicate by determining the amount of organochlorines and PCB congeners in target analytes-spiked liver, adipose tissues, and blood (Table II). Recoveries of organochlorines and PCB congeners ranged from 65.5 to 114.0% and 67.3~107.2%, respectively.

The concentration of  $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC,  $\delta$ -BHC, p,p'-DDE, p,p'-DDT, p,p'-DDE, aldrine, dieldrine, endrine, PCB 28, 52, 101, 118, 138, 153, and 180 in blood, adipose tissue, liver, kidney cortex, and lung in terms of gender is shown in Table III. In blood,  $\rho, \rho'$ -DDE was the most frequently detected (74%) and had the highest concentration value (0.5018 ± 0.6748 µg/g extracted fat basis). In 1997, Archibeque-Engle et al. reported that the serum concentration of p,p'-DDE's in 35 women of Connecticut was 967 ng/g (Archibeque-Engle et al., 1997). Compared to this report, the concentration of p,p'-DDE in our data was about the half of the reported value. The frequency of p,p'-DDE in adipose tissue was 100% and p,p'-DDT and p,p'-DDD were also detected in high frequencies. β-BHC was more frequently detected (69%) than other BHC isomers in adipose tissue. The obtained data was consistent with previous report which stated that, β-BHC is more persistent than other isomers (Kuts et al., 1991). In adipose tissues, PCB 118, 138, 153, and 180 were more frequently detected than PCB 28, 52, and 101. This was consistent with the Pauwels' report that described the level of PCB congeners in adipose tissue of Belgium women (Pauwels *et al.*, 2000). Mean concentration of  $\beta$ -BHC, p,p'-DDT, p,p'-DDE, and PCB congeners in human adipose tissues from various countries are shown in Table IV. Significant differences in the levels of PCB congeners between genders were found at PCB 118 in adipose tissue. In this result, PCB 118's concentration in male was higher than that of female. For the proper and accurate correlation study, further examination of larger sample is needed.

All of the determined organochlorines and PCB congeners, except PCB 52 were detected in liver. p,p'-DDE was detected in 96% of liver samples and it's mean concentration was  $0.3475 \pm 0.4451~\mu g/g$  (extracted fat basis). This level was lower than the concentration level in liver of Swedish and Greenland people (836 ng/g and 2,209 ng/g, respectively) (Weistrand *et al.*, 1998, Dewailly *et al.*, 1999). Significant differences in the levels of PCB congeners between genders were found for PCB 138 in liver. In the case of kidney cortex and lung,  $\delta$ -BHC, p,p'-DDT, PCB 52, 101, 138, 153, 180, and  $\alpha$ -BHC,  $\beta$ -BHC, dieldrine, PCB 52, 138, 153, and 180 were not detected, respectively.

Correlation coefficients of levels of organochlorines and PCB congeners in terms of age and tissues were also determined. The concentrations below 50% frequency were not accounted, as non-detected samples could also increase the statistical correlation. In adipose tissue, the levels of p,p'-DDE (r=0.225), p,p'-DDT (r=0.236), total DDTs (r=0.266), PCB 118 (r=0.361), PCB 138 (r=0.315), PCB 153 (r=0.286), and total PCBs (r=0.366); and in lung, the level of p,p'-DDE (r=0.416) significantly varied with age (p<0.05). This result shows that these compounds accumulated more heavily in aged people and eliminated

**Table IV.** Mean concentration of β-BHC, p,p'-DDT, p,p'-DDE, and PCB congeners in human adipose tissue as reported from various countries (unit :  $\mu g/kg$  lipid weight basis)

Country	Year	β-ВНС	p,p'-DDT	ho, ho'-DDE	PCBs	References
Canada	1991-1992	40	NA	765		Dewailly et al., 1994
Italy	1989	213	64	395	NA	Gallelli et al., 1995
Iran	1991-1992	728	190	2,450	NA	Burgaz <i>et al</i> ., 1995
Spain	1991	1,530	400	3,930	2,400	Gomez-Catalan et al., 1995
U.S.A.	1994-1996	37	28	913		Stellman et al., 1995
Vietnam	1991	30°		4,900 <sup>b</sup>	300	Nakamura et al., 1994
Mexico	1997-1998	143	1,224	4,355	NA	Waliszewski et al., 1999
Japan	1986-1987	1,800°		2,400 <sup>6</sup>		Kashimoto et al., 1989
Korea	1994-1995	190ª		1,100 <sup>b</sup>	400	Kang <i>et al.</i> , 1997
Korea	2000	23	27	153	NA	Yoo et al., 2001
Korea	2001	61	29	160	89°	Yoo et al., 2002

