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ScienceDirect
Chemosphere 67 (2007) S247–S255
CHEMOSPHERE
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Human intake of PCDDs, PCDFs, and dioxin like PCBs in Japan, 2001 and 2002

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Accepted 26 May 2006
Available online 9 February 2007

Abstract

PCDDs, PCDFs, and dioxin like PCBs (dioxin) surveillance results derived from regular environmental monitoring as well as other dioxin surveys by national and local governmental bodies in Japan were collected and analyzed. Several thousand data for air and soil in fiscal year 2001 (from 01/04/2001 to 31/03/2002) and 2002, water (from the sea, rivers and lakes), sediment (from the sea, rivers and lakes), ground water, aquatic organisms, purified water from water purification plants, raw water from water purification plants, human breast milk, and human blood in fiscal 2001, and total diet study (TDS) and various kinds of foodstuff in fiscal 1998–2002 were collected. Average human uptake of dioxin in Japan in fiscal 2001 was estimated at 1.68 pg-TEQ/kg-bw/day, while uptake in fiscal 2002 was estimated at 1.52 pg-TEQ/kg-bw/day. Diet accounted for more than 90% of the total intake. Contributions of inhalation and soil ingestion were relatively small. Age-group-specific contribution of various foodstuff to total dietary intake was also estimated. The estimates of intake through fish and shellfish accounted for approximately 45–70% of total dietary intake in each age group. Monte Carlo simulation was conducted, using the data of the air and soil concentrations in fiscal 2001 and the total diet study data in fiscal 1998–2001, in order to obtain information on the variability of dioxin intake; The estimated average, median, 5th percentile and 95th percentile of the intake distribution were 1.78, 1.69, 0.95 and 2.91 pg-TEQ/kg-bw/day, respectively. This study found that the average total intake estimates in Japan in both fiscal 2001 and 2002 were estimated to be below tolerable daily intake level (TDI) defined by the Ministry of Health, Labour and Welfare, Japan (*i.e.* 4 pg-TEQ/kg-bw/day). The 95th percentile of the dioxin intake distributions estimated with Monte Carlo simulation using the data of the air and soil concentrations in fiscal 2001 and TDS data in fiscal 1998–2001 was also below the Japanese TDI.

Keywords: Dioxin; Human intake; Japan; Monte Carlo simulation; Distribution

1. Introduction

Although the Japanese national PCDDs and PCDFs emissions in 2003 were reported to be 95% less than the 1997 levels (Ministry of the Environment (MOE), 2004), and the environmental levels of PCDDs, PCDFs and dioxin like PCBs (dioxin) were reported to be decreasing (Council of Ministries and Agencies on Dioxin Policy, Japan, 2003), our previous study, however, found that the average human intake of dioxin in 2000 was still at 37.5% (1.50 pg-TEQ/kg/day) of tolerable daily intake level

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0045-6535/$ - see front matter © 2006 Published by Elsevier Ltd.
doi:10.1016/j.chemosphere.2006.05.105

(TDI) defined by the Ministry of Health, Labour and Welfare, Japan(MHLW) (*i.e.* 4 pg-TEQ/kg-bw/day) (Suzuki et al., 2003). Therefore, it is necessary to maintain the observation of Japanese dioxin intake levels. Ministry of the Environment, Japan (MOE) estimates human intake level to dioxin every fiscal year. This manuscript presents the results of a study on human dioxin intake carried out by MOE in fiscal 2002, as well as part of its study in fiscal 2003. These studies were based on dioxin concentration data measured in fiscal 2001 and 2002 respectively. In order to conduct a comparison, the data were analyzed in the same way as fiscal 2000 where the average intake levels were estimated by a "point" estimate approach (*i.e.*, a single value derived from arithmetic means).

Diet is usually the predominant pathway of dioxin intake. Therefore, detail analysis on intake through diet is required. Food consumption behaviors differ depending on age (MHLW, 2003b). In Japan, older generations generally consume more fish than younger generations because they are more likely to prefer a traditional, seafood-rich Japanese diet. Also, children usually have different food consumption patterns from adults. In this study, estimation of age-group-specific contributions of various foodstuff to total dietary intake was also calculated by point estimation approach.

