

BACKGROUND DIOXIN LEVELS IN THE BLOOD OF RESIDENTS IN THE VICINITY OF A MUNICIPAL WASTE INCINERATOR IN SOUTHERN TAIWAN

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Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are among the most important toxic substances in the environment and ubiquitously distributed. They are unwanted by-products in a variety of industrial and thermal processes. Daily consumption of contaminated food is the main route of environmental exposure to PCDDs/PCDFs. Food consumption normally comprises 90% of the total daily intake of dioxins. The lipophilic PCDDs/PCDFs accumulate in human tissue in their parent forms rather than as corresponding metabolites. Dioxins have long half-lives of elimination in human tissue. Ryan and his coworkers reported half-lives of two to three years at the time of exposure for Yusho and Yu-Cheng patients, and of approximately ten years when the levels were lowered¹. Measurement of PCDD/PCDF levels in human tissue is useful to assess long-term dioxin exposure. The PCDD/PCDF concentrations, based on lipid weight, in human adipose tissue, blood, and milk are very similar².

Concerns over contamination from dioxins from waste incinerators and other sources have risen in recent years in Taiwan. We report here the blood level of PCDDs and PCDFs of fifty residents in the vicinity of a municipal waste incinerator before its operation in southern Taiwan. The concentrations were determined using high resolution gas chromatography high resolution mass spectrometry (HRGC-HRMS) combined with isotope dilution method. Results derived from this study are intended to estimate current background human exposure to PCDDs/PCDFs in the area and set a baseline for future monitoring.

Methods and Materials

Study Subjects and Serum Collection

The study was carried out in Tainan City of approximately 720,000 residents in southern Taiwan. An appeal with request to participate in the study was announced in 1997 to the residents in the vicinity of a municipal waste incinerator in Tainan City. The blood samples of 81 participants, 30 men and 51 women, were drawn in August and November of 1997, before the incinerator started its operation in August 1999. PCDD/PCDF levels of 50 blood samples were reported in this study. Some blood samples were not analyzed due to insufficient amount of specimen. Some results of analysis were not reported if the analytical data did not meet with quality control standards. A questionnaire was used to identify personal data, previous work experiences, and life habits of the participants.

Each study participant provided 50 mL of venous blood. Blood samples were drawn into 10-mL chemically clean tubes containing no anti-coagulants and allowed to coagulate followed by centrifugation to obtain serum samples. Serum samples were transferred under refrigeration to the laboratory and kept frozen at -70 °C until the time of analysis. The serum lipid contents were determined and ranged from 4.2 to 9.9 mg/mL-serum, with an average of 6.1 mg/mL-serum.

Sample Preparation and HRGC/HRMS Analysis of PCDDs and PCDFs

The sample enrichment and cleanup procedures used in this study were similar to the procedures reported by Chang³. The average of recovery rates for sample enrichment and

ORGANOHALOGEN COMPOUNDS

HUMAN EXPOSURE-POSTERS

cleanups was 64%. A Fison 8060 GC and a Micromass AutoSpec Ultima EBE tri-sector mass spectrometer were used for the HRGC/HRMS analysis. Samples were chromatographed on an 60-m Rtx-5MS capillary fused-silica column. The HRMS was operated in electron impact ionization mode. Isotope dilution HRGC/HRMS method was used to determine quantitatively the seventeen PCDD/PCDF congeners. Selected ion monitoring (SIM) was used to acquire $M/(M+2)$ or $(M+2)/(M+4)$ PCDD/PCDF ions for identification. Quality assurance/quality control protocols were established, according to those defined in USEPA method 1613, in our laboratory to ensure positive identification and the quality of the measurements.

Results and Discussion

The emission of PCDDs/PCDFs from waste incinerators may cause possible health hazard to nearby residents has been of great public concern. In the present study, the concentrations of PCDDs/PCDFs in the blood of fifty residents in the vicinity of the municipal waste incinerator in Tainan City, which is located in southern Taiwan, before its operation were determined. The information obtained from the questionnaire did not indicate any known occupational exposure to dioxins prior to the study. The serum levels of seventeen toxic 2,3,7,8-substituted PCDD/PCDF congeners of the 50 participants are tabulated in Table 1. The values are expressed on a lipid-adjusted basis.

