HUMAN EXPOSURE TO PCDDs, PCDFs, AND DIOXIN LIKE PCBs IN JAPAN,2001

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Introduction

In our previous study[1], we have estimated the level of human exposure to dioxins (PCDDs, PCDFs, and Dioxin like PCBs) in Japan based on dioxins monitoring data and results of total diet studies (TDS) in fiscal 2000 (April 2000- March 2001). It has been reported that the national PCDDs/DFs emission in 2001 against the 1997 level has been reduced by approximately 77%[2]. In addition, reduction of environmental levels was reported[2]. The enforcement of Japan's Law Concerning Special Measures against Dioxins has significant impact on the reduction of the average dioxins concentrations in the ambient air. Therefore, the transitions of Japanese dioxins exposure levels in recent years are worthy of attention. In order to determine exposure level in fiscal 2001, collection and compilation for surveillance results derived from the regular environmental monitoring under the law as well as other dioxins surveys by national and local governmental bodies were continued, and the data were analyzed. The exposure level in fiscal 2001 was 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. However, elaboration in curve fitting to the distribution of dioxins intake through diet wasn't completely achieved due to the limitation of TDS data size in fiscal 2000 (n=16) in our previous study[1]. In the present study, the curve fitting to diet were updated and elaborated, based on larger size of TDS data (n=54) by combining all the data in fiscal 1998-2001.

Methods

Data collection and compilation: Dioxins concentrations in the air, soil, water and sediment (from sea/river/lake), groundwater, foodstuff and TDS results were obtained from literature cited[3-6]. Data for aquatic organisms, purified and raw water from water purification plants, and human blood were supplied by Japanese local governmental bodies. Human breast milk data was from literature cited[7] and local governmental bodies. The WHO98-TEF was used in this study. Congeners under the quantitation limits in soil, human blood, TDS and foodstuff were treated as 0pg-TEQ, while those in the other media calculated as a half of the TEQ for 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. Estimates of exposure through inhalation were based on arithmetic means of dioxins concentrations in the air, using a body weight[1] of 50kg and a daily respiration volume[1] 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[1] of 100mg/day. Arithmetic mean of TDS results from fiscal 2001 (n=12) was used to estimate exposure through diet. Total exposure was obtained as follows: [Total exposure = {Air conc. x Respiration vol. (15m^3/day) / Body weight (50kg)} + {Soil conc. x Soil ingestion (100mg/day) / Body weight (50kg)} + {Intake through diet (TDS)}].

Estimation of age-group-specific contribution of various foodstuff to total dietary exposure: Dioxins concentration data of various foodstuff (fiscal 1998-2001) were classified into food groups according to the survey by the Ministry of Health, Labour and Welfare (MHLW) (2003)[8]. The age-group-specific contribution of each food group to total dietary exposure was then estimated. Body weight and dietary intake data of each age group were obtained from MHLW (2003)[8].

Monte Carlo simulation: The method for the Monte Carlo simulation was the same as for the point estimation, except that the dioxins concentrations in the air and soil, and exposure through diet were represented as probabilistic density functions. TDS data from fiscal 1998-2001 were combined to obtain adequate data size to elaborate curve fitting. When considering the standard deviation of TDS data, the dioxins concentrations in foods are 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. The range used in the Monte Carlo simulation was from zero to the maximum measurement result, including the vicinity of pollution sources (air: 0-1.7pg-TEQ/m^3; soil: 0-4600pg-TEQ/g; TDS: 0-7.01pg-TEQ/kg/day). Crystal Ball®2000 (Decisioneering Inc.) was used, with 5,000 trials.

Results and Discussion

Table 1 shows compiled data on dioxins concentrations in various media as well as TDS results. The dioxins levels were slightly less than or approximately same as those in fiscal 2000[1]. In air, soil and human blood, the statistics were derived from data excluding the vicinity of pollution sources. The statistics based on the entire data including those from the vicinity of pollution sources are shown in the parenthesises. The arithmetic mean of air concentration of entire data was approximately the same as the arithmetic mean calculated excluding the vicinity of pollution sources. Point exposure estimates through each pathway and in total are shown in Table2. Estimates based on the entire data set including those from the vicinity of pollution sources are shown in the parenthesises. Total exposure was estimated at 1.68pg-TEQ/kg-bw/day, with exposure through inhalation, soil ingestion, and diet at 0.042, 0.0064, and 1.63pg-TEQ/kg-bw/day, respectively. Exposure through diet accounted for more than 90% of the total exposure; the contributions through inhalation and soil ingestion were relatively small. Although, the total exposure were slightly higher than the estimate in fiscal 2000 (1.50pg-TEQ/kg-bw/day), it was not significant in considering the standard deviations of TDS data. Table3 shows dioxins concentrations in various foodstuff classified into food groups (in fiscal1998-2001). Concentrations in ‘fish and shellfish’ were high (average =0.94, range = 0-26 pg-TEQ/g). Furthermore, ‘fats and oils of animal origin’ and ‘meat and eggs’ were also high. According to the arithmetic means in Table3, age-group-specific contributions of various foodstuff to total dietary exposure were estimated and are shown in Figure1; the estimates of exposure through fish and shellfish accounted for approximately 45-70% of total dietary exposure in each age group. Contributions of fish and shellfish were higher in older age groups, while those of milk and dairy products were higher in younger age groups. Regarding Monte Carlo simulation, the maximum value of TDS (7.01 pg-TEQ/kg-bw/day) was regarded as abnormal value by Grubbs’ test (1%), therefore it was excluded from the data set for curve fitting. The histograms, curve fitting and P-P plots of the air, soil and

