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Temporal trends in concentrations and total serum burdens of organochlorine compounds from birth until adolescence and the role of breastfeeding

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ABSTRACT

Introduction: The aims of the present study are to assess the temporal trends of organochlorine compounds (OCs) concentrations and total serum burdens from birth until adolescence and the influence of breastfeeding in these temporal trends. *Methods*: In 1997 two birth cohort studies were set up in Ribera d'Ebre (N = 102) and the island of Menorca (N = 482), Spain. Concentrations (ng/mL) of OCs [pentachlorobenzene (PeCB), four isomers of hexachlorocyclohexane (HCH), hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (4,4′-DDT), dichlorodiphenyldichloroethylene (4,4′-DDE) and seven polychlorobiphenyl congeners (Σ_7PCBs)] were measured in cord blood and at the age of 4 and 14 years. The total serum burdens (ng) of these compounds were estimated based on the total blood volume (mL) of children at the different ages. We compared median concentrations and total serum burdens of these OCs at the different time-points of follow-up between children of Ribera d'Ebre and Menorca and between breastfed and non-breastfed children. *Results*: From birth until adolescence concentrations of all OCs drastically reduced. These reductions were mainly derived from the dilution of OCs, associated to an increase in total blood volume of children at the age of 4 and 14 years. Despite the reduction in OCs concentrations, the total serum burdens of 4,4′-DDE and Σ_7PCBs, were higher in adolescents than at birth. Increases in OCs total serum burden occurred both in breastfed and non-breastfed children, but were significantly higher in the first. *Conclusions*: Even after decades of banning OCs production and use, current young generations in westernized countries are still bioaccumulating these compounds. Given the potential health effects of OCs, especial attention should be paid in the control of secondary emissions in the environment and in the control of food production and contamination. In countries with endemic malaria it is important to work towards effective alternatives to the use of DDT.

1. Introduction

Organochlorine compounds (OCs) include a wide range of chemicals, such as polychlorobiphenyls (PCBs), dioxins, dichlorodiphenyldichloroethylene (DDE) or hexachlorobenzene (HCB). They are lipophilic synthetic chemicals and belong to the family of persistent organic pollutants (POPs) because they persist in the environment for years and bioaccumulate through the food chain in human and animal fatty tissues (Carpenter, 2011). Through the placenta, humans start being exposed to OCs during prenatal life. After birth, in the first months or years of life, mothers can transfer a certain amount of these compounds through breastfeeding because of the lipophilic properties of POPs

Abbreviations: 4,4′-DDE, dichlorodiphenyldichloroethylene; 4,4′-DDT, dichlorodiphenyltrichloroethane; HCB, hexachlorobenzene; HCH, hexachlorocyclohexane; OCs, organochlorine compounds; PeCB, pentachlorobenzene; PCBs, polychlorobiphenyls; POPs, persistent organic pollutants.

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(LaKind et al., 2004; Ribas-Fito et al., 2005). After breastfeeding, the child continues being exposed to OCs through diet, which currently is the most important source of exposure in the general population (Llop et al., 2010; Vrijheid et al., 2010). Because of their persistency and the health effects associated to OCs exposure, including neurotoxic, immunotoxic and endocrine and reproductive health effects, as well as cancer, the production and use of most OCs are currently banned in the majority of countries, which has led to a general reduction of the levels in the environment and human tissues (Carpenter, 2011). In Spain, these compounds were banned between the early 70s and the late 80s (Ribas-Fito et al., 2005). However, HCB has been unintentionally produced until now as a subproduct of industrial processes. This is the case of the chloro-alkali plant in the village of Flix, area of Ribera d'Ebre, Catalonia, Spain; in this village of around 5000 inhabitants, air and human serum samples collected in the early 90s had the highest levels of HCB ever reported worldwide (Grimalt et al., 1994; Sala et al., 1999, 2001), which made this case unique and particularly relevant at an international level (Ballester et al., 2000; Herrero et al., 1999; Ozalla et al., 2002; Ribas-Fito et al., 2003c; Sunyer et al., 2008). Furthermore, because of the high levels of HCB found in the area, a birth cohort study was set up in 1997 with the aim to study the health effects of prenatal and postnatal exposure to environmental pollutants, particularly OCs and mercury, in children (Guxens et al., 2012). In this same year, another birth cohort was set up in the island of Menorca, Spain, with the aim to study the effects of early life exposure to air-borne irritants and allergens on allergy and asthma (Guxens et al., 2012). In both sites OCs exposure was evaluated at birth and at the age of 4 years (Carrizo et al., 2007b, 2008; Sala et al., 2001) as well as the health effects associated (Ribas-Fito et al., 2002, 2003a,b, 2006a,b, 2007a,b). The inclusion of both birth cohorts in a common study was of interest because of the different exposure scenarios in each site: whereas Ribera d'Ebre was an industrial site with a high production of OCs, Menorca had a rural environment without industrials sites manufacturing this type of chemicals. Thus, children from the latter constituted an example of the background exposure to these pollutants in western countries (Carrizo et al., 2007b). In 2012, when children were around 14 years of age, a new follow-up with a common protocol was performed in both study areas and POPs were analyzed in serum samples of participant adolescents. To our knowledge, no other birth cohort study has assessed exposure to OCs at different ages from birth until adolescence and in two different settings with contrasted sources of exposure and levels of OCs. Thus, the aims of the present study are to assess 1) the current concentrations of OC exposure in adolescents of both birth cohort studies, 2) the temporal trends of OC concentrations and total serum burdens from birth until adolescence and 3) the influence of breastfeeding in these temporal trends. Because OC exposure concentrations at birth and at the age of 4 years and differences between the two birth cohorts were already assessed and discussed in a previous study (Carrizo et al., 2007b), in the present study we will focus on exposure concentrations at the age of 14 years and on the exposure temporal trends from birth up to adolescence.

