The survey of the exposure to dioxins and other chemical compounds in humans （Ⅲ）

- 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[1].

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 2013.

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 50 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, thyroidal 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, 100 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 2013, chemical compound concentration in blood and urine of 253 people (the mean of age 50.6 years old, ranging from 26 to 77 years old) from 9 survey regions was measured. In addition, the chemical compound intake from food was estimated for 45 people.

Blood analyses were performed as previously reported [2].

Results and discussion

The mean blood dioxin concentration of the 253 subjects was 12 pg-TEQ/g-fat, ranging from 0.40 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).

The mean dioxin intake from food was 0.56 pg-TEQ/kg/day, with a range of 0.035 to 2.4 pg-TEQ/kg/day. Dioxin intakes from food were calculated for the 45 subjects (Table 3).

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 environment. As a result, the blood dioxin concentration and intake from food has decreased.

Among the 253 subjects, 63 people had participated in the past surveys and had blood dioxins concentration analyzed. The dioxin concentrations in blood have decreased in most subjects (Table 4, Figure2).

The measurement results of chemical compounds besides dioxins are shown in Tables 5 and 6. The analysis is currently being conducted.

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Reference

1. Hasegawa et al., (2007) *Organohalogen Compounds*, **69**, 2001-2005
2. Matsumura et.al., (2007) *Organohalogen Compounds*, **69**, 1154-1157

Table 1. Chemical substance

classification	Chemical substance	blood	urine	diet
Polychlorinated dioxin	PCDDs, PCDFs, Co-PCBs	*		*
Polybrominated Dioxins	PBDDs, PBDFs	*		
Hydroxylated polychlorobiphenyl	5Cl-HO-PCBs, 6Cl-HO-PCBs, 7Cl-HO-PCBs	*		
Heavy metals	T-Hg, Pb, As, Cu, Se, Zn, Mn	*		*
	Me-Hg			*
	Cd	*	*	*
	Speciated As ((Ⅲ), (Ⅴ), arsenobetaine, methylarsonic acid, dimethylarsinic acid)		*	
Pesticide (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	*		*
Radioactive substances	Cesium-134, Cesium-137, Iodine-131, Pottassium-40	*	*	*
Others	Mono(2-ethylhexyl) phtalete, bisphenol A, PAHs(Polycyclic aromatic hydrocarbons) metabolites, Parabens, phytoestrogens, caffeine, cotinine, benzophenone3		*	

Table 2. Comparison with past survey result (Blood Dioxin)

TEQ	FY2002-FY2010 (n=2,264)	FY2011-FY2013 (n=253)
PCDDs+PCDFs (pg-TEQ/g-fat)	11±7.6 (9.8, 0.040 - 63)	7.4±5.2 (6.1, 0.013 - 28)
Co-PCBs (pg-TEQ/g-fat)	7.9±7.2 (5.6, 0.013 - 81)	5.1±4.4 (3.9, 0.054 - 36)
PCDDs+PCDFs +Co-PCBs (pg-TEQ/g-fat)	19±14 (16, 0.10 - 130)	12±8.9 (10, 0.40 - 56)

mean±SD
median, range

Table 3. Comparison with past survey result (Dioxin intake from food)

TEQ	FY2002-FY2010 (n=625)	FY2011-FY2013 (n=45)
PCDDs+PCDFs (pg-TEQ/kg/day)	0.35±0.35 (0.25, 0.015-3.8)	0.31±0.30 (0.21, 0.016-1.0)
Co-PCBs (pg-TEQ/kg/day)	0.47±0.58 (0.28, 0.016-4.2)	0.34±0.47 (0.17, 0.019-1.7)
PCDDs+PCDFs +Co-PCBs (pg-TEQ/kg/day)	0.82±0.86 (0.56, 0.031-6.2)	0.65±0.71 (0.39, 0.035-2.4)

mean±SD
median, range

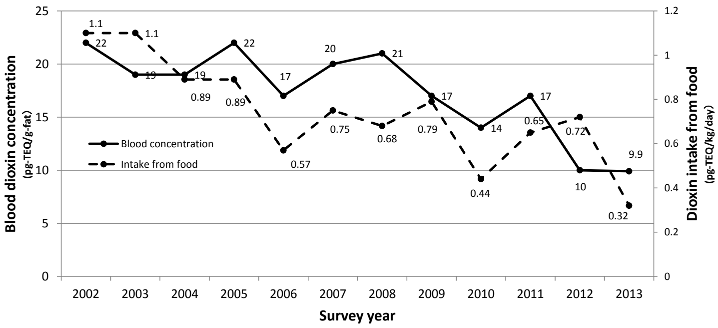

Figure 1. Chronological change in blood dioxin concentration and intake from food.

Table 4. Comparison with past survey result for the same subjects (Blood Dioxin)

