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Concentrations and 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin Toxic Equivalents of Non-*ortho* Coplanar PCBs in Adipose Fat of Poles

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The non-*ortho* chlorine substituted coplanar polychlorinated biphenyls, such as, 3,3',4,4'-T_4CB (IUPAC no. 77), 3,3',4,4',5-P_5CB (IUPAC no. 126) and 3,3',4,4',5,5'-H_6CB (IUPAC no. 169) are highly bioactive and toxicologically potent trace constitutents in technical PCB formulations. These congeners have been traced in a wide variety of environmental compartments including biota, food and human tissues (Tanabe et al. 1987). These non-*ortho* coplanar PCBs have been assigned a toxic equivalent factor (TEF) based on their toxicity relative to 2,3,7,8-tetrachloro dibenzo-*p*-dioxin (TCDD) which is assigned a TEF of 1. The normalisation based on the TCDD toxic equivalents has permitted a new perspective in assessing the inherent toxicity of these congeners and their likely biological impact. Since the AHH-active PCB congeners and PCDDs/PCDFs act through initial binding to the same receptor, an additive model is used to express toxicity of a mixture of these compounds (Safe 1990). The concentrations of toxic equivalents are expressed when assessing risks associated with exposure and to identify those congeners that pose greater health risks.

In our recent survey on organochlorine contamination in human adipose fat from Poland, total PCBs concentrations were found in the range of 0.75-1.9 μg/g on a lipid wt basis in 1979 and 0.76-4.7 μg/g in 1990 (Tanabe et al. 1993). Further studies have also shown the presence of considerably high concentrations of PCBs upto 10 μg/g in human adipose fat near Poland-Slovakia (Petrik et al. 1991). These values suggested the ongoing contamination by PCBs in this region and highlighted the need for monitoring toxic PCB isomers in foodstuffs and in human tissues. The present paper provides a baseline data on the non-*ortho* coplanar PCB concentrations in human adipose tissue from Poland in 1979 and 1990.

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MATERIALS AND METHODS

Human adipose tissue samples were collected during an autopsy from randomly selected donors in Skierniewice and Gdansk city (see Fig. 1.).

Figure 1. The map of Poland with indication of the sampling sites

The samples were wrapped in aluminium foil and kept deep frozen for a few days. The adipose fat samples originated from the Province of Skierniewice were taken in 1979. Lipid extracts from these samples (after Soxhlet extraction with light petroleum) were kept sealed in ampoules for further analysis. The samples taken in Gdansk city in 1990 were cut into small pieces, packed in clean polyethylene containers with a small amount of 10% formalin and stored at 4°C until analysis.

Non-*ortho* coplanar PCBs (IUPAC nos. 77,126 and 169) were analysed following the method described by Tanabe et al. (1987). Briefly, the method involved digestion of samples with 1N KOH-ethanol for 1 h followed by shaking with 100 ml of hexane in a separatory funnel for re-extraction and clean up using silica gel (Wako gel S-1) column chromatography. Non-*ortho* coplanar PCB members were separated from other PCB isomers and congeners using carbon column chromatography. The concentrated hexane layer was further cleaned up with a mixture of 5% fuming sulphuric acid in conc. H_2SO_4. A gas chromatograph/mass spectrometer (Hewlett Packard 5890 GC with 5970 mass selective detector) having an electron impact (EI) mode at 70eV and equipped with a fused silica capillary column coated with chemically bonded DB-1701 of 0.25 μm film thickness, was used for quantification. The operating conditions were as follows: injection port temperature-260°C, for transfer line-280°C, column initial temperature-180°C, rate 2°C/min, final temperature-250°C. For confirmation / quantification of three non-*ortho* coplanar PCBs, M^+ and $(M+2)^+$ cluster ions were monitored at *m/z* 290 and 292 for T_4CB (77), 324 and 326 for P_5CB (126) and 358 and 360 for H_6CB (169). The non-*ortho* coplanar PCB standards used were of 98-100% purity.

RESULTS AND DISCUSSION

The concentrations of non-*ortho* coplanar PCBs in adipose fat of Poles in 1979 and 1990 are presented in Table 1. Coplanar PCBs were detected in all the samples analysed at the following concentration ranges: 54-500 pg/g for CB no. 77, 41-850 pg/g for CB no. 126 and 50-390 pg/g for CB no. 169 on a fat wt basis.

