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THE SURVEY OF THE EXPOSURE TO DIOXINS AND OTHER CHEMICAL COMPOUNDS IN HUMANS (IV) - DIOXINS AND OTHER CHEMICAL COMPOUNDS CONCENTRATION IN HUMAN BODIES OF GENERAL PUBLIC IN JAPAN AND INTAKE SURVEY FROM FOOD -

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Introduction

In 2002, the Risk Assessment Office of the Ministry of the Environment, Japan, started a survey entitled "Survey of Accumulation of Dioxins in Humans", in order to obtain a general picture of the state of the accumulation of dioxins in the Japanese people.

The accumulation and uptake of dioxins in Japan were measured by analyzing dioxins in blood and dioxin intake from foods in this survey. In 2011, the survey was expanded as "Exposure Monitoring Survey of Dioxins and Other Chemical Compounds" to include chemical compounds other than dioxins. This report summarizes the results from 2011 to 2015.

Materials and methods

In this survey, three areas were selected for each year: two areas from coastal villages where blood dioxins concentration was relatively high, and one area from inland area as control. Twenty-five participants and 30 participants were recruited from high concentration area and control area, respectively. These areas were selected from areas where former dioxin survey was conducted. Priority was given to the recruitment of participants of former surveys.

A briefing session was held in each survey area, and 51 mL of venous blood was collected from each subject, after having obtained informed consent. Items analyzed besides chemical pollutants included general biochemical tests, blood counts, thyroid function, and four unsaturated fatty acids (DGLA, AA, EPA, and DHA). Also, an inquiry about dietary and health conditions was performed by community health nurses and nutritionists, to grasp the living and health conditions of the subjects. In addition, 200 mL urine was sampled in the morning of blood sampling.

Dietary surveys using duplicate portion analysis were conducted on approximately five subjects from each area, in addition to the blood survey. The meals for a total of whole three days were collected, to measure the chemical compounds concentration and to calculate chemical compounds intake via daily meals. When collecting meals, details of seasonings and ingredients were asked and recorded by a nutritionist.

From FY 2011 to FY 2015, chemical compound concentration in blood and urine of 410 people (the mean of age 50.1 years old, ranging from 24 to 77 years old) from 15 survey regions was measured. In addition, the chemical compound intake from food was estimated for 75 people.

Blood analyses were performed as previously reported .

Results and discussion

The mean blood dioxin concentration of the 410 subjects was 11 pg-TEQ/g-fat, ranging from 0.39 to 56 pg-TEQ/g-fat. The dioxins concentration was within the range of the former surveys conducted from FY 2002 to FY 2010 (Table 2).

Dioxin intake from food was calculated for the 75 subjects (Table 2).The mean dioxin intake from food was 0.50 pg-TEQ/kg/day, with a range of 0.035 to 2.4 pg-TEQ/kg/day.

Figure 1 shows chronological change of the blood dioxin concentration and intake from food. Enforcement of the "Act on Special Measures against Dioxins (January, 2000)" has greatly decreased emission of dioxins into the environment. As a result, the blood dioxin concentration and intake from food has decreased.

Among the 410 subjects, 77 people had participated in the past surveys, and had blood dioxins concentration analyzed. The dioxin concentrations in blood have decreased in most subjects (Figure2).

The measurement results of chemical compounds besides dioxins are shown in Tables 3 and 4. The analysis is currently being conducted.

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Reference
1. Hasegawa et al., (2007) Organohalogen Compounds, 69, 2001-2005
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Table 1. Chemical substance

Classification	Chemical substance	Blood	Urine	Diet
Polychlorinated dioxin	PCDDs, PCDFs, Co-PCBs	*		*
Polybrominated dioxins	PBDDs, PBDFs	*		
Organofluorine compounds	PFOS, PFOA, PFHxA, PFHpA, PFHxS, PFTeDA, PFNA, PFDA, PFUdA, PFDS, PFDoA, PFTrDA	*		*
Hydroxylated polychlorobiphenyls	5Cl-HO-PCBs, 6Cl-HO-PCBs, 7Cl-HO-PCBs	*		
Metals	T-Hg, Pb, As, Cu, Se, Zn, Mn	*		*
	Me-Hg			*
	Cd	*	*	*
	Speciated As ((Ⅲ), (Ⅴ), arsenobetaine, methylarsonic acid, dimethylarsinic acid)		*	
Pesticides (metabolites)	OP metabolites, Pyrethroid metabolites, Ethylenethiourea, Triclosan, Acephate, Methamidophos, 6-Chloronicotinic acid, 3-methyl-4-nitrophenol, p-nitrophenol, Deet		*	
POPs	PCB, DDT, Chlordane, Drins, HCB, Heptachlor, Toxaphene, Mirex, PBDE, Pentachlorobenzene, HCH, Chlordecone, Hexabromobiphenyl, Endsulfan, HBCD	*		*
Others	Mono(2-ethylhexyl) phtalete, Bisphenol A, PAHs(Polycyclic aromatic hydrocarbons) metabolites, Parabens, Iodine, Perchloric acid, Phytoestrogens, Caffeine, Cotinine, 8-OHdG,Benzophenone3		*	