2. Materials and methods

2.1. Study populations

A total of 102 singleton children born in the main hospital of the Ribera d'Ebre between 1997 and 1999 were included in the Ribera d'Ebre birth cohort study. OCs were measured at three different follow-ups: at birth (cord blood, $N = 73$), at the age of 4 years (serum, $N = 58$, years 2001–2003) and at the age of 14 years (serum, $N = 36$, year 2012). Additionally, at the age of 14 years, schoolmates of children of the original cohort were invited to participate in the study (all children attended the same high school in the village of Flix); 15 accepted to participate and to provide serum samples.

Between 1997 and 1998 the Menorca birth cohort study recruited all women presenting for antenatal care. In total, 482 children were enrolled and OCs were measured at three different follow-ups: at birth (cord blood, $N = 405$, years 1997–1998), at the age of 4 years of children (serum, $N = 285$, years 2001–2002) and at age 14 years (serum, $N = 43$, year 2012). Both birth cohort studies were approved by the ethical committee of the Institut Municipal d'Investigació Mèdica (IMIM, Barcelona) and the hospitals of each area.

2.2. Exposure assessment

OCs in serum samples were analyzed by gas chromatography (GC) with electron capture detection and GC coupled to chemical ionization negative-ion mass spectrometry. All of the analyses were carried out in the Department of Environmental Chemistry (IDAEA-CSIC). Details of the methodology have been reported elsewhere (Carrizo et al., 2006, 2007b; Ribas-Fito et al., 2003b). The limits of detection (LOD) and quantification (LOQ) ranged between 0.01 ng/mL and 0.05 ng/mL depending on the year of analysis and the compound analyzed. Compounds measured in all samples were pentachlorobenzene (PeCB), four isomers of hexachlorocyclohexanes (α-HCH, β-HCH, δ-HCH, γ-HCH), HCB, dichlorodiphenyltrichloroethane (4,4′-DDT), and its main metabolite 4,4′-DDE, and seven PCB congeners (28, 52, 101, 118, 138, 153 and 180), which were summed into one single exposure variable (Σ_7PCBs). The protocol and instruments used were the same in the three follow-ups, and the method performed satisfactorily in repeated international intercalibration exercises within the Arctic Monitoring and Assessment Program (Arctic Monitoring Assessment Programme, 2002).

At the age of 14 years, 23 children from Ribera d'Ebre (3 not belonging to the original cohort) and 43 from the Menorca birth cohort had information on total lipid serum levels. This information was used to calculate OC concentrations in ng/g lipid [based on the equation of (Phillips et al., 1989)] and compare these exposure concentrations with those of previous studies including adolescent population. However, because total lipid data was only available for some of the participants at the age of 14 years, the main results of the present study are based on non-lipid adjusted OC concentrations (ng/mL).

To calculate the total serum burden of OCs (ng) at each age, we estimated the total blood volume (mL) of each child and multiplied it by the concentrations of each OC (ng/mL). The total blood volume was estimated using the body weight and the gender of the child according to different references consulted (Booth; Green; Linderkamp et al., 1977; Stephen, 2011). At birth (cord blood) we estimated 85 mL of blood per kg of body weight, at age 4 years we estimated 75 mL per kg of body weight, and at age 14 years we estimated 65 mL per kg of body weight for girls and 70 mL per kg of body weight for boys.

2.3. Breastfeeding definition

At the age of 1 and 2 years of the child mothers were asked whether they had breastfed their child. In the present study, we classified children as breastfed children (any breastfeeding, independently of the duration) and non-breastfed children.

2.4. Data analysis

OCs exposure concentrations (ng/mL) at each follow-up were calculated for each study population separately (median and the 25th and 75th percentiles). Children of Ribera d'Ebre not belonging to the original cohort were treated separately in order to compare OC concentrations with those of the original cohort. We compared the total serum burden of OCs (median and the 25th and 75th percentiles) between birth cohort studies and between breastfed and not breastfed children. Statistical significant differences ($p \leq 0.05$) were tested with the Kruskal–Wallis test.