

The survey of the exposure to dioxins and other chemical compounds in humans □□□

- 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 people¹.

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 2014.

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 61 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 2014, chemical compound concentration in blood and urine of 334 people (the mean of age 50.3 years old, ranging from 26 to 77 years old) from 12 survey regions was measured. In addition, the chemical compound intake from food was estimated for 60 people.

Blood analyses were performed as previously reported².

Results and discussion

The mean blood dioxin concentration of the 334 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 2).

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 334 subjects, 69 people had participated in the past surveys and had blood dioxins concentration analyzed. The dioxin concentrations in blood have decreased in most subjects (Figure 2).

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
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	*		
Organofluorine compounds	PFOS, PFOA, PFHxA, PFHpA, PFHxS, PFTeDA, PFNA, PFDA, PFUdA, PFDS, PFDoA, PFTTrDA	*		*
Hydroxylated polychlorobiphenyl	5Cl-HO-PCBs, 6Cl-HO-PCBs, 7Cl-HO-PCBs	*		
Metals	T-Hg, Pb, As, Cu, Se, Zn, Mn	*		*
	Me-Hg	*		*
	Cd	*	*	*
	Speciated As ((□), (□), 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, 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 and Dioxin intake from food)

TEQ	Blood concentration (pg-TEQ/g-fat)		Intake from food (pg-TEQ/kg/day)	
	FY2002-FY2010 (n=2,264)	FY2011-FY2014 (n=334)	FY2002-FY2010 (n=625)	FY2011-FY2014 (n=60)
PCDDs+PCDFs	11±7.6 (9.8, 0.040 - 63)	7.0±4.8 (5.9, 0.013 - 28)	0.35±0.35 (0.25, 0.015-3.8)	0.31±0.30 (0.21, 0.016-1.0)
Co-PCBs	7.9±7.2 (5.6, 0.013 - 81)	4.8±4.1 (3.9, 0.054 - 36)	0.47±0.58 (0.28, 0.016-4.2)	0.34±0.47 (0.17, 0.019-1.7)
PCDDs+PCDFs +Co-PCBs)	19±14 (16, 0.10 - 130)	12±8.4 (9.9, 0.40 - 56)	0.82±0.86 (0.56, 0.031-6.2)	0.65±0.71 (0.39, 0.035-2.4)

mean±SD

median, range

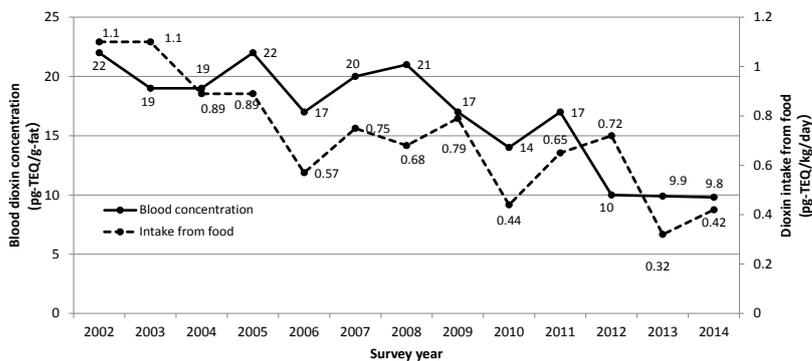


Figure 1. Chronological change in blood dioxin concentration and intake from food.

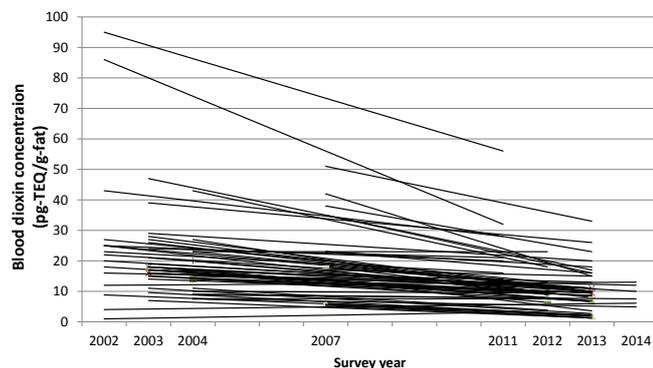


Figure 2. Decreased dioxin concentrations in blood of the same subjects.