Because dioxin intake is not clearly below the level at which it might be no matter of concern, it is highly important to characterize the variability quantitatively in intake assessments. Therefore, the "probabilistic" approach using a Monte Carlo simulation was also conducted. Monte Carlo simulation is a computer-based method of analysis that uses statistical sampling techniques in obtaining a probabilistic approximation to the solution of a mathematical equation (USEPA, 1997). In our previous study, elaboration in curve fitting to the observed distribution of dioxin intake through diet was not completely achieved due to the limitation of TDS data size in fiscal 2000 ($n = 16$) (Suzuki et al., 2003). In the present study, the curve fitting to diet was updated and elaborated, based on larger size of TDS data ($n = 54$) by combining all the data in fiscal 1998–2001 under the assumption that the intake via diet is constant over this period.

The scope of the present study is to estimate the average and the distribution of human dioxin intake under normal conditions in Japan.

2. Methods

2.1. Data collection

PCDDs, PCDFs and dioxin like PCBs (dioxin) concentration data in various media measured in fiscal 2001–2002 (April 1, 2001–March 31, 2003) and total diet study (TDS) data in fiscal 1998–2002 (April, 1998–March 31, 2003) were collected.

For fiscal 2001, dioxin concentrations in the air (1028 sites) and soil (3735 sites) were obtained from MOE (2002) (Table 1(a)). For fiscal 2002, data of dioxin concentrations in the air (989 sites) and soil (3300 sites) were taken from MOE (2003) (Table 1(b)). The air concentration data from 979 out of 1028 sites in fiscal 2001 and those from 966 out of 989 sites in fiscal 2002 were seasonal average values, as two or usually four measurements were taken per year. The concentrations in the air were measured at sites in general environment (764 sites in fiscal 2001; 732 sites in fiscal 2002), roadside (27 sites in fiscal 2001; 29 sites in fiscal 2002), and vicinity of the pollution sources (237 sites in fiscal 2001; 228 sites in fiscal 2002). The concentrations in the soil were measured at sites in general environment (2313 sites in fiscal 2001; 2282 sites in fiscal 2002) and vicinity of the pollution sources (1422 sites in fiscal 2001; 1018 sites in fiscal 2002). Basically, only data from the general environment and road side were compiled for further analysis to estimate the average and the distribution of human dioxin intake under normal conditions in Japan. The statistics derived from the entire data including those from the vicinity of the pollution sources were shown in parentheses (Tables 1(a) and 1(b)).

The water from rivers, lakes, marshes and the sea (2236 sites), sediment from rivers, lakes, marshes and the sea (1835 sites), and groundwater (1473 sites) in fiscal 2001 were obtained from MOE (2002) (Table 1(a)).

Data for aquatic organisms ($n = 18$) (Aichi prefecture, 2002; Fukushima prefecture, 2002; Gifu prefecture, 2002; Kitakyushu city, 2002a; Shiga prefecture, 2002), and purified and raw water from water purification plants ($n = 46$ and 34, respectively) (Fukuoka prefecture, 2002; Hiroshima city, 2002; Kagawa prefecture, 2002; Kitakyushu city, 2002b; Nara prefecture, 2002; Osaka city, 2002; Saitama prefecture, 2002a; Sapporo city, 2002) in fiscal 2001 were supplied by Japanese local governmental bodies (Table 1(a)). Human breast milk data ($n = 170$) in fiscal 2001 were from Tada et al. (2002) and a local governmental body (Saitama prefecture, 2002b) (Table 1(a)). Human blood data ($n = 59$) in 2001 were supplied from a local governmental body (Shimane prefecture, 2002). Regarding the human blood data, 48 samples were from the vicinity of the pollution sources, and 11 samples were from a controlled site (Table 1(a)).

Total diet study (TDS) data in fiscal 1998–2001 ($n = 54$) were taken from MHLW (1999, 2000, 2001, 2003a). TDS data in fiscal 2002 ($n = 36$) were also taken from MHLW (2004) (Table 1(b)).

Four years of dioxin concentration data in various foodstuff in fiscal 1998–2001 ($n = 1740$) were obtained from The Ministry of Agriculture, Forestry and Fisheries of Japan (1999, 2000, 2001, 2002), MHLW (1999, 2000, 2001, 2003a), and Fisheries Agency, Japan (2002) (Table 3).

