Biomonitoring polybrominated diphenyl ethers in human milk as a function of environment, dietary intake, and demographics in New Hampshire

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Article info
Article history:
Received 8 February 2010
Received in revised form 19 May 2010
Accepted 7 June 2010
Available online 10 July 2010

Keywords:
Biomonitoring
Human milk
Brominated flame retardants
Depuration
Polybrominated diphenyl ethers
PBDEs

Abstract
Human milk is a valuable biological specimen for biomonitoring lipid-soluble polybrominated diphenyl ethers (PBDEs). The purpose of this study was to determine the levels of PBDEs in human milk from New Hampshire and to examine potential relationships between PBDE levels in human milk and stage of lactation, maternal characteristics, living environment and dietary intake. Forty women provided up to three human milk samples at the end of their first, second and third month of breastfeeding for evaluation of day-to-day and month-to-month variation in PBDE levels. Participants completed four questionnaires, which provided maternal, living environment, and diet information. The PBDE concentrations in human milk over the 3-month collection period ranged from 6.5 to 166.7 ng g⁻¹ lipid. The median for the 3-month period was 29.7 ng g⁻¹. BDE-47 was the predominant congener, however, BDE-153 predominated in 20% of the participants’ samples. Day-to-day variation in PBDEs was negligible; there was no significant difference in mean PBDE levels from month-to-month. Positive associations were seen between BDE-153 and age, postpartum saturated fat consumption, and the home model. There was a negative association between PBDE levels and fruit consumption during the third trimester. Our results indicate that PBDE levels in human milk from New Hampshire are within the range that has been reported in the US, and levels are stable during the first 3-months of lactation. Our findings revealed a higher predominance pattern with BDE-153 compared to other studies, and indicate that PBDE levels are influenced by diet and the home environment.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) are synthetic flame retardants that are added to a wide range of consumer products (e.g. electronics, fabrics and foams) to reduce the likelihood of items catching on fire (Bromine Science and Environmental Forum [BSEF] 2000). PBDEs have been in use since the 1970s, but over the past 15 years they have captured the attention of scientists, policymakers, firefighters, and the general public, worldwide, due to their persistent, lipid-soluble characteristics resulting in bioaccumulation. Over time, PBDEs can diffuse out of consumer products, become airborne or enter dust, and disperse into the environment and into humans. PBDEs have been detected in air, dust, marine mammals, and humans – including human milk (She et al., 2002; Jones-Otazo et al., 2005; Allen et al., 2007; Wu et al., 2007). The appearance of PBDEs in humans raises concern given the potential for PBDEs to cause adverse health effects (Zhou et al., 2001; Lilien-thal et al., 2006; Viberg et al., 2006; Chao et al., 2007; Main et al., 2007). This concern has resulted in biomonitoring initiatives around the globe.

Since the late 1990s, human milk biomonitoring has enabled researchers to document and update human exposure information on PBDE levels in nursing mothers and estimate breastfeeding infants’ intake. In the US, a limited number of studies have reported PBDE levels in human milk, with no documentation to date from the state of New Hampshire. Two studies in the US have reported rates of PBDE elimination in human milk over time in small samples of nursing mothers from California and Pennsylvania (Hooper
et al., 2007; LaKind et al., 2009), and one has demonstrated relationships between PBDE levels in human milk and house dust, dairy intake, and meat intake (Wu et al., 2007).

Given the limited number of studies in the US, there is a need to further document PBDE levels in humans. Our research was designed to contribute to the current body of knowledge about PBDE levels in human milk of lactating women and to examine potential relationships between PBDE levels in human milk and maternal characteristics, stage of lactation, living/occupational environment, and dietary intake. In addition, our research provided new knowledge about PBDE levels in the state of New Hampshire.

2. Materials and methods

2.1. Recruitment

Participants were recruited from southeastern New Hampshire from November 2005 to July 2006 (see Supplemental Materials for additional location information). Women, ages 22–40, who were in the early stages of lactation (less than 2 weeks) and delivered a full-term healthy infant or in their last trimester of pregnancy and planned to breastfeed for at least the first 3-months postpartum, were eligible for the study. The study protocol was approved by the University of New Hampshire (UNH) Institutional Review Board (IRB # 3433) and all participants provided informed consent prior to their enrollment into the study.