Table 2. Comparison with past survey results (Blood Dioxin and Dioxin intake from food)

	Blood concentration (pg-TEQ/g-fat)		Intake from food (pg-TEQ/kg/day)	
TEQ	FY2002-FY2010 (n=2,264)	FY2011-FY2015 (n=410)	FY2002-FY2010 (n=625)	FY2011-FY2015 (n=75)
PCDDs+PCDFs	11±7.6 (9.8, 0.040 - 63)	6.7±4.6 (5.8, 0.013 - 28)	0.35±0.35 (0.25, 0.015-3.8)	0.21±0.18 (0.16, 0.016-1.0)
Co-PCBs	7.9±7.2 (5.6, 0.013 - 81)	4.7±4.1 (3.6, 0.052 - 36)	0.47±0.58 (0.28, 0.016-4.2)	0.29±0.35 (0.16, 0.019-1.8)
PCDDs+PCDFs +Co-PCBs)	19±14 (16, 0.10 - 130)	11±8.1 (9.5, 0.39 - 56)	0.82±0.86 (0.56, 0.031-6.2)	0.50±0.49 (0.34, 0.035-2.4)

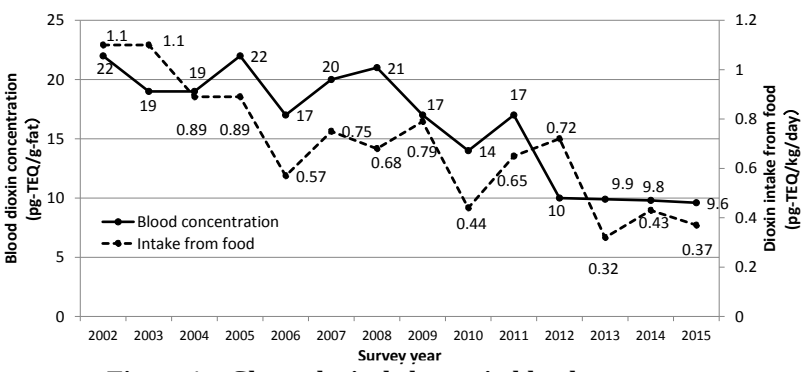

Figure 1. Chronological change in blood dioxin concentration and intake from food.

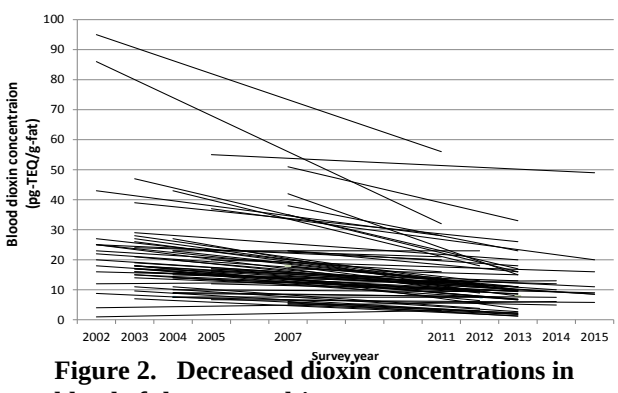

Figure 2. Decreased dioxin concentrations in blood of the same subjects.

Table 3. Concentrations of chemical substances in blood and intake from food（FY 2011-FY 2015）