Table 1. Non-*ortho* coplanar PCBs in human adipose tissue in Poland

Sample			Concentration (pg/g on a fat wt basis)			
Age	Sex	Fat(%)	CB 77	CB126	CB169	Total co-PCBs
Skierniewice (1979)						
54	F	NA	260	350	120	730
64	F	NA	330	850	380	1600
83	F	NA	240	160	170	570
Gdańsk (1990)						
35	M	71	54	41	50	150
42	M	76	420	280	98	800
60	M	68	500	530	380	1400
42	F	77	140	180	290	610
50	F	79	120	200	170	490
58	F	62	160	310	390	860
60	F	74	310	530	530	1400
67	F	68	130	320	210	660
68	F	67	110	270	220	600

F - female; M - male; NA - data not available

Table 2. 2,3,7,8-TCDD toxic equivalents of non-*ortho* coplanar PCBs in human adipose tissue in Poland (pg/g on a fat wt basis)

PCB	TEQ	Range	Mean
77	0.01	0.54-5.0	2.3
126	0.10	4.1-85	34
169	0.05	2.5-27	13
Total		7.1-107.3	49.3

The recorded concentrations were comparable to higher than those reported for the general populations in Japan (Kannan et al. 1988), Canada (Williams and LeBel 1991), the USA (Hong et al. 1993), the UK (Davidson et al. 1993) and Germany (Georgii and Brunn 1990). The adipose fat samples collected in the Province of Skierniewice in 1979 contained comparable concentrations of non-*ortho* coplanar congeners with those of donors in Gdańsk city sampled during 1990. As for total PCBs (see Tanabe et al. 1993), there exhibited no considerable variation in the concentrations of non-*ortho* coplanar PCBs between 1979 and 1990. This suggests

a slower elimination rate of coplanar congeners and/or a continuing exposure of the Polish population to PCBs. It seems that in the case of male citizens from Gdańsk, concentrations of coplanar congeners in the adipose tissue was age dependent, although the number of samples analysed are very small to obtain a valid conclusion. Although the adipose tissues collected from females contained slightly lower levels of coplanar PCBs than in males, in very old female samples, the concentrations were comparable to those of males reflecting the cessation of parturition and lactation in female results in the loss of elimination routes of toxic congeners.

The CB no. 77 occupied 30% and 28% of total non-*ortho* coplanar PCBs concentration in samples from Skierniewice and Gdańsk, respectively. Analogically for CB no. 126, it was 47% and 39% and for CB no. 169, it was 23% and 33%, suggesting a higher contribution of CB no. 126 in human tissues. The non-*ortho* coplanar PCBs composition found in the adipose fat of Poles differed largely from those observed in other environmental samples from Poland (Fig. 2.). The CB no. 77 constituted a major proportion in fish and other aquatic biota as well as in chlorofen (a Polish PCB formulation; refer Falandysz et al. 1992b), while in the human tissue its contribution was low. In contrast, CB no. 126 was maximum in the human tissue and minimum in fish and porpoise. A similar pattern of non-*ortho* coplanar PCBs composition has been observed for the human tissues in several other studies. It has been reported that the rates of biodegradation of non-*ortho* coplanar PCBs were in the order of #77 > #126 > #169 (Tanabe et al. 1989). Lower levels of #77 indicate the metabolic degradation of this congener by humans, although this isomer constitute more than 95% among the three non-*ortho* congeners in technical PCB formulations (Kannan et al. 1987), suggesting an higher exposure of #77 than #126 and #169. It worth indicating that fish and marine mammals which have lower metabolic potential comprised of non-*ortho* congeners in the order #77 > #126 > #169 (Tanabe et al. 1987).

The 2,3,7,8-TCDD toxic equivalents (TEQs) for three non-*ortho* coplanar PCBs in Poles fat were 49.3 pg/g (Table 2). In the UK, it was 18 pg/g (Davidson et al. 1993), 38 pg/g in Canada (Williams and LeBel 1991) and 41 pg/g in Japan (Kannan et al. 1988). The general population appears to possess dioxin and furan levels between 5 and 1,000 pg/g in its adipose tissue. In highly exposed individuals such as Yusho and Yucheng victims (Ryan et al. 1985), the concentrations of dioxins were 2 to 3 orders of magnitude higher than are now estimated in the human tissue from Poland. These epidemiological studies provide only the correlations with the observed concentrations of toxic substances and the resultant clinical symptoms. Therefore, it is essential that in the absence of adequate data on cause-effect linkage of dioxins/furans in humans, care should be taken to avoid excessive human exposures through contaminated foods. Among the three congeners, CB no. 126 accounted for 69% of total TEQs in Poland, while CBs no. 169 and 77 occupied 26% and 5% respectively. The CB no. 126 is the most toxicologically

