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PCB Residues in the Adipose Tissue of the Population of Barcelona (Spain)

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Polychlorinated biphenyls (PCBs) are a class of aryl halides widely distributed in the environment. Although production has virtually ceased, and most industrial applications (capacitors, transformers, hydraulic fluids, lubricants, etc) are severely restricted, they are one of the most ubiquitous and persistent environmental pollutants. Several toxic effects caused by PCBs have been described including: liver enlargement, hepatomegalocytosis, acne, lymphoid atrophy, immunosuppression, tumor promotion, porphyria, etc (Reggiani, 1982; McConnell, 1980).

This work continues our previous reports about organochlorine residues in human tissues of some Spanish populations, that showed high levels of some residues, specially of hexachlorobenzene (HCB) (To-Figueras et al, 1986; Camps et al, 1988). PCBs pattern and concentration in the adipose tissue of the inhabitants of an urban and industrial area (Barcelona) were determined.

MATERIALS AND METHODS

Specimens of abdominal adipose tissue (N=55) from necropsies carried out on randomly selected subjects were removed, frozen and stored at -70 C until analysis. Their ages ranged from 9 to 95 years (mean, 55 years), 28 were males and 27 females; all had lived in the area of Barcelona.

Tissues were ground (1 gram) with sodium sulphate anhydrous (Merck) and PCBs were extracted with hexane (Merck, for residue analysis) in a Soxhlet apparatus. A chemical cleanup with sulphuric acid was applied to extracts (Veierov and Aharonson, 1978); assays with an olive oil matrix showed quantitative recovery of Aroclor^R 1260 after cleanup. When analyses by GLC/MS were performed, a further cleanup by adsorbent column chromatography in silica-alumina was done (De Voogt, 1986).
Extracts were concentrated under a nitrogen stream and redisolved in undecane, in order to obtain a good solvent effect in split-less inyection with a high initial oven temperature. Analyses were performed in a Varian

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3700 gas chromatograph with electron capture detector and a SPB-5 capillary column (30m, 0.32 mm id, 0.30 um film thickness; Supelco) and split-less inyector. Chromatographic parameters: initial T, 160 C; initial time, 1 min; rate, 7 C/min; final T, 240 C; carrier, helium; column head pressure, 10 psi. Volum injected was always 0.3 ul.

GLC/MS analyses were performed in a HP5985 equipment, with EI ionization (70 eV), in SIM mode (m/z: 324, 326, 360, 394, 396, 428; dwell time 100 msec).

Chromatographic peaks were identified: first, determining the degree of chlorination by GLC/MS; second, comparing the retention times with those of Aroclor peaks and assuming the identification reported by Safe et al (1985) for Aroclor 1260 components.

Quantitation was done by calibration against Aroclor 1260 (Supelco); Aldrin was used as internal standard (Method A):

$$Q_{as} = (\Sigma\ A_i/A_o)_s/\Sigma\ (A_i/A_o)_c)*(Q_{os}*Q_{ac}/Q_{oc})$$

where Q are ng of compounds; A areas of peaks; subindex refer to: a, Aroclor; i, individual peaks; o, internal standard; s, sample; c, calibration standard.

Because of the discrimination effect of the injector and short linear range of the ECD, response factors were very sensitive to variations in peak areas; a multiple point calibration was done with a least squares adjustement of relative respose factors in function of Aroclor peak areas and internal standard area:

$$(A_i/A_o)*(Q_o/Q_a) = R_i*A_i + S_i*A_o + Z_i$$

where coefficients R, S and Z were optimized by least squares after injection of six different calibration standards. Total recalibration was done weekly; recalculation of Z coefficients was performed daily by injection of a single standard. The precision obtained, based in replicate injection of a real sample at different dilutions, was better than 8% RSD.

Furthermore, a specific quantitation of individual congeners was atempted by a method adapted from that of Webb and McCall (1973): assuming the Aroclor 1260 composition determined by Safe et al (1985), individual response factors were determined in our analytical system, adjusted by a least squares method as previously described, and applied to quantitation of individual peaks in samples. Thirty major peaks were considered (some of the peaks not belonging to pure congeners). As almost all sample peaks were present in Aroclor 1260, a quantitation of total PCBs can be performed by addition of individual concentrations (Method B). A BASIC routine was developped for the management of data.

RESULTS AND DISCUSSION

PCBs pattern in human adipose tissues was very repetitive; congeners with high degree of chlorination (penta- to octa Cl) predominating, as in Aroclor 1260; these findings are similar to those reported in other human populations and are the consequence of the major lipophilicity and resistance to degradation of the higher chlorinated congeners (Zell and Ballschmiter, 1980). This justifies the calculation of results as Aroclor 1260. Results obtained are summarized in Table I. Mean values in the order of 1 ppm were obtained (refered to extracted lipid). However, the distribution was not normal and had an asymmetry to the right; in fact, a major part of samples were under the mean, as indicated by the lower geometric mean. The mean values obtained are similar to those reported in other human populations.

Both quantification methods produced slightly different results, but an excellet correlation between them was found. Lower concentrations with method B may be due to the fact that some sample peaks were not present in Aroclor 1260 and some small ones were not taken into account. Major peaks, quantitated by method B, correspond to the congeners (Ballschmiter numbers): 153 (20%), 180 (14%), 138 (10%), 170 (9%), 118 (5%, roughly approximate because of insufficient resolution with 149 and low concentration present in Aroclor 1260), 187 (5%), 196+203 (5%), 201 (4%). Standard deviations of this percentages were less than 25%, indicating a very regular pattern in our population. As a whole, these major congeners amount to more than 72% of PCB residues; they have chlorine in positions 3 and/or 5 in both rings and in position 4 in at least one ring, but also they have substitutions in the *ortho* positions. Therefore, the planar conformation is disfavored and they dont have a "dioxine-like" structure. Congeners with the "dioxine-like" structure are the most actives as receptor Ah agonists and enzyme inducers, effects that might be related to its toxicity (Goldstein, 1980; Safe, 1987)

No significant differences in whole PCBs concentration were found between males and females. There was some correlation between PCBs concentration

Table 1
PCBs in human adipose tissue (expresed as μg/g lipid extracted).

	mean	SD	ESM	geo. mean	range
Method A (*)	1.138	0.669	0.090	0.934	0.19-2.70
Method B (*)	0.917	0.532	0.072	0.761	0.18-2.11
Correlation	$r = 0.986$		$N = 55$	$p < 0.001$	
Linear regression equation:	$B = 0.784\ A + 0.024$				

(*) See Material and Methods

and age: $r = 0.360$, $p < 0.01$. The scattering plot showed that correlation was produced by the low PCBs levels of individuals below 35 years old (0.73 ± 0.47 ; $N = 12$), whereas levels in older population were more dispersed (1.24 ± 0.70 ; $N=43$) and uncorrelated.

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