ORIGINAL ARTICLE

Polychlorinated dibenzo-*p*-dioxin and dibenzofuran concentrations in serum samples of workers at intermittently burning municipal waste incinerators in Japan

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Objectives: To find whether or not incinerator workers employed at intermittently burning municipal incineration plants are exposed to high concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs).
Methods: 20 Workers employed at three municipal waste incineration plants (incinerator workers) and 20 controls were studied. The previous job, dietary, smoking, and body weight and height were obtained from a questionnaire survey. Concentrations of PCDDs and PCDFs were measured in serum samples of the workers and the deposited dust of the plants. The influence of occupational exposure on concentrations of PCDDs and PCDFs in serum samples was examined by multiple regression analysis.
Results: Dust analysis showed that dominant constituents were octachlorodibenzo-*p*-dioxin (OCDD) and 1,2,3,4,6,7,8-heptachlorodibenzo-*p*-dioxin (HpCDD) among the PCDDs, and 1,2,3,4,6,7,8-heptachlorodibenzofuran (HpCDF) and octachlorodibenzofuran (OCDF) among the PCDFs. The toxicity equivalents (TEQs) of summed PCDDs and PCDFs in the deposited dust were 0.91, 33, and 11 ng TEQ/g, respectively, for plants I, II, and III. The means of TEQ in serum samples of summed PCDDs and PCDFs in the incinerator workers and controls were 22.8 and 16.4 pg TEQ/g lipid for area I, 29.4 and 19.3 pg TEQ/g lipid for area II, and 22.8 and 24.9 pg TEQ/g lipid for area III, which were almost the same as for the general population of Japan. No significant differences in the TEQ of PCDDs and TEQ of PCDDs and PCDDs were found between the incinerator workers and the controls. However, the TEQ of PCDFs was significantly higher among the incinerator workers in areas I and II, and the 1,2,3,4,6,7,8-HpCDF concentration was also significantly higher for all three areas. When the occupational exposure index for each constituent of PCDDs and PCDFs was defined as the product of the duration of employment at the incineration plant and the concentration of the constituent in the deposited dust, multiple regression analysis showed that the concentrations of HxCDF, HpCDF, and TEQ of PCDFs in serum samples increased with the occupational exposure index. The multiple regression analysis also suggested that significant factors affecting the concentrations in serum samples were area for HxCDD, age for TCDD, PeCDD, PeCDF, TEQ of PCDDs, TEQ of PCDFs, and TEQ of summed PCDDs and PCDFs, and BMI for HxCDD, HpCDD, and OCDD.
Conclusion: This study showed that incinerator workers employed at intermittently burning incineration plants were not necessarily exposed to high concentrations of PCDDs and PCDFs. However, the increases in the concentrations in serum of HxCDF, HpCDF and TEQ of PCDFs with the occupational exposure index suggest that the incinerator workers had inhaled dust containing PCDDs and PCDFs during their work.

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Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are chemically and biologically similar compounds and are also highly toxic chemicals. In Japan, the most important source of PCDDs and PCDFs is incinerators[1] because most solid waste is burned in municipal waste incinerators without sufficient measures to prevent the generation of these chemicals. The municipal incinerators were classified into the intermittently burning type and the 24 hour continuously burning type. Concentrations of summed PCDDs and PCDFs in the exhaust gas of municipal waste incinerators ranged from 0.04 to 990 ng toxicity equivalents (TEQ)/Nm^3 for the intermittently burning type and from <0.01 to 200 ng TEQ/Nm^3 for the continuously burning type in 1995–6.[2] Consequently, the emission rate of PCDDs and PCDFs was generally higher in the intermittently burning type than in the continuously burning type.

Previously, we conducted biological monitoring of PCDDs and PCDFs among workers employed at three municipal continuously burning incineration plants, and found that the serum concentration of 1,2,3,4,6,7,8-heptachlorodibenzofuran (HpCDF) increased significantly among the incinerator workers, but the TEQ of summed PCDDs and PCDFs in serum samples was not higher than that for the general population of Japan.[3] However, the Ministry of Labor performed a medical examination of workers employed at a municipal waste incineration plant equipped with intermittently burning incinerators (Toyono, Osaka Prefecture), and found that the mean

Abbreviations: PCDDs, polychlorinated dibenzo-*p*-dioxins; PCDFs, polychlorinated dibenzofurans; OCDD, octachlorodibenzo-*p*-dioxin; HpCDD, heptachlorodibenzo-*p*-dioxin; HpCDF, heptachlorodibenzofuran; OCDF, octachlorodibenzofuran; TEQs, toxicity equivalents; TCDD, tetrachlorodibenzo-*p*-dioxin; PeCDD, pentachlorodibenzo-*p*-dioxin; HxCDD, hexachlorodibenzo-*p*-dioxin; TCDF, tetrachlorodibenzofuran; PeCDF, pentachlorodibenzofuran: HxCDF, hexachlorodibenzofuran; BMI, body mass index

Table 1 Characteristics of incinerator workers and control workers at the three areas