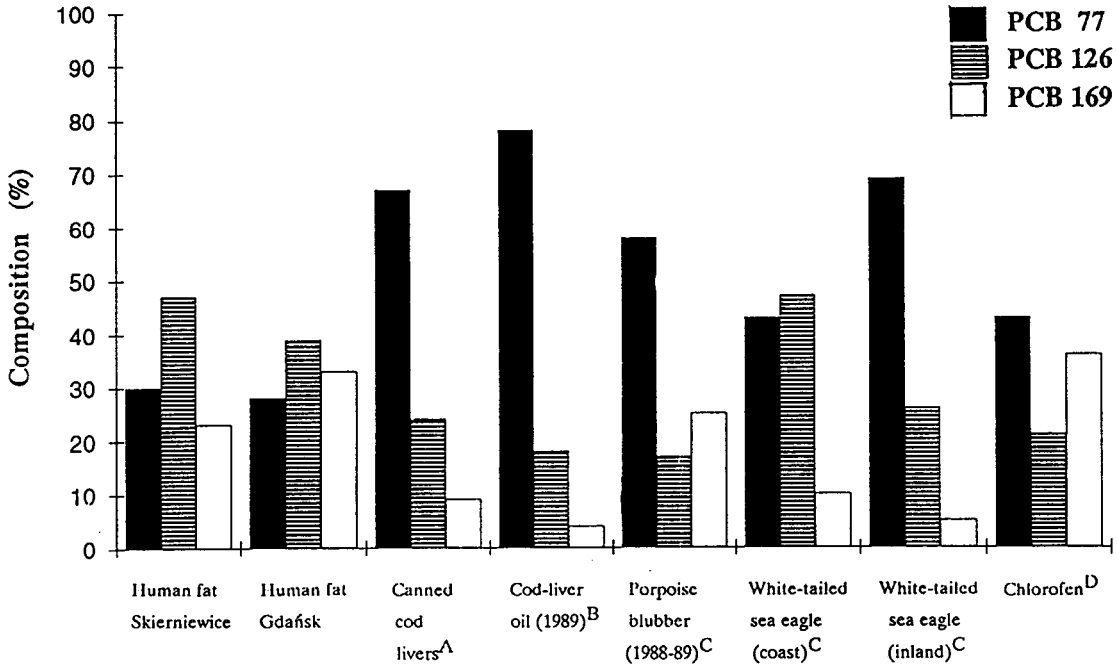

Figure 2. Composition (%) of non-*ortho* coplanar PCBs in human adipose fat and some other samples from Poland. A, B, C and D refers to references: Falandysz *et al.*, 1992a; Unpublished data; and Falandysz *et al.*, 1992b and c, respectively.

significant in humans because of its higher accumulation, greater potency relative to other congeners.

A comparison of non-*ortho* coplanar PCBs concentrations in human adipose tissues from Poland, UK, Canada and Japan suggests that the concentrations recorded in human tissues from Poland are higher than those reported for other developed nations and the rate of decline has been slow (Fig.3). It is probable that the human exposure to PCBs is still continuing in Poland. In addition to non-*ortho* coplanar PCBs, mono-*ortho* congeners such as, IUPAC nos. 105, 118 and 156 represent an important part of the toxicity of PCB and PCDD/PCDF mixtures. Therefore it is necessary to monitor these congeners for human body burden assessment.

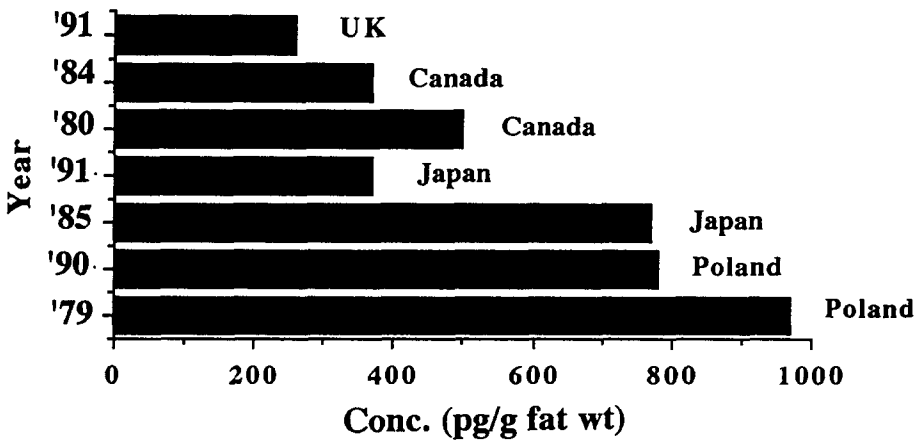

Figure 3. Comparison of the concentrations of non-*ortho* coplanar PCBs (IUPAC Nos. 77+126+169) in the human adipose fat in different countries (references are cited in the text)

It can be concluded that the concentrations of non-*ortho* coplanar PCBs in Poles fat are considerably high and the congener 126 contributes higher toxicity in humans. Continuous monitoring of foods and studies on cause-effect linkage of dioxins/furans are essential to undersatnd their toxic impacts and to avoid risks associated with excessive exposure to toxic chemicals.

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