<sup>\*:</sup>  $\alpha$ -BHC+ $\beta$ -BHC+ $\gamma$ -BHC, b: p,p'-DDE+p,p'-DDT+p,p'-DDD, c: Sum of 7 marker PCBs, NA : not available

more slowly in our environment. The levels of p,p'-DDE and  $\beta$ -BHC were significantly different between tissues. Positive correlation coefficients between tissues were detected in the following cases: p,p'-DDE in the liver/blood (p<0.01, r=0.376), lung/blood (p<0.01, r=0.436), liver/lung (p<0.01, r=0.732), liver/adipose tissue (p<0.01, r=0.382), liver/kidney cortex (p<0.01, r=0.367), kidney cortex/adipose tissue (p<0.05, r=0.313), and lung/adipose tissue (p<0.051, r=0.317) :  $\beta$ -BHC in the liver/adipose tissue (p<0.01, r=0.706). As a result, p,p'-DDE was more readily accumulated and slowly eliminated from liver than from blood, lung, adipose tissue, and kidney cortex.

## CONCLUSION

The levels of  $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC,  $\delta$ -BHC, p,p'-DDE, p,p'-DDE, aldrine, dieldrine, endrine, PCB 28, 52, 101, 118, 138, 153, and 180 in blood, adipose tissue, liver, kidney cortex, and lung in terms of gender were determined. On the whole, the values were relatively lower than the reported values from other countries, but we could estimate that these compounds were extensively distributed in Korean human bodies. This study would be useful to establish the risk assessment of persistent organochlorines and PCB congeners in Koreans.

## **ACKNOWLEDGEMENT**

This study was supported by the grant from the KFDA of Korea (ED 2003-405-2).

## REFERENCES

- Archibeque-Engle, S. L., Tessari, J. D., Winn, D. T., Keefe, T. J., Nett, T. M., and Zheng, T., Comparison of organochlorine pesticide and polychlorinated biphenyl residues in human breast adipose tissue and serum. *J. Toxicol. Environ. Health*, 52, 285-293 (1997).
- Burgaz, S., Afkham, B. L., and Karakaya, A. E., Organochlorine pesticide contaminants in human adipose tissue collected in Tebriz(Iran). *Bull. Environ. Contam. Toxicol.*, 54, 546-553 (1995).
- Colborn, T., Vom Saal, F., and Soto, A. M., Developmental effects of endocrine disrupting chemicals in wildlife and humans. *Environ. Health Perspect.*, 101, 378-384 (1993).
- Dewailly, E., Dodin, S., Verreault, R., Ayotte, P., Sauve, L., Morin, J., and Brisson, J., High organochlorine body burden in women with estrogen receptor-positive breast cancer. *J. Natl. Cancer Inst.*, 86, 232-234 (1994).
- Dewailly, E., Mulvad, G., Pedersen, H. S., Ayotte, P., Demers, A., Weber, J. P., and Hansen, J. C., Concentration of organochlorines in human brain, liver, and adipose tissue autopsy samples from Greenland. *Environ. Health Perspect.*,