The data collection was conducted nationwide. Only data generated in accordance with QA/QC protocol by MOE were compiled and used for further analysis.

The results were shown as toxic equivalents (TEQs). WHO98-TEF (Van den Berg et al., 1998) was used as the toxicity equivalent factor (TEF) of isomers in this study.

Table 1(a)
PCDDs, PCDFs and dioxin like PCBs concentrations in various media and total diet study results in Japan, fiscal 2001

	Fiscal year	n	Min.	25th %tile	Median	75th %tile	Max.	Arithmetic mean	Geometric[b] mean	S.D.
Air[a] (pg-TEQ/m^3)	2001	791 (1028)	0.0072 (0.0072)	0.054 (0.051)	0.093 (0.090)	0.18 (0.18)	1.7 (1.7)	0.14 (0.13)	0.095 (0.092)	0.13 (0.14)
Soil[a] (pg-TEQ/g)	2001	2313 (3735)	0 (0)	0.055 (0.085)	0.35 (0.66)	1.8 (3.0)	240 (4600)	3.2 (6.2)	0.29 (0.48)	11 (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[c] (pg-TEQ/g-fat)	2001	170	–	–	–	–	–	20	–	–
Human blood[a,c] (pg-TEQ/g-fat)	2001	11 (59)	3.5 (1.0)	–	–	–	9.8 (20)	7.6 (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

[a] For air, soil, and human blood data, values shown without parentheses were calculated by excluding the data from the vicinity of pollution sources. Values shown in parentheses were calculated by all data including those from the vicinity of sources. For other media, values were calculated using all collected data.
[b] Geometric mean were calculated excluding 0pg-TEQ/g data. The fraction of zero data were less than 1%.
[c] The missing values indicated by hyphens were not obtained from the original literatures.

The methods of calculating TEQ were as follows, depending on the media:

1) *Soil, human blood, foodstuff and TDS*:
For values below the lowest quantitative limit, they were regarded as 0 in the calculation of TEQ for each isomer.
2) *Other media*:
For values below the lowest detection limit, only half the lowest detection limit was used in the calculation of TEQ for each isomer for the values that were over the lowest detection limit and below the lowest quantitative limit, the measured values were applied as they were in the calculation.

For all the media, the total TEQs were derived by adding up these values.

2.2. Point estimation

The average human intake levels in Japan in 2001 and 2002 were estimated with point estimation. The estimations were conducted taking into account three intake pathways: inhalation, soil ingestion, and diet.

The equation used to estimate dioxin intake due to inhalation was

$$E_{\mathrm{air}} = (C_{\mathrm{air}} \times \mathrm{IR}_{\mathrm{air}})/\mathrm{BW} \quad (1)$$

where

E_{air} = Daily intake of dioxin through inhalation (pg-TEQ/kg-bw/day);
C_{air} = Concentration of PCDDs, PCDFs, and dioxin like PCBs in air (pg-TEQ/m^3);
$\mathrm{IR}_{\mathrm{air}}$ = Inhalation rate (m^3/day);
BW = Body weight (kg).

The equation used to estimate dioxin intake due to soil ingestion was

$$E_{\mathrm{soil}} = (C_{\mathrm{soil}} \times \mathrm{IR}_{\mathrm{soil}})/\mathrm{BW} \quad (2)$$

where

E_{soil} = Daily intake of dioxin through soil ingestion (pg-TEQ/kg-bw/day);
C_{soil} = Concentration of PCDDs, PCDFs, and dioxin like PCBs in soil (pg-TEQ/g);
$\mathrm{IR}_{\mathrm{soil}}$ = Soil ingestion rate (g/day);
BW = Body weight (kg).