	I		II		III	
	Incinerator workers	Controls	Incinerator workers	Controls	Incinerator workers	Controls
Workers (n)	7	7	7	7	6	6
Age (y, mean (SD))	44.1 (5.5)	43.0 (5.7)	40.6 (11.1)	41.6 (9.9)	38.7 (8.3)	41.7 (8.1)
Duration of employment at incineration plant (year, mean (SD))	11.0 (5.1)	0	10.3 (3.4)	0	15.7 (7.3)	0
Previous job with potential for exposure to dioxin in agriculture (workers, n)	2	1	2	3	0	2
Dietary habit (times/week, mean (SD)):						
Fish	9.1 (3.6)	7.3 (2.7)	6.6 (2.4)	6.4 (2.6)	10.2 (4.5)	8.0 (3.2)
Meat	4.6 (3.5)	7.6 (3.5)	8.1 (3.7)	8.1 (3.1)	4.7 (2.4)	5.7 (4.2)
Milk	2.4 (2.1)	5.1 (10.1)	4.0 (4.6)	3.9 (3.2)	5.0 (5.1)	5.8 (3.9)
Current smoking:						
Non-smokers (workers, n)	1	3	3	2	1	3
Smokers (workers, n)	6	4	4	5	5	3
Smorking habit index (cigarettes/day×year, mean (SD))	699 (367)*	274 (336)	327 (440)	263 (296)	141 (140)	89 (143)
Body mass index (kg/m^2, mean (SD))	23.9 (3.6)	24.7 (3.7)	22.4 (2.3)	24.2 (2.1)	24.6 (3.8)	22.1 (2.8)

*p<0.05.

concentration of summed PCDDs and PCDFs in plasma was 323 pg TEQ/g lipid among highly exposed workers.[4 5] This value was about 15 times as high as that of the general population. The finding raised the question whether workers employed at other intermittently burning incineration plants were exposed to PCDDs and PCDFs of a similar level. In this study, serum concentrations of PCDDs and PCDFs were measured for workers employed at three intermittently burning municipal waste incineration plants, and the influence of occupational exposure to PCDDs and PCDFs was examined. This study was approved by the Osaka Prefectural Institute of Public Health.

SUBJECTS AND METHODS

Subjects

Three municipal waste incineration plants I, II, and III equipped with intermittently burning incinerators were selected in Nagasaki (area I), Chiba (area II) and Nagano (area III) prefectures in Japan. One incinerator was operated for 8 hours/day in plant I, and two incinerators were operated for 16 hours/day in plants II and III. The incinerators were fluid bed types for plants I and II, and a stoker type for plant III. To remove particulate matter from the flue gas stream, the incinerator of plant I was equipped with an electrostatic precipitator until 1997 and with a bag filter since then. The incinerators of plants II and III were equipped with electrostatic precipitators. The incinerated volumes were 20, 40, and 60 tonnes/day, respectively, at plants I, II, and III. In 1996–9, concentrations of summed PCDDs and PCDFs in the exhaust gas of the incinerators were 0.011–590, 11–260, and 11–53 ng TEQ/Nm^3, respectively.

The numbers of workers employed at plants I, II, and III were seven, 10, and 16, respectively. Three, seven, and six of the workers were employed in the operation of the incinerator for 1 year or more and they agreed to participate in this study after a full explanation of it. These workers usually operated and inspected the incinerators, and periodically cleaned the inside of the cooling towers between the incinerator and bag filter or electrostatic precipitator. The cleaning work was performed wearing a dust mask. In plant I, three workers who were employed in the treatment of incombustible waste sometimes supported the incinerator workers, and they also participated in this study. A worker who had been employed at plant I until 1994 and had since been employed at a neighbouring crematorium also participated in this study. Thus, the total number of subjects at plant I was seven. For comparison, seven, seven, and six male clerical workers matched for age (±5 years) were selected from the municipal governmental employees in areas I, II, and III, respectively. Statistical power could have been increased with more controls, but we did not use more because of the ethics of the invasive blood test. The age of incinerator workers ranged from 24 to 59 and of controls from 27 to 58. The means of age and employment duration at the incineration plants are shown in table 1.

Questionnaire survey

The workers were questioned about their previous job, numbers of meals of fish, meat, and cow's milk in a week, smoking, and body weight and height, because these variables could have confounding effects.

Measurement of PCDDs and PCDFs

Reagents

Standard solutions of PCDDs and PCDFs were purchased from Wellington Laboratories (Canada). Silica gel and aluminium oxide were obtained from Merck (Germany) for cleaning up. Other reagents were obtained from Wako Pure Chemical Industries (Japan).

Serum samples

The workers were asked not to eat breakfast, and a blood sample (about 100 ml) was collected from each person on a morning in 1999 for plants I and II and in 2000 for plant III. The serum samples were separated by centrifugation and collected in a chemically clean container. After addition of $^{13}C_{12}$-2,3,7,8-substituted PCDDs/PCDFs as internal standards, the sample was kept frozen at −30°C until the time of analysis.

