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Levels and profiles of brominated and chlorinated contaminants in human breast milk from Thessaloniki, Greece

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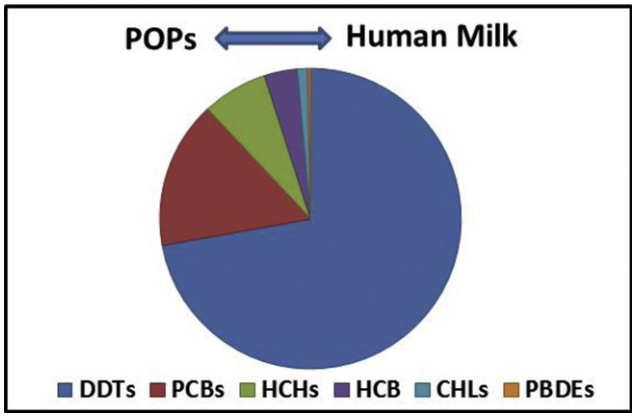
GRAPHICAL ABSTRACT



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ABSTRACT

Human breast milk samples ($n = 87$) collected between July 2004 and July 2005 from primipara and multipara mothers from Thessaloniki, Greece were analysed for six groups of persistent organic pollutants (POPs): polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane and its metabolites (DDTs), chlordane compounds (CHLs), hexachlorocyclohexane isomers (HCHs) and hexachlorobenzene (HCB). DDTs [median: 410 ng/g lipid weight (lw)], PCBs (median: 90 ng/g lw) and HCHs (median: 40 ng/g lw) were the predominantly identified compounds in all the breast milk samples. Levels of PBDEs (median: 1.5 ng/g lw) in human breast milk samples from Thessaloniki, Greece were lower compared to other countries. Maternal age had a positive correlation with most compounds, but not with PBDEs. Women with a higher occupational exposure to PBDEs (i.e., working in office environments) had higher PBDE concentrations than all others and showed strong correlations, especially for BDE 47 and BDE 153. None of the analysed compounds showed any correlation with parity. Based on these levels, the daily intake of each group of POPs via

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human milk was calculated and compared with the tolerable daily intakes (TDI) or the reference doses (RfD). For the majority of samples (85 out of 87) a higher daily intake of PCBs than the TDI was calculated, while 11 out of 87 samples had a higher HCB intake than the TDI. The TDI and the RfD were not exceeded for DDTs and PBDEs, respectively. This is the first report of brominated flame retardants in human breast milk from Greece.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a group of brominated flame retardants (BFRs) that are incorporated into a variety of consumer products, such as furniture foam padding, plastics and textiles to slow down combustion. They have a high degree of lipophilicity and are known to be persistent and bioaccumulative through the food chain (Klosterhaus et al., 2012). PBDEs are not chemically bound to the polymers to which they are added. As a consequence of substantial long-term use, PBDEs have contaminated humans, wildlife, air, water, soils, and sediments, even in remote areas (Hale et al., 2003; de Wit et al., 2006; Law et al., 2006; Covaci et al., 2011; Kalantzi and Siskos, 2011). In recent years, strict bans have been imposed on the worldwide use of Penta- and Octa-BDE formulations and components of these mixtures have been added to the persistent organic pollutants (POPs) list of the Stockholm Convention (Ashton et al., 2009). Being persistent chemicals, PBDEs accumulate in the human body, in breast milk and adipose tissue (Malarvannan et al., 2009, 2013a; Covaci et al., 2008). Despite regulations, PBDEs continue to leach from existing products that are in service or have been disposed of in landfills. Based on a number of recent studies, it has been observed that human non-occupational exposure to PBDEs occurs mainly via a combination of diet, ingestion of indoor dust, and inhalation of indoor air (Roosens et al., 2009; Harrad et al., 2010). The exact contribution of these three pathways varies substantially on a compound-specific basis and between individuals and within national populations (Roosens et al., 2009; Covaci et al., 2011).