Table 3. Blood concentrations and intake from food □ FY 2011-FY 2014 □

classification	Chemical substance	Blood concentration			Intake from food		
		median(range)	unit	rate of detection	median(range)	unit	rate of detection n
Polybrominated Dioxins	PBDDs	all N.D.	pg/g-fat	0/84	□	□	□
	PBDFs	all N.D.	pg/g-fat	0/84	□	□	□
	PFOS	3.8 □ 0.29 □ 17 □		250/250	0.53 □ N.D. □ 1.7 □	ng/kg-weight/day	10/15
	PFOA	1.9 □ 0.27 □ 13 □		250/250	0.62 □ N.D. □ 2.9 □	ng/kg-weight/day	12/15
	PFHxA	All N.D.		0/164	□	□	□
	PFHpA	N.D. □ N.D. □ 1.2 □		21/164	□	□	□
Organofluorine compounds	PFHxS	0.46 □ N.D. □ 1.8 □		162/164	□	□	□
	PFTeDA	N.D. □ N.D. □ 0.41 □	pg/mL	13/164	□	□	□
	PFNA	1.5 □ 0.35 □ 7.7 □	pg/mL	164/164	□	□	□
	PFDA	0.57 □ 0.092 □ 2.7 □		164/164	□	□	□
	PFUdA	1.3 □ 0.29 □ 6.4 □		164/164	□	□	□
	PFDS	N.D. □ N.D. □ 0.065 □		3/164	□	□	□
	PFDoA	0.17 □ N.D. □ 0.89 □		139/164	□	□	□
	PFTrDA	0.39 □ N.D. □ 2.7 □		161/164	□	□	□
Hydroxylated polychloro-biphenyls	5Cl-HO-PCBs	24 □ 1.2 □ 120 □		30/30	□	□	□
	6Cl-HO-PCBs	30 □ 1.6 □ 200 □	pg/g	30/30	□	□	□
	7Cl-HO-PCBs	23 □ 4.0 □ 130 □		30/30	□	□	□
Metals	T-Hg	8.7 □ 1.7 □ 41 □	ng/mL	253/253	0.062 □ N.D. □ 0.30 □		44/45
	Me-Hg	□	□	□	0.059 □ N.D. □ 0.29 □		43/45
	Cd	1.1 □ 0.25 □ 3.5 □		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 □	μg/kg-weight/day	30/30
	Cu	830 □ 590 □ 1,400 □	ng/mL	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 4. Urine concentrations (Creatinine corrected) □FY 2011-FY 2014□

classification	Chemical substance	median(range)	unit	rate of detection	
Pesticide metabolites	OP metabolites	DMP	3.1□N.D.□140□	μg /g Cr	102/105
		DEP	3.6□N.D.□520□		81/105
		DMTP	6.8□N.D.□110□		78/105
		DETP	N.D.□N.D.□8.3□		21/105
	Pyrethroid metabolites	PBA	0.24□N.D.□3.4□		74/105
		DCCA	N.D.□N.D.□13□		16/105
	Carbamate metabolite	Ethylenethiourea	N.D.□N.D.□0.50□		6/105
	Triclosan		1.2□0.15□380□		105/105
	Acephate		N.D.□N.D.□1.9□		8/60
	Methamidophos		N.D.□N.D.□0.058□		2/60
	Imidacloprid metabolite	6-Chloronicotinic acid	N.D.□N.D.□1.8□		8/60
	Fenitrothion metabolite	3-methyl-4-nitrophenol	N.D.□N.D.□3.6□		26/60
	p-nitrophenol		0.67□N.D.□4.6□		58/60
	Deet		All N.D.		0/60
Others	Mono(2-ethylhexyl) phtalete	MBP	17□3.7□5,200□	263/263	
		MEHP	2.8□0.23□22□	263/263	
		MEHHP	9.7□1.8□59□	263/263	
		MEOHP	6.0□1.0□35□	263/263	
		MBzP	0.56□N.D.□38□	248/263	
	Bisphenol A		0.33□N.D.□31□	231/263	
	PAHs	1-Hydroxypyrene	0.14□N.D.□4.7□	59/60	
		1&9- ydroxyphenanthrene	0.12□N.D.□0.69□	56/60	
		2- Hydroxyphenanthrene	0.080□N.D.□0.46□	48/60	
		3- Hydroxyphenanthrene	0.16□N.D.□0.65□	57/60	
		4- Hydroxyphenanthrene	N.D.□N.D.□0.20□	20/60	
	Parabens	Methylparaben	82□1.3□2,500□	60/60	
		Ethylparaben	2.7□N.D.□410□	48/60	
		Propylparaben	1.2□N.D.□77□	36/60	
		Butylparaben	N.D.□N.D.□87□	21/60	
		Benzylparaben	All N.D.	0/60	
	phytoestrogens	Genistein	1,300□74□5,700□	60/60	
		Daidzein	1,800□97□19,000□	60/60	
		Equol	180□N.D.□28,000□	59/60	
	Caffein		2,000□0.36□22,000□	60/60	
Cotinine		0.34□N.D.□1,600□	45/60		
Benzophenone3		N.D.□N.D.□190□	10/60		
Metals	As	As (□)	N.D.□N.D.□2.9□	75/263	
		As (□)	1.4□N.D.□6.9□	230/263	
		methylarsonic acid	1.9□N.D.□13□	249/263	
		dimethylarsinic acid	31□6.2□170□	263/263	
		arsenobetaine	39□2.1□2,300□	263/263	
	Cd		0.81□0.11□3.9□	263/263	