The average total level of seventeen congeners ranges from 480 to 5930 pg/g-lipid with an average of 1620 pg/g-lipid. These concentrations can be regarded as an indicator of background human exposure to PCDDs/PCDFs in the area. To our knowledge, the measurements of PCDDs/PCDFs in Table 1 are the first background human dioxin exposure data in Taiwan with sufficiently large number of study subjects. The toxicity equivalent values (TEQ) were also calculated, according to the international toxic equivalency factors (TEFs), to give an average of 47.3 pg-TEQ/g-lipid with a range from 13.5 to 104 pg-TEQ/g-lipid. Figure 1 shows the distribution pattern of seventeen PCDD/PCDF congeners in this study ($n = 50$), expressed in mass and in TEQ. The patterns are very similar to those described in literature for background dioxin exposure. Three congeners, 2,3,4,7,8-PeCDF, 2,3,7,8-TCDD, and 1,2,3,7,8-PeCDD, contribute most of the toxicity among seventeen congeners, representing 60 % of the total TEQ.

Among industrialized countries, dioxin blood concentrations are considered to be geographically fairly uniform and TEQs in the general population are between 20-40 pg-TEQ/g-lipid^{4,5}. Nonetheless, dioxin blood concentrations in the general population in some countries have been decreasing gradually. Papke² reported the background human PCDD/PCDF levels in Germany showed a distinct decline over an observed period of ten years, from 1986 to 1996. The mean TEQ value was 42 pg-TEQ/g-lipid in 1988 and decreased to roughly 10 pg-TEQ/g-lipid in 1996. Schecter⁶ reported that the mean blood level of 2,3,7,8-TCDD for the U.S. general population was approximately 3.5 pg-TEQ/g-lipid in 1996. Tainan is a mixed residential-industrial city in southern Taiwan. The observation suggests there could have been some possible industrial sources of dioxin exposure in the area.

One of the goals for this study is to utilize the data in Table 1 to set a current baseline of human exposure in the area for future monitoring of dioxins. The incinerator has been in operation since August 1999. The blood PCDD/PCDF levels of the residents can be monitored in later years. If elevated levels should occur, possible exposure due to emissions originating from the incinerator may be indicated.

HUMAN EXPOSURE-POSTERS

Table 1. Serum PCDD/PCDF concentrations (pg/g-lipid)

PCDD/PCDF Congeners	Male (n=16)		Female (n=34)		All (n=50)
	Range	Mean ± SD	Range	Mean ± SD	Mean ± SD
2,3,7,8-TCDF	8.8-57.1	39.7±22.3	ND-132	42.9±28.8	41.9±26.8
1,2,3,7,8-PeCDF	ND-37.4	10.9±13.4	ND-442	21.5±77.1	17.2±62.1
2,3,4,7,8-PeCDF	9.4-62.9	29.2±16.8	5.8-51.9	22.3±12.3	24.5±14.1
1,2,3,4,7,8-HxCDF	8.6-48.7	23.5±12.5	5.0-64.8	17.2±10.7	19.2±11.6
1,2,3,6,7,8-HxCDF	7.5-42.4	18.4±9.6	3.7-46.6	14.2±8.4	14.8±9.3
2,3,4,6,7,8-HxCDF	4.8-38.2	16.5±11.4	3.7-39.8	13.5±8.8	13.6±10.0
1,2,3,7,8,9-HxCDF	ND-41.5	14.9±14.6	ND-45.9	12.7±10.7	12.7±12.1
1,2,3,4,6,7,8-HpCDF	11.0-180	58.4±50.3	12.1-353	49.0±60.1	52.0±56.8
1,2,3,4,7,8,9-HpCDF	2.7-60.4	18.9±18.7	ND-104	16.7±19.6	17.4±19.2
OCDF	24.8-2750	295±683	ND-515	147±124	198±401
2,3,7,8-TCDD	2.7-16.0	8.1±4.6	ND-45.8	10.0±9.2	8.8±8.1
1,2,3,7,8-PeCDD	6.0-42.2	16.9±10.7	ND-36.4	13.5±8.6	14.3±9.5
1,2,3,4,7,8-HxCDD	ND-28.1	11.9±9.5	ND-31.1	10.6±7.1	9.8±8.2
1,2,3,6,7,8-HxCDD	15.4-72.5	29.1±15.0	7.2-45.5	24.9±10.2	24.6±13.7
1,2,3,7,8,9-HxCDD	7.5-50.6	21.5±15.4	2.7-43.3	16.6±9.7	18.2±11.9
1,2,3,4,6,7,8-HpCDD	27.5-177	92.6±41.5	29.4-344	93.6±68.1	93.3±60.4
OCDD	478-5000	1280±1160	111-5120	935±834	1070±950
Total PCDF	145-3000	525±709	121-1160	357±236	441±445
Total PCDD	573-5240	1460±1180	225-5550	1100±890	1230±993
Total PCDD/PCDF	719-5680	1980±1560	483-5930	1460±975	1670±1200
Total TEQ	19.7-104	51.4±26.1	13.5-94.5	45.7±20.6	47.3±22.4

