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Risk analysis of persistent chlorinated organic compounds, dioxins and PCBs in Japan

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Abstract

As a reference value to protect humans from possible deleterious effects of dioxins (PCDD/Fs and co-PCBs), tolerable intake has been established as 4 pg-TEQ/kg-bw/day in Japan. For the general Japanese public, the primary source of exposure to dioxins is food, mostly through fish and its products. Co-PCBs contribute to nearly half the total dioxin intake on TEQ (2,3,7,8-TCDD equivalents) basis. The current level of exposure to dioxins for the general public is estimated to be less than the above-mentioned tolerable intake. A steep yearly decline in dioxin release into the environment is shown in the emission inventory largely due to the reduction in waste incineration, which is the predominant source on the inventory. On the other hand, the levels of existing dioxin reservoirs, the soil and sediment contaminated by the past use of herbicide, are anticipated to remain largely constant for years to come. Consequently, dioxin levels in inland and coastal fish may as well remain stable, hence keeping the dietary intake levels from a further rapid decline.

PCB production was banned in 1972 and its use was limited to closed systems in 1974, while the disposal of PCBs and PCB containing appliances were stipulated by law as late as in 2001 ordering all PCBs be destructed within 15 years. PCB levels in environmental media, food, or breast milk all exhibited a substantial decrease since 1970s. The general intake level for PCBs is well below the provisional tolerable intake, which was set at 5 μg/kg-bw/day based upon liver toxicity as the endpoint. One should notice, however, that significant amounts of PCBs had been released into the environment during the last 30 years of storage and that environmental exposure to PCBs as well as dioxins may disrupt neurological, hormonal or immunological functions of children at much lower doses than they elicit liver toxicity.

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1 Introduction

A serious PCB poisoning, known as *Yusho*, took place in Japan in 1968. Over 13,000 persons were poisoned by rice oil contaminated with PCBs (It was later elucidated that the toxicity was mainly due to PCDFs and quarterphenyls generated in PCBs by heat)[1]. PCB production and use in open-system were banned in 1972 and 1974 respectively. Disposal of PCBs, however, was deadlocked by NIMBY attitude of the public fearing the causal chemical of *Yusho*. In 2001, Special Measures Law on Promotion of Waste PCB Management was finally established and all the PCB products are destined for complete degradation for disposal within the next 15 years.

In February 1999, a popular TV news program broadcasted the detection of dioxins in green vegetable (TV Asahi dioxin report), which evoked serious concerns in the public over the safety of consuming agricultural products from and living close to incinerators. The joint dioxin risk assessment committee under Ministry of Health, Labour and Welfare (MHLW) and Ministry of the Environment (MOE) working at the time established the tolerable daily intake (TDI) of 4 pg-TEQ/kg-bw/day in June 1999 [2]. The Special Measures Law on Dioxin Control stipulating environmental standards and release limits derived from this TDI value was enacted in July and went into force in January 2000. The law was made by a strong motion from diet members and highly appreciated for its quick formation responding to grass-root's campaigns. The law prescribes for the constant monitoring of the environment by municipal governments and compliance to the stricter emission limits of the incinerators and furnaces, which will cost heavily on both government and private sectors.

Here we summarize the dioxin and PCB risk assessment and give the overview of these issues in Japan. In describing contamination levels, we try to adopt long-range studies to show the trends. Data from publicized ministerial reports were used whenever possible. The former Ministry of Health and Welfare was reformed into MHLW, while Environment Agency was reformed into MOE in January 2002. The current names were used throughout.

2 Dioxins and dioxin-like PCBs

2.1 Tolerable Intake

In 1997, MHLW established the tolerable intake of 10 pg I-TEQ/kg-bw/day for dioxins extrapolating from the administered dose producing cancer and hyperplasia in the liver of rats. In the same year, MOE established the health risk guideline value of 5 pg I-TEQ/kg-bw/day, by incorporating the dose on endometriosis in rhesus monkeys into consideration.