The arithmetic mean of dioxin concentrations in the air for each year was used for C_{air}. The arithmetic mean of the concentrations in soil for each year was used for C_{soil}. Because this study aims to estimate dioxin intake under normal conditions, only data from the sites in general environment and roadside were used for the estimation. However, estimations using all the collected data including those from the vicinity of the pollution sources were also

Table 1(b)
PCDDs, PCDFs and dioxin like PCBs concentrations in the air and soil (fiscal 2002) and total diet study results (fiscal 1998–2002) in Japan

	Fiscal year	n	Min.	25th %tile	Median	75th %tile	Max.	Arithmetic mean	Geometric[b] mean	S.D.
Air[a] (pg-TEQ/m^3)	2002	761 (989)	0.0066 (0.0066)	0.039 (0.036)	0.073 (0.070)	0.12 (0.12)	0.84 (0.84)	0.093 (0.093)	0.069 (0.067)	0.083 (0.089)
Soil[a] (pg-TEQ/g)	2002	2282 (3300)	0 (0)	0.096 (0.13)	0.47 (0.69)	2.0 (2.8)	250 (250)	3.4 (3.8)	0.39 (0.53)	12 (12)
Total diet study (pg-TEQ/kg/day)	2002	36	0.57	0.98	1.40	1.84	3.40	1.49	1.36	0.65
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

[a] For air, soil, and human blood data, values shown without parentheses were calculated by excluding the data from the vicinity of pollution sources. Values shown in parentheses were calculated by all data including those from the vicinity of sources. For other media, values were calculated using all collected data.

[b] Geometric mean were calculated excluding 0 pg-TEQ/g data. The fraction of zero data were less than 1%.

conducted to represent a more conservative estimation. Inhalation rate (IR_{air}), soil ingestion rate (IR_{soil}), and body weight (BW) were assumed as 15 m^3/day, 0.1 g/day, and 50 kg, respectively (MOE, 2000).

Total intake level was obtained as follows:

$$E_{total} = (E_{air} + E_{soil})/E_{diet} \qquad (3)$$

where

E_{total} = Total daily intake level of dioxin (pg-TEQ/kg-bw/day);
E_{air} = Daily intake of dioxin through inhalation (pg-TEQ/kg-bw/day);
E_{soil} = Daily intake of dioxin through soil ingestion (pg-TEQ/kg-bw/day);
E_{diet} = Daily intake of dioxin through diet (pg-TEQ/kg-bw/day).

The arithmetic mean of TDS results for each year (fiscal 2001 ($n = 12$), and 2002 ($n = 36$)) was used to estimate the intake through diet (E_{diet}).

2.3. Estimation of age-group-specific contribution of various foodstuff to total dietary intake

The contribution of various foodstuff to total dietary intake was estimated in point estimation approach. The difference of food consumption patterns of different age groups was taken into account. The estimation was conducted for the following age groups: 1–2, 3–5, 6–8, 9–11, 12–14, 15–17, 18–29, 30–49, 50–69, and 70+ years old. Dioxin concentration data for various foodstuffs (fiscal 1998–2001) were classified into 13 food groups according to the survey by MHLW (2003b). The food groups were as follows: rice and rice products; cereals, seeds and potatoes; sugars and confectioneries; fats and oils of animal origin; fats and oils of vegetable origin; pulses; fruits; green vegetables; other vegetables, mushrooms and seaweed; beverage, sauce and seasoning; fish and shellfish; meat and eggs; and milk and dairy products (Table 3). The food consumption volumes for food groups depending on age were calculated from MHLW (2003b). In order to obtain dioxin intake through each food group, the arithmetic mean of dioxin concentration in each food group was multiplied with the consumption volume. The age-specific total dietary intake was obtained as the sum of dioxin intakes through the 13 food groups. The age-group-specific contribution (percentage) of each food group to total dietary intake was then estimated.

2.4. Probabilistic approach by Monte Carlo simulation

The equations for the Monte Carlo simulation were the same as for the point estimation (Eqs. (1)–(3)). The dioxin concentrations in the air (C_{air}) and soil (C_{soil}), and intake through diet (E_{diet}) were represented as probabilistic density functions, in order to take into account of the variability associated with each parameter. Inhalation rate (IR_{air}), soil ingestion rate (IR_{soil}), and body weight (BW) were assumed as to be constant (IR_{air}: 15 m^3/day, IR_{soil}: 0.1 g/day, and BW: 50 kg) (MOE, 2000).

Four years of TDS data from fiscal 1998–2001 were combined to obtain adequate data size to elaborate curve fitting. The data in fiscal 2001 were used for the dioxin concentrations in the air and soil. The TDS levels for the four years were almost the same when considering the uncertainty as indicated by the large standard deviations of the data (Table 4) (MHLW, 1999, 2000, 2001, 2003a).