2.2. Surveys

Participants were asked to visit with us on four occasions over the 3-month study period, and to complete a total of four surveys. The first survey, Maternal Characteristics Survey, examined potential relationships between PBDE levels in human milk and personal characteristics such as measured body weight, calculated body mass index (BMI), breastfeeding history, physical activity level, and smoking status.

The second survey, a food frequency questionnaire, developed by the Nutrition Assessment Shared Resource (NASR) of Fred Hutchinson Cancer Research Center (FHCRC) (Patterson et al., 1999) was administered twice during the study – at the study’s outset to assess dietary intake during the last trimester and at 3-months postpartum to assess dietary intake during the first 3-months of lactation. One of us (RLD) showed food models to the participants to assist in estimating portion sizes, and completed the questionnaire with the participants on both occasions. The completed food frequency questionnaires were sent to FHCRC for analysis. Prior to analysis, a mixture of three 13C-PBDE injection standards (BDE-77, 139 and 205) was added to give a final volume of 20 μL.

Samples (2 μL) were analyzed for PBDEs on a Hewlett Packard 5890 GC coupled to a VG Autospec mass spectrometer (Micromass, Beverly, MA) using cool-on-column injection onto a 30 m DB-5ms column (0.25 mm internal diameter, 0.25 μm film thickness) and detection at a resolution of >5000 in the selective ion monitoring mode (Huwe and Smith, 2007).

A method blank (water) or spiked sample was run with each set of samples (three human milk samples from one participant) to provide quality control data. Because method blanks contained detectable amounts of several common PBDE congeners, all results were blank subtracted. The limits of detection (LOD) for each congener was calculated from method blanks as three times the standard deviation of the blanks; all values below the detection limit were treated as nondetects (nd) and were set to zero. Typical LODs for BDE-47 and BDE-209 were 20.1 pg g⁻¹ milk and 1759 pg g⁻¹ milk, respectively, and a list of other LODs is provided in the Supplemental Materials. Spiked samples of cow’s milk (n = 14) or water (n = 11) were run interspersed with the samples and demonstrated an accuracy within 17% (relative standard deviations = 7–17%) and precision within 14% (% errors = −12% to +14%) for the major PBDE congeners.

2.3. Human milk sampling

Participants used a personal use electric breast pump to collect human milk samples. One sample was the equivalent of one complete expression from one breast. The collected volume of human milk samples differed from participant to participant based upon the mother’s milk supply. An emphasis was placed on collecting a complete expression to ensure that hind milk was included in the sample, which enabled us to examine lactation-specific maternal characteristics (e.g. percent lipid content in human milk) on whole-weight PBDE levels in human milk. In addition, milk expression occurred between the hours of 6 a.m. and 9 a.m. on the determined collection day with a minimum two hour time frame since the previous feeding from that breast; this protocol standardized the sample collection period to reflect the daily average lipid concentration in human milk. Once milk expression was completed, participants transferred the sample into a four ounce amber glass storage container with a Teflon fitted lid that had been detergent-washed and then rinsed with analytical grade solvents (toluene followed by hexane). To evaluate day-to-day variation in PBDE levels, the first 17 participants provided one sample from three successive mornings at the end of months one, two, and three. Month-to-month variation in PBDE levels was evaluated on all participants by analyzing one sample at the end of months one, two, and three.

2.4. Analytical methods

Analysis of each human milk sample was performed using a modification of previously published methods (Huwe and Smith, 2007; Ryan, 1991; US Environmental Protection Agency, 2003). Forty-six PBDE congeners were quantified in 150 human milk samples by an isotope dilution method using high resolution gas chromatography/high resolution mass spectrometry (HRGC/MS). Prior to extraction, a mixture of 10 13C-PBDE recovery standards (BDE-28, 47, 99, 100, 153, 154, 183, 197, 207, and 209) was added to each sample; at least one recovery standard in each homolog group (tri, tetra, penta, hexa, hepta, octa, nona, and deca). All standards were obtained from either Wellington Laboratories (Guelph, Ontario, Canada) or Cambridge Isotope Laboratories (Andover, MA).

