POLYCHLORINATED DIBENZO-*p*-DIOXINS, DIBENZOFURANS, AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS IN INFANT FORMULA MILK

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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 foods. Formula milk is one of the major food sources in infant period. A survey on the PCDD/PCDFs and DL-PCBs levels in formula milk was conducted. The levels of 17 PCDD/PCDFs and 12 DL-PCBs in 10 formula milk samples ranged from 0.849 to 1.23 pg WHO_{98}-TEQ/g lipid, with a mean value of 1.04 ± 0.20 pg WHO_{98}-TEQ/g lipid (mean ± S.D.). This is the first report of a global survey for 17 PCDD/PCDF and 12 DL-PCB levels in formula milk manufactured in seven countries, including America, Australia, Denmark, Germany, Netherlands, New Zealand, and Taiwan. The data are valuable for the future studies on evaluating PCDD/PCDFs and DL-PCBs exposure to infants or adults due to formula milks consumptions.

Introduction
Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) are widespread and persistent environment pollutants[1]. Although extensive efforts have been made to reduce emission of these fat-soluble compounds to environment, foods are still contaminated. Furthermore, food is the major source for PCDD/PCDFs accumulation in the human body[2]. In infant period, breast milk formula milk is one of the major food sources. However, most of the dioxin exposure in infants are focused on the levels of PCDD/PCDFs and DL-PCBs in human milk, but only a few of the study investigates the PCDD/PCDF levels in formula milk for infant[3,4]. In the present study, the 17 PCDD/PCDFs and 12 DL-PCBs levels in formula milk samples purchased from stores in southern Taiwan were reported.

Materials and Methods
Approximately, a hundred brands of formula milk can be purchased in Taiwan and 99% of the formula milk brands are imported from other countries. The information, brands of formula milk which were used to feed their infants, was obtained by telephone interviews for 118 lying-in women in southern Taiwan. The ten brands of the formula milk were selected in this study according to results of the telephone interviews and all of the ten brands of the formula milk. As a national distribution chain applies to formula milk, the same products would have been on sale throughout the Taiwan. The locations are selected for the convenience of those buying the samples. The formula milk samples were purchased from stores in southern Taiwan during 2000-2001. Three cans of formula milk samples with the same brand were purchased in different stores and the produced dates for the three cans of formula milk samples were difference. Three cans of formula milk samples with the same brand were mixed to one sample of the brand and the formula milk sample was stored in room temperature until the time of analysis.
The 17 PCDD/PCDFs and 12 DL-PCBs levels in formula milk 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[5] and Method 1668A[6] with the minor modifications. Quality assurance/quality control (QA/QC) protocols are established, according to those defined in USEPA Method 1613B[5] and Method 1668A[6], in ALTEP-NCKU laboratory to ensure positive identification and the quality of the measurements.

Results and Discussion
The telephone interviews result showed that 22 brands of formula milk were most commonly used in Taiwan and

73% of mothers used 10 brands among the 22 ones. The ten brands of formula milk samples were collected from stores and measured the levels of 17 PCDD/PCDFs and 12 DL-PCBs levels. The levels of 17 PCDD/PCDF and 12 DL-PCB congeners in these formula milk samples were summarized in Table 1. According to the information labeled on the packages of the formula milk, the origins of formulas included seven countries, America (N=2), Australia (N=1), Denmark (N=1), Germany (N=1), Netherlands (N=2), New Zealand (N=2), and Taiwan (N=1). Levels of total 17 PCDD/PCDFs and 12 DL-PCBs in the 10 formula milk samples ranged between 0.849 and 1.23 pg WHO_{98}-TEQ/g lipid, with a mean value of 1.04 ± 0.20 pg WHO_{98}-TEQ/g lipid (mean ± S.D.). The PCDD/PCDF and DL-PCBs levels in the formula milk samples are below the maximum guideline value for milk and milk products, 3 pg WHO_{98}-TEQ/g lipid, set by the European Community (EC) Regulation[7]. The formula milks are reconstituted from cow milks during manufacturing process. However, PCDD/PCDF levels in the cow milk[8,9] is higher than the levels in formula milk reported in the present study. Infant formulas have a special role to play in the diets of infants because they are often the only source of nutrients for infants. For this reason, the formula milk is reconstituted from cow milk so most lipids of animal origins with relative higher PCDD/PCDF levels are intentionally removed. Formula milk contains added lipids of vegetable origins with relatively lower PCDD/PCDF levels, and therefore it has lower PCDD/PCDF levels than the cow milk.