The air and soil concentration data from the vicinity of the pollution sources were excluded to obtain the probabilistic density functions because intake under abnormal conditions such as the vicinity of pollution sources is not the scope of this study, and the large sizes of the data from the vicinity of pollution sources might lead to different types or parameters of probabilistic density functions from those for normal environments. Also, Grubbs' test was conducted to remove abnormal data from the data set for TDS curve fitting. Although the air and soil data from the vicinity of the pollution sources and the statistically abnormal data of TDS were not used for curve fitting, the ranges of the probabilistic density functions were defined taking account of the actual measurement results of the entire data including those from the vicinity of pollution sources (*i.e.* from zero to the actual maximum measurement results [air: 0–1.7 pg-TEQ/m^3; soil: 0–4600 pg-TEQ/g; TDS: 0–7.01 pg-TEQ/kg/day].). The fit of the probabilistic density functions to the observed data was examined graphically with histograms and probability–probability plots (P–P plots).

After characterizing the variability of the parameters, the variability of the total intake was estimated. The total intake calculation equation (Eq. (3)) was solved 5000 times using a commercial software for Monte Carlo simulation (Crystal Ball® 2000 by Decisioneering Inc., USA).

3. Results and discussion

Table 1(a) shows compiled data on dioxin concentrations in various media as well as TDS results in fiscal 2001. Table 1(b) shows dioxin concentrations data in the air and soil in fiscal 2002, as well as TDS results in fiscal 2002 and 1998–2001. In the air, soil, and human blood, the statistics were derived from data excluding results from the vicinity of pollution sources, and the statistics based on the entire data including those from the vicinity of pollution sources are shown in parentheses. The arithmetic mean of the air concentration of the entire data was approximately the same as the arithmetic mean calculated excluding data from the vicinity of pollution sources. The dioxin concentrations in the air and TDS result in fiscal 2002 were lower than those in fiscal 2001, while those in soil were almost at the same level. A declining trend of dioxin concentrations in environmental media during our survey (fiscal 2000–2002) could be suggested (Tables 1(a), 1(b) and Suzuki et al. (2003)), when considering the reported declining trend between fiscal 1999 and 2001 (Council of Ministries and Agencies on Dioxin Policy, Japan, 2003). The Japanese government has restricted dioxin emissions from waste incinerators since 1997, and the ‘‘Law Concerning Special Measures against Dioxin’’ was enacted in 1999. These efforts might have resulted in the reduction tendencies in the various media.

Point intake estimates for each pathway and in total for fiscal 2001 and 2002 were shown in Table 2. This table also contains estimates for fiscal 2000 from our previous study (Suzuki et al., 2003). Estimates based on the entire data

Table 2
Estimates of intake of PCDDs, PCDFs and dioxin like PCBs in Japan, fiscal 2000, 2001 and 2002

	Dioxins exposure (pg-TEQ/kg-bw/day)				
	Fiscal 2000[a]	Fiscal 2001		Fiscal 2002	
Inhalation	0.042	0.042	(0.039)[b]	0.028	(0.028)[b]
Soil ingestion	0.0092	0.0064	(0.012)[b]	0.0068	(0.0076)[b]
Diet	1.45	1.63		1.49	
Total	1.50	1.68	(1.68)[b]	1.52	(1.53)[b]

[a] Quoted from Suzuki et al. (2003).
[b] Estimates in parentheses were based on dioxin concentrations including data from the vicinity of pollution sources.

Table 3
PCDDs, PCDFs and dioxin like PCBs concentrations in foodstuff (fiscal 1998–2001)[a]

Foodstuff	Dioxins concentrations (pg-TEQ/g)								
	n	Min.	25 %tile	Median	75 %tile	Max.	Arithmetic mean	Geometric[b] 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 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

[a] 4 years of data in fiscal 1998-2001 were combined.
[b] Geometric mean were calculated excluding 0 pg-TEQ/g data. The fraction of zero data were as follows: Rice and rice products = 25/166; cereals, seeds, and potatoes = 25/67; sugars and confectioneries = 3/6; pulses = 13/35; fruits = 54/112; green vegetables = 51/153; other vegetables, mushrooms, and seaweed = 94/205; beferage, sauce, and sesoning = 15/17; fish and shellfish = 7/644; meat and eggs = 12/243; milk and dairy products = 1/81.