The analytical method was described in our previous paper.[3] Briefly, a serum sample was extracted with aqueous saturated ammonium sulfate, ethanol, and hexane.[6] The hexane extract was washed with deionised water and dried, and the lipid was measured gravimetrically. Next, the lipid was decomposed in KOH water/ethanol solution, and PCDDs and PCDFs were extracted with hexane.[7] The hexane extract was cleaned with a multistage column of silica gel coated with $AgNO_3$, H_2SO_4, and KOH, and an aluminium oxide column.[7] The effluent was dried and reconstituted to 10 μl with nonane

containing $^{13}C_{12}$-1,2,3,4-tetrachlorodibenzo-*p*-dioxin (TCDD). Finally, 2,3,7,8-substituted PCDDs and PCDFs were measured with a high resolution gas chromatograph (HP5890II, Hewlett Packard, USA)/high resolution mass spectrometry (JMS700, JEOL, Japan) connected with capillary columns of SP2331 (0.25 mm internal diameter×60 m, Supelco, USA) and DB5-MS (0.25 mm internal diameter×30 m, J and W, USA). The limits of measurement were 1 pg/g lipid for TCDD, pentachlorodibenzo-*p*-dioxin (PeCDD), hexachlorodibenzo-*p*-dioxin (HxCDD), heptachlorodibenzo-*p*-dioxin (HpCDD), tetrachlorodibenzofuran (TCDF), pentachlorodibenzofuran (PeCDF), hexachlorodibenzofuran (HxCDF) and HpCDF, and 2 pg/g lipid for octachlorodibenzo-*p*-dioxin (OCDD) and octachlorodibenzofuran (OCDF). Half of the limit value was assigned as an undetectable value. The TEQ value was calculated as directed by the World Health Organisation (WHO, 1997).[8]

Dust

In each incineration plant, to assess the mean concentration of PCDDs and PCDFs in deposited dust, five dust samples were taken from five beams at height of about 1 m, and one analyzed sample was made by mixing them equally. Fine particles are potentially inhalable, because they can be easily scattered by disturbance and remain suspended in air for a long time, whereas coarse particles cannot. The dust sample was passed through a sieve of 32 μm, and the concentrations of PCDDs and PCDFs were measured as described in our previous paper. Briefly, the sample was first treated with HCl for better extraction efficiency, and the solid content was separated with a filter and then dried at room temperature. The PCDDs and PCDFs in the liquid passing through the filter were extracted with toluene, and those in the solid were Soxhlet extracted with toluene. The two toluene extracts were combined and washed with deionised water, and the internal standard was added. Procedures of purification and quantitative measurement were almost the same as for the analysis of serum samples.

Statistical analysis

Differences of serum concentrations of PCDDs and PCDFs between the incinerator workers and controls were examined by the Mann-Whitney *U* test, because many non-detectable cases were found for some PCDDs and PCDFs.

For HxCDD, PeCDF, HxCDF and HpCDF, the numbers of non-detectable cases decreased by summing 2,3,7,8-substrated isomers, so that these values were used in the following relation analyses. The occupational exposure index of each constituent of PCDDs and PCDFs was defined as the product of the duration of employment at the incineration plant and the concentration of the constituent in the deposited dust. The dietary index was defined as the sum of numbers of meals of fish, meat, and cow's milk (times/week). The smoking index was defined as the product of smoking degree (cigarettes/day) and smoking duration (years). The relations between logarithms of serum concentrations of PCDDs and PCDFs and the seven factors of area, age, occupational exposure index, dietary index, smoking index, body mass index (BMI), and previous job with potential exposure to PCDDs and PCDFs were tested, first in a univariate way and subsequently with a multiple linear regression model. For the univariate analyses, analysis of variance (ANOVA) was used to test differences among the three areas, and the *t* test was performed to test differences between two groups classified by other factors. To reduce the number of variables in the multivariate model, only those prognostic factors that were related to the serum PCDD/PCDF concentration in the univariate analysis ($p<0.10$) were included in the model. In the multivariate analyses, two dummy variables, II/I (area II: 1, other areas: 0) and III/I (area III: 1, other areas: 0), were used for area. Data analyses were performed with the STATISTICA system (StatSoft, USA).

RESULTS

Table 1 shows the workers' previous jobs with potential for exposure to PCDDs and PCDFs, diet, current smoking, and BMI for the incinerator workers and controls. Four of the incinerator workers and six of the controls had engaged in agriculture. Because some herbicides contained PCDDs and PCDFs as impurities,[9] they may have been exposed to these chemicals. However, they did not remember the name of the herbicides used, which made it difficult to clarify whether they had been exposed to PCDDs and PCDFs in their previous jobs. The smoking index was the only factor that was significantly different between the two groups in area I.

Table 2 shows the concentrations of 2,3,7,8-substituted PCDDs and PCDFs in the deposited dust. The TEQ values of summed PCDDs and PCDFs were 0.91, 33, and 11 ng TEQ/g, respectively, for incineration plants I, II, and III. Dominant constituents were OCDD and 1,2,3,4,6,7,8-HpCDD for PCDDs, and 1,2,3,4,6,7,8-HpCDF, and OCDF for PCDFs.

Tables 3, 4, and 5 show the concentrations of summed PCDDs and PCDFs in the serum samples. The numbers of non-detectable cases were six (one, three, and two, respectively, for areas I, II, and III), 15 (six, eight, and one), 16 (seven, three, and six), 35 (14, nine, and 12), 30 (11, seven, and 12) and 37 (14, 11, and 12) for 2,3,7,8-TCDD, 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,7,8,9-HpCDF, and OCDF.