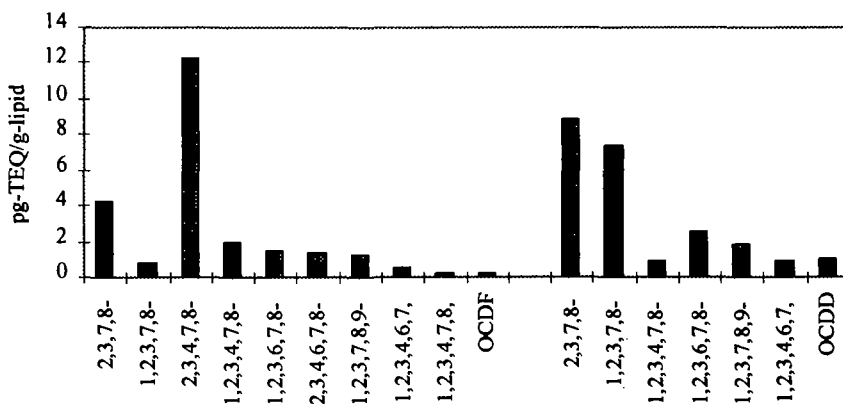


Figure 1. Distribution pattern of seventeen toxic 2,3,7,8-substituted PCDD/PCDF congeners in this study (n = 50), expressed in mass (A) and in TEQ (B).

It has been reported that nursing infants lowers the body burden of dioxins in women⁷. On the other hand, some literature data suggest the difference in PCDD/PCDF levels between

ORGANOHALOGEN COMPOUNDS

HUMAN EXPOSURE-POSTERS

male and female has been considered insignificant⁴. In table 1, the average PCDD/PCDF concentration was found to be higher in males (1980 ± 1560 pg-/g-lipid, $n = 16$) than in females (1460 ± 975 pg-/g-lipid, $n = 34$), however, the difference was not significant ($\alpha = 0.05$).

Conclusions

The blood PCDD/PCDF concentrations of fifty residents in the vicinity of a municipal waste incinerator before its operation in southern Taiwan were determined by HRGC-HRMS. The average ($n=50$) total level of seventeen toxic 2,3,7,8-substituted PCDD/PCDF congeners was, on a lipid-adjusted basis, 1670 (range 483 ~ 5930) pg/g-lipid, or 47.3 (range 13.5 ~ 104) pg-TEQ/g-lipid expressed in international toxic equivalent (TEQ). Three congeners (2,3,4,7,8-PeCDF, 2,3,7,8-TCDD, and 1,2,3,7,8-PeCDD) contribute 60 % of the total toxicity among seventeen congeners. These concentrations can be regarded as an indicator of background human exposure to dioxins in the area. To our knowledge this is the first report describing such background human exposure data in Taiwan. The data indicate that the level of dioxin exposure of the study area in Taiwan is among the highest levels observed for general population in industrialized countries.

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