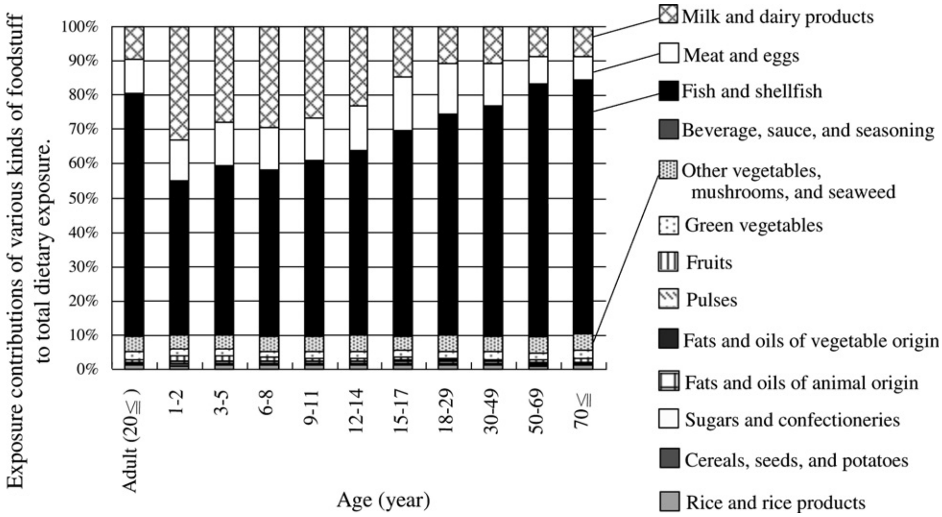

Fig. 1. Age-group-specific contribution of various foodstuffs to total dietary exposure.

set including those from the vicinity of pollution sources are shown in parentheses. The air data were obtained from more than 700 sites, and the soil data were from more than 2000 sites for each year. These sampling sites cover all the 47 prefectures in the nation. Moreover, regarding the TDS data, 120 food items were analyzed for one TDS sample, as taking their ingestion rates into account. Also, the TDS surveys were strategically conducted throughout seven districts which cover the whole nation. The large data size and the coverage of the nation ensured that the estimates were representative. In fiscal 2001, total intake was estimated at 1.68 pg-TEQ/kg-bw/day, with intake through inhalation, soil ingestion, and diet at 0.042, 0.0064, and 1.63 pg-TEQ/kg-bw/day, respectively. Also, total intake in fiscal 2002 was estimated at 1.52 pg-TEQ/kg-bw/day, with intake through inhalation, soil ingestion, and diet at 0.028, 0.0068, and 1.49 pg-TEQ/kg-bw/day, respectively. The total intake estimates in fiscal 2001 and 2002 based on the entire data including those from the vicinity of the pollution sources were 1.68 and 1.53 pg-TEQ/kg-bw/day respectively. Intake through diet accounted for more than 90% of the total intake; the contributions through inhalation and soil ingestion were relatively small. The average total intake estimates in both fiscal 2001 and 2002 were below the tolerable daily intake level (TDI) defined by MHLW (*i.e.* 4 pg-TEQ/kg-bw/day). The intake estimates in fiscal 2000 and 2002 were almost at the same level, when considering the standard deviations of TDS data (Tables 2 and 4). It is expected that reduction of dioxin concentrations in foods will be slower than those in environmental media such as the air and water due to dioxin's hydrophobicity and environmental persistence. Therefore, a long-term observation is required to clarify the transition of human intake levels.

Table 3 shows the dioxin concentrations in various foodstuff classified into food groups (in fiscal 1998–2001). Concentrations in "fish and shellfish" were remarkably high (average = 0.94, range = 0–26 pg-TEQ/g). Furthermore, "fats and oils of animal origin" and "meat and eggs" were also high. Age-group-specific contributions of various foodstuff to total dietary intake were estimated and are shown in Fig. 1; the estimates of intake through fish and shellfish accounted for approximately 45–70% of total dietary intake 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.

