POLYCHLORINATED DIBENZO-*p*-DIOXINS, DIBENZOFURANS, AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS IN GENERAL POPULATION OF TAIWAN

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Abstract
Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) are widespread and persistent environment pollutants and can be found in blood, milk, and adipose tissue of human. It is important to establish background exposure data in Taiwan to provide a scientific basis for setting up dioxin monitoring and regulatory programs. The background blood PCDD/PCDF and DL-PCB levels of 18~45 years old adults in six geographical areas (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) were surveyed in Taiwan during 2001~2006. The 17 PCDD/PCDFs level in blood of 18~45 years old adults in Taiwan was 11.7 pg WHO_{98}-TEQ/g lipid on median (range: 4.95~23.4 pg WHO_{98}-TEQ/g lipid). The 12 DL-PCBs level in blood of 18~45 years old adults in Taiwan was 5.09 pg WHO_{98}-TEQ/g lipid on median (range: 1.74~14.4 pg WHO_{98}-TEQ/g lipid). The contribution of 12 DL-PCBs level to total 17 PCDD/PCDFs and 12 DL-PCBs level was 32% on median (range: 17%~49%). The blood level of 17 PCDD/PCDFs and 12 DL-PCBs was positive associations with age ($r = 0.23$, $p = 0.002$). There were statistical differences in the median blood 17 PCDD/PCDFs and 12 DL-PCBs levels associated with gender and geographical areas.

Introduction
Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) are widespread and persistent environment pollutants[1]. Epidemiologic evidence indicated that 2,3,7,8-TCDD caused various kinds of cancer and the International Agency for Research on Cancer (IARC) concluded that 2,3,7,8-TCDD is a human carcinogen[2]. It is remains unclear whether background exposure to dioxin-like compounds is associated with increased risk of health effects[3]. Therefore, it is important to establish background exposure data in Taiwan to provide a scientific basis for setting up dioxin monitoring and regulatory programs. A survey on 17 PCDD/PCDFs and 12 DL-PCBs levels in blood of 18~45 years old adults in Taiwan was carried out during 2001~2006.

Materials and Methods
All of the subjects were volunteers living in six geographical areas (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) of Taiwan. The announcement of recruitments was announced by poster or internet system in school. To be eligible for participating in this survey, volunteers should have resided in the sampling area for the previous 5 years and did not have other potential exposures of PCDD/PCDFs, such as occupational environment. There were 283 volunteers recruited in this survey. Blood samples were drawn from 283 volunteers during 2001~2006. Each participant provided 80 mL of venous blood. Blood samples were drawn to chemically clean tubes containing no anti-coagulants and allowed to coagulate. The coagulated blood was then centrifuged to obtain serum samples, which were kept frozen at –70°C until analysis. Sample containers were tested to verify that they contained no detectable levels of PCDD/PCDF and DL-PCBs contamination.
The 17 PCDD/PCDFs and 12 DL-PCBs levels in blood were measured by the Analytical Laboratory for Trace Environmental Pollutant at National Cheng Kung University (ALTEP-NCKU) in Taiwan. The isotope dilution high resolution gas chromatography-high resolution mass spectrometry (HRGC-HRMS) method was used to quantitatively determine the 17 PCDD/PCDFs and 12 DL-PCBs congeners. The analytical procedures were adopted from USEPA Method 1613B[4] and Method 1668A[5] with the minor modifications. Quality assurance/quality control (QA/QC) protocols are established, according to those defined in USEPA Method 1613B[4] and Method 1668A[5], in ALTEP-NCKU laboratory to ensure positive identification and the quality of the measurements.

Results and Discussion

The 17 PCDD/PCDFs levels in 283 adults aged from 18 to 45 years and 12 DL-PCBs levels in 181 adults aged from 18 to 45 years were measured. An assessment on the quality of analytical data indicates 272 blood samples of 17 PCDD/PCDFs levels and 180 blood samples of 12 DL-PCBs should be included to establish the background blood dioxin-like compound levels in Taiwan. The 17 PCDD/PCDFs level in blood of 18~45 years old adults living in six areas of Taiwan (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) was 11.7 pg WHO_{98}-TEQ/g lipid on median. The 12 DL-PCBs level in blood of 18~45 years old adults living in six areas of Taiwan was 5.09 pg WHO_{98}-TEQ/g lipid on median. The total 17 PCDD/PCDFs and 12 DL-PCBs level in blood of 18-45 years old adults living in six areas of Taiwan was 16.4 pg WHO_{98}-TEQ/g lipid on median. The contribution of 12 DL-PCBs level to total 17 PCDD/PCDFs and 12 DL-PCBs level was 32% on median. These values of blood dioxin levels in Taiwanes are not found to be significantly higher than those reported in other countries.

