Environmental Research 111 (2011) 10–16

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

# Background levels of polychlorinated biphenyls in the population of the Canary Islands (Spain)

Luis A. Henríquez-Hernández [a], Octavio P. Luzardo [a], Maira Almeida-González [a], Eva E. Álvarez-León [b], Lluis Serra-Majem [c], Manuel Zumbado [a], Luis D. Boada [a,*]

[a] Toxicology Unit, Dept. of Clinical Sciences, Universidad de Las Palmas de Gran Canaria, and Instituto Canario de Investigación del Cáncer (ICIC), P.O. Box 550, 35080 Las Palmas de Gran Canaria, Spain
[b] Preventive Medicine Service, Complejo Hospitalario Materno Insular de Gran Canaria, Canary Health Service, and Instituto Canario de Investigación del Cáncer (ICIC), Avda. Marítima s/n, 35016 Las Palmas de Gran Canaria, Spain
[c] Preventive Medicine and Public Health Unit, Dept. of Clinical Sciences, Universidad de Las Palmas de Gran Canaria, and Instituto Canario de Investigación del Cáncer (ICIC), P.O. Box 550, 35080 Las Palmas de Gran Canaria, Spain

ARTICLE INFO

*Article history:*
Received 23 April 2010
Received in revised form
6 November 2010
Accepted 10 November 2010
Available online 23 November 2010

*Keywords:*
Canary Islands
Polychlorinated biphenyls (PCBs)
PCB markers
Dioxin-like PCBs
TEQs
Human serum

ABSTRACT

Polychlorinated biphenyls (PCBs) are persistent and toxic compounds that have been detected in human serum or tissues worldwide. The objective of our study was to determine serum PCB levels in a representative sample of the general population of the Spanish Archipelago of the Canary Islands (607 serum samples from subjects aged between 6 and 75 years) in order to establish the main causes of this contamination and to evaluate the potential risks posed by these chemicals on the population through the use of toxicity equivalence to dioxins (TEQs). PCB congeners (28, 52, 77, 81, 101, 105, 114, 118, 123, 126, 138, 153, 156, 157, 167, 169, 180, and 189) were measured by gas chromatography–mass spectrometry (GC/MS). Our results showed that PCB residues were found in 76% of serum samples analyzed, with the congeners 153 and 180 being the most frequently detected and having the highest median values (21.8 and 6.7 ng/g lipid, respectively). Serum levels of non-dioxin-like PCBs increased with age, body mass index (BMI), urban habitat, and smoking. The median concentration of the sum of PCBs considered as markers of environmental contamination by these chemicals (M-PCBs) was 46.4 ng/g lipid. Levels of the sum of dioxin-like PCBs (DL-PCBs) were 48.5 ng/g lipid in the 95th percentile, and were also positively associated with age. As a consequence, age seemed to be positively associated with TEQs levels, reaching values as high as 58.6 pg/g lipid in the serum samples from oldest people. Our results indicate that the inhabitants of the Canary Archipelago show levels of PCB contamination lower than other populations present on the Spanish mainland, as well as many populations from developed countries. Nevertheless, as these compounds may induce adverse health effects even at very low doses, our findings should be considered by local Public Health authorities in order to establish measures for diminishing the exposure of the population of these islands to PCBs.



## 1. Introduction

Polychlorinated biphenyls (PCBs) are polyhalogenated aromatic hydrocarbons that have been widely used in closed systems such as electrical transformers and capacitors, as well as in a large number of other applications (Safe, 1990; Headrick et al., 1999). PCB production decreased and eventually ceased in the 1970s (Tanabe, 1988). In fact, they were banned in most Western countries (including Spain) in the late 1980s (Kimbrough, 1987; Kimbrough, 1995). However, an exception has been made to allow continued use of PCB-containing equipment until 2025. Because of this, in addition to their ubiquitous presence in the environment, PCBs could also reach soil and waterways as a result of leaks, spills, and improper disposal. The volatility of these compounds results in their evaporation from water surfaces and their movement through the atmosphere, resulting in widespread dispersal into the environment (Headrick et al., 1999). Furthermore, due to their fat solubility and resistance to chemical and biological degradation, ingestion of PCBs by animals leads to bioaccumulation throughout their lives and also to biomagnification in the food chain (Safe, 1994).

