New Guideline for Controlling Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDDs/DFs) in MSW Management in Japan

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Abstract
The "Guideline for Controlling PCDDs/DFs in Municipal Waste Management - PCDDs/DFs Reduction Program (New Guideline)" was established in Japan in January, 1997. This action was instigated by the following decision; in 1996 the Ministry of Health and Welfare set a Tolerable Daily Intake (TDI) of 10 pg TEQ/kg/day as the acceptable level. The amount of PCDDs/DFs from MSW incineration was 4,300 g TEQ/y according to the 1996 data. At present, MSW incineration is still the biggest source of the PCDDs/DFs emission. The new guideline recommended, in principle, the use of full-continuous-feed incineration and high-technology measures, and provided PCDDs/DFs concentration standards for full-continuous-feed incineration, 0.1ng TEQ/Nm^3 for new incinerators and 0.5 ~ 5ng TEQ/Nm^3 for existing incinerators, according to the facility type. It also showed various schemes like wide-area type incinerators or RDF facilities for basic waste management systems. Through these steps, the amount of PCDDs/DFs in emission gases has to be reduced to 590g TEQ/y, or by 86%, in 5 years, and to approximately 20g TEQ/y, or by 99%, in 20 years. The guideline set the target for the total amount of PCDDs/DFs, not only from emission gas but also from residues, at below 5μg TEQ/waste-ton.

1. Introduction

When we find evidence that environmental pollutants, especially hard-to-decompose organic pollutants like Persistent Organic Pollutants (POPs), have an irreversible effect on human beings and the ecosystem, it is very important what preventive measures we can take. PCDDs/DFs, generated from MSW incineration, are exactly one of the hardest-to-decompose organic pollutants and urgent measures must be taken to reduce them. Efforts for MSW generation control and recycling are bearing fruit in Japan, but on the other hand, MSW incineration has also an important role from the view of public health and energy recycling. In 1993, the total amount of MSW was 50.3 mio. tons, the recycling rate was 8.0% and the incineration rate was 74.3%. As, in principle, refuse has to be disposed of within the boundaries of each ward, the number of incineration facilities amounts to 1,854 (full-continuous type: 433/batch type: 1421). It is quite a lot. This report explains the "Guideline for Controlling PCDDs/DFs in MSW Management - PCDDs/DFs Reduction Program" (hereafter called "the new guideline"), which was issued in January, 1997.

2. Recent Development and Conditions in Japan

MSW incinerators in Japan have been regarded as the largest source of PCDDs/DFs. According to authors' estimation, MSW incinerators generated PCDDs/DFs of 3,100 ~ 7,400g TEQ/y in 1990[1)], and the Ministry of Health and Welfare reported that they generated PCDDs/DFs of 4,300g TEQ/y in 1996. Although all sources has not been officially surveyed, it is certain that MSW incineration is the biggest source of the PCDDs/DFs emission at the present stage. Control measures for PCDDs/DFs in Europe are far ahead of the rest of the world. Germany and the Netherlands have reduced PCDDs/DFs levels to one hundredth of those of 1980's, through recent combustion or waste gas control measures, and now PCDDs/DFs are produced only in some grams TEQ/y order from the biggest source, MSW incineration.[2), 3)]

"Japan's Guideline for Controlling Dioxins and Dibenzofurans (PCDDs/DFs) in Municipal Waste Treatment (the old guideline)", which was established by the Ministry of Health and Welfare in December, 1990, incorporated reduction measures for PCDDs/DFs as much as were technologically feasible in those days, based on research results and international trends.[4] The guideline set a target of 0.5ng TEQ/Nm^3 for new incinerators and regulated operating conditions such as combustion temperatures, concentrations of CO and oxygen and dust collector inlet temperatures.

