HUMAN EXPOSURE TO PCDDs, PCDFs, AND DIOXIN LIKE PCBs IN JAPAN, FROM 1999 TO 2005

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Abstract
The levels of human exposure to dioxins (PCDDs, PCDFs, and Co-PCBs) in Japan were determined as part of efforts to tackle the dioxins issues. To determine exposure levels, the survey results derived from the regular environmental monitoring of dioxins under Japan's Law Concerning Special Measures against Dioxins (Dioxins Law) were collected and compiled, as well as other dioxin surveys by national governmental bodies in fiscal year 1999-2005. Particular attention was given to the transitions of the exposure levels in Japan from FY 1999 to 2005. In a "point" estimate (i.e., a single value derived from arithmetic means) approach based on the compiled data, the exposure levels were on a declining trend however this trend is not statistically significant through FY 1999 to 2005. Even using a Monte Carlo simulation method for "probabilistic" approach, the exposure levels (the estimated average of the exposure distribution) were also on a declining trend however this trend is not statistically significant through FY 1999 to 2005. In FY 2000-2005, the 95th percentile of the human exposure to dioxin distributions in Japan estimated were below the tolerable daily intake level (TDI) defined by the Ministry of Health, Labour and Welfare (WHLW), Japan (i.e., 4pg-TEQ/kg-bw/day). In conclusion, improvements of the levels of human exposure to dioxins are considered to be the effect of regulation concerning countermeasures against dioxin.

Introduction

Based on Dioxins Law, local public authorities have been monitoring dioxins (PCDDs, PCDFs, and Dioxin like PCBs) levels in air, water (including the bottom sediment) and soil to grasp the actual conditions of dioxins pollution throughout Japan. [1)] The precision and accuracy of these monitoring data have been managed and inspected according to the technical guideline for the analyses of dioxins in environmental samples. [2)] The Dioxins Law has significantly effected to the reduction of the average dioxins concentrations in the ambient air. The reduction of national PCDDs/DFs emission in 2004 against the 1997 level to approximately 96% has been reported.[3)] Therefore, the transitions of Japanese dioxins exposure levels in recent years are worthy of attention and Ministry of the Environment has been conducting researches and surveys on human exposure assessment for grasping the amount of dioxins taken by human via various routes.[4) 5)] To determine exposure levels, collection and compilation of surveillance results derived from the regular environmental monitoring under the law as well as other dioxins surveys by national governmental bodies, and the data were analyzed.[6) 7)] The exposure levels were estimated by a "point" estimate (i.e., a single value derived from arithmetic means) approach based on the collected data. Because dioxins exposure is not clearly below the level of concern, an emphasis is placed on the importance of quantitatively characterizing the variability in exposure assessments. Therefore, the "probabilistic" approach using a Monte Carlo simulation was also conducted. In this study, the exposure levels were estimated in both ways during from FY 1999 to 2005.

Materials and Methods

Data collection and compilation: Dioxins concentrations in the air, soil and total diet study (TDS) results were obtained from literature cited. [6) 7)] The WHO-TEF1998 was used in this study. Each congeners under the quantitation limits in soil and TDS were treated as 0pg, while those in air calculated as a half of their quantitation limits.

Point estimation: The air and soil concentration data from the vicinity of the pollution sources were not used, because a few abnormally high data points could cause an improperly high arithmetic means and an overestimation of average exposure level. Point estimation was conducted taking into account three intake pathways: inhalation, soil ingestion, and diet. Estimations of exposure through inhalation were based on arithmetic means of dioxins concentrations in the air, using a body weight of 50kg and a daily respiration volume of $15m^3$/day. Estimates of exposure through soil ingestion were obtained using arithmetic means of the dioxins concentration in the soil, and an assumed daily ingestion rate of soil of 100mg/day. [8] Arithmetic means of TDS results from each year (FY 1999-2005(n=12~36)) were used to estimate exposure through diet. Total exposure was obtained as follows: [Total exposure (pg-TEQ/kg/day) = {Air conc. (pg-TEQ/m^3) x Respiration vol. ($15m^3$/day) / Body weight (50kg)} + {Soil conc. (pg-TEQ/g) x Soil ingestion (100mg/day) / Body weight (50kg)} + {Intake through diet (TDS) (pg-TEQ/kg/day)}].

