CONTROL OF SOURCES OF PCDDs/DFs AND THEIR ENVIRONMENTAL MONITORING IN JAPAN

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Introduction

It is well known that polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/DFs) cause various toxic effects through the Ah receptor mechanism. In addition to PCDDs/DFs, some coplanar congeners of polychlorinated biphenyl (Co-PCBs) are categorized as dioxin-like compounds. There are a variety of emission sources of PCDDs/DFs: primary sources include impurities in pesticides, which are byproducts of the manufacturing process, and byproducts of combustion reactions, such as occur during the incineration of municipal solid wastes (MSW); secondary sources appear when PCDDs/DFs from these primary sources are transported and accumulate elsewhere. To decrease the exposure of humans and the environment to PCDDs/DFs, it is indispensable to take appropriate measures to reduce the emissions from these sources; therefore, it is critical to grasp the kinds of sources and the quantities of emissions from each source. While PCDDs/DFs released into the environment are transported chiefly via air and water, human exposure is mostly from food. In Japan, intake from fish accounts for more than 70 % of human PCDD/DF exposure, and approximately 20% is from meat and dairy products. Because of the high rates of PCDDs/DFs occurring in particulate form, we need to pay further attention to the transfer of PCDDs/DFs regionally, although we also need to become aware of their transfer on a global scale.

With these features of PCDDs/DFs in mind, we focus on an inventory of sources in Japan and the measures that have been taken so far to control emissions. Moreover, the results of environmental monitoring and details of exposure conditions are reported in order to evaluate the efficacy of such control measures.

Source Inventory

An inventory of amounts of PCDDs/DFs emitted to the environment in Japan was investigated according to source, including industrial processes (Table 1).[1)] This inventory is a summary based on data officially measured and calculated by the Japanese Government and made public annually since 1997. In 1997, emissions to the air from MSW incineration facilities amounted to approximately 5000 g-TEQ/y and emissions from incineration facilities for industrial wastes amounted to about 1500 g-TEQ/y. In industrial processes other than waste treatment, emissions were about 230 g-TEQ/y from electric furnaces for steel manufacturing and 135 g-TEQ/y from sintering processes in the steel industry. Other examples are 47 g-TEQ/y from zinc recovery and 25 g-TEQ/y from aluminum alloy manufacturing. The total emissions were approximately 7500 g-TEQ/y, of which waste incineration accounted for over 85 %. In 2000, after the implementation of full-scale control of PCDDs/DFs, emissions had decreased to a total of about 2200 g-TEQ/y, a reduction of nearly 70 % compared to 1997 emissions. The emissions from MSW incineration, which used to be the greatest source, had decreased from approximately 5000 g-TEQ/y to about 1000 g-TEQ/y, a decrease of nearly 80 %. The

total emissions are on target for a 90 % decrease by the end of fiscal 2002. In European countries such as Germany and the Netherlands where measures against PCDDs/DFs have preceded those in Japan, the greatest amounts of PCDD/DF emissions were initially from waste incineration; after having taken necessary measures against waste incineration, the majority of PCDDs/DFs are now released by industrial processes, such as steel manufacturing utilizing steels and recycled, non-ferrous materials.[2,3)]

Table 1. PCDD/DF Source Inventory in Japan

Sources	Emission Amounts (g-TEQ/y)			
	1997	1998	1999	2000
(I) Emission to the air				
General waste incineration facilities	5000	1550	1350	1019
Industrial waste incineration facilities	1500	1100	690	555
Small waste furnaces	340–591	368–619	307–509	353–370
Industrial sources	459	324	296	259
Electric furnaces for steel manufacturing	229	140	142	131
Steel industry sintering process	135	114	101	70
Zinc recovery	47	25	22	27
Aluminum alloy manufacturing	25	23	17	17
Other industries	23	22	14	15
Others	3.8–6.4	3.9–6.6	3.9–6.7	3.9–6.7
(II) Emission to water				
Waste incineration facilities	5.3	5.3	5.3	2.5
Industrial sources	6.1	5.6	5.7	4.7
Sewerage final treatment facilities	1.2	1.2	1.2	1.2
Final disposal facilities	0.1	0.1	0.1	0.1
Total	7343–7597	3358–3612	2659–2864	2198–2218
Reduction from 1997 rates	100 %	51 %–56 %	61 %–65 %	70 %–71 %