Since November, 1994, a Ministry of Health and Welfare scientific research group has been discussing the toxicity evaluation of PCDDs/DFs. They made an interim report in June, 1996 that, for the moment, the TDI should be 10pg TEQ/kg/day. The main points of the report were as follows;

(1) the NOAEL (No Observed Adverse Effect Level) was set at 1ng TEQ/kg/day, comprehensively judging from the toxicity evaluation results,

(2) applying a safety factor, or one hundredth of NOAEL, the TDI became 10pg TEQ/kg/day, and

(3) I-TEF (International Toxicity Equivalent Factors) were used as the coefficient of toxicity equivalent.

Meanwhile the Environment Agency established "Risk Evaluation Standards for Human Health" of 5pg TEQ/kg/day as a desirable PCDDs/DFs toxicity level to protect human health, in accordance with the interim report of the PCDDs/DFs risk evaluation committee. The Environment Agency has focused on the risk evaluation value of 1pg TEQ/kg/day[6], which was set by the Health Council in the Netherlands, based on the results of research on Rhesus monkeys by Rier et. al.[5] The agency concluded that further investigation would be necessary if the value of 1pg TEQ/kg/day is used in Japan. Therefore, they established the risk evaluation index of 5pg TEQ/kg/day, twice as safe as 10pg TEQ/kg/day.

With the investigation of PCDDs/DFs toxicity evaluation, "The Advisory Committee for Controlling PCDDs/DFs in MSW Management", chaired by Hiraoka, was established in June, 1996, and made a final report in January, 1997. The histogram of PCDDs/DFs concentration from MSW incineration in 1996 showed that concentrations of PCDDs/DFs from new incinerators regulated by the old guideline was considerably low. It was realised that the old guideline had a certain effect. In the case of the batch-type operation, however, a lot of PCDDs/DFs were produced in starting up and shutting down the operation (Table 1). It became obvious that high concentrated PCDDs/DFs were still emitted from many incineration facilities. This evidence moved us to establish the new guideline.

Table 1 Concentration of PCDDs/DFs Discharged at Starting up and Shutting down of Semi-continuous-feed Incinerator

[unit: ng-TEQ/Nm^3]

Incinerator	Capacity	Steady operation	Starting up	Shutting down	Stock fire
Semi-continuous-feed and stoker type	30t/16h x 2	77.3	344	120	726
Semi-continuous-feed and stoker type	65h/16h x 2	2.5	4.2	1.5	-
Semi-continuous feed and stoker type	31t/16h x 2	27.6	25.5	63.5	-
Semi-continuous feed and fluidized bed type	48t/16h x 2	5.6	92.7	6.5	12.9

Note: 1. These data were measured at the outlet of dust collector.
2. "Stock fire" means the status that operation keeps a certain temperature for the next smooth starting up with kindling matter.
3. These data were reported by Japan Waste Management Foundation.

3. New Guideline - PCDDs/DFs Reduction Program[7]

3.1 Basic Ideas of the New Guideline and Emergency Measures

PCDDs/DFs controlling measures of the new guideline are classified principally into "Permanent Measures", which reduce the total PCDDs/DFs emission as much as possible systematically and "Emergency Measures" which handle PCDDs/DFs if concentrations exceed the standard at the existing facilities. The "Emergency Measures" mean concrete PCDDs/DFs reduction measures are considered and carried out when the concentrations of PCDDs/DFs in the emission gases is beyond the limit of 80ng TEQ/Nm^3. Even if the concentration is not beyond the standard, the new guideline attempts to control the combustion situation and carry out the permanent measures systematically. Table 2 details emergency measures for the existing incinerators. Compared to the old guideline for the existing incineration facilities in 1990, the new guideline listed the facility operation. The new guideline tries to control the PCDDs/DFs which are produced most at starting up and shutting down incinerators by sifting semi-continuous-type and batch-type incinerators to full-continuous-type ones and eliminating the stock fire system. In addition, it clearly indicates that incinerators should be operated under a proper load because the operation under an excessive load, exceeding the design specifications, generates a large amount of PCDDs/DFs. From the technological aspect, the measures for gas cooling system are added to the old guideline measures for incineration and waste gas treatment facilities. The control of the resynthesis of PCDDs/DFs in waste heat boilers or air heaters is also important. As for incineration facilities, the control value of CO, which is an incineration control indicator, was lowered to 50ppm from 100ppm for the full-continuous type and from 200ppm for the batch type. For a standard of stable combustion, the frequency of CO concentration over 500ppm should be less than 5 times/h. In waste gas treatment facilities, the dust collector inlet temperature was lowered to 200 ~ 280℃, from 250 ~ 280℃ for electric precipitators and to an appropriate level below 200℃ from 250 ~ 280℃ for fabric filters.