Monte Carlo simulation: Total personal exposure levels to dioxins were estimated by Monte Carlo simulation method, since the exposure to dioxins from various environmental media or diet intake has statistical distribution (shown in Table 1). TDS data from the past 4 years were combined to obtain adequate data size to elaborate curve fitting. When considering the standard deviation of TDS data, the dioxins concentrations in foods were decreasing so slowly such that no significant error is introduced by combining the data sets. The air and soil concentrations data from the vicinity of the pollution sources were excluded to obtain the probabilistic density functions, because a few abnormal data could make them shift to unreasonably high. Grubbs' test was conducted to remove abnormal data in TDS from statistical point of view (FY 2001). The range used in the Monte Carlo simulation was from zero to the maximum measurement result, including the vicinity of pollution sources (shown in Table 1). Crystal Ball®2000 (Decisioneering Inc.) was used, with 5,000 trials.

Table 1: Parameters of all the variables (air, soil, and total diet study) used in the Monte Carlo Simulation method through FY 1999 to 2005.

	Air (pg-TEQ/m^3)			Soil (pg-TEQ/g)			Total diet study (pg-TEQ/kg/day)[3]		
	appropriate probabilistic function[1]	range of probabilistic distribution		appropriate probabilistic function[1]	range of probabilistic distribution		appropriate probabilistic function[1]	range of probabilistic distribution	
		Min.	Max.[2]		Min.	Max.[2]		Min.	Max.[2]
1999	Weibull distribution	0	1.5	log-normal distribution	0	180	logistic distribution	0	7.01
2000	Weibull distribution	0	1.0	log-normal distribution	0	1200	logistic distribution	0	2.01
2001	log-normal distribution	0	1.7	log-normal distribution	0	4600	log-normal distribution	0	3.40
2002	log-normal distribution	0	0.84	log-normal distribution	0	250	log-normal distribution	0	3.40
2003	log-normal distribution	0	0.72	log-normal distribution	0	1400	log-normal distribution	0	3.05
2004	log-normal distribution	0	0.55	log-normal distribution	0	250	log-normal distribution	0	2.93
2005	log-normal distribution	0	0.61	log-normal distribution	0	2800	log-normal distribution	0	3.56

1) Most appropriate probabilistic funtction was applied in comparison to all types of probabilistic functions (i.e. normal, Weibull, logistic, extreme value, gamma, beta, exponential, uniform, and Pareto distribution).

2) The maximum value in the Monte Carlo Simulation method was the maximum measurement result in each variables from FY 1999 to 2005, including the vicinity of pollution sources.

3)Total diet study data from past 4 fiscal yours were combined to obtain adequate data size to elaborate curve fitting (Except in FY 1999 and 2000, total diet study data from single fiscal year were used to elaborate curve fitting.).

Results and Discussion

Table 2 shows complied data of dioxins concentrations in air, soil and TDS results. In FY 2005, these dioxins levels were less than half compared to those in FY 1999. However in TDS results, the dioxins levels from FY 2000 to 2005 were approximately the same levels. The statistics based on the entire data including those from the vicinity of pollution sources were shown in the parenthesizes. The arithmetic means were calculated excluding the vicinity of pollution sources.

Table 2: PCDDs/DFs and DL-PCBs concentrations in air, soil and total diet study results (FY 1999 – 2005) in Japan.

	1999	2000	2001	2002	2003	2004	2005
Air (pg-TEQ/m^3) 1)							
n	453 (690)	733 (961)	791 (1028)	761 (989)	756 (986)	759 (964)	695 (898)
Min.	0.0065 (0.0065)	0.0073 (0.0073)	0.0072 (0.0072)	0.0066 (0.0066)	0.0056 (0.0056)	0.0083 (0.0082)	0.0045 (0.0039)
Max.	0.82 (1.5)	0.76 (1.0)	1.7 (1.7)	0.84 (0.84)	0.50 (0.72)	0.38 (0.55)	0.61 (0.61)
Arithmetic mean	0.20 (0.21)	0.14 (0.15)	0.14 (0.13)	0.0093 (0.093)	0.064 (0.065)	0.059 (0.058)	0.051 (0.051)
Geometic mean 2)	0.14 (0.15)	0.10 (0.10)	0.095 (0.092)	0.069 (0.067)	0.051 (0.050)	0.047 (0.046)	0.043 (0.042)
Soil (pg-TEQ/g) 1)							
n	763 (811)	1943 (3051)	2313 (3735)	2282 (3300)	2128 (3059)	1983 (2618)	1314 (1782)
Min.	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
Max.	180 (180)	280 (1200)	240 (4600)	250 (250)	360 (1400)	250 (250)	73 (2800)
Arithmetic mean	5.6 (6.1)	4.6 (6.9)	3.2 (6.2)	3.4 (3.8)	2.6 (4.4)	2.2 (3.1)	2 (5.9)
Geometic mean 2)	0.41 (0.58)	0.38 (0.67)	0.29 (0.48)	0.39 (0.53)	0.23 (0.36)	0.23 (0.32)	0.23 (0.34)
Total diet study (pg-TEQ/kg/day)							
n	16	16	12	36	33	27	27
Min.	1.19	0.84	0.67	0.57	0.58	0.48	0.47
Max.	7.01	2.01	3.40	3.40	3.05	2.93	3.56
Arithmetic mean	2.25	1.45	1.63	1.49	1.33	1.41	1.20
Geometic mean 2)	1.98	1.40	1.50	1.36	1.22	1.25	1.07