The cleanup of the milk samples is briefly described here. Each human milk sample (5, 7, or 15 mL) was deproteinized with potassium oxalate (20 mg g⁻¹ milk) then partitioned with equal volumes of ethanol and diethyl ether:hexane (2:3, v:v) (Ryan, 1991). After exchange into hexane, the organic fraction was further purified on a Power Prep instrument (Fluid Management Systems, Wallingford, MA) using triphasic silica gel and basic alumina oxide columns. Prior to analysis, a mixture of three 13C-PBDE injection standards (BDE-77, 139 and 205) was added to give a final volume of 20 μL.

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The lipid content of the milk samples was determined gravimetrically after extraction. Results are reported on a lipid-adjusted basis, and as needed, on a whole-weight basis. Using the lipid-adjusted basis allows for normalizing concentrations of PBDEs both within samples from one mother and among samples from many mothers (Needham and Wang, 2002). Additionally, these lipid-adjusted concentrations could be used for infant intake estimates.

2.5. Statistical analysis

PBDE data was log-transformed prior to statistical analysis, and normality of PBDE concentrations was assessed using normal probability plots and Shapiro–Wilk’s test. PBDE concentrations in human milk (monthly means and 3-month means) were evaluated for associations with dietary habits (last trimester and postpartum), maternal characteristics, and living/occupational environments. Potential associations were explored using scatter plots, correlation analysis, and backward regression. Differences in mean PBDE values over time and mean percent lipid levels from months one, two, and three were evaluated using repeated-measures analysis of variance. Regression analysis was used to evaluate the relationship between PBDE levels in human milk and maternal characteristics, living/occupational environments, and dietary intake. Potential confounders were evaluated for associations with exposure variables, and by examining the changes in the β coefficients when excluded variables were added back into the reduced model. Regression coefficients were converted back to the original scale, which provides the fold change (or percent increase or decrease) in human milk PBDE concentrations per unit of exposure. Statistical analyses were performed using the Statistical Package for Social Science (SPSS) 13.0 version with statistical significance set at 0.05.

3. Results

3.1. Participant characteristics

A total of 40 women completed the study with an average age of 31 years (range 22–40). Participants’ current residential locations represented 16 different towns from the southeastern area of New Hampshire (see Supplemental Materials). The highest percentage (25%) of participants resided in Dover, New Hampshire. There was limited diversity among participants; 39 out of 40 participants were Caucasian, and 80% of participants had completed a 4-year college degree or higher (Table 1).

3.2. PBDE levels in human milk

BDE-209 levels varied widely in method blanks resulting in a detection limit that prevented reliable quantitation of BDE-209 in samples. Therefore, BDE-209 was not included in the sum of PBDEs (ΣPBDE). The ΣPBDE congeners found in human milk was defined as: BDE-28/33, 37, 47, 85, 99, 100, 153, 154, and 183. On average, these eight congeners comprised 95.4 ± 6.9% of the total PBDEs quantitated. BDE-47 was the predominant congener for each month and correlated with ΣPBDEs (r = 0.78 p < 0.001). BDE-47 was correlated with BDE-28/33, BDE-85, BDE-99, BDE-100, and BDE-154; however, BDE-47 was not correlated with BDE-153 and BDE-183. BDE-153 was correlated with ΣPBDEs (r = 0.66 p < 0.001) and BDE-100. Log-transformed ΣPBDEs, BDE-47, and BDE-153 levels were evaluated over time (stage of lactation) and for associations with maternal characteristics, dietary habits, and living/occupational environments.