The contributions of 7 PCDD, 10 PCDF, and 12 DL-PCB congeners to total TEQ (17 PCDD/PCDFs and 12 DL-PCBs) in formula milk samples were showed in Figure 1. Seven PCDD levels accounted for 30% ± 8% (mean ± S.D.) of total TEQ (17 PCDD/PCDFs and 12 DL-PCBs), ten PCDF levels accounted for 42% ± 9%, and twelve DL-PCB levels accounted for 33% ± 6%. The congener profiles of 17 PCDD/PCDFs and 12 DL-PCBs in formula milk samples were demonstrated in Figure 2A and 2B, respectively. For PCDD/PCDFs levels in the formula milk, The congeners with highest percentages were 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDD, and 2,3,7,8-TCDD. For DL-PCBs levels in the formula milk, the congeners with highest percentages were PCB114, PCB156, and PCB 157.

This is a first report of a global survey for 17 PCDD/PCDF and 12 DL-PCB levels in formula milk manufactured in seven countries, including America, Australia, Denmark, Germany, Netherlands, New Zealand, and Taiwan. The data are valuable for the future studies on evaluating PCDD/PCDFs and DL-PCBs exposure to infants or adults due to formula milks consumptions.

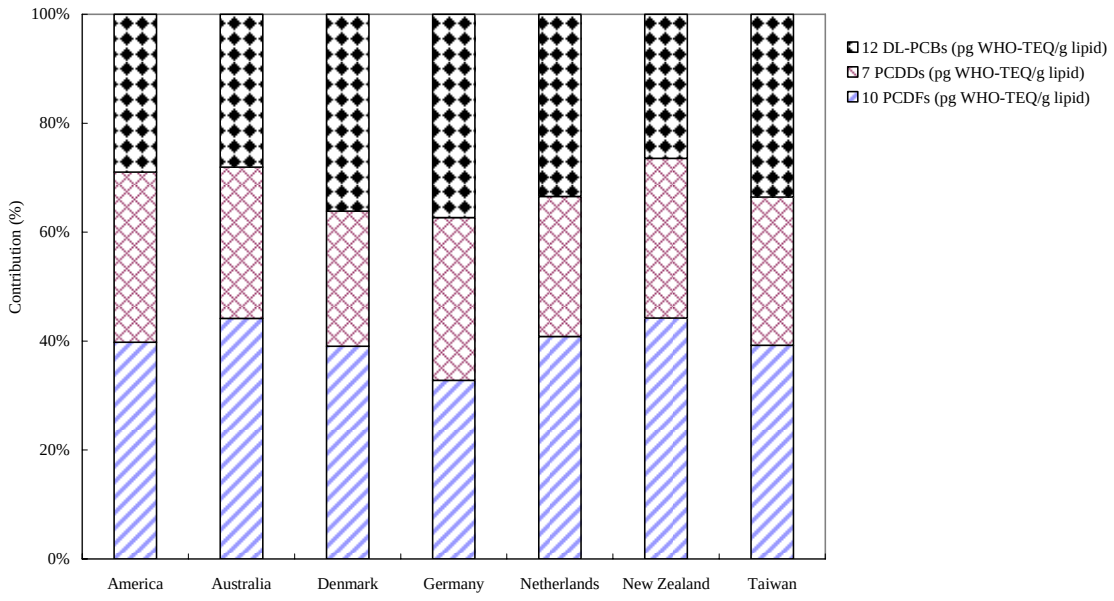

Figure 1. Contribution of PCDD/PCDFs and dioxin-like PCBs levels to total TEQ in formula milk