The means of TEQs of summed PCDDs and PCDFs for the incinerator workers and controls were 22.8 and 16.4 pg TEQ/g lipid, respectively, for area I, 29.4 and 19.3 pg TEQ/g lipid for area II, and 22.8 and 24.9 pg TEQ/g lipid for area III. No significant differences in the TEQ of PCDDs and in the TEQ of summed PCDDs and PCDFs were found between the incinerator workers and the controls, but the TEQ of PCDFs in the incinerator workers was significantly higher than that in the controls in areas I and II.

In area I, 2,3,7,8-TCDD, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF and 1,2,3,4,6,7,8-HpCDF were significantly higher among the incinerator workers than the controls. In

Table 2 Concentrations of PCDDs and PCDFs in the deposited dust on beams in the incineration plants (ng/g)

	I	II	III
2,3,7,8-TCDD	0.02	0.01	0.38
1,2,3,7,8-PeCDD	0.11	1.8	3.2
1,2,3,4,7,8-HxCDD	0.14	5.3	4.2
1,2,3,6,7,8-HxCDD	0.26	53	11
1,2,3,7,8,9-HxCDD	0.17	31	8.5
1,2,3,4,6,7,8-HpCDD	1.9	790	73
OCDD	3.4	1800	150
2,3,7,8-TCDF	0.14	0.72	0.65
1,2,3,7,8-PeCDF	0.58	5.0	2.0
2,3,4,7,8-PeCDF	0.57	7.4	2.5
1,2,3,4,7,8-HxCDF	0.87	13	4.4
1,2,3,6,7,8-HxCDF	0.80	17	4.9
1,2,3,7,8,9-HxCDF	0.11	2.4	1.8
2,3,4,6,7,8-HxCDF	1.6	54	9.2
1,2,3,4,6,7,8-HpCDF	3.4	120	32
1,2,3,4,7,8,9-HpCDF	0.85	45	4.4
OCDF	3.0	210	25
TEQ-PCDD	0.20	19	6.7
TEQ-PCDF	0.71	14	3.8
TEQ-PCDD/PCDF	0.91	33	11

The dust samples were analyzed after being passed through a sieve of 32 μm.

Table 3 PCDDs and PCDFs (pg/g lipid) in serum samples of incinerator workers and controls in area I

	Incinerator workers (n=7)			Controls (n=7)		
	Mean	Median	(Range)	Mean	Median	(Range)
2,3,7,8-TCDD	1.8	1.6*	(1.2–3.2)	1.1	1.1	(ND–1.5)
1,2,3,7,8-PeCDD	8.4	7.7	(4.4–15.9)	6.1	6.9	(4.2–8.2)
1,2,3,4,7,8-HxCDD	3.1	3.3	(1.7–4.6)	2.8	2.3	(1.8–4.9)
1,2,3,6,7,8-HxCDD	19.1	19.5	(10.8–26.4)	18.4	18.3	(10.7–27.3)
1,2,3,7,8,9-HxCDD	4.0	4.0	(2.8–5.8)	4.1	3.4	(2.2–9.8)
1,2,3,4,6,7,8-HpCDD	31.5	14.7	(8.9–126)	31.1	22.9	(16.6–92.4)
OCDD	365	137	(79.6–1670)	344	170	(81.2–1460)
2,3,7,8-TCDF	1.4	1.3	(ND–2.7)	0.9	0.5	(ND–1.8)
1,2,3,7,8-PeCDF	0.9	1.0	(ND–1.9)	1.0	0.5	(ND–1.8)
2,3,4,7,8-PeCDF	15.5	15.3*	(8.8–26.8)	9.8	9.1	(6.5–13.4)
1,2,3,4,7,8-HxCDF	6.2	6.1*	(3.5–8.9)	4.1	3.5	(2.3–8.6)
1,2,3,6,7,8-HxCDF	7.0	6.7*	(5.1–8.9)	4.6	4.6	(2.2–10.1)
1,2,3,7,8,9-HxCDF	0.5	0.5	(ND–ND)	0.5	0.5	(ND–ND)
2,3,4,6,7,8-HxCDF	2.9	2.9	(2.1–3.8)	2.6	2.3	(1.3–4.7)
1,2,3,4,6,7,8-HpCDF	7.5	7.5*	(6.0–8.9)	4.8	3.7	(2.9–9.7)
1,2,3,4,7,8,9-HpCDF	0.8	0.5	(ND–1.8)	0.7	0.5	(ND–1.7)
OCDF	1.0	1.0	(ND–ND)	1.0	1.0	(ND–ND)
TEQ-PCDD	13.1	12.5	(7.3–22.8)	10.1	10.8	(6.9–15.0)
TEQ-PCDF	9.7	9.6*	(5.7–16.0)	6.3	6.0	(4.0–9.3)
TEQ-PCDD/PCDF	22.8	21.3	(13.1–38.8)	16.4	17.2	(10.9–24.3)

*p<0.05, Mann-Whitney *U* test.

area II, 1,2,3,7,8-PeCDF, 1,2,3,4,7,8-HxCDF 1,2,3,6,7,8-HxCDF 1,2,3,7,8,9-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, and OCDF were higher among the incinerator workers. In area III, 1,2,3,4,6,7,8-HpCDF was higher among the incinerator workers. Thus, 1,2,3,4,6,7,8-HpCDF was significantly higher among the incinerator workers than the controls in all three areas.