### Table 1

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>n = 40</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maternal age, years (mean, SD)</td>
<td>31 ± 4.5 (range 22–40)</td>
</tr>
<tr>
<td>Calculated BMI (kg m⁻²), starta (mean, SD)</td>
<td>27.3 ± 4.1 (range 20.8–35.9)</td>
</tr>
<tr>
<td>Calculated BMI (kg m⁻²), endb (mean, SD)</td>
<td>26.5 ± 4.4 (range 19.2–36.6)</td>
</tr>
<tr>
<td>Ethnicity (%)</td>
<td></td>
</tr>
<tr>
<td>Caucasian</td>
<td>97.5</td>
</tr>
<tr>
<td>Asian</td>
<td>2.5</td>
</tr>
<tr>
<td>Level of education (%)</td>
<td></td>
</tr>
<tr>
<td>Less than high school graduate</td>
<td>0</td>
</tr>
<tr>
<td>High school diploma or equivalent</td>
<td>2.5</td>
</tr>
<tr>
<td>Some college or technical school</td>
<td>15</td>
</tr>
<tr>
<td>College graduate, 2-year degree</td>
<td>2.5</td>
</tr>
<tr>
<td>College graduate, 4-year degree</td>
<td>42.5</td>
</tr>
<tr>
<td>Masters degree</td>
<td>32.5</td>
</tr>
<tr>
<td>Doctoral degree</td>
<td>5</td>
</tr>
<tr>
<td>Total household income (%)</td>
<td></td>
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<tr>
<td>Less than $20,000</td>
<td>2.5</td>
</tr>
<tr>
<td>$20,000–$34,999</td>
<td>2.5</td>
</tr>
<tr>
<td>$35,000–$49,000</td>
<td>2.5</td>
</tr>
<tr>
<td>$50,000 and higher</td>
<td>92.5</td>
</tr>
<tr>
<td>Smoking status (%)</td>
<td></td>
</tr>
<tr>
<td>Current smoker</td>
<td>0</td>
</tr>
<tr>
<td>Ex-smoker</td>
<td>10</td>
</tr>
<tr>
<td>Lives with smoker</td>
<td>2.5</td>
</tr>
<tr>
<td>First time mothers (%)</td>
<td>77.5</td>
</tr>
<tr>
<td>Parity (years)</td>
<td>1.4</td>
</tr>
<tr>
<td>Exclusively breast feeding, starta (%)</td>
<td>87.5</td>
</tr>
<tr>
<td>Exclusively breast feeding, endb (%)</td>
<td>80</td>
</tr>
<tr>
<td>Vegetariansa (%)</td>
<td>12.5</td>
</tr>
</tbody>
</table>

Abbreviations: n, number of samples; SD, standard deviation; BMI, body mass index.

### Table 2

<table>
<thead>
<tr>
<th>Collection period</th>
<th>Minimum ng g⁻¹ lipid</th>
<th>Maximum ng g⁻¹ lipid</th>
<th>Mean ng g⁻¹ lipid</th>
<th>Median ng g⁻¹ lipid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Month 1</td>
<td>6.8</td>
<td>166.7</td>
<td>37.3a</td>
<td>28.6</td>
</tr>
<tr>
<td>Month 2</td>
<td>6.6</td>
<td>133.7</td>
<td>35.1a</td>
<td>28.1</td>
</tr>
<tr>
<td>Month 3</td>
<td>6.5</td>
<td>116.4</td>
<td>34.1a</td>
<td>32.8</td>
</tr>
</tbody>
</table>

Abbreviations: n, number of samples.