Table 2 Emergency Measures for Existed Incineration Facilities (1997)

	Full-continuous type incinerator	Semi-continuous type and mechanical batch type
Management	①Long-term continuous operation ②Adequate loading operation, and homogenization of waste quality ③Annual measurement of PCDDs/DFs concentration	①Shift to continuous operation from intermittent one ②Adequate loading operation, and homogenization of waste quality ③Annual measurement of PCDDs/DFs concentration
Combustion equipment	①Combustion temperature: over 800℃ (over 850℃ is desirable.) ②CO concentration: below 50ppm (below 100ppm on the old GL) ③Stable combustion: Momentary CO concentration does not reach the peak of 500 ppm (less than 5 times/hr).	①Combustion temperature: over 800℃ (over 850℃ is desirable.) ②CO concentration: below 50ppm (below 200ppm on the old GL) ③Stable combustion: Momentary CO concentration does not reach the peak of 500 ppm (less than 5 times/hr).
Gas cooling equipment	①Controlling dust on the heating surface of boiler, temperature of outlet waste gas of boiler is lowered. ②Remodeling the gas cooling chamber fixed on the top of furnace. ③Controlling dust piled in air preheater.	①Remodeling the gas cooling chamber fixed on the top of furnace. ②Controlling the dust piled in air preheater.
Waste gas treatment equipment	①Lowering the temperature of inlet waste gas of electric precipitator (200-280℃) (250-280℃ for the old GL). ②Substituting electric precipitator for fabric filter. ③Lowering the temperature of inlet waste gas of fabric filter (below 200℃).	①Lowering the temperature of inlet waste gas of electric precipitator (200-280℃) (250-280℃ for the old GL). ②Substituting electric precipitator for fabric filter. ③Lowering the temperature of inlet waste gas of fabric filter (below 200℃). ④Substituting multi-cyclone for fabric filter.

The "Permanent Measures" mean stabilized combustion, which produces as little PCDDs/DFs as possible, can be carried out, reducing the quantity of waste through waste discharge control and recycling. The following are given as concrete examples; remodeling facilities for stable combustion and high technological waste gas treatment, integrating small-sized facilities, shifting from semi-continuous operation to full-continuous and shifting to RDF (Refuse-Derived Fuel) facilities. For new incineration facilities, the PCDDs/DFs concentration in emission gases must be regulated to be less than 0.1ng TEQ/Nm^3 as are European and American ones. The guideline is characterized as follows;

(1) high technologies should be applied to curb the generation of PCDDs/DFs,
(2) measures to shift the whole system, e.g. shifting to full-continuous-feed type or RDF facilities, are important, and
(3) measures for residues, like a high technological waste gas treatment, are specified.

The new guideline is expected to work as a social control unit in order to prevent the environmental accumulation of PCDDs/DFs.

3.2 Applying High Technology to PCDDs/DFs Control

The measures for new incineration facilities are shown in Table 3.