From the univariate analysis, the five variables of area, age, occupational exposure index, smoking index, and BMI were selected for multivariate analysis (table 6). Table 7 shows the square of adjusted multiple correlation (R^2) and standardised slope (β) estimated by the multiple linear regression analysis. The β value is not interpretable in the unit of the variable, but in a standardised unit. Multivariate analysis suggested that serum concentrations of HxCDF, HpCDF, and TEQ of PCDFs increased with the occupational exposure index ($p<0.01$ or $p<0.001$). The other significant variables ($p<0.01$ or $p<0.001$) were area for HxCDD, age for TCDD, PeCDD, PeCDF, TEQ of PCDDs, TEQ of PCDFs, and TEQ of summed PCDDs and PCDFs, and BMI for HxCDD, HpCDD, and OCDD.

DISCUSSION

The incinerators studied were the intermittently burning type, which generate larger amounts of PCDDs and PCDFs than the continuously burning type.[2] In 1996, the concentrations of summed PCDDs and PCDFs in the exhaust gas were 590 and

Table 4 PCDDs and PCDFs (pg/g lipid) in serum samples of incinerator workers and controls in area II

	Incinerator workers (n=7)			Controls (n=7)		
	Mean	Median	(Range)	Mean	Median	(Range)
2,3,7,8-TCDD	1.4	1.6	(ND–2.5)	1.5	1.4	(ND–2.5)
1,2,3,7,8-PeCDD	8.1	8.5	(3.9–13.0)	6.7	7.2	(3.5–10.4)
1,2,3,4,7,8-HxCDD	4.5	4.2	(1.5–8.4)	3.0	2.8	(1.9–4.5)
1,2,3,6,7,8-HxCDD	24.8	22.1	(14.0–38.7)	24.2	22.4	(15.9–39.4)
1,2,3,7,8,9-HxCDD	6.7	6.3	(2.3–12.2)	5.2	5.2	(2.5–9.7)
1,2,3,4,6,7,8-HpCDD	46.2	32.7	(13.0–118)	18.2	19.9	(10.4–27.5)
OCDD	249	148	(74.5–890)	298	193	(102–962)
2,3,7,8-TCDF	1.7	0.5	(ND–4.8)	0.7	0.5	(ND–1.1)
1,2,3,7,8-PeCDF	2.6	2.1**	(1.3–5.4)	0.9	1.0	(ND–1.7)
2,3,4,7,8-PeCDF	20.7	19.5	(10.5–44.3)	13.5	15.2	(7.9–22.1)
1,2,3,4,7,8-HxCDF	13.4	13.5**	(6.9–25.2)	5.0	4.5	(2.0–9.2)
1,2,3,6,7,8-HxCDF	22.5	23.2**	(11.3–41.7)	7.7	6.7	(4.9–11.8)
1,2,3,7,8,9-HxCDF	1.5	1.7**	(ND–2.5)	0.5	0.5	(ND–ND)
2,3,4,6,7,8-HxCDF	21.8	20.2**	(4.9–58.6)	3.2	3.0	(2.3–4.0)
1,2,3,4,6,7,8-HpCDF	50.0	48.7**	(17.1–94.0)	6.2	6.5	(3.6–7.7)
1,2,3,4,7,8,9-HpCDF	4.7	3.7**	(2.0–10.5)	0.5	0.5	(ND–ND)
OCDF	2.6	1.0	(ND–9.6)	1.0	1.0	(ND–ND)
TEQ-PCDD	13.6	14.8	(6.5–21.3)	11.6	12.2	(6.4–16.7)
TEQ-PCDF	17.1	15.9*	(8.4–36.3)	8.6	8.8	(5.2–13.6)
TEQ-PCDD/PCDF	30.7	29.1	(14.9–57.6)	20.2	21.1	(12.1–30.3)

*p<0.05; **p<0.01, Mann-Whitney *U* test.

Table 5 PCDDs and PCDFs (pg/g lipid) in serum samples of incinerator workers and controls in area III