### Table 3

<table>
<thead>
<tr>
<th>Stage of lactation and sum of PBDEs</th>
</tr>
</thead>
<tbody>
<tr>
<td>There was no significant difference in mean ΣPBDE levels from months one, two, and three (Table 2). The relationship between ΣPBDE levels and time did not change based on age, BMI, or number of other prior children breastfed. The ΣPBDE concentrations in human milk over the 3-month collection period ranged from 6.5 to 166.7 ng g⁻¹ lipid. The mean for the 3-month period was 35.5 ng g⁻¹ with a median value of 29.7 ng g⁻¹ lipid.</td>
</tr>
</tbody>
</table>

Day-to-day variation in PBDE levels was analyzed early on in the first five out of 17 participants (nine samples per participant). The variation from day-to-day revealed that 41 out of 45 samples had less than a 20% change in the ΣPBDE concentrations from day one to day two, day two to day three, and day one to day three. Because methodological variation was 20%, it was concluded that day-to-day variation was minimal. From this point forward, one sample per month (day one) from each participant was collected and analyzed.
3.4. Stage of lactation and congeners

The median congener pattern of human milk samples from month one was 47% BDE-47, 17% BDE-153, 9% BDE-100, 8% BDE-99, and 19% other congeners. The median congener pattern from month two was 44% BDE-47, 15% BDE-153, 9% BDE-99, 7% BDE-100, and 25% other congeners. Lastly, the median congener pattern from month three was 38% BDE-47, 14% BDE-153, 8% BDE-99, 6% BDE-100, and 34% other congeners (Fig. 1). There were no significant differences in mean BDE-47, BDE-153, or BDE-100 levels from months one, two, and three. The relationship between the congener levels and time did not change based on age, BMI, or number of other prior children breastfed. BDE-47 was the predominant congener in 80% of the samples. However, BDE-153 was the predominant congener in 20% of the participants’ samples from each month. A summary of PBDE concentrations are shown in Table 3.

3.5. Percent lipid content of human milk

The mean lipid content in human milk for month one was 3.3% (range 1.6–5.0%), for month two was 3.3% (range 1.1–8.5%), and for month three was 3.0% (range 0.9–6.8%). There was no significant difference in mean percent lipid values from months one, two, and three. The relationship between the congener levels and time did not change based on age, BMI, or number of other prior children breastfed. BDE-47 was the predominant congener in 80% of the samples. However, BDE-153 was the predominant congener in 20% of the participants’ samples from each month. A summary of PBDE concentrations are shown in Table 3.

3.6. PBDE levels in human milk and maternal characteristics

Some of the data from the Maternal Characteristics Survey indicated limited variability among participants, including education, ethnicity, total household income, and health history information. There were no associations between \( \Sigma \)PBDE levels and age, parity, body mass index, pre-pregnancy weight, number of other children breastfed, physical activity level, weight lost on a diet, vegetarian status, smoking history, or the estimated amount of supplemental formula provided to the infant during the study. Similar findings were also seen with congener (BDE-47 and BDE-153) levels and maternal characteristics, however, an association was seen between BDE-153 and age (\( r = 0.36 \), \( p = 0.02 \), participant 3-month means). For each unit (year) increase in age, there was a 1.1-fold change (or 10% increase) in BDE-153; however, the association between age and BDE-153 weakened after adjusting for change in BMI from the start of the study until the end (\( r = 0.36 \), \( p = 0.07 \)).

3.7. PBDE levels in human milk and dietary intake

There were no significant associations between PBDE levels (\( \Sigma \), BDE-47, and BDE-153) and the majority of nutrients consumed during the last trimester or 3-month postpartum period. However, there was a small, but significant association between BDE-153 and saturated fat consumption during the 3-month postpartum period. For each unit (gram) increase in daily saturated fat intake, there was a 1.04-fold change (or 4% increase) in BDE-153 levels (monthly means and participant 3-month means) (\( r = 0.31 \), \( p = 0.05 \), participant 3-month means) (Fig. 2). The association between saturated fat intake during the postpartum period and BDE-153 persisted after adjusting for total meat consumption during the postpartum period (\( r = 0.41 \), \( p = 0.03 \), participant 3-month means) (Table 4). There was no association between saturated fat consumption and BDE-153 during the last trimester.

### Table 3

Summary of PBDE congener concentrations (pg g\(^{-1}\) lipid) in 40 samples of human milk, first month collections.