Table 3 Measures for Newly Established Waste Incineration Facilities

	Full-continuous type incinerator	Semi-continuous type and mechanical batch type
Management	①Long-term continuous operation. ②Adequate loading operation, and homogenization of waste quality. ③Annual measurement of PCDDs/DFs concentration.	②Adequate loading operation, and homogenization of waste quality. ③Annual measurement of PCDDs/DFs concentration.
Receiving and feeding equipment	①Sufficient capacity of refuse pit, agitation and quantitative feed. ②Homogenizing the quality of waste by milling and crushing, quantitative feeding and enough controlling.	①Sufficient capacity of refuse pit, agitation and quantitative feed. ②Homogenizing the quality of waste by milling and crushing, quantitative feeding and enough controlling.
Combustion equipment	①Combustion temperature: over 850℃ (over 900℃ is desirable.) (800 or 850℃ for the old GL.) ②Residence time: more than 2 sec. (On the old GL, 2 sec. is required for full-boiler type.) ③CO concentration: below 30ppm (below 50ppm on the old GL) ④Stable combustion: Momentary CO concentration does not reach the peak of 100 ppm.	①Combustion temperature: over 850℃ (over 900℃ is desirable.) (800 or 850℃ for the old GL.) ②Residence time: more than 2 sec. (On the old GL, 2 sec. is required for full-boiler type.) ③CO concentration: below 30ppm (below 50ppm on the old GL) ④Stable combustion: Momentary CO concentration does not reach the peak of 100 ppm. ⑤Reduction of start-up time, use of a burner for secondary combustion, and stopping the operation after complete combustion
Gas cooling equipment	①Making cooling tubes of boiler long enough to reach combustion chamber wall. ②Controlling dust on the heating surface of boiler. ③Lowering the heat boiler outlet temperature and reducing the passing-through time.	①In principle, gas cooling chamber is separated from the furnace. ②For gas cooling chamber is fixed on the top of furnace, it is designed not to give a bad effect for secondary combustion.
Waste gas treatment equipment	①Lowering the temperature of dust collector (below 200℃). ②Absorptional removal: blowing powder activated carbon, absorption tower of activated carbon. ③Destructive removal by oxidation catalysis.	①Lowering the temperature of dust collector (below 200℃). ②Absorptional removal: blowing powder activated carbon, absorption tower of activated carbon. ③Destructive removal by oxidation catalysis.

In comparison with the old guideline, the new one sets the combustion temperature at over 850℃, preferably 900℃, and sets the residence time at more than two seconds, regardless of boiler type. These regulations have reduced CO concentrations, by combustion standards, to 30ppm, from 50ppm under the old guideline. The establishment of the new indicator for gas cooling equipment aims at the

control of de novo synthesis, as dose the old indicator. As for the waste gas treatment system, the dust collector temperature was specified to be below 200℃ and more positive ways to remove PCDDs/DFs, including activated carbon adsorption and oxidation catalyst destruction, were recommended. High-technology waste gas treatment is almost fully developed and is expected to be ready for practical use in the near future. To implement these measures the following concentrations were given as standards;

- New full-continuos-feed incinerator: 0.1ng TEQ/Nm^3
- Existing old guideline-applied full-continuous feed incinerator : 0.5ng TEQ/Nm^3
- Existing full-continuous-feed incinerator (old guideline are not applied): 1ng TEQ/Nm^3
- Existing semi-continuous-feed or batch-type incinerator (long-term continuous operation): 1ng TEQ/Nm^3
- Existing semi-continuous-feed or batch-type incinerator (intermittent operation): 5ng TEQ/Nm^3

In the future, environment-oriented and highly regulated incineration facilities will be required. It is important that technologies are well-balanced and not lean only toward the control of PCDDs/DFs. The technologies have to be effective controlling other hazardous components, like heavy metals, and be selected not to give significant influence to other environmental impact elements including energy load.

3.3 Systematic PCDDs/DFs Control Measures

High-technology measures are based on the reduction of the quantity of waste requiring incineration treatment through waste discharge control and recycling. Moreover, the new guideline suggests substituting wide-area type incineration systems for small-scale incineration (Figure 1).