	Incinerator workers (n=6)			Controls (n=6)		
	Mean	Median	(Range)	Mean	Median	(Range)
2,3,7,8-TCDD	1.5	1.4	(ND–2.8)	1.8	1.7	(1.2–2.3)
1,2,3,7,8-PeCDD	8.1	8.1	(3.5–13.3)	8.5	8.3	(7.1–11.2)
1,2,3,4,7,8-HxCDD	4.3	4.4	(2.6–6.9)	3.7	3.8	(1.6–5.1)
1,2,3,6,7,8-HxCDD	24.4	25.9	(11.8–30.9)	25.5	26.1	(18.2–31.5)
1,2,3,7,8,9-HxCDD	6.6	6.4	(5.2–8.4)	4.7	4.4	(2.8–7.5)
1,2,3,4,6,7,8-HpCDD	24.3	25.6	(8.5–34.6)	20.2	21.5	(9.0–26.6)
OCDD	253	262	(139–351)	248	209	(161–430)
2,3,7,8-TCDF	1.9	1.2	(ND–3.7)	2.2	1.8	(1.2–4.7)
1,2,3,7,8-PeCDF	1.5	1.2	(ND–3.0)	1.8	0.5	(ND–6.6)
2,3,4,7,8-PeCDF	14.3	13.7	(6.8–23.2)	17.1	14.2	(12.1–27.4)
1,2,3,4,7,8-HxCDF	5.4	5.3	(2.3–8.4)	6.1	5.4	(4.1–10.8)
1,2,3,6,7,8-HxCDF	7.8	8.0	(5.0–11.2)	8.3	7.5	(4.5–14.6)
1,2,3,7,8,9-HxCDF	0.5	0.5	(ND–ND)	0.5	0.5	(ND–ND)
2,3,4,6,7,8-HxCDF	4.5	4.6	(2.4–6.8)	5.7	4.5	(2.4–11.5)
1,2,3,4,6,7,8-HpCDF	12.0	10.2*	(7.8–18.7)	7.1	5.1	(4.9–15.1)
1,2,3,4,7,8,9-HpCDF	0.5	0.5	(ND–ND)	0.5	0.5	(ND–ND)
OCDF	1.0	1.0	(ND–ND)	1.0	1.0	(ND–ND)
TEQ-PCDD	13.4	13.8	(6.3–20.9)	13.9	13.4	(11.6–18.1)
TEQ-PCDF	9.4	9.2	(4.7–14.9)	11.0	8.8	(8.1–18.4)
TEQ-PCDD/PCDF	22.8	22.9	(11.1–35.8)	24.9	22.8	(20.0–31.6)

*p<0.05 by Mann-Whitney *U* test.

260 ng TEQ/Nm^3, respectively, for plants I and II, and the values were very high among municipal waste incinerators in Japan.[2] The concentration of summed PCDDs and PCDFs in the exhaust gas was 53 ng TEQ/Nm^3 for plant III in 1996, and this value was about medium in Japan.[2] Thereafter, because the equipment was improved in all plants, the concentration decreased to 0.22–0.52, 11–20, and 11–32 ng TEQ/Nm^3, respectively, for plants I, II, and III in 1997–2000. However, this study shows that the concentrations of summed PCDDs and PCDFs in the deposited dust were 0.91, 33, and 11 ng TEQ/g, respectively, which were about 1000 times as high as those in general soil.[10]

Work at the incineration plant was classified into the usual operation of the incinerator and the periodic cleaning of the inside of the equipment, including the incinerator, cooling tower, electrostatic precipitator, and bag filter. The usual operation consists of waste weighing, crane operation, incinerator operation, carrying of slag and fly ash, and daily inspection and maintenance. Because the incinerator workers did not wear any respiratory protection during the usual operation, they inhaled the suspended dust in the plants. In the periodic cleaning, because a large amount of ash and slag was scattered in the air due to the sweeping process, the workers may have been exposed to high concentrations of PCDDs and PCDFs. However, the periodic cleaning and repair of the incinerator, electrostatic precipitator, and bag filter for the three plants were done by workers from other companies. The periodic cleaning of the cooling tower was done by the cleaning workers for plant III, but by the incinerator workers for plants I (once a month) and II (once a week). Although the incinerator workers wore dust masks during the cleaning work, they probably inhaled some dust due to leaks in the dust masks, because they complained of nostril pollution after the cleaning work. When the hands and skin of workers are contaminated with the deposited dust, hand to mouth ingestion and dermal absorption of PCDDs and PCDFs can occur.

However, the mean TEQ values of summed PCDDs and PCDFs for the incinerator workers were 23 to 29 pg TEQ/g lipid, which were not significantly higher than the controls. Because the mean TEQ values of summed PCDDs and PCDFs in the general population have been reported to be 16–29 pg TEQ/g lipid

Table 6 Significance of seven variables by univariate analyses (p value)

	Area	Age	Occupational exposure index	Dietary index	Smoking index	BMI	Agriculture
TCDD	0.791	<0.001***	0.879	0.999	0.310	0.711	0.982
PeCDD	0.516	<0.001***	0.265	0.987	0.976	0.934	0.999
HxCDD	0.033*	0.115	0.604	0.266	0.872	0.065†	0.183
HpCDD	0.773	0.028*	0.322	0.850	0.022*	0.026*	0.178
OCDD	0.733	0.033*	0.703	0.685	0.417	0.061†	0.131
TCDF	0.035*	0.088†	0.363	0.506	0.029*	0.667	0.754
PeCDF	0.205	0.017*	0.089†	0.531	0.297	0.406	0.472
HxCDF	0.003**	0.499	0.005**	0.858	0.122	0.821	0.991
HpCDF	0.003**	0.890	<0.001***	0.988	0.546	0.971	0.614
OCDF	0.106	0.131	0.131	0.655	0.844	0.618	0.913
TEQ-PCDD	0.450	0.001***	0.330	0.793	0.777	0.548	0.686
TEQ-PCDF	0.062†	0.044*	0.026*	0.680	0.268	0.583	0.553
TEQ-PCDD/PCDF	0.232	0.005**	0.086†	0.879	0.439	0.962	0.902

*p<0.05; **p<0.01; ***p<0.001; †p<0.10. Area I, II, and II; age 24–42 y (n=20) and 43–59 y (n=20); occupational exposure index 0 y.ng/g (n=20) and >0 y.ng/g (n=20); dietary index ≤18 times/week (n=23) and >18 times/week (n=17); BMI <24 kg/m^2 (n=24) and ≥24 kg/m^2 (n=16); agriculture yes (n=10) and no (n=30).