Development of Regulatory Measures in Japan (Table 2)

With the Guideline for PCDDs/DFs Control in MSW Management, which was promulgated in 1990, Japan began to promote substantial measures for waste management. Under historical waste management policy, in principle, each small administrative unit was supposed to deal with and manage wastes within its municipalities. Japan had approximately 2000 MSW incineration facilities at that time. Full-scale measures were initiated in 1997 in response to many requests for more efficient measures. The then Ministry of Health and Welfare revised the guidelines in 1997 and decided to incorporate some further desirable legal measures, and under the Waste Management Law strengthened the standards for the structures of incineration systems and for the maintenance and management of incineration facilities. In connection with this, the Environment Agency (currently Ministry of the Environment) designated PCDDs/DFs as a 'specific substance' and specified facilities subject to regulation under the Air Pollution Control Law. Regulatory standards were set at 0.1–5 ng-TEQ/m^3 for newly installed waste incineration facilities and 1–10 ng-TEQ/m^3 for existing facilities, depending on their capacities. Regulatory standards were also established for sintering processes in the steel industry, with criteria set at 0.1 ng-TEQ/m^3 for newly installed facilities and 1 ng-TEQ/m^3 for existing facilities. Furthermore, the law placed restrictions on flue gas emissions from electric furnaces for the steel industry, the zinc recovery industry, and aluminum alloy manufacturing.

Table 2. Development of Policies to Control PCDDs/DFs in Japan

System	Enactment	Jurisdiction	Outline
Guideline for PCDDs/DFs Control in MSW Management (The First)	1990	Ministry of Health and Welfare	1. Specification of technical measures for combustion and waste gas treatment facilities 2. Regulation of standard methods for analyzing PCDDs/DFs 3. Expectation value of flue gas: < 0.5 ng-TEQ/m^3
Guideline on Waste Management (The Second) Amendment of Waste Management Law	1997	Ministry of Health and Welfare	1. Reinforcement of standards for structure and maintenance Combustion: 800 °C, > 2 s, CO 100 ppm, Ignition Loss < 10% Waste gas: Precipitation temperature < 200 °C Advanced dust control function 2. Proper management of fly ash and incinerator ash Melting and dechlorination Total emission target:> 5 mg-TEQ/waste-ton 3. Licensable facilities Change from 5 t/d to > 200 kg/h or an over-fire lattice area of = 2 m^2
Amendment of Air Pollution Control Law	1997	Environment Agency	1. Set up control standards for 'specified substances' 2. Facility: Waste incineration facilities Newly installed: 0.1–5 ng-TEQ/m^3 (according to capacity) Existing: 1–10 ng-TEQ/m^3 (Within 5 years, according to capacity) Urgent measure: = 80 ng-TEQ/m^3
Special Measures Law on Dioxins Control	1999	Environment Agency	1. Set up TDI and environmental standards (air, water quality, and soil) 2. Control waste gas and waste water 3. Draw up reduction plan (target: 90% reduction) 4. Impose limits on incineration fly ashes, and contaminated soil 5. Relegate duty of survey/measurement of pollution
Special Measures Law on Promotion of Waste PCB Management	2001	Ministry of the Environment	1. Notification of stock, manifest system 2. Set up PCB management plan and manufacturers' responsibilities 3. Fund for PCB management and management business by Japan Environment Corporation 4. Completion of PCB destruction within 15 years

In 1998, a high PCDD/DF concentration of 8500 pg-TEQ/g was detected in the soil surrounding the Toyono Clean Center in Nose-cho, Osaka Prefecture. This issue gave rise to considerable controversy, and the cause of the soil pollution was investigated. The investigation detected an extremely high concentration of PCDDs/DFs in the water circulation system of the wet spray tower. This presumably happened because combustion was not complete in the furnace, and emission gas containing high concentrations of PCDDs/DFs was generated in the process of electrostatic precipitation. In addition, the recycling of the waste water from emission gas scrubbing resulted in secondary production and concentration of PCDDs/DFs, of which a portion became mist from the open cooling tower and was released into the environment. In a separate incident in 1999, a TV news program reported that PCDDs/