Table 1. Seventeen PCDD/PCDF and twelve dioxin-like PCB levels in formula milk

origin of formula milk by country	America (N=2)	Australia (N=1)	Denmark (N=1)	Germany (N=1)	Netherlands (N=2)	New Zealand (N=2)	Taiwan (N=1)	mean ± S.D.
	Unit: pg/g lipid							
2,3,7,8-TCDF	0.159	0.204	0.122	0.107	0.079	0.214	0.087	0.139 ± 0.055
1,2,3,7,8-PeCDF	0.434	0.490	0.676	0.413	0.364	0.506	0.247	0.447 ± 0.133
2,3,4,7,8-PeCDF	0.458	0.496	0.353	0.415	0.307	0.564	0.325	0.417 ± 0.095
1,2,3,4,7,8-HxCDF	0.460	0.466	0.306	0.333	0.347	0.484	0.333	0.390 ± 0.076
1,2,3,6,7,8-HxCDF	0.536	0.526	0.244	0.447	0.300	0.488	0.363	0.415 ± 0.114
1,2,3,7,8,9-HxCDF	0.488	0.525	0.281	0.360	0.313	0.564	0.303	0.405 ± 0.117
2,3,4,6,7,8-HxCDF	0.507	0.550	0.268	0.347	0.391	0.530	0.430	0.432 ± 0.104
1,2,3,4,6,7,8-HpCDF	0.418	0.374	0.222	0.367	0.260	0.412	0.300	0.336 ± 0.076
1,2,3,4,7,8,9-HpCDF	1.32	0.475	0.222	0.367	0.330	0.509	0.300	0.503 ± 0.373
OCDF	1.04	1.43	1.22	0.667	0.801	1.47	0.333	0.995 ± 0.419
2,3,7,8-TCDD	0.079	0.057	0.089	0.080	0.061	0.054	0.073	0.071 ± 0.013
1,2,3,7,8-PeCDD	0.157	0.117	0.053	0.171	0.051	0.147	0.041	0.105 ± 0.056
1,2,3,4,7,8-HxCDD	0.464	0.555	0.259	0.377	0.300	0.634	0.400	0.427 ± 0.134
1,2,3,6,7,8-HxCDD	0.403	0.425	0.216	0.280	0.287	0.408	0.380	0.343 ± 0.081
1,2,3,7,8,9-HxCDD	0.409	0.434	0.221	0.387	0.278	0.522	0.360	0.373 ± 0.100
1,2,3,4,6,7,8-HpCDD	1.48	0.488	0.238	0.433	0.281	0.327	0.333	0.512 ± 0.436
OCDD	0.763	0.502	1.17	1.33	0.966	0.768	0.333	0.834 ± 0.354
PCB 77	163	216	168	182	98.7	161	119	158 ± 39.1
PCB 81	163	124	169	179	103	126	118	140 ± 29.4
PCB 105	155	141	195	214	128	159	128	160 ± 32.9
PCB 114	182	249	122	210	96.4	168	145	167 ± 52.2
PCB 118	198	270	191	213	131	206	142	193 ± 46.6
PCB 123	192	261	196	202	128	203	143	189 ± 43.7
PCB 126	0.095	0.052	0.127	0.030	0.164	0.108	0.023	0.086 ± 0.053
PCB 156	151	136	124	236	116	148	128	148 ± 40.5
PCB 157	145	34.9	117	204	127	111	131	124 ± 50.1
PCB 167	277	241	963	227	404	475	153	392 ± 275
PCB 169	0.093	0.049	0.013	0.030	0.370	0.076	0.023	0.093 ± 0.125
PCB 189	134	31.8	167	206	101	102	135	125 ± 55.3
10 PCDF*	0.483	0.508	0.337	0.395	0.321	0.545	0.333	0.417 ± 0.093
7 PCDD*	0.379	0.320	0.214	0.360	0.202	0.361	0.231	0.295 ± 0.077
17 PCDD/PCDF*	0.862	0.828	0.551	0.755	0.522	0.905	0.564	0.713 ± 0.163
12 dioxin-like PCBs*	0.352	0.323	0.312	0.450	0.263	0.326	0.285	0.330 ± 0.060
17 PCDD/PCDF and 12 dioxin-like PCBs*	1.21	1.15	0.864	1.20	0.786	1.23	0.849	1.04 ± 0.199

