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Differential contribution of animal and vegetable food items on persistent organic pollutant serum concentrations in Spanish adults. Data from BIOAMBIENT.ES project

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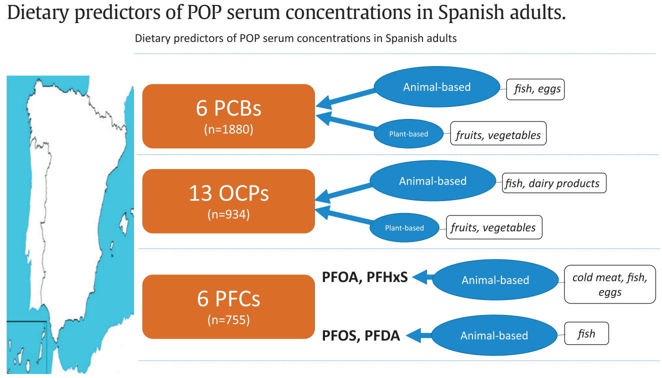
HIGHLIGHTS

- We studied the effect of diet on 26 POP serum levels in Spanish adult population.
- Animal-based food had a 2.1–4.0× stronger effect on most POPs than plant-based food.
- Fish was the main animal-based contributor to OCPs, PCBs, PFOS, and PFDA.
- Cold meat was the main animal-based contributor to PFOA and PFHxS.
- Fruits and vegetables were the main plant-based contributors to PCBs and OCPs.

GRAPHICAL ABSTRACT

Dietary predictors of POP serum concentrations in Spanish adults.



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ABSTRACT

Diet is considered the main source of Persistent Organic Pollutant (POP) exposure in the general population, although there are still several gaps of knowledge regarding the differential contribution of main food groups. The aim of this study was to identify dietary patterns that contribute to human exposure to organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and Perfluoroalkyl Substances (PFASs).

Study population (n = 1880, 18–65 years old) was recruited during 2009–2010 in all the main geographical areas of Spain. For this work, exposure was estimated by chemical analyses of serum levels of 6 PCBs (n = 1880), 13 OCPs (n = 934), and 6 (PFASs) in a subsample of 755 (n = 755). Dietary habits and covariates were gathered via self-administered questionnaires. Data analyses were performed by means of multivariable linear regression and weighted quantile sum regression.

Both the consumption of animal-based and plant-based food were positively associated with the individual concentrations of *p,p´*-DDE, hexachlorobenzene, and PCB-congeners −138, −153, and − 180. The contribution of

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animal-based products was 2.1–4.0× stronger except in *p,p´*-DDE, to which both patterns had similar contributions. In PFASs only animal food was positively associated with the exposure levels. The main animal-based contributors to PCB exposure were fish (49–64%) and eggs (19–36%), while OCP concentrations were mainly influenced by dairy products (32–48%) and fish (47–48%). PFOA and PFHxS were mainly explained by cold-meat (34–37%), fish (25–26%), and eggs (19–21%), while PFOS and PFDA were primarily influenced by fish consumption (44–77%). In the case of plant-based items, fruits (25–82%) and vegetables (18–63%) accounted for the majority of the variability of PCB and OCP concentrations.
Our results highlight the relevance of dietary POP exposure as well as the need for the consideration of nutritional interventions in public health programs aiming to reduce POP exposure in the general population.

1. Introduction

Persistent Organic Pollutants (POPs) constitute a heterogeneous group of chemicals (mostly of man-made origin), which are usually highly resistant to degradation, tend to bioaccumulate in living organisms, and have been used throughout 20th and 21st centuries for very diverse purposes (Porta et al., 2002).

Organochlorine pesticides (OCPs), which are among the most well-known POPs, have been used for both agriculture and public health interventions. One of the most studied OCPs is dichlorodiphenyltrichloroethane (DDT), which has been used extensively worldwide in agriculture and for vector control since 1939 (Turusov et al., 2002). *p,p´*-Dichlorodiphenyldichloroethylene (*p,p´*-DDE) is the main metabolite of DDT, and it is still found as predominant OCP in the general population, and it is even more persistent than the parent compound (Ramos et al., 2017). Hexachlorobenzene (HCB), was used as a fungicide, but other sources of exposure include the emission of chemical industry by-products, combustion reactions, wood preservation agents, and metallurgical processes (Barber et al., 2005). Lindane (γ-HCH) has been used both as an insecticide as well as pharmaceutical treatment for lice and scabies (Vijgen et al., 2011). Polychlorinated biphenyls (PCBs) include a wide group of 209 congeners which have been used in multitude of industrial applications, e.g., in thermal insulation or as coolant (ATSDR, 2000). Although banned in Spain since 1986, they are commonly found in the environment, and living organisms, including non-occupationally-exposed populations. Concentrations of three of these congeners, i.e. PCB-138, PCB-153, and PCB-180, are frequently used as biomarkers of the total exposure of PCBs (Huetos et al., 2014).

