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PBDE levels in breast milk are decreasing in California

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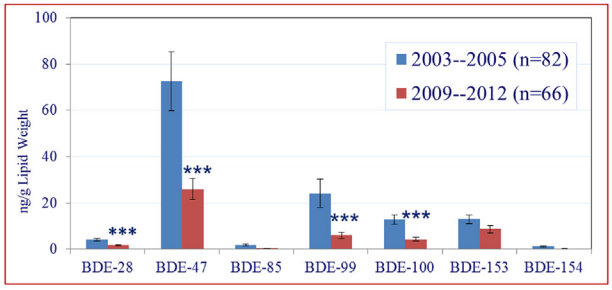
HIGHLIGHTS

- We found significant declines in PBDE and PCB levels between 2003–2005 and 2009–2012 in breast milk of California women.
- We found highly correlated levels and identical congener profiles in breast milk, maternal, and cord blood.
- We confirmed that PBDE levels are decreasing in California after PBDE ban.
- Almost 30% of breastfed babies would be exposed to BDE-47 above the USEPA RfD.

GRAPHICAL ABSTRACT



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ABSTRACT

To assess the efficacy of the bans in reducing PBDE levels, we recruited 67 California first time mothers (sampled during 2009–2012) and collected cord blood at birth (n = 31), breast milk (n = 66) and maternal blood (n = 65) at 3–8 weeks postpartum. Using the same sample extraction procedures and analytical instrumentation method (GC-HRMS), we compared PBDE as well as PCB levels in these breast milk samples to those from our previous study (n = 82, sampled during 2003–2005) and found that the sum of PBDEs over the ~7 year course declined by 39% (GeoMean = 67.8 ng/g lipid in 2003–2005; 41.5 ng/g lipid in 2009–2012) and that the sum of PCBs declined by 36% (GeoMean = 71.6 ng/g lipid in 2003–2005; 45.7 ng/g lipid in 2009–2012). This supports our earlier finding of a PBDE decline (39%) in blood. We also found that the PBDE concentrations and congener profiles were similar in breast milk and their matched maternal/cord blood: BDE-47 was the dominant congener, followed by BDE-153, -99, and -100. Similar levels and congener profiles of PBDEs in these matrices suggest that they are at equilibrium. Therefore, we propose that maternal serum levels may be used to predict an infant's daily dose of PBDE exposure from breastfeeding when breast milk levels are not available. In addition, our study confirmed that breastfeeding babies are still exposed to high levels of PBDEs, even though PBDE levels are decreasing.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been widely used

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as flame retardants (FRs) in commercial and household products since the 1970s. Due to their bioaccumulation, persistence and long range transport, PBDEs have been detected worldwide in humans and in wildlife (McKinney et al., 2011; Andersen et al., 2015; Dorneles et al., 2015; Linares et al., 2015; Wu et al., 2015). Numerous studies have shown PBDEs to cause health problems such as thyroid homeostasis disruption, neurodevelopmental deficits, reproductive changes, and even cancer in animals (Shaw et al., 2010; Abbasi et al., 2015; Linares et al., 2015) as well as lower birth weight, behavioral problems in youth and altered reproductive function in adult humans (Bradman et al., 2012; Eskenazi et al., 2013). Due to these health concerns, PBDE commercial mixtures were banned from all applications in 2004 (penta-BDE and octa-BDE) or 2013 (deca-BDE) in the European Union, the United States and elsewhere (Ward et al., 2008; USEPA, 2009; Ravnum et al., 2012; BSEF, 2015).

As new governmental regulatory policies on PBDEs have been implemented in California (CA) and globally for more than 10 years, serum PBDE levels declined 39% from 2008–2009 to 2011–2012 in CA women (Zota et al., 2013). Declines were also observed in other communities in Australia, European and Asian countries (Toms et al., 2012; Airaksinen et al., 2014; Sun et al., 2015). However, the downward temporal trend was not found in breast milk from a small UK study of cohabiting couples (Bramwell et al., 2014). Moreover, temporal trends have not yet been reported in breast milk in CA.

For several years, we have been studying body burdens of persistent organic pollutants (POPs) in CA women. In 2003–2005 (1st time period), we collected and analyzed breast milk and found PBDE levels to be among the highest in the world (Park et al., 2011). For the current study we collected cord blood at birth, along with maternal blood and breast milk postpartum in 2009–2012 (2nd time period). Our primary objective was to compare changes in the levels of PBDEs and PCBs in CA breast milk from 2003 to 2012. We also compared levels and congener profiles in breast milk and maternal/cord blood.