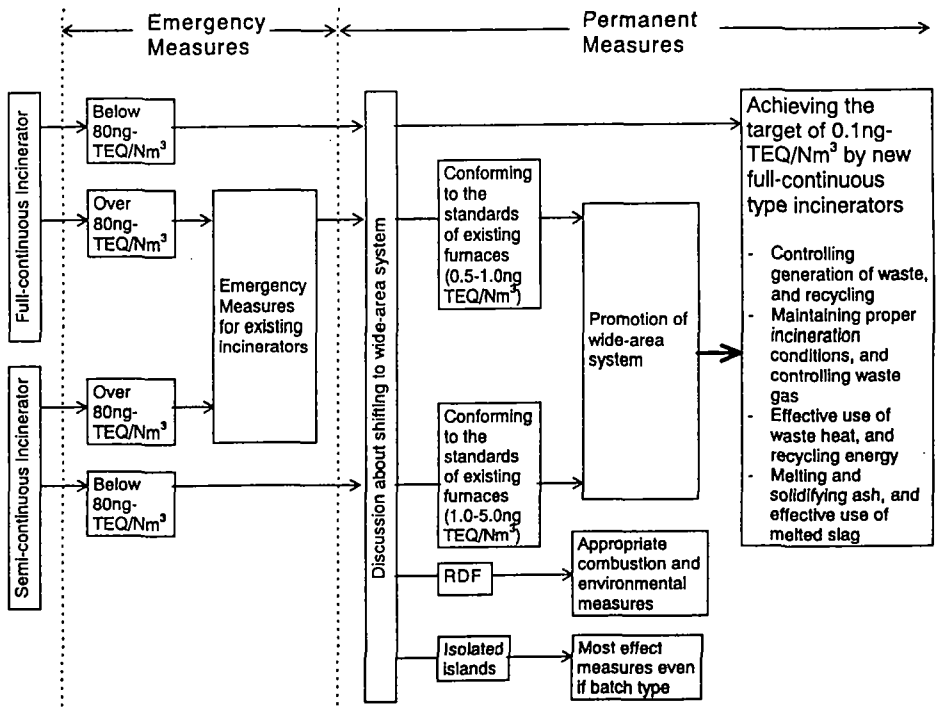

Figure 1. Waste Management System in the Future

It is also one of the characters of the new guideline that systematic measures are specified as well as technological measures. That is because 24-hour continuous operation generates less PCDDs/DFs than intermittent operation. On the other hand, it is difficult to shift from the batch type to the full-continuous type in small-population municipalities because of the small amount of waste. Therefore, integration into larger-scale full-continuous-feed incinerators and wide-area type facilities are required. Feasible wide-area type facilities are as follows;

(1) the existing small-sized system expands to the full-continuous-type incinerator, and
(2) all semi-continuous types are replaced by RDF facilities in every municipality, and integrate RDF into one facility. Power or heat, generated from full-continuous operation, is used as an energy source.

If the RDF system is introduced and waste is burnt in a fulidized bed incinerator, energy can be recovered by the continuous combustion, even though it is a small scale. From the view point of efficiency and economization of waste treatment, however, the capacity of full-continuous incinerators should be at least 100 ton/day, and if possible, over 300 ton/day. Although the wide-area system is partly opposed to the principle of the waste administration that waste should be disposed of in each municipality, taking into account the present situation that requires a cyclic viewpoint of disposal administration, small-population community should pay attention to reduction of waste, recycling, generation of waste in easier form for incineration and comprehensive recovery of energy. Considering the greenhouse effect, gas control and the effective use of energy and resources, they would be right directions.

RDF system has some problems, such as energy-consumption during each process of crushing, selecting and casting, and the treatment of selected nonflammable substances. It has the following advantages however;

(1) uniform quality of RDF and even heat levels make the control of combustion standards easy,
(2) the long-term storage of RDF is possible because lime is mixed with RDF in the process of casting, and
(3) If RDF, from which heavy metals have been removed, is incinerated and the hydrogen chloride is neutralized by lime, the amount of PCDDs/DFs is reduced in comparison to the mass burning process (Table 4).