Table 1: PCDDs/DFs and DL-PCBs concentrations in various media (fiscal 2001) and total diet study results (fiscal 1998 – 2001) in Japan.

	fiscal year	n	Min.	25th%tile	Median	75th%tile	Max.	Arithmetic mean	Geometric[2)] mean	S.D.
Air[1)]		791	0.0072	0.054	0.093	0.18	1.7	0.14	0.095	0.13
pg-TEQ/m^3	*2001*	(1028)	(0.0072)	(0.051)	(0.090)	(0.18)	(1.7)	(0.13)	(0.092)	(0.14)
Soil[1)]		2313	0	0.055	0.35	1.8	240	3.2	0.29	11
pg-TEQ/g	*2001*	(3735)	(0)	(0.085)	(0.66)	(3.0)	(4600)	(6.2)	(0.48)	(79)
Water (sea/river/lake) pg-TEQ/L	*2001*	2236	0.0028	0.073	0.12	0.27	27	0.25	0.14	0.64
Sediment (sea/river/lake) pg-TEQ/g	*2001*	1835	0.012	0.32	1.1	5.6	540	8.5	1.5	32
Ground water pg-TEQ/L	*2001*	1473	0.00020	0.049	0.065	0.075	0.92	0.074	0.061	0.064
Aquatic organisms pg-TEQ/g	*2001*	18	0.49	0.87	1.5	1.5	3.0	1.5	1.3	0.78
Purified water (water purification plant) pg-TEQ/L	*2001*	46	0.0013	0.0039	0.0083	0.017	0.065	0.017	0.0092	0.021
Raw water (water purification plant) pg-TEQ/L	*2001*	34	0.011	0.072	0.12	0.22	0.54	0.15	0.11	0.12
Human breast milk pg-TEQ/g-fat	*2001*	170	-	-	-	-	-	20	-	-
Human blood[1)]		11	3.5	-	-	-	9.8	7.6	-	-
pg-TEQ/g-fat	*2001*	(59)	(1.0)	-	-	-	(20)	(8.2)	-	-
Total diet study pg-TEQ/kg/day	*2001*	12	0.67	1.27	1.57	1.99	3.40	1.63	1.50	0.71
Total diet study pg-TEQ/kg/day	*1998-2001*	54	0.67	1.40	1.63	2.00	7.01	1.83	1.67	0.96

1) For air, soil and human blood data, values shown without parenthesises 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. 6/2313 of soil data were 0pg-TEQ/g (Including the vicinity of pollution sources, 9/3735 were 0pg-TEQ/g).

Table 2: Estimates of exposure to PCDDs/DFs and DL-PCBs in Japan, fiscal 2000 and 2001.
(pg-TEQ/kg-bw/day)

	fiscal 2000[1)]	fiscal 2001	
Inhalation	0.042	0.042	(0.039)[2)]
Soil ingestion	0.0092	0.0064	(0.012)[2)]
Diet	1.45	1.63	
Total	1.50	1.68	(1.68)[2)]

1)Quoted from Suzuki *et.al.* (2003)[1].
2)Estimates in parenthesises were based on dioxins concentrations, in the air and soil, including data from the vicinity of pollution sources.

TDS were shown in Figure 2, 3 and 4, respectively. Lognormal distribution was the most appropriate to all the variables (air: [geometric mean (GM), geometric standard deviation (GSD) = 0.095, 2.4pg-TEQ/m^3], soil: [GM, GSD = 0.29, 13pg-TEQ/g], foods: [GM, GSD = 1.63, 1.42pg-TEQ/kg-bw/day]) in comparison with other types of probabilistic functions (i.e. normal, Weibull, logistic, extreme value, gamma, beta, exponential, uniform, and Pareto distribution). Figure 5 shows the results of an exposure distribution using the Monte Carlo simulation method. The estimated average, median, 5th percentile and 95th percentile of the exposure distribution were 1.78, 1.69, 0.95 and 2.91pg-TEQ/kg-bw/day, respectively. The elaborated exposure distribution had longer tail in upper side, and the average and the medium have shifted slightly higher than the previous exposure distribution[1]. In conclusion, this study found that both the average and the 95th percentile of the dioxins exposure distributions in Japan were estimated to be below the World Health Organisation's tolerable daily intake levels (i.e., 4pg-TEQ/kg-bw/day). This study indicated that the variability of dioxins intake from diet in Japan fiscal 1998-2001 was in lognormal distribution. There was no significant difference, comparing exposure level in fiscal 2001 with fiscal 2000. The transitions of exposure levels should be addressed in the future.

