PCDD/PCDF AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS IN BLOOD OF AN ADULT POPULATION

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Introduction

Polychlorinated dibenzodioxins and dibenzofurans (PCDD/PCDF) and dioxin-like Polychlorinated Biphenyls (dl-PCB) are persistent organic pollutants, biaccumulate in the biotic environment and in humans. In the last years a decreasing tendency could be observed in human body burden, which was due to extensive efforts reducing the exposure against these compounds. Nevertheless, due to their toxicological properties they are still of health concern. The main route of exposure was diet, but beyond other sources like elastic indoor sealants have to be taken into account.

The purpose of the present study was to quantify the internal exposure to Dioxins / Furans and Polychlorinated Biphenyls in the general population of Germany. The analyses were part of the Integrated Exposure Assessment Survey (INES), which aims to estimate the exposure to different substances with health concern, combining exposure assessment by different routes such as residential indoor air, house dust and duplicate diet with quantification of internal exposure using biomarkers.

Materials and Methods

Study population

The investigation was carried out in an adult population of 26 female and 22 male healthy subjects occupationally not exposed to the target analytes. They are living in the city of Munich (Germany) and the nearby suburban and rural areas in the southern parts of Germany. The volunteers, between 14 and 60 (median: 35) years of age, having a body weight of 42 to 107 kg (median: 70 kg) were recruited between April 2005 and October 2005.

Sampling and laboratory analysis

Whole blood was collected once in our laboratory after venipuncture for each participant and stored after preservation with EDTA at -20°C until laboratory analysis.

A total of 48 whole blood samples were analysed for 17 PCDD/F and 12 dioxin-like PCBs congeners for which WHO-TEF has been assigned.

Sample preparation

Each whole blood sample was spiked with all 17 $^{13}C_{12}$-labeled PCDD/PCDF and all 12 $^{13}C_{12}$-labeled dioxin-like (dl-) PCB congeners, which are e.g. described in annex of Commission Regulation (EC) No 199/2006, as internal standard. First blood fat was extracted with a mixed Isolut/sodiumchloride column. Fat and non-acid stable compounds were removed on a silica gel column coated with sulfuric acid. The pentane fraction containing native and internal standard PCDD/PCDF and dl-PCB was carefully concentrated by rotary evaporation at 40°C under reduced pressure and subsequently applied to a Carbopack/Celite column to separate dl-PCB (eluted with hexane, cyclohexane and dichlormethane) from PCDD/PCDF (eluted with toluene). While the PCDD/PCDF fraction was further cleaned up on a Florisil column (1% water), the dl-PCB eluate was applied to an aluminium oxide/sodium sulfate anhydrous column to separate the mono-ortho compounds from the non-

ortho-PCB fraction. The resulting three final extracts were separately analyzed by HRGC/HRMS on a Micromass AutoSpec system at a resolution of R = 10,000.

Statistical analysis
For the estimation of correlations the non-parametric Spearman rank correlation coefficient was derived, the difference in median was tested with the Wilcoxon rank sum test .

Results and Discussion
The statistical parameters of the calculated PCDD/F and PCB TEQ-values of all subjects are given in Table 1 and their median share on total TEQ-values were expressed in figure 1. As seen from figure 1 the pattern between PCDD/F and PCB only slightly changed using the different TEF schemes of the WHO. On the other side it is observable that the proportion of mono-ortho PCB decreased from 25 % to only 5 %. Compared with another German study[1] conducted in 2000-2003 the account of PCBs on total TEQ (40 %) was lower than observed in our study (49 %).
No significant difference was found between females and males. We observed a significant associations of non-ortho and mono-ortho PCBs with body weight and body mass index (r values between 0.37 and 0.40), whilst this relation could not be observed with regard to PCDDF/F. As described from numerous other studies before we found increasing concentrations of the target substances according to age (Figure 2).

Our results are in line with previously findings from Germany[1], where total PCDD/F and PCB TEQ-values of 26.4 pgTEQ/g lipid base were observed in a study of 169 participants conducted 2000-2003. In a recent study from Greece[2], conducted in an urban area 2002-2004 TEQ-values of 10.0 pg/g were seen depending mainly on low PCB concentrations. Studies performed earlier in the ends of nineties in Finland[3] and Belgium[4] observed considerably higher total TEQ-values of 49.7 and 72.7 pg/g blood fat. Otherwise, investigations conducted in Asia[5] and Australia[6] resulted in the same or lower levels in comparison to our study.

Table 1: Levels of PCDD, PCDF, and dioxin-like PCBs expressed as WHO-TEQ (pg/g lipid base) in the blood of 48 participants (TEQ values in brackets using WHO 2005 TEFs)

Compound	**5th percentile**	**Median**	**95th percentile**	**Maximum**	**Arithmetic mean**
PCDD	0.48 (0.50)	3.37 (3.38)	11.00 (11.08)	12.91 (13.04)	4.29 (4.33)
PCDF	0.10 (0.09)	5.65 (3.72)	19.45 (11.87)	22.55 (13.68)	7.09 (4.52)
PCDD/PCDF	0.78 (0.79)	10.09 (7.74)	24.95 (19.97)	29.10 (20.87)	11.41 (8.87)
Non-ortho PCB	1.58 (1.69)	4.21 (5.22)	14.88 (16.32)	17.97 (19.48)	5.02 (5.82)
Mono-ortho PCB	1.08 (0.17)	4.48 (0.63)	14.24 (1.70)	17.98 (2.21)	6.03 (0.72)
PCB	2.52 (1.84)	9.45 (5.80)	26.17 (17.36)	33.50 (21.70)	11.05 (6.54)
Sum of all TEQ	4.10 (2.90)	23.13 (14.35)	50.73 (34.20)	54.95 (39.14)	22.49 (15.40)

non-ortho substituted PCBs: PCB 77, PCB 81, PCB 126, PCB 169; **mono-ortho substituted PCBs:** PCB 105, PCB 114, PCB 118, PCB 123, PCB 156, PCB 157, PCB 167, PCB 189

