# DIETARY HABITS AND BLOOD POLYCHLORINATED DIBENZO-p-DIOXINS, DIBENZOFURANS AND COPLANAR PCB LEVELS AMONG JAPANESE RESIDENTS

Sachiko Nagasaka<sup>1</sup>, Chihiro Nagata<sup>1</sup>, Eri Ohwa<sup>1</sup>, Mika Ozaki<sup>1</sup>, Sayaka Yanai<sup>1</sup>, Yuriko Kikuchi<sup>1</sup>, Jamshid Hosseinpour<sup>2</sup> and <u>Shaw Watanabe<sup>1</sup></u>

<sup>1</sup>Department of Applied Bioscience, Tokyo University of Agriculture, Setagaya, Tokyo 156-8502, Japan, <sup>2</sup>The Bayreuth Institute for Environmental Research, Bernecker Str. 17-21, 95448 Bayreuth, Germany

### Introduction

Background exposure of dioxins among Japanese residents was 21.5 pgTEQ/g lipid in average. This value is almost the similar value of most developed countries. In this study, we focused the origin of these exposure with special reference to the dietary habits.

## **Subjects and Methods**

Japanese residents in 17 different areas participated the study on health and dioxin exposure between 1998-2002. Participants were all volunteers aged from 40s' to 60s'. They received detailed explanation of the study and gave us the written informed consent.



Life habits and dietary habits were collected by questionnaire and checked by trained dieticians. The questionnaire included dietary habits, smoking and drinking habits, residential and work environment, physical activity, past history of diseases and treatments, reproductive history, etc.

About 30 ml of blood was divided to tubes to perform peripheral blood tests. These tests were performed by the Serum Research Laboratory (Tokyo).

Blood PCDD/PCDF/Co-PCB was measured by a modification of Patterson's method. Lipids were extracted from 70-80 g of whole blood with a solution of 30 ml saturated ammonium sulfate and 80 ml of ethanol:hexane (1:3) solution after the addition of an internal standard of <sup>13</sup>C- labeled mixed dioxin congener solution (Cambridge Isotope Laboratories, Massachusetts, USA)). Clean up was achieved by a multilayer silica column. Analysis of PCDD/PCDF/Co-PCB was carried out by gas chromatography-high resolution mass spectrometry (GC-MS). Details are described in the previous paper<sup>1</sup>.

The toxicity of the dioxins was calculated by the WHO TEF method (1997) and is expressed as TEQ/g lipid and body burden. Estimated intake of foods was calculated from food-frequency questionnaire with portion size. Validation study of this questionnaire has been separately done. Statistical analysis. SPSS version 10 was used for the statistical analyses. Correlation analysis was performed between PCDD/PCDF/Co-PCB and various variables.

#### Results

Dioxin levels of 740 participants who completed the study were 10.6+/-7.6 (median 8.4) pgTEQ/g lipid of PCDDs and 7.2+/-5.4 (median 5.9) pgTEQ/g lipid of PCDFs, 5.6+/-6.2 (median 3.6) pgTEQ/g lipid of PCBs, and 23.3+/-15.6 (median 19.1) pgTEQ/g lipid of total in males. In females, these were 9.6+/-6.8 (median 7.5) pgTEQ/g lipid of PCDDs and 6.1+/-3.9 (median 5.3) pgTEQ/g lipid of PCDFs, 3.9+/-4.2 (median 2.6) pgTEQ/g lipid of PCBs, and 19.7+/-11.5 (median 17.2) pgTEQ/g lipid of total. Body burden was 4.5+/-3.8 (median 5.4) in males, and 4.7+/-3.4 (median 5.5) in females.