1) For air and soil data, values shown without parentheses were caluclated by excluding the data from the vicinity of pollution sources. Values shown in parenthesises were caluculated by including all collected data (both near and away from pollution sources). For all other media, values were caluculated from all collected data.

2) "Geometric mean" were caluculated excluding 0pg-TEQ/g. 1/763, 15/1943, 6/2313, 5/2282, 4/2128, 8/193, and 71/1314 of soil data were 0pg-TEQ/g in each FY 199-2005 (Including the vicinity of pollution sources, 1/811, 15/3051, 9/3735, 5/2300, 5/3059, 10/2618, 71/1782 were 0pg-TEQ/g in each FY 1999-2005).

Table 3: Estimates of exposure to PCDDs/DFs and DL-PCBs in Japan, from FY 1999 to 2005 (pg-TEQ/kg-bw/day).

	1999	2000	2001	2002	2003	2004	2005
Inhalation	0.06	0.042	0.042	0.028	0.019	0.018	0.015
Soil ingestion	0.011	0.0092	0.0064	0.0068	0.0052	0.0044	0.0041
Diet	2.25	1.45	1.63	1.49	1.33	1.41	1.2
Total	2.32	1.50	1.68	1.52	1.35	1.43	1.22

Point exposure estimates through each pathway and in total from FY 1999 to 2005 are shown in Table 3. Total exposures to dioxins were estimated as 2.32, 1.50, 11.68, 11.52, 1.35, 1.43 and 1.22pg-TEQ/kg-bw/day in each FY 1999-2005. Although it wasn't significant, the trend of the levels of human exposure to dioxins in a "point" estimate approach was declining with age. Exposure through diet accounted for more than 90% of the total exposure throughout seven years from FY 1999 to 2005; the contributions through inhalation and soil ingestion were on declining trend. Table 1 shows the parameters used in the Monte Carlo simulation in each FY 1999-2005. Log-normal distribution was the most appropriate to almost all variables except air in FY 1999 (Weibull distribution [L, s, m=0.01, 0.21, 1.3pg-TEQ/m^3]) and 2000 (Weibull distribution [L, s, m=0.01, 0.15, 1.3pg-TEQ/m^3]), and total diet study in FY 1999 (logistic distribution [m, s=1.98, 0.74pg-TEQ/kg-bw/day]) and 2000 (logistic distribution [m, s=1.46, 0.22pg-TEQ/kg-bw/day]). The range of each probabilistic distribution was from zero to the maximum measurement result, including the data from the vicinity of pollution sources (air: 0-0.61~1.7pg-TEQ/m^3; soil: 0-180~4600pg-TEQ/g; total diet study: 0-2.20~7.01pg-TEQ/kg-bw/day). Figure 1 shows the results of an exposure distribution using the Monte Carlo simulation method. The estimated average of the exposure distribution were 2.22, 1.45, 1.78, 1.62, 1.45, 1.43 and 1.36pg-TEQ/kg-bw/day in each FY 1999-2005 and the estimated median of the exposure distribution were 2.13, 1.47, 1.69, 1.51, 1.36, 1.32 and 1.25pg-TEQ/kg-bw/day in each FY 1999-2005. Although in FY 1999, the estimated average of the exposure and the estimated median of the exposure was approximately the same level, the differences gradually occurred over time, since the reduction in the estimated average of the exposure was more slightly than the reduction in the estimated median of the exposure. The elaborated exposure distribution was almost symmetrical in FY 1999 and 2000, but it has been changed to have longer tail in higher side of the exposure distribution in recent year. The 95th percentile of the dioxins exposure distributions were 4.25, 1.96, 2.91, 3.12, 2.51, 2.59, 2.57pg-TEQ/kg-bw/day in each FY1999-2005. The average and the 95th percentile of the human exposure to dioxins distributions in Japan estimated were below the tolerable daily intake level (TDI) by WHLW, Japan (i.e., 4pg-TEQ/kg-bw/day) in FY 2000 or later. In conclusion, improvements of the levels of human exposure to dioxins were considered to be the effect of regulation concerning countermeasures against dioxin. However the exposure through diet accounted for more than 90% of the total exposure and the dioxins tend to accumulate in adipose tissue. Therefore it is important to continue addressing the transitions of the exposure levels in the future.