In the Monte Carlo simulation, four year of TDS data (fiscal 1998–2001) were combined to elaborate the curve fitting for intake estimate through diet. When considering the standard deviation of TDS data, the dioxin concentrations in foods were decreasing so slowly that no significant error was introduced by combining the data sets (Table 4). The maximum value of TDS (7.01 pg-TEQ/kg-bw/day) was regarded as an abnormal value by Grubbs' test (1%). The histograms, curve fitting and P–P plots of the air, soil and TDS are shown in Figs. 2–4, respectively. Lognormal distribution was the best fit for all the variables (air: [geometric mean (GM), geometric standard deviation (GSD) = 0.095, 2.4 pg-TEQ/m^3], soil: [GM, GSD = 0.29, 13 pg-TEQ/g], foods: [GM, GSD = 1.63, 1.42 pg-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 distribu-

Table 4
Transition of the result of total diet study by the Ministry of Health, Labour and Welfare, Japan (MHLW)[a]

	Results of the total diet study by MHLW (pg-TEQ/kg-bw/day)			
	Fiscal 1998	Fiscal 1999	Fiscal 2000	Fiscal 2001
Arithmetic mean	2.01	2.25	1.45	1.63
Range (min.–max.)	1.22–2.77	1.19–7.01	0.84–2.01	0.67–3.40
Standard deviation	0.49	1.50	0.38	0.71

[a] Quoted from MHLW (1999, 2000, 2001, 2003a).

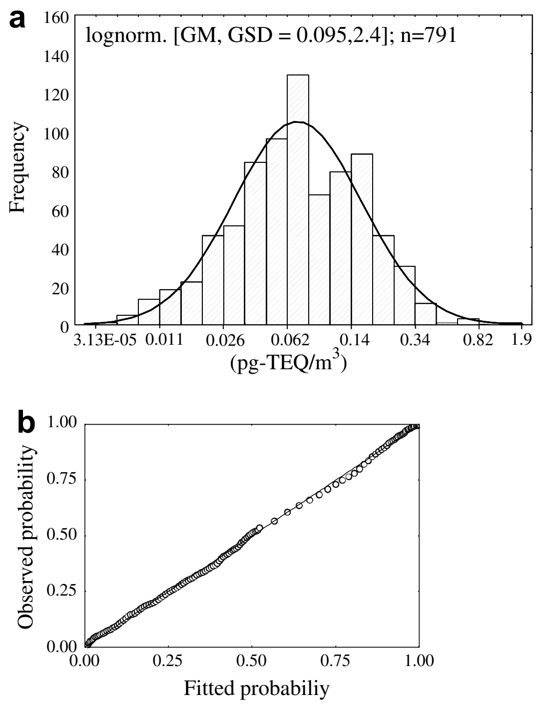

Fig. 2. (a) Histogram and curve fitting of dioxin concentrations in air (fiscal 2001). *X*-axis is in natural logarithmic scale. Data from the vicinity of pollution sources were excluded. (b) the P–P plots of air.

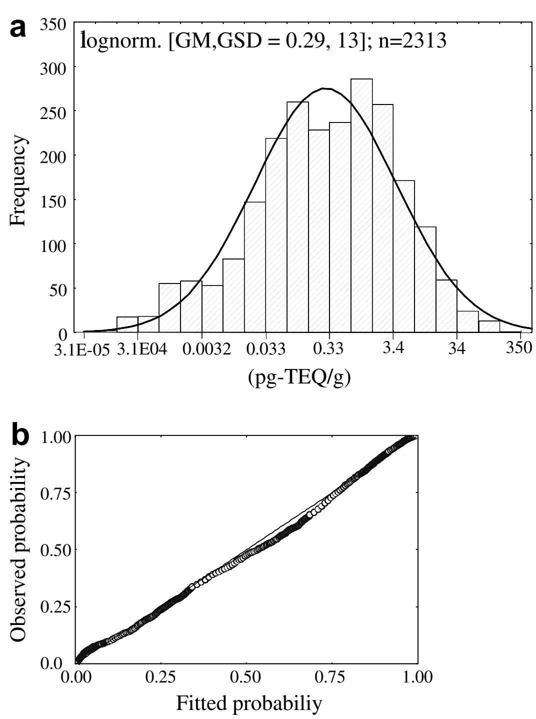

Fig. 3. (a) Histogram and curve fitting of dioxin concentrations in soil (fiscal 2001). *X*-axis is in natural logarithmic scale. Data from the vicinity of pollution sources were excluded. (b) the P–P plots of soil.