In 1998, WHO recommended to reduce the TDI value to 1-4 pg-TEQ/kg-bw/day from the previous 10 pg I-TEQ/kg-bw/day and use the new TEF system that includes the toxicity of co-PCBs [3]. Taking this into account, the joint dioxin risk assessment committee under MHLW and MOE established the TDI of 4 pg-TEQ/kg-bw/day in June 1999. The value was derived from the body-

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burden of 86 ng/kg in rats employing the affects in reproductive system of male offspring as endpoint. Although the value of 4 pg-TEQ/kg-bw/day is at the high end of WHO's recommendation, it is a substantial reduction from the former values even from the MOE's guideline level of 5 pg/kg-bw/day because the intake level may well double when calculated using WHO TEF system including the toxicity of co-PCBs.

In 2001, new dioxin health assessment results were reported by European Commission's Scientific Committee on Food (SCF)[4], Joint FAO/WHO Expert Committee on Food Additives (JECFA)[5], and UK Food Standard Agency [6] recommending lower tolerable intake levels (Table 1). The working groups under MHLW and MOE each studied these reports and concluded to maintain the current TDI of 4 pg-TEQ/kg-bw/day, with an emphasis on the need of continued investigations including kinetics of isomers and trans-generational effects at low doses.

It should be noted that tolerable intake values are the estimates of no effect dose from life-time exposure and are not the permissible levels. WHO stated clearly that the ultimate goal is to reduce the exposure to less than 1 pg-TEQ/kg-bw/day.

Table 1. Current tolerable intake levels for dioxins and dioxin-like PCBs

	Daily (pg-TEQ/kg/day)	Weekly (pg-TEQ/kg/week)	Monthly (pg-TEQ/kg/month)
WHO (1998)	**1 - 4**		
Japan (1999)	**4**	28[a)]	120[a)]
EC SCF (May, 2001)		**14**[b)]	
JECFA (June, 2001)			**70**[b)]
UK FSA (Oct., 2001)	**2**[c)]		

[a)] The number is shown only for the purpose of comparison.
[b)] Tolerable intakes are expressed on weekly or monthly basis taking into account the long half-lives (e.g. 7.5 years for TCDD) of the compounds in human body and that the excess in intake on any particular day may not significantly alters the body burden.
[c)] UK FSA takes precedence of daily basis for its better acquaintance.

2.2 Standards and limits

The Special Measures Law on Dioxin Control (Law 0150, 1999) was enacted in December 1999, and put into force in January 2000. The law established TDI value as 4 pg-TEQ/kg-bw/day on the basis of dioxin-related measures, and the environmental standards for air, water, sediment and soil were laid down accordingly. It also prescribes for regular monitoring of environmental dioxins (air, water, sediment and soil) by municipal governments and the compliance to the emission limits by the specified facilities. Table 2 summarizes the standards and limits established or amended based upon the law. Although phased approach was employed for some limits for three years of time, all the current limits are at their ultimate value after January 2002.

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Table 2. Standards and limits under the Special Measures Law on Dioxin Control

Environmental Standards:		
Ambient air	0.6 pg-TEQ/m^3 (max. annual average)	
Water	1 pg-TEQ/L (max. annual average)	
Sediment	150 pg-TEQ/g (max.)	
Soil	1,000 pg-TEQ/g (max.) (Action level: 250 pg-TEQ/g)	
Emission to air:		(Unit: ng-TEQ/m^{3}N)
Waste incinerators	New Facilities	0.1-5 (specified by capacity)
	Existing Facilities	1-10 (specified by capacity)
Furnaces and industrial processes		
	New Facilities	0.1-1 (specified by type)
	Existing Facilities	1-10 (specified by type)
Emission to water:	10 pg-TEQ/L (max.)	
For the specified facilities, including bleaching facilities using chlorine, waste PCB resolving facilities, waste gas cleansing facilities, wet dust collecting facilities.		
Terminal waste disposal facility discharge must comply with the same value.		