Since the early 1970s, most countries have banned or severely restricted the production, handling, and disposal of several (although not all) OCPs and PCBs, due to their high persistence in the environment and their proven or suspected adverse effects, including reproductive disorders, teratogenicity, endocrine disruption, immunosuppression, and carcinogenicity (Arrebola et al., 2014; Luo et al., 2017; Porta et al., 2002; Schæbel et al., 2017). Therefore, exposure levels in the general populations have declined over the last decades (Schoeters et al., 2017). However, virtually all humans have detectable levels of some of these chemicals, and there still exist a wide variety in both inter- and intra-population exposure levels (CDC, 2017), indicating that an important part of the exposure might be modifiable.

Perfluoroalkyl Substances (PFASs) are a class of chemicals with both hydrophobic and oleophobic properties which are used in a variety of consumer products and industrial processes, e.g. liquid repellant, industrial surfactants, or firefighting foams (Schultz et al., 2003). Their ubiquity, persistence and bioaccumulation result in their widespread presence in the environment and the general population, as a consequence of direct emissions during manufacturing, use, or disposal of products as well as transformation of other precursors into PFASs (Armitage et al., 2009; CDC, 2017). It has been suggested that human long-term exposure to low doses of PFASs, particularly at critical stages of development, may increase the risk of certain chronic conditions, e.g. metabolic and thyroid disruption, or inmunotoxicity (Corsini et al., 2014; Heindel et al., 2017; Luo et al., 2017), although results are still controversial.

On the other hand, diet (particularly food from animal-origin) is considered the most important contributor of POP body burden in the general population (Darnerud et al., 2006). However, epidemiological studies have reported controversial findings on which specific food items have the highest contribution to POP exposure, probably related to the specific characteristics of each population, including different life-style and nutritional habits (Boada et al., 2014).

In 2008, the Spanish Ministry of Agriculture, Food and the Environment promoted a national Human Biomonitoring program (HBM). The purpose of this program was to enhance the current understanding of the distribution of priority environmental pollutants, such as metals, pesticides, flame retardants, perfluorinated compounds, and PCBs, in the Spanish population and to establish reference values (Bartolomé et al., 2015; Cañas et al., 2014; Huetos et al., 2014; López-Herranz et al., 2016; Ramos et al., 2017). In this regard, we designed BIOAMBIENT.ES project, a nationwide cross-sectional study, aimed at obtaining a representative sample of the Spanish occupied population.

The aim of the present study was to identify dietary patterns that contribute to human exposure to OCPs, PCBs, and PFASs in a sample of adults representing all the main Spanish regions.

2. Materials and methods

2.1. Study population

BIOAMBIENT.ES is a nationwide cross-sectional epidemiological study with a stratified cluster sampling designed to cover all geographical areas, sex and occupational sectors, and aimed to obtain a representative sample of the Spanish active workforce. The design of the study has been extensively described elsewhere (Pérez-Gómez et al., 2013). In brief, volunteers were consecutively selected among occupationally active population > 16 years, residents in Spain for at least 5 years, which underwent their annual occupational medical check-up between March 2009 and July 2010 in the health facilities of the following Societies for Prevention: IBERMUTUAMUR, MUTUALIA, MC-PREVENCIÓN, MUGATRA, UNIMAT PREVENCIÓN, and PREVIMAC.

A total of 113 Prevention Health Centers were available for the project. Those centers are distributed across the whole country; they provide their services to >436,000 companies in Spain in all activity sectors, with 3,600,000 workers employed within a large spectrum of occupations and occupational categories, and perform >650,000 occupational health exams per year. This high number of annual surveys, as well as its wide geographical coverage could allow us to obtain a fairly representative sample of the Spanish workforce. A sample of 38 Health Prevention Centers was randomly selected across 12 previously pre-defined geographical areas, following a proportional distribution according to data from the Spanish Active Population Survey 2007 (Instituto Nacional de Estadística, 2009). The sample obtained included a total of 1880 men and women aged between 18 and 65. PCBs were analyzed in the 1880 individuals (Huetos et al., 2014), while a subset of 934 were included in the OCPs study (Ramos et al., 2017), and 755 in the PFASs analysis (Bartolomé et al., 2017). The subsamples were