The PCDDs/DFs concentration from RDF combustion was regulated to the same level as that of permanent measures (0.1ng TEQ/Nm^3) in order to prevent the generation of PCDDs/DFs. RDF, from which heavy metals like copper are removed, is expected to be an effective material because it includes less catalysts for de novo synthesis. Recycling of metals is also recommended. When introducing RDF system, an adequate selecting equipment is essential in order to remove metals and nonflammable matter.

The systematic measures were not specified in the old guideline and their further development is expected. The new guideline showed the follow-up of these measures to see the effect. PCDDs/DFs concentration should be measured regularly (once a year) under the ordinary load and control situation. It is recommended that results are available to the public.

3.4 Function as Social Control Unit to Prevent the Environmental Accumulation

The new guideline also showed more detailed measures for the treatment of incinerator residues, like fly ash and bottom ash. This is because PCDDs/DFs of approximately 0.3ng TEQ/g are contained in bottom ash and 1 ~ 50ng TEQ/g in fly ash. The total amount of PCDDs/DFs in incinerator residues is more than that in emission gases, and amounts to 80 ~ 90% of the total. Since gases move easier than solids, we should make efforts to control emission gases. However, it is also important to focus on the control of residues in solid phase. As mentioned-above, if the PCDDs/DFs from the incineration process can be controlled, the content of PCDDs/DFs in residues would be automatically reduced. The

measures to control PCDDs/DFs in emission gases from incinerator are essential. The guideline showed technologies to decompose PCDDs/DFs contained in incinerator residues, melting and solidification[8], and heating dechlorination[9]. These measures can reduce the content of PCDDs/DFs in total, not only in emission gases but in treated residues, to the level below 5μg TEQ/waste-ton. Reducing the emission gases load and the residue load, we should reduce the environmental load of PCDDs/DFs step by step.

Table 4. PCDDs/DFs Concentration from RDF Combustion

Furnace type	Incineration capacity (kg/hr)	Gas temperature (℃)		PCDDs/DFs Conc. (ng-TEQ/Nm^3)		Remarks
		Inlet of fabric filter	Outlet of fabric filter	Inlet of fabric filter	Outlet of fabric filter	
Stoker	125	223 (176-270) *cyclone outlet	67 (37-86)	16.6 (11.9-22.1)	0.65 (0.05-2.0)	- Cyclone + cooler + fabric filter - Five kinds of RDF was burnt. [Processes toward RDF] - Removing metals by a magnetic separator. - Sending dry waste to an air classifier, and removing unfit matters for fuel (earth and sand, metallic debris, leathers, etc.). - Crushing into pieces in the size of less than 2 cm. - Adding slaked lime (2% of solid waste amount).
Fluidized bed	Approx. 70	173 (157-186)	142 (137-149)	2.5 (1.1-3.7)	0.20 (0.09-0.28)	- Ten kinds of RDF were burnt. [Processes toward RDF] - Almost same as stoker's.
	Approx. 300	157 (156-157)	127 (126-128)	3.5 (3.1-3.8)	0.88 (0.66-2.1)	- Two kinds of RDF were burnt. [Processes toward RDF] -Removing metals by a magnetic separator and aluminum separator. - Crushing into pieces in the size of 15mm. - Removing nonflammable materials and dry. - Adding slaked lime (3% of inlet waste amount) and molding.