2.3 Inventory

Figure 1 shows the estimation of dioxin emission including dioxin-like PCBs (WHO TEQ) by MOE. The data shows that as much as 94% in 1997 and 88% in 2001 of the yearly emission was from incinerators in total. During these years, the emission estimates to water declined from 12.8 (1997) to 4.6 (2001) g-TEQ/year and constituted 0.2 and 0.3% of the yearly emission respectively. For other even smaller emissions, cigarette smoke (max. 0.2 g-TEQ/year) and car exhaust (1.59 g-TEQ/year) are posted on the inventory.

The government of Japan stated in the Basic Guidelines for the Measures against Dioxins that it aims at 90% reduction in total dioxin emission within 4 years starting in 1999. Two years later in 2001, the emission estimate in the inventory pointed to more than 75% reduction from 1999. Marked regression was estimated between 1997 and 1998, probably reflecting the reduction due to the measures against the waste incinerators that had been stipulated amending existing Air Pollution Control Law in 1997. The effects of more stringent emission limits after 2002 on dioxin emission reduction are yet to be counted.

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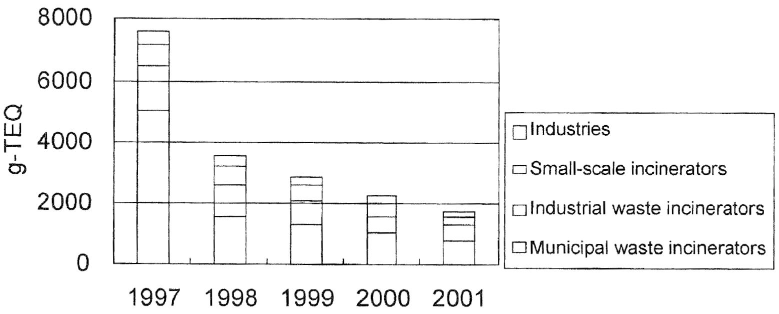

Figure 1: Dioxin emission trends in Japan shown in an inventory by MOE.

2.4 Exposure

Environmental media: The results from the dioxin monitoring of general environment by the national and municipal governments in 2000 are summarized in Table 3. Environmental standard was surpassed in 4 air and 83 water samples. 215 air samples from the vicinity of roads or identified emission sources were also analyzed and 6 surpassed the standard. Among 1,089 soil samples from the vicinity of identified emission sources, 1 (1,200 pg-TEQ/g) surpassed the standard.

Table 3. Dioxin levels in environmental media (excluding the vicinity of roads and identified emission sources)

Medium	n	min.	25%	med.	75%	max.	mean	s.d.
Air ($pg\text{-}TEQ/m^3$)	707	0.0073	0.065	0.12	0.19	0.76	0.14	0.11
Soil (pg-TEQ/g)	1,943	0	0.067	0.44	2.7	280	4.6	18
Water (pg-TEQ/L)	2,128	0.012	0.08	0.14	0.32	48	0.31	1.2
Sediment (pg-TEQ/g)	1,838	0.0011	0.47	1.4	6.1	1,400	9.6	46

Food: Fish is the most significant dioxin source in food for Japanese adults. More than 70% of dioxin intake through food is form fish. Meat is the second largest source and constitutes about 10% of intake. Figure 2 shows the food survey data by MHLW (2000). The contamination level of fish and fish products was the highest with the average of 1.6 pg-TEQ/wet-g. The highest value of 6.8 pg-TEQ/wet-g was from one sample of conger eel.

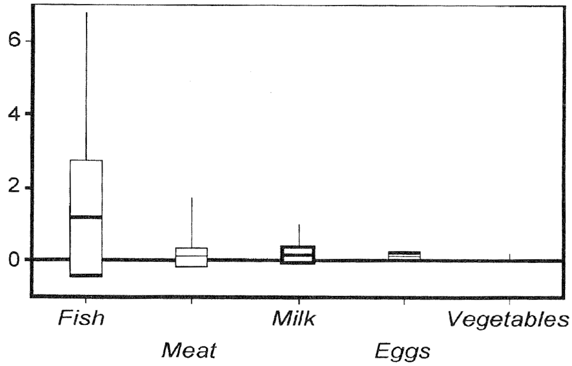

Figure 2: Dioxin contamination levels of food (pg-TEQ/wet-g).
The shaded box shows the range of standard deviations with the average as the central horizontal line. The vertical line shows the range of distribution. Fish, meat and milk include their corresponding products. Vegetables include grain crops.