2. Materials and methods

2.1. Study population and sample collection

We recruited first time mothers who lived in CA for at least three years before giving singleton birth. The 82 participants from the 1st time period (2003–2005) were from birthing and lactation centers in 24 Northern CA communities and the 67 participants from the 2nd time period (2009–2012) were from the Women's Health and Birth Center in the Northern CA city of Santa Rosa. All participants signed an informed consent form at recruitment and completed an exposure questionnaire. This study was approved by the Committee for the Protection of Human Subjects, CA Department of Health Services.

Each participant provided two breast milk samples (~100 mL each) by hand expression (1st time period (Park et al., 2011)) or electric pump (2nd time period) at 3–8 weeks postpartum. Breast milk was collected at one time or over a period of 1–2 weeks and was directly collected or transferred from the pump bottles to 4 oz. solvent rinsed wide mouth amber glass jars (VWR, USA). The breast milk samples were frozen and stored at −20 °C until analysis. During the 2nd time period, we also collected cord blood (n = 31) at birth and maternal blood (n = 65) during breast milk collection. All blood samples were collected in 10 mL red-top blood collection tubes (BD® Vacutainer, USA) by clinic personnel and the serum portions were then separated, frozen, and stored at −20 °C. All breast milk and blood samples were transported to and analyzed by the Environmental Chemistry Laboratory of the CA Department of Toxic Substances Control.

2.2. Sample extraction and instrumental analysis

Breast milk samples: We followed the same extraction and analysis procedures described previously (Park et al., 2011; Zota et al., 2011) and measured 19 PBDEs and 15 PCBs. Briefly, around 100 mL of milk were freeze-dried (Freezemobile, 12SL, VirTis, USA) and 2–3 g of freeze-dried milk were fortified with a panel of ^{13}C labeled surrogate standards (Table S1), extracted by Accelerated Solvent Extraction (ASE 200, Dionex, USA), cleaned-up through acidified silica and gel permeation columns (GPC 717, Envirogel™ cleanup columns: 19 mm × 300 mm and 19 mm × 15 mm with Envirogel™ guard column: 4.6 mm × 30 mm, Waters, USA), and analyzed by gas chromatography/high resolution mass spectrometry using isotope dilution (GC–HRMS, DFS, ThermoFisher, Bremen, Germany). PBDEs and PCBs were measured using two different injections and separated on a DB-5MS column (15 m × 0.25 mm I.D. × 0.1 μm film thickness, Agilent J&W, USA) and a HT8-PCB column (60 m × 0.25 mm I.D × 0.25 μm film thickness, SGE International Pty Ltd., Australia & Pacific Region), respectively. Duplicates, pooled breast milk (QCP), and standard reference material (NIST SRM 1954) were used as QA/QC samples. The method detection limit (MDL) was defined as 3 times the standard deviation of the QCP samples.

Blood samples: We followed the sample extraction and analysis procedure described previously (Whitehead et al., 2015) and measured 5 PBDEs and 18 PCBs. Briefly, 2 mL of serum were fortified with surrogate standards, extracted with Oasis®HLB cartridges (3 cc, Waters, USA) and cleaned up using manually packed acidified silica cartridges (3 cc) on a Biotage® RapidTrace system (Biotage, Sweden). All analytes were separated on a DB-5MS column (30 m × 0.25 mm I.D. × 0.25 μm film thickness, Agilent J&W, USA) and simultaneously measured on a gas chromatograph coupled with a triple quadrupole mass spectrometer (Agilent, USA) using electron ionization and multiple reaction monitoring. Bovine serum pre-spiked with known amounts of target analytes and standard reference material (NIST SRM 1958) were used as QA/QC samples. The method detection limit (MDL) was defined as 3 times the standard deviation of the blank concentrations.

2.3. Lipid measurement

Breast milk: Total lipid content was determined gravimetrically from a portion of the extract (lipid (g) = net weight of fat (g) × 100/(fraction of extract used for fat determination × dry weight of sample extracted (g))).

Blood: All samples were sent to CERLab of Boston Children's Hospital where Cholesterol (CHOL) and triglycerides (TG) were measured using enzymatic methods (Allain et al., 1974; Stinshoff et al., 1977) to calculate the lipid content based on Philips' formula (Phillips et al., 1989).

2.4. Thyroid Stimulating Hormone and thyroxine measurements

Thyroid Stimulating Hormone (TSH) and Thyroxine (Free T4) in serum were measured at Boston Children's Hospital.

2.5. Data analysis

For concentrations below the MDL, we used MDL values divided by the square root of 2 as the estimates. We calculated for all compounds measured: mean and standard error (SE), median and range, geometric mean (GM) and geometric standard error (GSE). Due to their skewed distributions, all measurement data were log-transformed to achieve normal (or more symmetric) distributions as shown by the Chi-square goodness of fit test. Lipid normalized