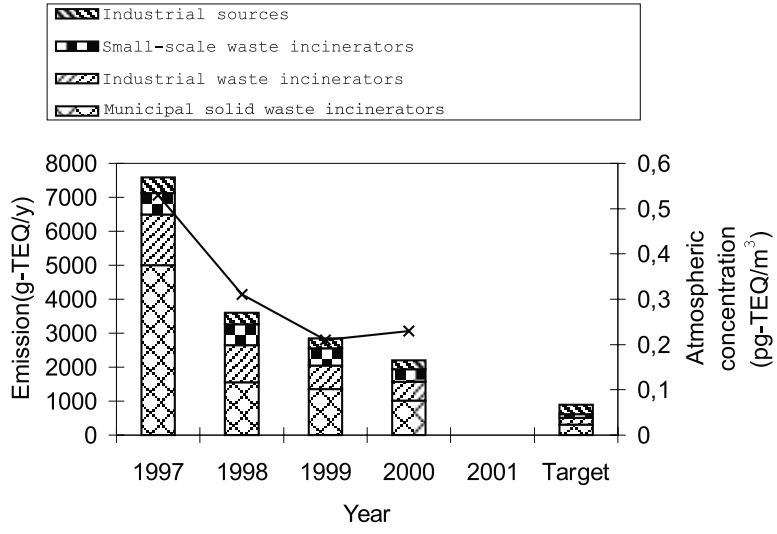

Figure 1. Changes in atmospheric concentration and total emissions of PCDDs/DFs

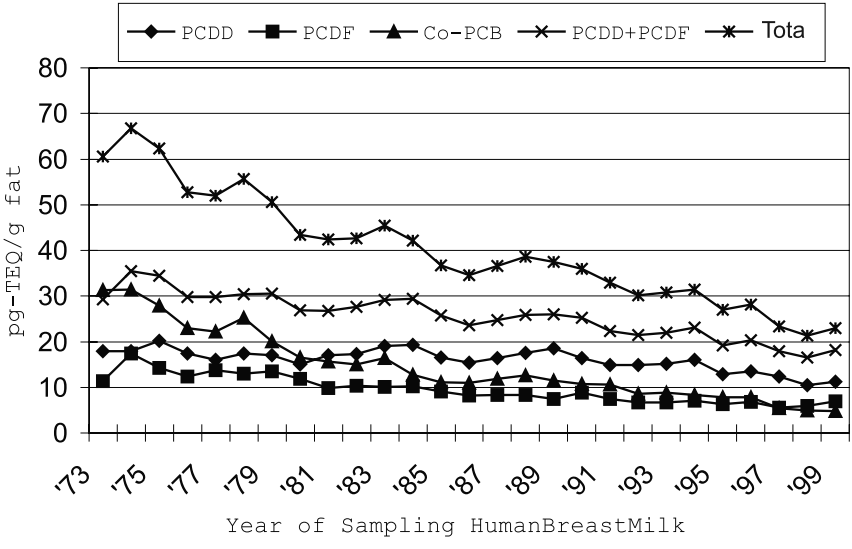

Figure 2. Changes in concentrations of PCDDs/DFs in stored samples of human breast milk

DFs had been detected in the leaves of spinach and tea cultivated in Tokorozawa City, Saitama Prefecture, which led to serious public concern. Following these two incidents, the Japanese government established the Special Measures Law on Dioxin Control in 1999 from the viewpoint that dioxin control measures had to be undertaken urgently. The tolerable daily intake (TDI) was set at 4 pg-TEQ/kg/d, and environmental standards for air, water quality, and soil were, respectively, 0.6 pg-TEQ/m^3, 1 pg-TEQ/L, and 1000 pg-TEQ/g. The government policy was developed with the following

priorities: firstly to decrease emission sources, secondly to establish more satisfactory measurement systems, and thirdly to evaluate the actual reduction effects by environmental monitoring.