Table 7 Multiple correlation coefficients (R) and estimated standardised slope (β) by multiple regression analysis

		Standardised slope (β) (95% CI)					
		Area					
	Adjusted R^2	II/I	III/I	Age	Occupational exposure index	Smoking index	BMI
TCDD	0.357	0.07 (−0.27 to 0.42)	0.26 (−0.18 to 0.69)	0.59 (0.29 to 0.88)***	−0.02 (−0.38 to 0.33)	0.07 (−0.25 to 0.40)	0.16 (−0.14 to 0.47)
PeCDD	0.344	0.18 (−0.18 to 0.53)	0.44 (0.01 to 0.86)*	0.51 (0.21 to 0.81)**	−0.02 (−0.36 to 0.32)	0.22 (−0.11 to 0.54)	0.19 (−0.11 to 0.49)
HxCDD	0.388	0.50 (0.11 to 0.88)*	0.63 (0.26 to 0.99)**	0.24 (−0.04 to 0.53)	0.06 (−0.27 to 0.39)	0.23 (−0.09 to 0.55)	0.41 (0.12 to 0.70)**
HpCDD	0.425	−0.13 (−0.50 to 0.25)	−0.05 (−0.40 to 0.29)	0.20 (−0.08 to 0.47)	0.45 (0.12 to 0.78)*	−0.12 (−0.43 to 0.18)	0.55 (0.27 to 0.84)***
OCDD	0.330	0.15 (−0.25 to 0.56)	0.29 (−0.08 to 0.67)	0.30 (0.00 to 0.60)*	−0.05 (−0.42 to 031)	0.10 (−0.23 to 0.43)	0.51 (0.21 to 0.82)**
TCDF	0.367	−0.14 (−0.50 to 0.22)	0.23 (−0.17 to 0.63)	0.36 (0.07 to 0.66)*	0.10 (−0.21 to 0.42)	−0.33 (−0.65 to 0.00)*	0.04 (−0.25 to 0.33)
PeCDF	0.449	0.25 (−0.11 to 0.60)	0.29 (−0.06 to 0.64)	0.51 (0.24 to 0.78)***	0.31 (0.01 to 0.61)*	0.06 (−0.24 to 0.36)	0.04 (−0.24 to 0.31)
HxCDF	0.598	0.30 (−0.01 to 0.61)	0.20 (−0.10 to 0.50)	0.16 (−0.08 to 0.39)	0.62 (0.36 to 0.89)***	0.05 (−0.21 to 0.30)	0.14 (−0.09 to 0.38)
HpCDF	0.722	0.19 (−0.07 to 0.45)	0.16 (−0.09 to 0.40)	−0.02 (−0.21 to 0.18)	0.78 (0.55 to 1.00)***	0.18 (−0.03 to 0.39)	0.13 (−0.07 to 0.33)
OCDF	0.428	0.26 (−0.12 to 0.63)	0.22 (−0.13 to 0.56)	0.26 (−0.02 to 0.53)	0.42 (0.09 to 0.75)*	0.36 (0.05 to 0.66)*	0.23 (−0.05 to 0.51)
TEQ-PCDD	0.364	0.22 (−0.17 to 0.61)	0.45 (0.07 to 0.83)*	0.49 (0.19 to 0.78)**	0.07 (−0.26 to 0.40)	0.21 (−0.11 to 0.53)	0.29 (−0.01 to 0.58)
TEQ-PCDF	0.501	0.28 (−0.07 to 0.62)	0.30 (−0.03 to 0.63)	0.43 (0.17 to 0.69)**	0.43 (0.14 to 0.73)**	0.09 (−0.20 to 0.37)	0.08 (−0.19 to 0.34)
TEQ-PCDD/PCDF	0.413	0.26 (−0.11 to 0.63)	0.37 (0.01 to 0.73)*	0.48 (0.20 to 0.76)**	0.28 (−0.03 to 0.60)	0.15 (−0.16 to 0.45)	0.17 (−0.11 to 0.46)

*p<0.05; **p<0.01; ***p<0.001. II/I (area II=1, others areas=0) and III/I (area III=1, other areas=0) are dummy variables.

for Japan,[3 11–14] 14 to 43 pg TEQ/g lipid for Europe,[15–23] and 19–27 pg TEQ/g lipid for North America,[24–27] those values in the incinerator workers were almost the same as those for the general population in the industrialised countries. This agrees with our previous findings for three municipal continuously burning incineration plants in Japan.[3] Schecter *et al* also found that the mean TEQ values of summed PCDDs and PCDFs in workers employed in two German incineration plants were not higher in controls.[23] In the general population, the dietary intake of PCDDs and PCDFs predominates over other routes and differs greatly among people.[28 29] Probably because of such confounding factors, the occupational exposure at these plants did not lead to a detectable increase in the TEQs of summed PCDDs and PCDFs in serum samples.