Classification	Chemical substance	Blood concentration Median(range)	Unit	Rate of detection	Intake from food Median(range)	Unit	Rate of detection
Polybrominated dioxins	PBDDs	all N.D.	pg/g-fat	0/84	—	—	—
	PBDFs	all N.D.		0/84	—	—	—
Organofluorine compounds	PFOS	3.5（0.29～17）	ng/mL	326/326	0.53（N.D.～1.7）	ng/kg-weight/day	10/15
	PFOA	1.8（0.27～13）		326/326	0.62（N.D.～2.9）		12/15
	PFHxA	All N.D.		0/240	—	—	—
	PFHpA	N.D.（N.D.～1.2）		28/240	—	—	—
	PFHxS	0.37（N.D.～1.8）		229/240	—	—	—
	PFTeDA	N.D.（N.D.～0.41）		16/240	—	—	—
	PFNA	1.4（0.30～7.7）		240/240	—	—	—
	PFDA	0.52（0.092～2.7）		240/240	—	—	—
	PFUdA	1.2（0.13～6.4）		240/240	—	—	—
	PFDS	N.D.（N.D.～0.065）		3/240	—	—	—
	PFDoA	0.15（N.D.～0.89）		189/240	—	—	—
	PFTrDA	0.39（N.D.～2.7）		236/240	—	—	—
Hydroxylated polychloro-biphenyls	5Cl-HO-PCBs	27（1.2～120）	pg/g	60/60	—	—	—
	6Cl-HO-PCBs	30（1.6～200）		60/60	—	—	—
	7Cl-HO-PCBs	26（4.0～200）		60/60	—	—	—
Metals	T-Hg	8.3（1.5～41）	ng/mL	410/410	0.060（N.D.～0.30）	µg/kg-weight/day	68/75
	Me-Hg	—	—	—	0.055（N.D.～0.29）		67/75
	Cd	1.1（0.25～6.2）	ng/mL	324/324	0.23（0.059～0.57）		75/75
	Pb	11（4.3～37）		324/324	0.074（N.D.～0.28）		74/75
	As	5.3（0.70～110）		324/324	2.2（0.54～14）		60/60
	Cu	850（590～1,500）		324/324	16（8.2～38）		60/60
	Se	190（110～480）		324/324	1.2（0.56～2.5）		60/60
	Zn	6,400（3,800～8,600）		324/324	120（65～220）		60/60
	Mn	13（5.8～53）		240/240	58（28～140）		45/45

Table 4. Concentrations of chemical substances in urine (Creatinine corrected) （FY 2011-FY 2015）

Classification		Chemical substances	Median(range)	Unit	Rate of detection
Pesticide metabolites	OP metabolites	DMP	2.8（N.D.～140）	μg /g Cr	172/182
		DEP	3.1（N.D.～520）		146/182
		DMTP	4.7（N.D.～110）		125/182
		DETP	N.D.（N.D.～16）		36/182
	Pyrethroid metabolites	PBA	0.27（N.D.～8.7）		139/182
		DCCA	N.D.（N.D.～21）		37/182
	Carbamate metabolite	Ethylenethiourea	N.D.（N.D.～0.50）		6/120
	Triclosan		1.3（0.12～380）		182/182
	Acephate		N.D.（N.D.～11）		12/75
	Methamidophos		N.D.（N.D.～0.19）		3/75
	Imidacloprid metabolite	6-Chloronicotinic acid	N.D.（N.D.～1.8）		15/137
	Fenitrothion metabolite	3-methyl-4-nitrophenol	N.D.（N.D.～4.2）		32/75
	p-nitrophenol		0.67（N.D.～4.6）		71/75
	Deet		N.D.（N.D.～0.087）		1/75
Others	Mono(2-ethylhexyl) phtalete	MBP	16（3.7～5,200）		340/340
		MEHP	2.7（0.23～22）		340/340
		MEHHP	8.8（1.2～59）		340/340
		MEOHP	5.6（0.55～35）		340/340
		MBzP	0.53（N.D.～38）		323/340
	Bisphenol A		0.31（N.D.～31）		298/340
	PAHs	1-Hydroxypyrene	0.12（N.D.～4.7）		70/75
		1&9- ydroxyphenanthrene	0.12（N.D.～0.69）		69/75
		2- Hydroxyphenanthrene	0.095（N.D.～0.46）		63/75
		3- Hydroxyphenanthrene	0.17（N.D.～0.65）		72/75
		4- Hydroxyphenanthrene	N.D.（N.D.～0.20）		21/75
	Parabens	Methylparaben	82（1.3～2,500）		75/75
		Ethylparaben	4.3（N.D.～410）		59/75
		Propylparaben	1.1（N.D.～110）		43/75
		Butylparaben	N.D.（N.D.～87）		26/75
		Benzylparaben	All N.D.		0/75
	phytoestrogens	Genistein	1,100（74～7,600）		75/75
		Daidzein	1,600（27～19,000）		75/75
		Equol	190（N.D.～28,000）		72/75
	Caffein		1,800（0.36～22,000）		75/75
	Cotinine		0.29（N.D.～3,000）		52/75
	8-OHdG		5.0（1.2～7.7）		15/15
	Benzophenone3		N.D.（N.D.～190）		11/75
Metals	As	As (Ⅴ)	N.D.（N.D.～2.9）		92/340
		As (Ⅲ)	1.4（N.D.～6.9）		301/340
		methylarsonic acid	1.8（N.D.～13）		319/340
		dimethylarsinic acid	30（6.2～170）		340/340
		arsenobetaine	44（2.1～2,300）		340/340
	Cd		0.79（0.11～4.7）		340/340