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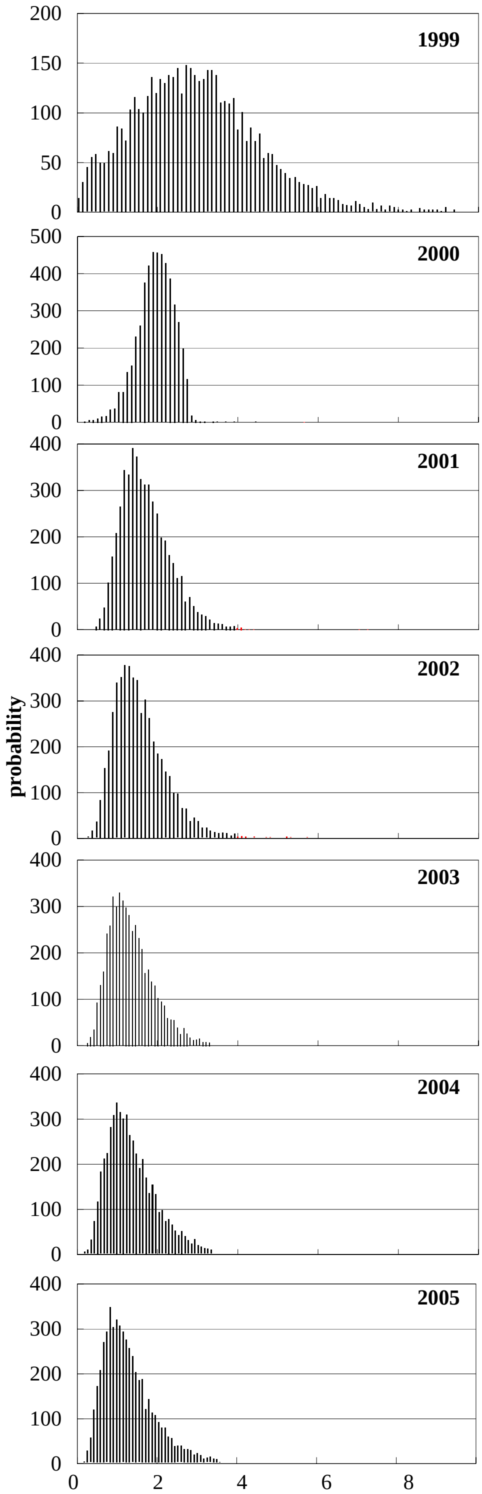

Figure 1: Distribution of PCDDs/PCDFs and DL-PCBs exposure in Japan from FY 1999 to 2005, estimated by Monte Calro simulation.

References

1. Ministry of the Environment, Japan (1999) *Law Concerning Special Measures against Dioxins*
2. Ministry of the Environment, Japan (1999) *Guidelines on Quality Assurance/Quality Control for The Environmental Measurements of Dioxins*
3. Ministry of the Environment, Japan (2005) *Information Brochure Dioxins 2005*
4. Suzuki N, Ishikawa N, Takei T, Mato Y, Nakayama S, Uchiyama I, Katatani N, Kadokami K, Nakano T.,Miyata H.,and Morita M. *Organohalogen Comp* 2003;64:67.
5. Mato Y, Suzuki N, Kadokami K, Katatani N, Nakano T, Matsuoka T, Takei T, Nakayama S, Uchiyama I, Miyata H, Toyoda M, Morita M. *Organohalogen Comp* 2004;66:2428.
6. Ministry of the Environment, Japan (2000, 2001, 2002, 2003, 2004, 2005, and 2006) *Environmental Survey of Dioxins FY1999, 2000, 2001, 2002, 2003, 2004, and 2005 Results*
7. Toyoda M, Iida T, Yoneya T, Tsutsumi T, Amakura Y, and Sasaki K. (1999, 2000, 2001, 2003, 2005, 2006 and 2006) *Reports on the results of daily intake study of dioxins from foods in Japan (FY1998,1999,2000, 2001, 2002, 2003, 2004 and 2005)* (in Japanese)
8. Ministry of the Environment, Japan (2000) *Detailed Study of Dioxin Exposure: Findings of the Fiscal 1999 Survey*