However, this study found a significant increase in the serum concentration of 1,2,3,4,6,7,8-HpCDF for all three plants. In the previous study,[3] we obtained the same result in the three continuously burning incineration plants. Schecter *et al*[23] reported that the increase in the serum concentration of HpCDF was most marked at an old incineration plant. HpCDF was one of the dominant constituents among 2,3,7,8-subtituted PCDFs in the deposited dust. Consequently, we concluded that the increase in serum concentration of HpCDF in incinerator workers was caused by the inhalation of the dust during work. This conclusion was confirmed by the multiple regression analysis that suggested that serum concentration of HpCDF increased with the occupational exposure index after the effects of the confounding factors were eliminated.

The multiple regression analyses also found that serum concentrations of HxCDF increased with the occupational exposure index in the present study, and that serum concentrations of OCDF increased in our previous study.[3] Schecter *et al*[23] found an increase in the serum concentrations of HxCDF in incinerator workers. On the other hand, the Ministry of Labor found that the serum concentrations of TCDF and PeCDF as well as HxCDF, HpCDF, and OCDF increased among workers employed at the Toyono incineration plant, where the concentrations of PCDDs and PCDFs in the suspended dust were probably extremely high because the concentration in the deposited dust in the toxic gas scrubber was 96 000 ng TEQ/g.[4 5] These findings suggest that the serum concentrations of HxCDF, HpCDF, and OCDF increase at incineration plants with low contamination, and serum concentrations of all furans increase at incineration plants with high contamination. This is reasonable, because concentrations of HpCDF and OCDF in the deposited dust were the highest among 2,3,7,8-substitued PCDFs, and then HxCDF, PeCDF, and TCDF in that order (table 2).

The OCDD and HpCDD were dominant constituents among 2,3,7,8-substitued PCDDs in the deposited dust, but serum concentrations of these dioxins were not significantly higher among the incinerator workers in all three areas. Because variability between individual serum concentrations of these dioxins were much higher than those of HpCDF in the controls (tables 3, 4, and 5), the increases of these dioxins due to occupational exposure would be more difficult to detect because of other confounding factors at incineration plants with low contamination. On the other hand, serum concentrations of TeCDD to OCDD were increased at the Toyono incineration plant,[4 5] which suggests that the increases of all 2,3,7,8-substituted PCDDs in serum samples can be detected at incineration plants with high contamination.

The multiple regression analysis also suggested that the positive association between age and serum concentrations of TCDD, PeCDD, PeCDF, TEQ of PCDDs, TEQ of PCDFs, and TEQ of summed PCDDs and PCDFs, and between BMI and serum concentrations of HxCDD, HpCDD, and OCDD. This agrees with previous knowledge.[17 27 30] This analysis showed that the area was a factor affecting serum concentrations of HxCDD. As the importance of area is not known in Japan, further study is necessary. Ten of the workers were previously engaged in agriculture, but the concentrations of PCDDs and PCDFs in their

Main messages

- Serum TEQs of summed PCDDs and PCDFs in the workers employed at three intermittently burning municipal waste incineration plants were almost the same as in the general population of the industrialised countries.
- Concentration of 1,2,3,4,6,7 8-HpCDF in serum samples of workers increased at all the incinerator plants. The multiple regression analysis showed that the serum concentrations of HxCDF and HpCDF significantly increased with the occupational exposure index.
- These findings suggest that incinerator workers employed at intermittently burning municipal incineration plants in Japan were not necessarily exposed to high concentrations of PCDDs and PCDFs, but that they had inhaled dust containing PCDDs and PCDFs during their work.

Policy implications

- The result of the present study can lessen anxiety about body burden of PCDDs and PCDFs among incinerator workers.
- The result recommends that the incinerator workers take measures to decrease exposure to PCDDs and PCDFs during work.

serum samples were not higher than those of the other workers (table 6). This suggests that they had not used herbicides containing PCDDs and PCDFs or if they had used them, the exposure was low.

CONCLUSION

The TEQs of summed PCDDs and PCDFs in serum samples in all of the incinerator workers and controls were almost the same as in the general population of the industrialised countries. Comparison between the incinerator workers and controls did not find differences in the TEQs of summed PCDDs and PCDFs in serum samples, but the TEQs of PCDFs for the two areas and 1,2,3,4,6,7,8-HpCDF for all three areas had increased among the incinerator workers. The multiple regression analysis showed that the serum concentrations of HxCDF and HpCDF significantly increased with the occupational exposure index. These findings suggest that incinerator workers employed at intermittently burning municipal incineration plants of Japan were not necessarily exposed to high levels of PCDDs and PCDFs, but that they had inhaled dust containing PCDDs and PCDFs during their work.

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