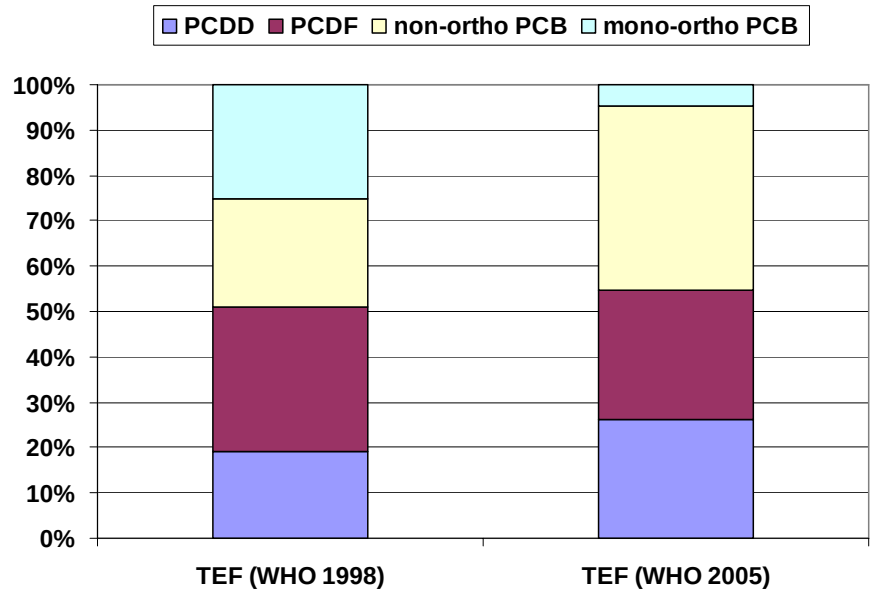

Figure 1: Share of PCDD, PCDF, non-ortho and mono-ortho PCB on total TEQ using diferent TEF schemes

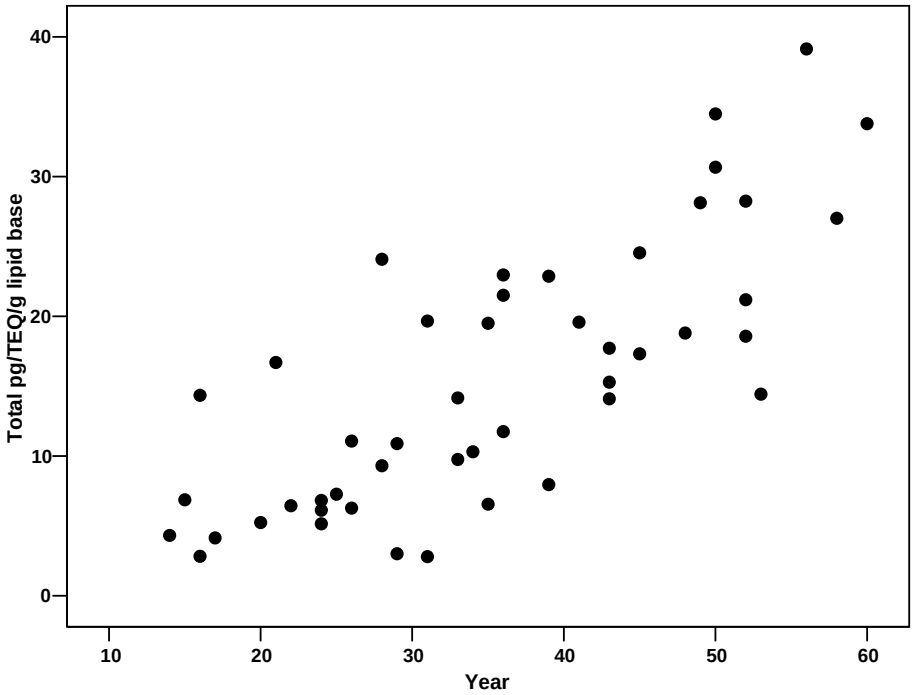

Figure 2: Age-dependence of Dioxin and PCBs concentration (expressed as WHO-TEQ) in human blood

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References

1. Wittsiepe J, Fürst P, Schrey P, Lemm F, Kraft M, Eberwein G, Winneke G, Wilhelm M. *Chemosphere* 2007; 67: S286-S294
2. Costopoulou D, Vassiliadou I, Papadopoulos A, Makropoulos V, Leondiadis L. *Chemosphere 2006*; 65:1462-1469.
3. Kiviranta H, Tuomisto JT, Tuomisto J, Tukiainen E, Vartiainen. *Chemosphere* 2005; 60: 854-869.
4. Koppen G, Covaci A, Van Cleuvenbergen R, Schepens P, Winneke G, Nelen V, van Larebeke N, Vlietinck R, Schoeters G. *Chemosphere* 2002; 48: 811-825.
5. Kim B-H, Ikonomou MG, Lee S-J, Kim H-S, Chang Y-S. *Sci Total Environ* 2005 ; 336: 45-56.
6. Harden F, Müller J, Toms L. *Dioxins in the Australian population: levels in blood.* National Dioxin Program. Technical Report No. 9. Department of Environmental and Heritage. Canberra, Australia. 2004.