Table 3: PCDDs/DFs and DL-PCBs concentrations in foodstuff (fiscal 1998-2001). (pg-TEQ/g)

Foodstuff	n	Min.	25%tile	Median	75%tile	Max.	Arithmetic mean	Geometric[1)] mean	S.D.
Rice and rice products	166	0	0.0000080	0.00023	0.0011	0.13	0.0039	0.00030	0.014
Cereals, seeds, and potatoes	67	0	0	0.000026	0.00040	0.047	0.0027	0.00025	0.0092
Sugars and confectioneries	6	0	0	0.00050	0.0025	0.019	0.0038	0.0038	0.0075
Fats and oils of animal origin	8	0.090	0.28	0.42	0.70	0.98	0.49	0.39	0.31
Fats and oils of vegetable origin	3	0.0030	0.0030	0.0030	0.0045	0.0060	0.0040	0.0038	0.0017
Pulses	35	0	0	0.000034	0.0010	0.060	0.0026	0.00041	0.010
Fruits	112	0	0	0.0000050	0.0012	0.35	0.0078	0.00076	0.041
Green vegetables	153	0	0	0.00050	0.027	0.36	0.030	0.0052	0.062
Other green vegetables, mushrooms, and seaweed	205	0	0	0.0000070	0.00098	2.7	0.028	0.00059	0.22
Beverage, sauce, and seasoning	17	0	0	0	0	0.0010	0.00012	0.0010	0.00033
Fish and shellfish	644	0	0.087	0.34	1.0	26	0.94	0.27	1.9
Meat and eggs	243	0	0.0050	0.042	0.12	1.8	0.12	0.047	0.25
Milk and dairy products	81	0	0.025	0.050	0.10	0.62	0.092	0.047	0.12

1) “Geometric mean“ were caluculated excluding 0pg-TEQ/g. The ratio of 0pg-TEQ/g in each category of foodstuff was as follows: ’Rice and rice products’ 25/166 ; ’Cereals, seads, and potatoes’ 25/67; ’Sugars and confectioneries’ 3/6; ’Pulses’ 13/35; ’Fruits’ 54/112; ’Green vegetables’ 51/153; ’Other green vegetables, mushurooms, and seaweed’ 94/205; ’Beverage, sauce, and seasoning’ 15/17; ’Fish and sellfish’ 7/644; ’Meat and eggs’ 12/243; and ’Milk and Milk products’ 1/81, respectively.

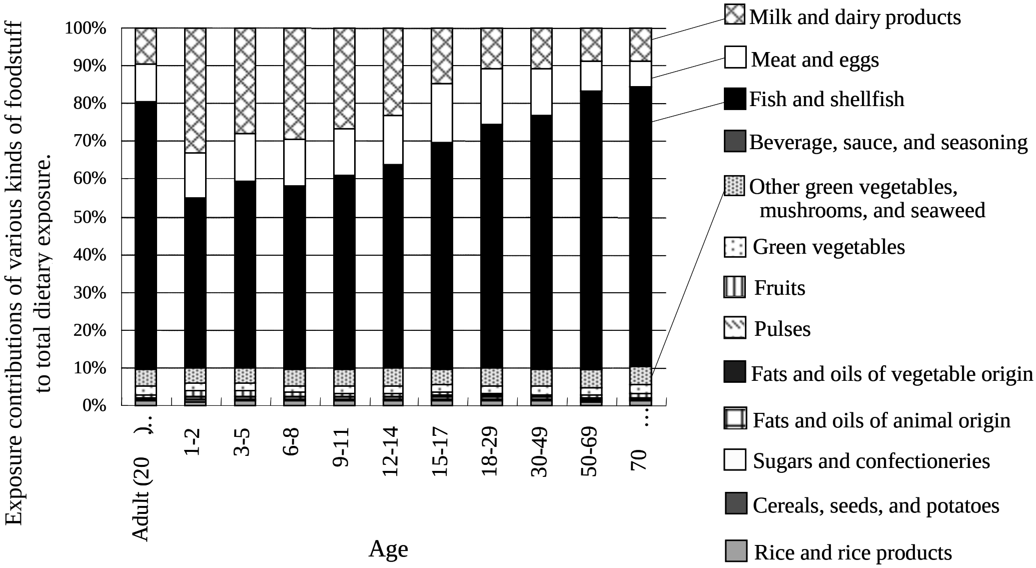

Figure 1: Age-group-specific contribution of various foodstuffs to total dietary exposure.