A series of total diet studies by MHLW shown in Figure 3 depicts that during these years dioxin intake from food had been less than tolerable intake levels. Average intakes other than from food were estimated to be 0.042 pg-TEQ/kg-bw/day from air and 0.0092 pg-TEQ/kg-bw/day from soil (MOE, 2003).

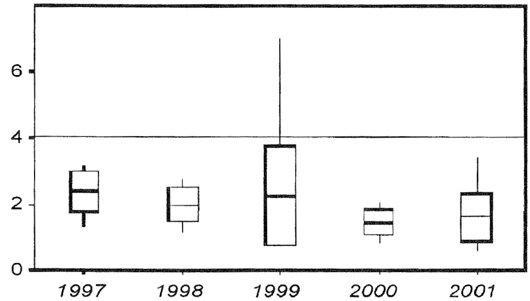

Figure 3: Dioxin intake from food for average Japanese (pg-TEQ/kg-bw/day).

Breast milk: Dioxin level in breast milk specimens provided from mothers living in Osaka (Figure 4, MHLW) shows a marked decline till the mid 1980s probably due to a decrease in co-PCB level during 1970s and in the first half of 1980s.

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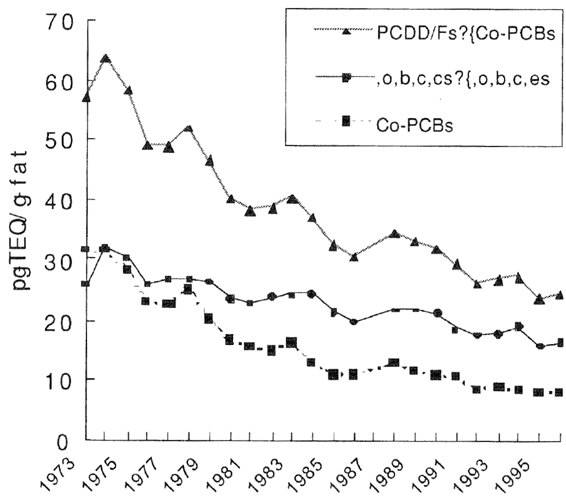

Figure 4: Dioxin levels in breast milk.

3 Non-dioxin-like PCBs

3.1 Tolerable Intake

The tolerable intake for PCBs of 5 μg/kg-bw/day was derived from the NOAEL of 10 ppm for hepatotoxicity in rats (Monsanto, 1972)[7].

3.2 Standards and limits

The values on Table 4 were determined based on the tolerable intake level. In 2001, Special Measures Law on Promotion of Waste PCB Management was established and all the PCB products are destined for destruction within the next 15 years.

Table 4. PCB standards and limits

Work area		0.1 mg/m^3
Ambient air (provisional)		0.5 μg/m^3
Emission gas (provisional)		0.15 mg/m^3
Emission from PCB incineration (provisional)		0.10 mg/m^3
Maximum level in Food:	coastal fish	3 ppm
	pelagic fish	0.5 ppm
	meat	0.5 ppm
Sediment (for removal, provisional)		10 ppm (dry)
Water		no detection
Soil		no detection
Waste water		0.003 mg/L

3.3 Inventory

PCBs were produced during the period between 1954 and 1972 and approximately 54,000 tons were used in Japan. Although little is known about

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the PCB inventory, two main suspected sources are the PCB products of the past and the non-intentionally formed by-products in certain industrial processes.

3.4 Exposure

Environmental media: PCBs were found in all samples (air, water, sediment and wildlife) in the nation wide survey by MOE (2000). The levels were: 0.091-2.3 ng/m^3 in air, 0.095-8.4 ng/L in water, 0.042-750 ng/g in sediment and 3.8-350 ng/g-wt in wildlife.