Note: These data were provided by Japan Waste Research Foundation

4. Conclusion

If appropriate emergency measures are taken in the incineration facilities, the PCDDs/DFs concentration in the emission gases from where is over 80ng TEQ/Nm^3, it is possible to reduce the total emission from the present situation of 4,300g TEQ/y to 2,800g TEQ/y, or by approximately 35%, in 2 or 3 years (Figure 2). In addition, permanent measures including the establishment of new incinerators, shifting to wide-area type incineration facilities from small-sized facilities and introduction of RDF facilities are adopted, it would be possible to reduce the total amount of PCDDs/DFs emission gases to 590g TEQ/y, i.e., by 86%, in 5 years, and to approx. 20g TEQ/y (by 99%) in 20 years. To solve the PCDDs/DFs problem in MSW incineration, the waste management system, which was launched as the first 5-year project in 1963, is ought to be radically reformed. Furthermore, to achieve the accepted risk evaluation indicators for human health, adequate measures to reduce PCDDs/DFs should be applied to other sources of pollution including industrial waste facilities and various manufacturing processes in addition to MSW incinerators.

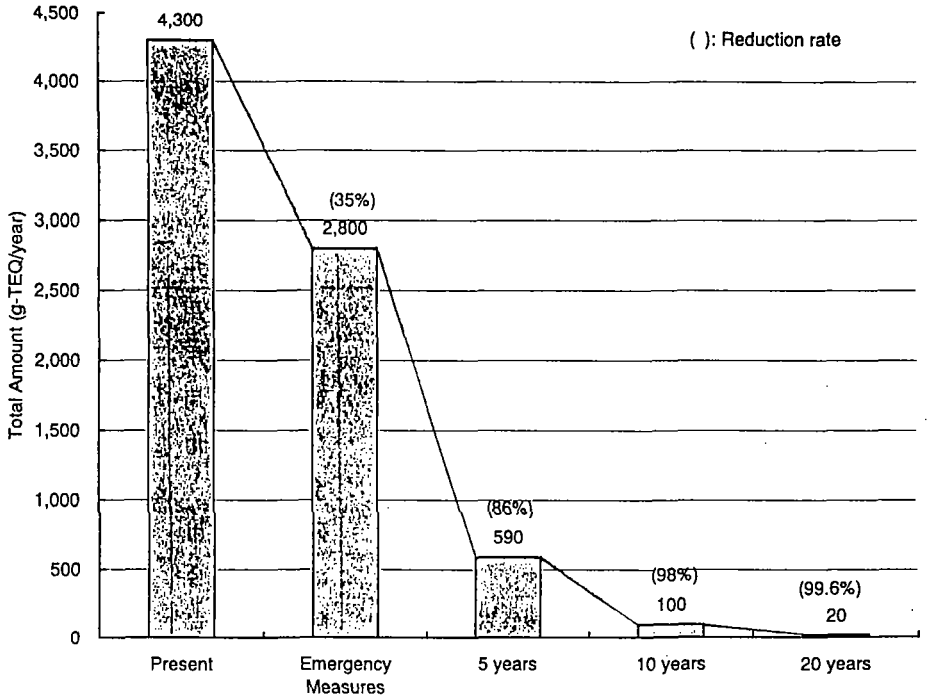

Figure 2. Expected PCDDs/DFs Emission by New Guidelines

References

1) Hiraoka, M.; Okajima, S. Organohalogen Compounds, 1994, 19, 275-291
2) Fiedler, H. Organohalogen Compounds, 1993, 11, 221-228
3) Born, J. Organohalogen Compounds, 1996, 27, 46-49
4) Hiraoka, M.; Sakai, S.; Yoshida, H. Chemoshere, 1992, 25, 1393-1398
5) Rier, S.; Martin, D.; Bowman, R; Dmowski, W.; Becker, J. Fundamental & Applied Toxicology, 1993, 21, 433-441
6) Health Council of the Netherlands. Dioxins, 1996
7) The Advisory Committee for Controlling PCDDs/DFs in MSW Management. Guideline for Controlling PCDDs/DFs in MSW Management, 1997
8) Sakai, S.; Hiraoka, M. WASCON'97, 1997
9) Hagenmaier, H.; Kraft, M.; Burnner, H.; Haag, R. Environ. Sci. Technol. 1987, 21, 1080-1084