Greece depends largely on agriculture, but at the same time has experienced a rapid degree of urbanization since the 1980s. Previous studies have indicated that the general population of Greece has been exposed to legacy POPs such as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) (Schinas et al., 2000; Costopoulou et al., 2006). However, data on POPs levels in humans in Greece is very limited compared to other European countries, and is mostly available for human serum and hair (Kalantzi and Siskos, 2011; Vafeiadi et al., 2014; Covaci et al., 2002; Tsatsakis et al., 2008). Very little information exists on PBDEs in humans in Greece and there is no previous report of PBDEs in human breast milk of Greek women.

In this study, we investigated the levels of PBDEs in human breast milk from Thessaloniki (the second largest urban centre in Greece) in order to assess their contamination status, examine relationships between contaminant levels and parity/age of the mothers and assess intake of contaminants by infants through breast milk consumption. The study also includes data on POPs such as PCBs, dichlorodiphenyltrichloroethane and its metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), chlordane compounds (CHLs) and hexachlorobenzene (HCB) to give a comprehensive picture on the organohalogen contaminants found in breast milk in Greece.

2. Materials and methods

2.1. Sample collection

Human breast milk samples ($n = 87$) from primipara ($n = 34$) and multipara ($n = 53$) mothers from Thessaloniki, Greece were collected between July 2004 and July 2005 and analysed for PBDEs, PCBs, DDTs, CHLs, HCHs and HCB. The mean age of the mothers was 30 years old and ranged from 18 to 43 years of age. About 20 mL of breast milk was collected using a breast pump to transfer the milk into pre-washed glass containers prepared for every individual. The samples were shipped frozen to the laboratory and stored at −20 °C until analysis. Informed consent was obtained from all the donors and ethical approval for this study was obtained from the Ethics Committee of the University of Thessaloniki. Table 1 presents the demographic characteristics of the cohort.

The distributions of milk lipid content were similar in the two groups (primipara and multipara mothers).

2.2. Chemical analysis

The following POPs were targeted: 21 PCB congeners (IUPAC nrs. 99, 101, 105, 118, 146, 153, 138, 187, 183, 128, 174, 177, 171, 156, 180, 170, 199, 196/203, 194, 206 and 209), dichlorodiphenyltrichloroethane and its metabolite (DDTs), three chlordanes (oxychlordane (OxC), trans-nonachlor (TN) and cis-nonachlor (CN), three hexachlorocyclohexane isomers (α-, β-, and γ-HCH), hexachlorobenzene (HCB) and 7 PBDEs (BDE 28, 47, 99, 100, 153, 154 and 183). Analyses of POPs in human breast milk samples were performed according to the methods described elsewhere (Covaci et al., 2008; Malarvannan et al., 2013a) with slight modifications. Breast milk samples (1 to 3 mL) were weighed, mixed with anhydrous sodium sulphate (Na_2SO_4) and then transferred to a mortar and mixed until dry. The samples were transferred to thimbles and were spiked with internal standards (CB 143, BDE 77 and ε-HCH), followed by a 2 h extraction by hot Soxhlet with 100 mL hexane/acetone (1:2, *v*/*v*). The lipid content was determined gravimetrically on an aliquot of the extract (105 °C, 1 h), while the rest of the extract was cleaned on ~8 g acidified silica (44%, *w*/*w*) and eluted with 20 mL hexane:dichloromethane (1:1, *v*/*v*). The cleaned extract was concentrated with a rotary evaporator, further evaporated under a gentle nitrogen stream to incipient dryness and reconstituted in 100 μL of iso-octane. The mixture was transferred to an injection vial for GC–MS analysis. Quantification of POPs was done using GC–MS operated in electron-capture negative ionization mode (Covaci et al., 2008; Malarvannan et al., 2013a) (see Supporting information for technical details, Table SI-1). Abbreviations are expressed as follows:

Table 1
General demographic characteristics of the breast milk donors who participated in this study.

		n	% of total
Maternal age	<20	7	8
[Mean (SD) = 30.2 (6.2) years]	21–25	14	18
	26–30	19	21
	>30	47	53
	Urban	67	77
Area of residence	Rural	20	23
	Housewife	45	52
	Office worker	35	40
Occupation	Other	7	8
	Primiparous	34	39
Parity	Multiparous	53	61
	Omnivore	86	99
Diet	Vegetarian	1	1
	Yes	13	15
Smoking status	No	74	85
	<40 weeks	50	57
Duration of pregnancy	≥40 weeks	37	43