Food: PCB levels in common market fish specimens declined from 0.1-0.9 ppm in 1971-1975 to less than 0.1 ppm in 1991-1994 [8].

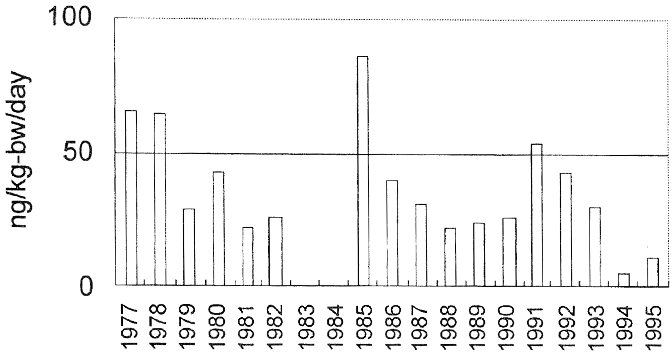

Figure 5: PCB intake from food for Osaka residents.

Figure 5 shows PCB intake levels from the total diet study in Osaka [9]. The similar study in Tokyo resulted in the same range of intake: 62, 50, and 18 ng/kg-bw/day in 1979, 1984, and 1994 respectively, demonstrating the decline in PCB intake level since the 1970s. Fish was the primary source of PCB in diet constituting 60-90% of the total intake through the study period [8].

Breast milk: PCB levels of breast milk from the mothers in Osaka shows the decline to less than 20% of the 1970s levels in recent years (Figure 6)[10].

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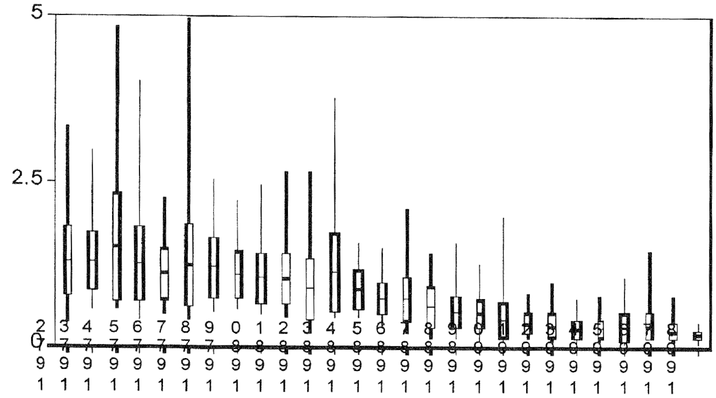

Figure 6: PCB levels in breast milk (μg/g-fat)

4 Discussion

After the ban of PCB production in 1972, following the drop of the levels in environmental media and in food, human tissue levels as seen in breast milk also declined greatly (Figure 6). Dioxins in terms of TEQ shows the similar drop during the same time period, although the PCDDs and PCDFs went down much gradually compared with the co-PCBs (Figure 4). The difference in declination trends may be due to the fact that PCBs were produced and used in large quantity before 1972, and the exposure levels of general public at the time must had been much higher than in later years. In recent years, the slopes seem to have leveled off at the low end, which may suggest that the measures taken were effective in reducing hazardous exposure, however, that further reduction is more difficult to archive. For both dioxins and PCBs, *de novo* synthesis and release into the environment is much smaller compared with the amounts already released and reside in the environment. While PCBs were manufactured as products, dioxins had been deposited in 1960s and 1970s as impurity in herbicides used in quantity in rice fields (Figure 7)[11]. For the both compounds sediment serves as the reservoir that contributes to the contamination of fish and shellfish, the main source of exposure in regular Japanese diet. Atmospheric deposition of dioxins, on the other hand, is shown to be the main source of contamination in some vegetables. There also are reports to show that cows grazing closer to incinerators tend to produce milk with higher TEQ levels. The current source-directed measures seem to be of great importance in keeping contamination levels low in certain farm products.