- 107, 823-828 (1999).
- Gallelli, G., Mangini, S., and Gerbino, C., Organochlorine residues in human adipose and hepatic tissues from autopsy sources in northern Italy. J. Toxicol. Environ. Health, 46, 293-300 (1995).
- Gomez-Catalan, J., Lezaun, M., To-Figueras, J., and Corbella, J., Organochlorine residues in the adipose tissue of the population of Navarra(Spain). *Bull. Environ. Contam. Toxicol.*, 54, 534-540 (1995).
- Hileman, B., Concerns broaden over chlorine and chlorinated hydrocarbons. *Chem. Eng. News*, 71, 11-20 (1993).
- Jensen, G. E. and Clausen, J., Organochlorine compounds in adipose tissue of Greenlanders and southern Danes. *J. Toxicol. Environ Health*, 5, 617-629 (1979).
- Kang, Y. S., Matsuda, M., Kawano, M., Wakimoto, T., and Min, B. Y., Organochlorine pesticides, polychlorinatedbiphenyls, polychlorinated dibenzo-p-dioxines and dibenzofurans in human adipose tissue from western Kyungnam, Korea. *Chemosphere*, 35, 2107-2117 (1997).
- Kutz, F. W., Wood, P. H., and Battimore, D. P., Organochlorine pesticide and polychlorinated biphenyls in human adipose tissue. Rev. Environ. Contam. Toxicol., 120, 71-82 (1991).
- Nakamura, H., Matsuda, M., Quynh, H. T., Cau, H. D., Chi, H. T. K., and Wakimoto, T., Levels of polychlorinated dibenzo-p-dioxines, dibenzofurans, PCBs, DDTs, and HCHs in human adipose tissue and breast milk from the south of Vietnam. *Organohalogen compounds*, 21, 71-76 (1994).
- Pauwels, A., Covaci, A., Weyler, J., Delbeke, L., Dhont, M., De Sutter, P., D'Hooghe, T., and Schepens, P. J. C., Comparison of persistent pollutant residues in serum and adipose tissue in a female population in Belgium, 1996-1998. *Arch. Environ. Contam. Toxicol.*, 39, 265-270 (2000).
- Schecter, A., Fürst, P., Krüger, C., Meemken, H. A., Groebel, W., and Constable, J. D., Levels of polychlorinated dibenzofurans, dibenzodioxines, PCBs, DDT, and DDE, hexachlorobenzene, dieldrin, hexachlorocyclohexanes, and oxychlordane in human breast milk from the United States, Thailand, Vietnam, and Germany. *Chemosphere*, 18, 445-454 (1989).
- Soto, A. M., Sonnenschein, C., Chung, K. L., Fernandez M. F., Olea, N., and Serrano, F. O., The E-SCREEN assay as a tool to identify estrogens: An update on estrogenic environmental pollutants. *Environ. Health Perspect.*, 103, 113-122 (1995).
- Stellman, S. D., Djordjevic, M. V., Muscat, J. E., Gong, L., Bernstein, D., Citron, M. L., White, A., Kemeny, M., Busch, E., and Nafziger, A. N., Relative abundance of organochlorine pesticides and polychlorinated biphenyls in adipose tissue and serum of women in Long Island, New York [see comments]. Cancer Epidemiol. Biomarkers Prev., 7, 489-496 (1998).
- Yoo, Y. C., Lee, S. K., Lee, S. Y., Yang, J. Y., In, S. W., Kim, K. W., and Chung, K. H., Distribution of Organochlorine

- Pesticides in Korean Human Tissues. *Yakhak Hoeji*, 45, 366-377 (2001).
- Yoo, Y. C., Lee, S. K., Lee, Kim, K. W., Lee, S. Y., Yang, J. Y., Kim, Y. S., Oh, S. M., and Chung, K. H., Distribution of Organochlorins and PCB Congeners in Korean Adipose Tissue, Liver, and Whole Blood. *Yakhak Hoeji*, 46, 247-257 (2002).
- Waliszewski, S. M., Aguirre, A. A., Infanzon, R. M., Benitez, A., and Rivera, J., Comparison of organochlorine pesticide levels in adipose tissue and human milk of mothers living in Veracruz, Mexico. *Bull. Environ. Contam. Toxicol.*, 62, 685-
- 690 (1999).
- Weistrand, C. and Noren, K., Polychlorinated naphthalenes and other organochlorine contaminants in human adipose and liver tissue. *J. Toxicol. Environ. Health*, *Part A*, 53, 293-311 (1998).
- WHO: Assessment of health risks in infants associated with exposure to PCBs, PCDDs, and PCDFs in breast milk. *Environ. Health Series*, 29 (1988).
- Wolff, M. S., Fischbein, A., and Selikoff, I. J., Changes in PCB serum concentrations among capacitor manufacturing workers. *Environ. Res.*, 59, 202-216 (1992).