TEQ	FY 2002-2004 (n=63)	FY 2011- 2013 (n=63)
PCDDs+PCDFs (pg-TEQ/g-fat)	14±8.2 (12, 0.87 - 48)	6.8±4.4 (6.1, 0.69 - 20)
Co-PCBs (pg-TEQ/g-fat)	8.8±9.2 (6.4, 0.080 - 59)	5.4±5.3 (4.0, 0.092 - 36)
PCDDs+PCDFs +Co-PCBs (pg-TEQ/g-fat)	23±16 (18, 0.96 - 95)	12±9.1 (9.9, 1.2 - 56)

mean±SD
median, range

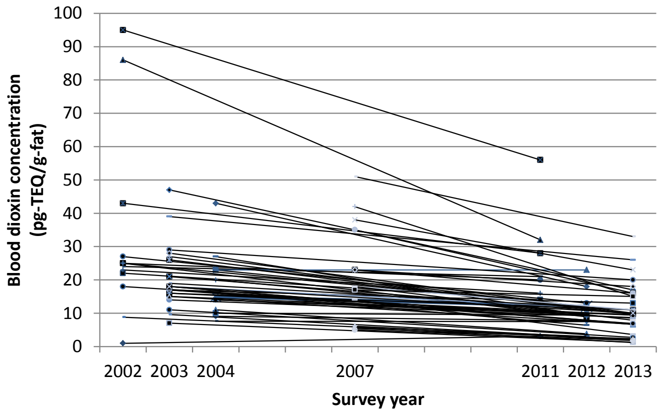

Figure 2. Decreased dioxin concentrations in blood of the same subjects.

Table 5. Blood concentrations and intake from food（FY 2011-FY 2013）

classification	Chemical substance	Blood concentration			Intake from food		
		median(range)	unit	rate of detection	median(range)	unit	rate of detection
Polybrominated Dioxins	PBDDs	all N.D.	pg/g-fat	0/84	—	—	—
	PBDFs	all N.D.		0/84	—	—	—
Hydroxylated polychloro-biphenyls	5Cl-HO-PCBs	24（1.2～120）	pg/g	30/30	—	—	—
	6Cl-HO-PCBs	30（1.6～200）		30/30	—	—	—
	7Cl-HO-PCBs	23（4.0～130）		30/30	—	—	—
heavy metals	T-Hg	8.7（1.7～41）	ng/mL	253/253	0.062（N.D.～0.30）	μg/kg-weight/day	44/45
	Me-Hg	—	—	—	0.059（N.D.～0.29）		43/45
	Cd	1.1（0.25～3.5）	ng/mL	167/167	0.24（0.059～0.57）		45/45
	Pb	11（4.8～31）		167/167	0.093（0.024～0.28）		45/45
	As	4.5（1.2～35）		167/167	2.4（0.76～14）		30/30
	Cu	830（590～1,400）		167/167	18（8.2～26）		30/30
	Se	180（110～480）		167/167	1.3（0.64～2.5）		30/30
	Zn	6,400（4,700～7,800）		167/167	140（80～190）		30/30
	Mn	13（7.4～25）		83/83	66（38～110）		15/15

Table 6. Urine concentrations (creatinine corrected) （FY 2011-FY 2013）

classification		Chemical substance	median(range)	unit	rate of detection
Pesticide metabolites	OP metabolites	DMP	3.3 （N.D.～140）	μg /g cr	74/75
		DEP	4.0 （N.D.～520）		60/75
		DMTP	7.3 （N.D.～7.3）		57/75
		DETP	N.D. （N.D.～8.3）		15/75
	Pyrethroid metabolites	PBA	0.19 （N.D.～3.4）		49/75
		DCCA	N.D. （N.D.～13）		9/75
	Carbamate metabolite	Ethylenethiourea	N.D. （N.D.～0.21）		5/75
	Triclosan		1.2 （0.15～380）		75/75
	Acephate		N.D. （N.D.～1.9）		3/45
	Methamidophos		N.D. （N.D.～0.0058）		2/45
	Imidacloprid metabolite	6-Chloronicotinic acid	N.D. （N.D.～1.8）		8/45
	Fenitrothion metabolite	3-methyl-4-nitrophenol	N.D. （N.D.～2.8）		19/45
	parathion metabolite	p-nitrophenol	0.73 （0.23～4.6）		45/45
	Deet		All N.D.		0/45
others	Mono(2-ethylhexyl) phtalete	MBP	19 （5.5～5,200）	μg /g cr	182/182
		MEHP	3.2 （0.54～22）		182/182
		MEHHP	11 （2.7～59）		182/182
		MEOHP	6.8 （1.1～35）		182/182
		MBzP	0.61 （N.D.～38）		178/182
	Bisphenol A		0.37 （N.D.～31）		162/182
	PAHs	1-Hydroxypyrene	0.15 （N.D.～0.76）		44/45
		1&9-Hydroxyphenanthrene	0.12 （0.029～0.60）		45/45
		2- Hydroxyphenanthrene	0.11 （N.D.～0.39）		42/45
		3- Hydroxyphenanthrene	0.20 （N.D.～0.65）		43/45
		4- Hydroxyphenanthrene	N.D. （N.D.～0.20）		14/45
	Parabens	Methylparaben	55 （1.3～2,500）		45/45
		Ethylparaben	2.7 （N.D.～410）		35/45
		Propylparaben	1.2 （N.D.～77）		27/45
		Butylparaben	N.D. （N.D.～64）		13/45
		Benzylparaben	All N.D.		0/45
	phytoestrogens	Genistein	1,300 （190～5,700）		45/45
		Daidzein	1,800 （97～19,000）		45/45
		Equol	370 （N.D.～28,000）		44/45
	caffein		2,000 （0.36～22,000）		45/45
	Cotinine		0.36 （N.D.～1,600）		37/45
	benzophenone3		N.D. （N.D.～190）		9/45
Heavy metals	As	As (Ⅴ)	N.D. （N.D.～2.9）	μg /g cr	62/182
		As (Ⅲ)	1.6 （N.D.～6.9）		167/182
		methylarsonic acid	2.1 （N.D.～13）		176/182
		dimethylarsinic acid	32 （6.7～170）		182/182
		arsenobetaine	36 （2.1～640）		182/182
	Cd		0.80 （0.11～3.9）		182/182