Despite the preceding measures, wastes containing PCBs were still not adequately treated in Japan. In order to organize necessary management systems and to promote reliable and proper treatment of wastes, the Special Measures Law on Promotion of Waste PCB Management was established in June 2001. This law requires that:

1. Manufacturers shall notify their municipalities of the conditions of stored PCB wastes and their disposal. It shall be the responsibility of the manufacturers to maintain and dispose of their stocks of PCBs safely and properly,
2. The government, prefectures, and large cities designated by ordinance shall each draw up a plan for PCB waste management and take the necessary steps to treat the wastes decisively and appropriately, and
3. All stored PCB wastes shall be disposed of within 15y.

Monitoring of Changing Environmental Levels and Human Exposure

To evaluate the efficacy of PCDD/DF control measures to decrease environmental loads, we conducted research on two incineration facilities—an old batch-operation MSW incinerator that had been in operation for nearly 20 years and a new, continuously operating MSW incineration facility built adjacent to it—to obtain comparative data on PCDD/DF emissions during operation and PCDD/DF concentrations in the environment around each facility.[4)] During the operation of the older facility, the concentration of PCDDs/DFs in ambient air ranged from 0.042 to 0.41 pg-TEQ/m^3. The concentration peaked directly below the stack and tended to decrease as the distance from the stack increased. During the operation of the new facility, the concentration range in ambient air was 0.023–0.17 pg-TEQ/m^3, with most values lower than those during operation of the older facility. In contrast to the values from the older facility, there was little tendency for concentrations to decrease at sampling points farther away from the point just below the stack. Concentrations at a point 10 km west of the facilities were 0.042 pg-TEQ/m^3 for the older facility in operation and 0.023 pg-TEQ/m^3 for the new facility. These data suggest that these values are the background values in this area. The concentrations of PCDDs/DFs in atmospheric depositions during the operation of the older facility ranged from 19 to 1900 pg-TEQ/m^2/d, and showed the same trend as data for ambient air. However, the peak adjacent to the facility was more striking than the peak value for ambient air. Concentrations of PCDDs/DFs during the operation of the new facility ranged from 20 to 25 pg-TEQ/m^2/d, almost equivalent to the value of the older facility at the point 10 km west (19 pg-TEQ/m^2/d), which was selected as the background value, suggesting that the values vary little with distance.

The average concentrations of PCDDs/DFs in the atmosphere throughout Japan, along with annual changes in total emissions, have been monitored since 1997 (Fig. 1). The atmospheric concentrations shown in the figure are the average concentrations of 43 continuously monitored sampling points. Figure 1 proves that atmospheric concentrations have decreased in response to reducing the total emissions of PCDDs/DFs. Changes of PCDDs/DFs in human breast milk were also evaluated since 1973 (Fig. 3). This result suggests that the PCDD/DF concentrations in human breast milk decreased from the 1970s until the 1990s [5)] . The success in dealing with air and water pollution since the 1970's imply the long term reduction and the recent specific measures on PCDDs/DFs could be reducing PCDDs/DFs as well.

Conclusion

In this report we examined environmental policies to control PCDDs/DFs in Japan, and we evaluated their efficacy as indicated by environmental monitoring. The main points are:

REGULATORY ASPECTS

1. While total emissions of PCDDs/DFs were approximately 7500 g-TEQ/y in 1997, they decreased to one-third, 2200 g-TEQ/y, in 2000. The greatest source, MSW incineration, decreased by nearly 80% from approximately 5000 g-TEQ/y to about 1000 g-TEQ/y.

2. Reduction of PCDDs/DFs was mostly due to advanced control technologies at waste incineration facilities; this was accelerated by the Special Measures Law on Dioxins Control established in 1999. In addition, the Special Measures Law on Promotion of Waste PCB Management, worked out in 2001, aimed at promoting measures for controlling dioxin-like PCBs, which are regarded as PCDDs/DFs. The target is to finalize PCB treatment within 15 years.

3. To evaluate the effects of these PCDD/DF control policies, we examined the annual changes in continuously monitored atmospheric concentrations. The average atmospheric concentration of 43 continuously monitored sampling points decreased to half in 2000 compared to the concentration in 1997. Concentrations of PCDDs/DFs in human breast milk were on the decrease as well.

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