PCB exposure has been associated with adverse effects on human health, especially dioxin-like PCBs (DL-PCBs), those PCB congeners that exhibit toxic actions similar to dioxins (Van den Berg et al., 1994).

Several studies on animals show that PCBs are neurotoxic even at low doses (Tilson et al., 1990; Rice, 1999). It has been found that

* Corresponding author. Fax: +34 928 451 461.
*E-mail address:* ldominguez@dcc.ulpgc.es (L.D. Boada).

0013-9351/$ - see front matter © 2010 Elsevier Inc. All rights reserved.
doi:10.1016/j.envres.2010.11.005

PCB exposure could be related to the incidence of neurocognitive and endocrine disorders (Hagmar, 2003; Longnecker et al., 2003; Donato et al., 2008; Fitzgerald et al., 2008; Philibert et al., 2009). Since 1987, PCBs are considered as probable carcinogens to humans (Group 2A) by the International Agency for Research on Cancer (IARC). In fact, it has been proposed that there may be a potential relationship between PCB levels and the risk for non-Hodgkin's lymphoma (Engel et al., 2007; Bertrand et al., 2010). Additionally, several PCB congeners are known to be estrogenic and have been linked to reductions in fertility (Buck Louis et al., 2009).

The population involved in the present study has been extensively studied for levels of contamination by persistent organic pollutants (POPs). Thus, in 1998, more than 600 serum samples were obtained for a Nutritional Survey (Canary Islands Nutrition Survey, ENCA) as being representative of the total population of the Canary Islands and were used to evaluate the level of contamination by environmentally persistent organochlorine compounds. Firstly, we evaluated the level of contamination by organochlorine pesticides (Zumbado et al., 2005; Luzardo et al., 2006). In this study, we build upon previously published data and evaluate the level of contamination by PCBs in those population samples and explore potential determining factors that might affect serum PCB levels.

## 2. Materials and methods

### 2.1. Study group and sample collection

The ENCA nutritional survey was conducted in 1997–1998 in the seven major islands of the Canary Archipelago. The population of the Canary Islands between 6 and 75 years old consisted, at the time of the study, of approximately 1.3 million people. The sample was stratified in two stages and it was representative of the population between 6 and 75 years of age for both genders. About 1747 subjects participated in the first part of the study. This part consisted of two face-to-face interviews with questions about dietary variables, life habits, and health condition. The participants had blood samples extracted after 12-h fasting in order to determine biochemical and nutritional parameters and the presence of residues of POPs. 783 subjects participated in the biochemical part (participation rate, 44.8%) (Serra-Majem et al., 2000; Zumbado et al., 2005). The PCBs were measured in 607 subjects. The distribution of their sociodemographic characteristics are shown in Table 1.

Approximately 40 ml of blood was collected from each individual by venipuncture in a vacuum system tube, refrigerated, and centrifuged at 4 °C (15 min × 3000 rpm) within 4 h, to obtain serum. Serum was distributed in aliquots of 2–3 ml and immediately frozen at −80 °C. The serum aliquots stored at −80 °C were used to determine biochemical (including lipid profile) and nutritional parameters. Once these initial analyses were completed, the remaining serum was kept frozen at −80 °C for further chemical analyses. All collection and handling equipment in contact with serum specimens were tested for possible PCB contamination. No contaminating materials were identified.

The local ethics committee approved the design of this study, and informed consent was obtained from the study participants.

### 2.2. Analytical procedures

The aliquots of serum were subjected to solid-phase extraction and analyzed by gas chromatography/mass spectrometry (GC/MS) using appropriate internal standards. The analytes included in this study were the PCB congeners with IUPAC numbers #28, 52, 77, 81, 101, 105, 114, 118, 123, 126, 138, 153, 156, 157, 167, 169, 180, and 189.

#### 2.2.1. Sample preparation

Two-milliliter aliquots of serum were applied to 100 mg (1 ml) Chromabond® C18 cartridges (Macherey-Nagel, Germany) that were mounted on a vacuum manifold (Waters Corporation, USA). Before the application of samples, the cartridges were cleaned and conditioned as indicated by the manufacturer. Samples were then passed through the cartridge by gravity flow. The adsorbed PCBs were eluted with 4 × 250 μl of *n*-hexane. The solvent of the extracts was then evaporated under gentle nitrogen stream. The extracted analytes were solubilized in 200 μl of *n*-hexane and used for chromatographic analysis (Petropoulou et al., 2006). The recovery rates were higher than 85% for all of the PCB congeners studied.