Table 1. PCDD/PCDFs and dioxin-like PCBs levels in blood in general population in Taiwan

	blood levels (unit: pg WHO_{98}-TEQ/g lipid)			
	N	Median	Mean ± SD	(range)
17 PCDD/PCDFs	270	11.7	12.2 ± 3.55	(4.92 ~ 23.4)
12 DL-PCBs	179	5.09	5.60 ± 2.56	(1.74 ~ 14.4)
17 PCDD/PCDFs + 12 DL-PCBs	173	16.4	17.6 ± 5.82	(6.74 ~ 35.8)
ratio of 12 DL-PCB to 17 PCDD/PCDFs + 12 DL-PCBs	173	32%	31% ± 7%	(17% ~ 49%)

Linear regression and correlation were performed to determine the degree of linear association (r) between 17 PCDD/PCDFs and 12 DL-PCBs levels versus age (Figure 1). The correlation coefficient (r) was 0.223 ($p = 0.002$). The blood level of 17 PCDD/PCDFs and 12 DL-PCBs was low degree of positive associations with age ($r = 0.23$, $p = 0.002$). Schuhmacher and co-workers reported 17 PCDD/PCDFs level in blood of a general population in Spain was positive associations with age ($r = 0.565$, $p < 0.01$)[6].

The blood PCDD/PCDF and DL-PCB levels in the subjects living in six geographical areas (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) of Taiwan were plotted in Figure 2. The area with the lowest levels was Hualien area, following by Hsinchu, Taichung, Kaohsiung, Taipei, and Tainan areas. Using Mann-Whitney U test, it was found that the PCDD/PCDF and DL-PCB levels in subjects living Hualien area was significantly different from those in Kaohsiung, Taipei, and Tainan areas ($p < 0.01$).

The blood PCDD/PCDF and DL-PCB levels in the male and female subjects were plotted in Figure 3. The blood PCDD/PCDF and DL-PCB levels in the female subjects were higher than male subjects.
Using Mann-Whitney U test, it was found that the levels in female subjects were significantly different from male subjects ($p = 0.003$ for PCDD/PCDFs and $p = 0.028$ for DL-PCBs).

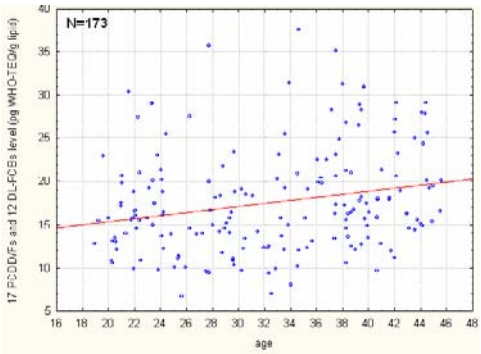

Figure 1. Relationship between age and blood levels of 17 PCDD/PCDFs and 12 DL-PCBs in adults of Taiwan