[*] Unit: pg WHO_{98}-TEQ/g lipid

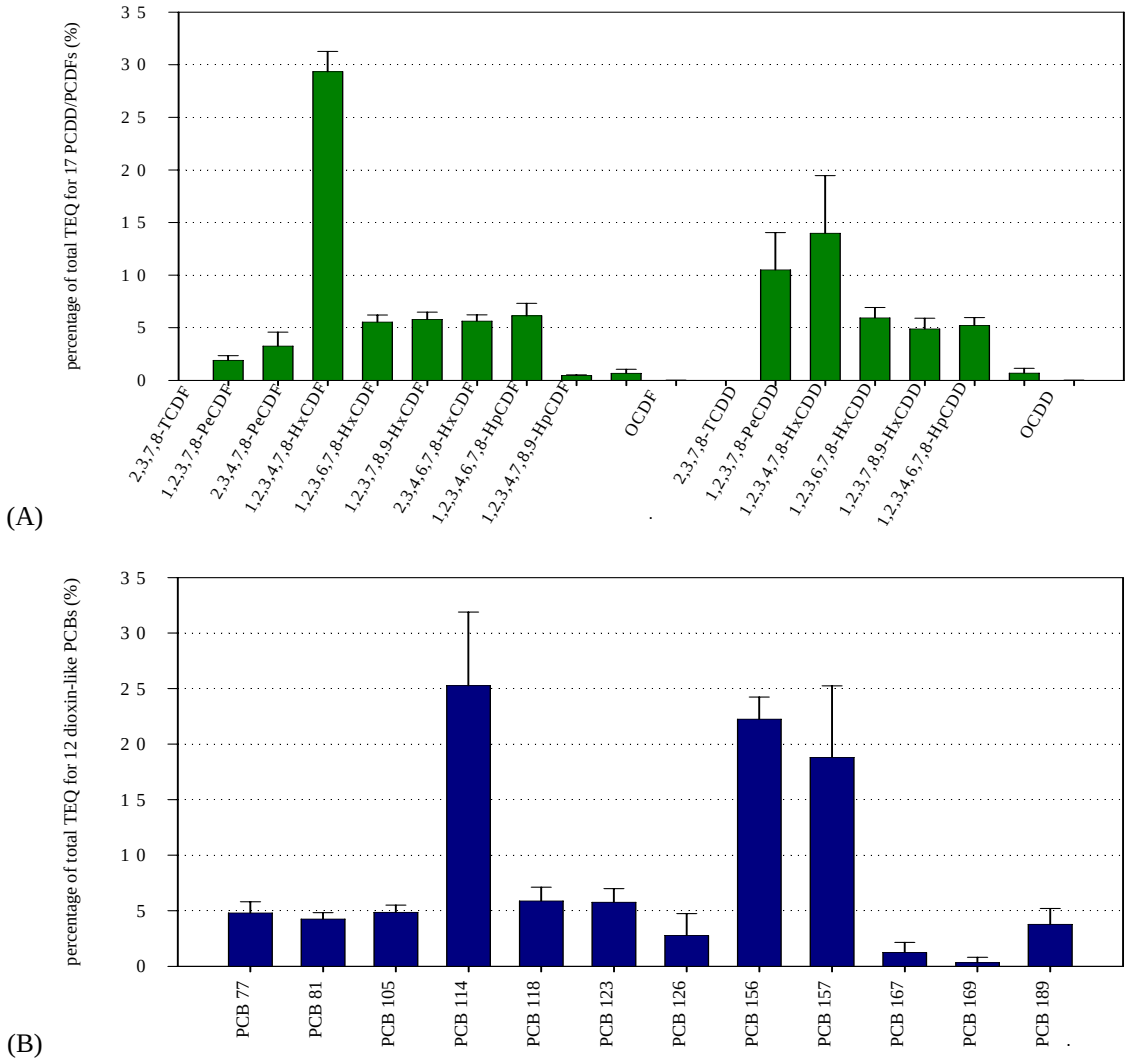

Figure 2. (A) 17 PCDD/PCDF congener profiles in formula and breast milk samples, (B) 12 dioxin-like PCB congener profiles in formula and breast milk samples.

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