tion). Fig. 5 shows the results of an intake distribution using the Monte Carlo simulation method. The elaborated intake distribution with larger data size had a longer tail in

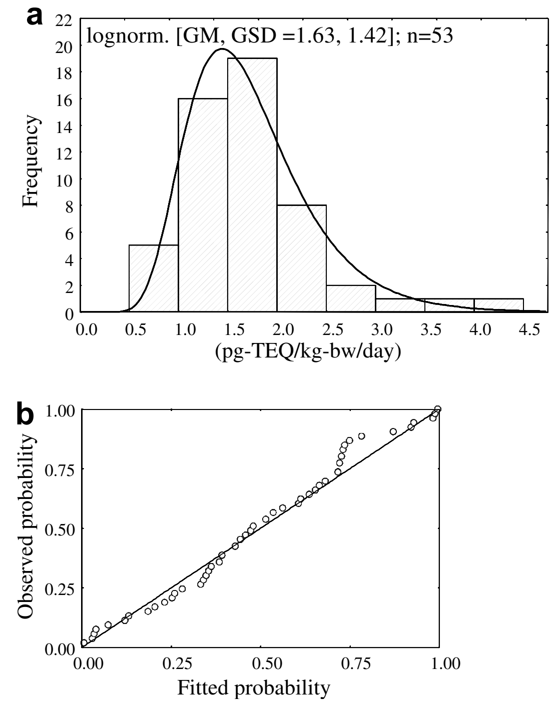

Fig. 4. (a) Histogram and curve fitting of dioxins intake from diet according to total diet studies (fiscal 1998–2001). The maximum value, 7.01 pg-TEQ/kg-bw/day, is excluded from this figure, because it was regarded as an abnormal value by Grubbs's test. (b) the P–P plots of diet.

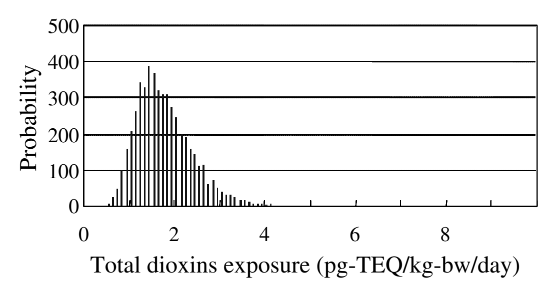

Fig. 5. Distribution of PCDDs/DFs and dioxin like PCBs intake in Japan, estimated by Monte Carlo simulation.

upper side, and the average and the medium have shifted slightly higher than the previous intake distribution in our study. The estimated average, median, 5th percentile and 95th percentile of the intake distribution were 1.78, 1.69, 0.95 and 2.91 pg-TEQ/kg-bw/day, respectively. The 95th percentile of the distribution was also below the TDI by MHLW (*i.e.* 4 pg-TEQ/kg-bw/day).

4. Conclusions

Although a decline of dioxin concentrations in environmental media in Japan between 1997 and 2001 were reported, there was no significant difference in human intake estimates by point estimate approach between fiscal 2000 and 2002. The transitions of intake levels should be addressed in the future. The average dioxin intake estimates in Japan in fiscal 2001 and 2002 were below the current tolerable daily intake level (TDI) by MHLW, Japan (*i.e.*, 4 pg-TEQ/kg-bw/day) in point estimation. Intake though diet accounted for more than 90% of the total intake. The estimates of intake through fish and shellfish accounted for approximately 45–70% of total dietary intake in each age group. A probabilistic approach by Monte Carlo simulation found that both the average and the 95th percentile of the dioxin intake distributions in Japan were estimated to be below the TDI by MHLW, Japan.

Acknowledgements

This research was conducted as a part of a project by Ministry of the Environment, Japan. We thank Prof. Iwao Uchiyama (Kyoto University), Prof. Hideaki Miyata (Setsunan University) and Masatake Toyoda (Jissen Women's University) for valuable comments on this study. We also thank Japanese local governmental bodies for supplying PCDDs, PCDFs, and dioxin like PCBs concentration data. Several members of the staff of Japan Environmental Sanitation Center provided welcome assistance with compilation of the data. We are grateful to Mr. Tommy Ka Kit Ngai (Massachusetts Institute of Technology) for revision of our manuscript to improve the readability.

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