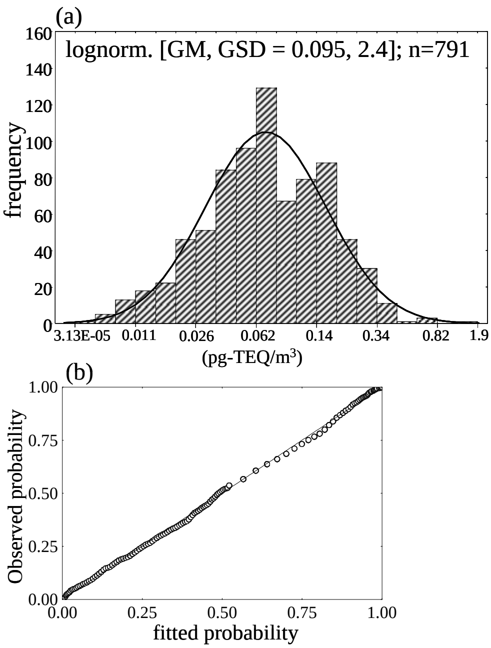

Figure 2: (a)Histogram and curve fitting of dioxin concentration in air (fiscal 2001). X-axis is in natural logarithmic scale. Data from the vicinity of pollution sources were excluded. (b) the P-P plots of air.

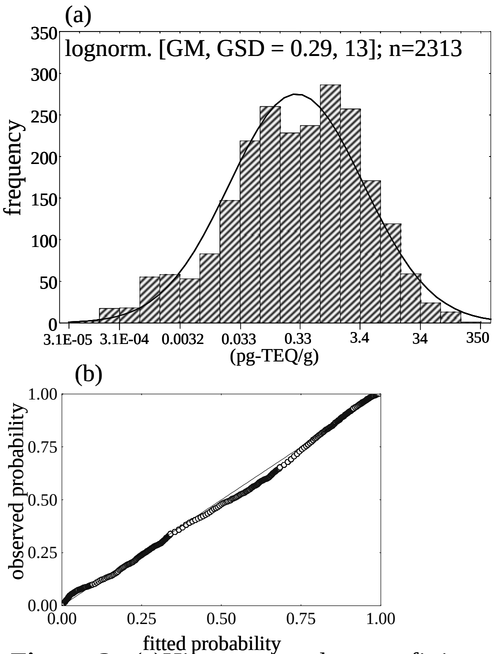

Figure 3: (a)Histogram and curve fitting of dioxins concentration in soil (fiscal 2001). X-axis is in natural logarithmic scale. Data from the vicinity of pollution sources were excluded. (b) the P-P plots of soil.

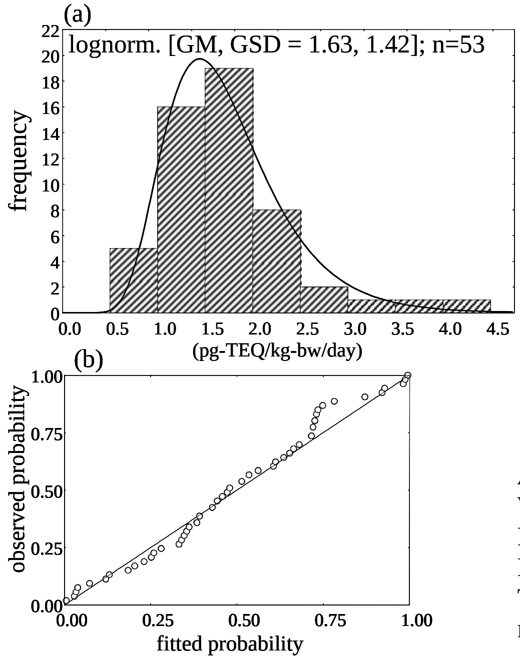

Figure 4: (a)Histogram and curve fitting of dioxins intake from diet according to total diet studies (fiscal 1998-2001). The maximum value, 7.01pg-TEQ/kg-bw/day, is excluded from this figure, because it was regarded as abnomal value by Grubbs's test. (b) the P-P plots of diet.

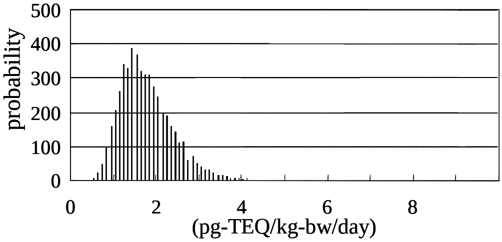

Figure 5: Distribution of PCDDs/DFs and DL-PCBs exposure in Japan, estimated by Monte Carlo simulation.

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