Reliability of food frequency questionnaire (FFQ) was confirmed by the correlation between the foods and meals. Multiplying the food frequency and portion size yielded the reasonable weight of food consumption. Total amount of daily intake was 848+/-265 g in males and 810+/-227 g in females. Dioxin contents of these foods were obtained from the tables made by Koseikagaku-Kenkyu supported from the Ministry of Health, Labor and Welfare, and the Dioxin contents table of fishes and clams reported by the Agency of Fishery, Ministry of Agriculture and Fishery. Doses of dioxin intake from foods by individual participants to the study were thus calculated (Table 1). Total amount of intake was 40.3+/-22.8 pg TEQ/day in males, and 39.8+/-20.4 pg TEQ/day in females. It was 0.6+/-0.3 pg TEQ/kg body weight/day in males 0.75+/-0.4 pg TEQ/kg body weight/day in females. Largest source of dioxins were from fishes and clams occupying 73% in males and 70% in females. Next was meat and egg (12.8% in males and 12.4% in females), and diary products (9.9% in males and 11.8% in females). Individual food intake frequency showed various correlation with blood dioxin levels (Table 2). Beef and eggs showed significant correlation with PCDD, and beef, pork and chicken showed it with PCB.

	Tapie I. I	stimated 1	ntaké of l	DIOXINS IT	COM FOODS		
	Males			Females			
Foods	PCDD/DF	Co-PCB	Total	PCDD/DF pgTEQ/da	Co-PCB	Total	
	pgTEQ/day	pgTEQ/day	pgTEQ/day	У У	pgTEQ/day	pgTEQ/day	
Beef	1.99±2.25	0.30±0.34	2.29±2.59	1.73±2.1 5 0.03±0.0	0.26±0.322	2.00±2.47	
Pork	0.03±0.02	0.07 <b>±</b> 0.06	0.10±0.08		0.08±0.720	0.11±0.10	
Chicken	0.32±0.36	0.30±0.32	0.62±0.68		0.34±0.29(	0.71±0.60	
Ham	0.12±0.11	0.01±0.01	0.13±0.01		0.01±0.01(	0.01±0.02	
Bacon	0.00±0.00	0.01±0.01	0.01±0.01		0.01±0.01(	0.01±0.02	
Egg	0.68±0.53	1.32±1.01	2.00±1.54		1.37±0.952	2.09±1.45	
Milk	1.91±1.92	1.06±1.06	2.97±2.98	2.04±1.8 4 0.48±0.6	1.12±0.133	3.16±2.86	
Yogurt	0.25±0.5	0.14±0.27	0.39±0.78		0.26±1.01(	0.74±1.00	
Cheese	0.19±0.33	0.14±0.25	0.33±0.58		0.17±0.360	0.39±0.60	
Butter	0.17±0.31	0.14±0.26			0.18±0.260	0.40±0.75	
Fish coast	4.10±3.99	9.52±9.28	13.62±13. 27	-	9.52±8.75	L3.62±12.51	
Fish ocean	1.78±1.89	6.62±7.03	8.40±8.92		5.88±5.73	7.46±7.27	
Squid Octopus	1.05±1.12	0.74±0.52	1.79±1.9	4 1.47±2.1	0.57±0.52	L.38±1.27	
Crab	1.48±2.03	1.05±1.52	2.53±3.45	6 0.96±0.9	1.04±1.522	2.51±3.68	
Shrimp	0.94±1.07	0.65±0.63	1.59±1.80	2 0.02±0.0	0.66±0.631	L.62±1.55	
Fish past	0.02±0.03	0.04±0.05	0.06±0.08	1.09±1.0	0.05±0.05(	0.07±0.08	
Clam	1.14±1.04	0.29±0.27	1.43±1.31		0.28±0.27	L.37±1.30	
Gy vegetable	0.99±1.01	0.12±0.16	1.11±1.14	1.46±1.2 9 0.02±0.0	0.18±0.161	L.64±1.45	
Carrot	0.01±0.01	0.00±0.00	0.01±0.01		0.00±0.00(	0.02±0.01	
Tofu	0.07±0.06	0.00±0.00	0.07±0.06		0.00±0.00(	0.07±0.06	