#### 2.2.2. Chemical analysis

Chromatographic analysis was performed using a Thermo-Finnigan TRACE DSQ GC/MS instrument as previously reported (Petropoulou et al., 2006; Luzardo et al., 2009). A fused silica capillary BPX5 column (Crosslinked 5% phenyl methylpolysiloxane, SGE Inc., USA) with a length of 30 m, an i.d. of 0.25 mm, and a film thickness

**Table 1**
Characteristics of the population, *N* (%)

| | Total | Age (years) | | | | |
|---|---|---|---|---|---|---|
| | | < 20 | 20–34 | 35–50 | 51–65 | > 65 |
| **Characteristic** | 607 (100) | 166 (27.3) | 102 (16.8) | 144 (23.7) | 127 (20.9) | 68 (11.2) |
| **Gender** | | | | | | |
| Male | 284 (46.8) | 86 (51.8) | 48 (47.1) | 58 (40.3) | 58 (45.7) | 34 (50.0) |
| Female | 323 (53.2) | 80 (48.2) | 54 (52.9) | 86 (59.7) | 69 (54.3) | 34 (50.0) |
| **BMI (kg/m$^2$)** | | | | | | |
| < 18.5 | 59 (9.7) | 52 (31.2) | 4 (4.1) | 3 (2.2) | 0 (0.0) | 1 (1.5) |
| 18.5–24.99 | 240 (39.5) | 85 (51.3) | 63 (61.2) | 55 (38.0) | 27 (21.3) | 11 (16.7) |
| 25–29.99 | 199 (32.8) | 27 (16.2) | 28 (27.6) | 59 (40.9) | 55 (43.4) | 29 (42.4) |
| > 30 | 109 (18.0) | 2 (1.3) | 7 (7.1) | 27 (19.0) | 45 (35.2) | 27 (39.4) |
| **Habitat** | | | | | | |
| < 10.000 | 299 (49.3) | 96 (57.9) | 57 (55.4) | 61 (42.6) | 53 (41.3) | 33 (48.5) |
| 10–100.000 | 164 (26.9) | 38 (22.6) | 33 (32.7) | 47 (32.6) | 30 (23.8) | 15 (22.7) |
| > 100.000 | 144 (23.7) | 32 (19.5) | 12 (11.9) | 36 (24.8) | 44 (34.9) | 20 (28.8) |
| **Island** | | | | | | |
| Gran Canaria | 232 (38.2) | 69 (41.6) | 40 (39.2) | 50 (34.7) | 50 (39.4) | 23 (33.8) |
| Lanzarote | 23 (3.8) | 7 (4.2) | 4 (3.9) | 5 (3.5) | 6 (4.7) | 1 (1.5) |
| Fuerteventura | 36 (5.9) | 8 (4.8) | 8 (7.8) | 8 (5.6) | 8 (6.3) | 4 (5.9) |
| Tenerife | 158 (26.1) | 23 (13.9) | 29 (28.4) | 43 (29.9) | 41 (32.3) | 22 (32.4) |
| La Palma | 82 (13.5) | 32 (19.3) | 10 (9.8) | 21 (14.6) | 12 (9.4) | 7 (10.3) |
| La Gomera | 45 (7.4) | 13 (7.8) | 10 (9.8) | 9 (6.3) | 8 (6.3) | 5 (7.4) |
| El Hierro | 31 (5.1) | 14 (8.4) | 1 (1.0) | 8 (5.6) | 2 (1.6) | 6 (8.8) |
| **Smoking habits (> 14 years old, *n*=513)** | | | | | | |
| Non-smokers | 257 (50.2) | 52 (72.5) | 43 (42.6) | 57 (40.0) | 69 (54.0) | 35 (51.7) |
| Ex-smokers | 103 (20.0) | 8 (11.6) | 11 (10.6) | 26 (17.8) | 33 (26.2) | 25 (36.7) |
| Smokers | 153 (29.8) | 12 (15.9) | 48 (46.8) | 61 (42.2) | 25 (19.8) | 8 (11.7) |