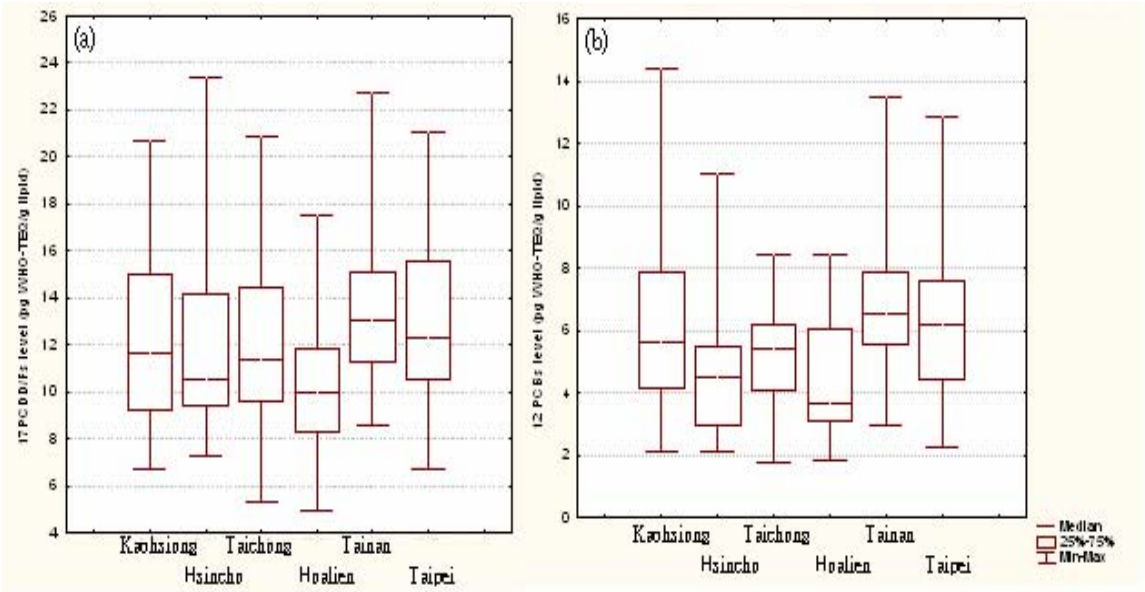

Figure 2. (a) The blood PCDD/PCDFs levels in the subjects living in six geographical areas (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) of Taiwan. (b) The blood DL-PCBs levels in the subjects living in six geographical areas (Taipei, Hsinchu, Taichung, Tainan, Kaohsiung, and Hualien) of Taiwan

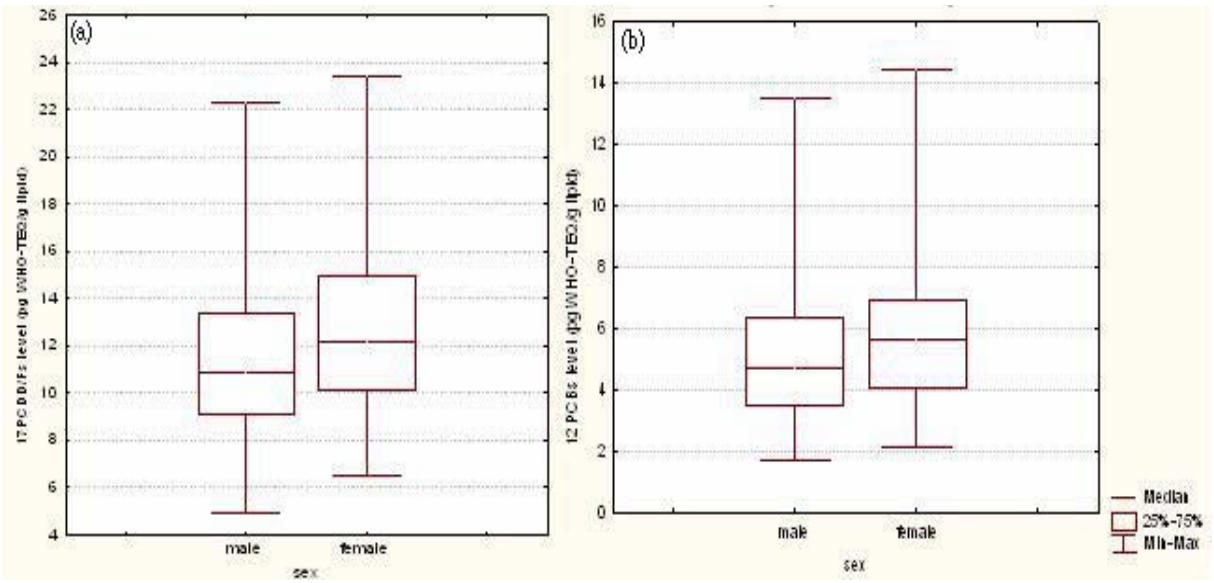

Figure 3. (a) The blood PCDD/PCDFs levels in the male and female subjects. (b) The blood DL-PCBs levels in the male and female subjects.

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