Table 1. Estimated Intake of Dioxins from Foods

Organohalogen Compounds, Volumes 60-65, Dioxin 2003 Boston, MA

				0.01±0.0	)	
Natto	0.01±0.01	0.00±0.00	0.01±0.01	1	0.00±0.0	00.01±0.01
				0.00±0.0	)	
Mushroom	0.00±0.00	0.00±0.00	0.00±0.00	1	0.00±0.0	00.00±0.01
				0.05±0.0	1	
Fruits	0.03±0.04	0.00±0.00	0.03±0.04	5	0.00±0.0	00.05±0.05
				0.38±0.1		
Rice	0.47±0.19	0.00±0.00	0.47±0.19	3	0.00±0.0	00.38±0.13
Total						
Intake(pgTEQ/	d	22.52±14.6	540.27±22.	17.83±8.	21.98±12	•
ay)	17.75±8.79	9 6	83	04	94	39.82±20.39
Intake(pgTEQ/	k			0.34±0.1		
g/day)	0.27±0.13	0.33±0.22	0.60±0.34	6	0.41±0.2	50.75±0.40

## Discussion

In Japan, 90% of the daily intake of PCDDs, PCDFs, and other dioxin-like compounds is estimated to come from food. The participants in this study remained to consume dioxins less than 1 pg TEQ/kg body weight/day. Positive correlation between the foods and dioxin levels was present in meats, dairy products and fishes as reported earlier.

Level					
					L_TOT_TE
Food		L_D_TEQ	L_F_TEQ	L_PCB_TEQ	Q
	Pearson's				
Beef	CC	0.128**	0.093*	0.167**	0.153**
	Pearson's				
Pork	CC	-0.013	-0.015	0.155**	0.05
	Pearson's				
Chicken	CC	0.065	0.024	0.097*	0.085*
	Pearson's				
Egg	CC	0.118**	0.005	0.031	0.078*
	Pearson's				
Cheese	CC	0.005	0.013	0.100**	0.044
	Pearson's				
Butter	CC	0.066	0.059	0.06	0.076*
	Pearson's				
Margarine	CC	0.139**	0.089*	0.075*	0.144**
	Pearson's				
Fish coast	CC	0.080*	0.118**	0.045	0.092*
	Pearson's				
Fish ocean	CC	-0.009	0.107**	0.171**	0.080*
	Pearson's				
Squid Octopus CC		0.112**	0.142**	0.054	0.129**
Fish paste	Pearson's	0.078*	0.003	-0.027	0.040

Table 2. Correlations between Food Intake and Blood Dioxin

Organohalogen Compounds, Volumes 60-65, Dioxin 2003 Boston, MA

	CC				
	Pearson's				
Clam	CC	0.071	0.083*	0.071	0.087*
	Pearson's				
Tofu <sup>1</sup>	CC	-0.028	-0.139**	-0.082*	-0.092*
	Pearson's				
Natto <sup>2</sup>	CC	-0.130**	-0.116**	0.000	-0.093*
	Pearson's				
Rice	CC	-0.020	0.008	-0.010	-0.06
*p<0.05, ** p<0.01, <sup>1</sup> Tofu; soy bean card, <sup>2</sup> Natto; Fermented soy					
bean.					

It is interesting that the soybean products, tofu and natto, showed negative correlation with blood dioxin levels. Our previous study on the half-life of dioxins in the body was shorter than that reported in the western countries. It suggested that the cumulated dioxins in the body could be eliminated by the control of dietary habits.

#### References

1) <u>Watanabe S</u>, Kitamura K, Kikuchi Y, Sunaga M, Iida T, Waechter G, Yamamoto F. (2000) Health effects of chronic exposure to polychlorinated dibenzo-p-dioxins, dibenzofurans and coplanar PCB around municipal waste incineraters. Organohalogen Compounds 48: 199-202.

2) Kitamura K, Kikuchi Y, <u>Watanabe S</u>, Waechter G, Sakurai H, Takada T. (2000) Health effects of chronic exposure to polychlorinated dibenzo-p-dioxin (PCDD), dibenzofurans (PCDF), and coplanar PCB (Co-PCB) of municipal waste incinerator workers. J Epidemiol 10: 262-270.