<table>
<thead>
<tr>
<th>PBDE number</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Mean</th>
<th>Median</th>
</tr>
</thead>
<tbody>
<tr>
<td>28</td>
<td>305.7</td>
<td>5375.1</td>
<td>1764.2</td>
<td>1495.0</td>
</tr>
<tr>
<td>47</td>
<td>3652.5</td>
<td>59001.4</td>
<td>17466.8</td>
<td>13362.5</td>
</tr>
<tr>
<td>85</td>
<td>nd</td>
<td>902.8</td>
<td>180.5</td>
<td>114.4</td>
</tr>
<tr>
<td>99</td>
<td>nd</td>
<td>6576.1</td>
<td>2299.2</td>
<td>2016.2</td>
</tr>
<tr>
<td>100</td>
<td>395.0</td>
<td>15323.1</td>
<td>3384.5</td>
<td>2538.5</td>
</tr>
<tr>
<td>153</td>
<td>745.5</td>
<td>109601.3</td>
<td>11897.7</td>
<td>4949.8</td>
</tr>
<tr>
<td>154</td>
<td>nd</td>
<td>620.7</td>
<td>139.8</td>
<td>102.4</td>
</tr>
<tr>
<td>183</td>
<td>nd</td>
<td>2345.9</td>
<td>116.4</td>
<td>nd</td>
</tr>
<tr>
<td>% Lipid</td>
<td>1.60%</td>
<td>5.01%</td>
<td>3.33%</td>
<td>3.27%</td>
</tr>
</tbody>
</table>

Abbreviations: nd, not detected (below the limit of detection).
The frequency of meat, fish, dairy product, egg, fat/oil, and vegetable consumption during the last trimester and 3-month postpartum period was not associated with PBDE levels (P, BDE-47, and BDE-153). However, the strongest association between PBDE levels and diet was seen with total fruit consumption in the last trimester of pregnancy (Fig. 3). For each unit (serving) increase in fruit, there was a 0.87-fold change (or 13% decrease) in PBDE levels (monthly means and participant 3-month means) (r = 0.36 p = 0.02, participant 3-month means). The association persisted between fruit consumption during the last trimester and PBDE levels after adjusting for total calorie and carbohydrate consumption during the last trimester (r = 0.56 p = 0.02, participant 3-month means) (Table 4). Similar findings were also seen with BDE-47 and fruit consumption in the last trimester. The association was not seen with total fruit consumption in the 3-month postpartum period.

3.8. PBDE levels in human milk and maternal environment

There were no associations between PBDE levels in human milk and geographical location (e.g. town vs. city), subject occupation, transportation (e.g. year automobile was purchased, hours per week in the car), and other possible PBDE exposures (e.g. hobbies that involved using fabric). Usage of household appliances and electronic equipment was not associated with PBDE levels in human milk, nor was their year of purchase. The sum of cushioned furniture in the home was not associated with PBDE levels in human milk, nor was the year of purchase. Similar findings were also seen with congener (BDE-47 and BDE-153) levels and living/occupational characteristics; however, an association was seen between BDE-153 and the home model (r = 0.51 p = 0.004) which included the total number of rooms in the home with curtains/valences and total number of rooms in the home with carpeting. As the number of rooms with curtains/valences increased, there was a 1.24-fold change (or 24% increase) in PBDE levels (participant 3-month means) and as the number of rooms with carpeting increased, there was a 0.86-fold change (or 14% decrease) in PBDE levels (participant 3-month means). The association persisted between the home model and BDE-153 after adjusting for total rooms in the home and total months at current residence (r = 0.53 p = 0.02, participant 3-month means).

4. Discussion

4.1. BDE-153: congener patterns

Even though BDE-47 was the predominant congener in most of our participants’ human milk samples, a predominance of BDE-153 was observed in 20% of the study population – the highest percentage in the US compared to 7% (Wu et al., 2007), 8% (She et al., 2007), and 10.5% (Sjödin et al., 2008) in earlier studies. Two European studies that have documented BDE-153 as the predominant congener in human milk samples report high seafood consumption in their subjects (Fängström et al., 2005; Ingelido et al., 2007); however, an association between BDE-153 and seafood was not seen in these two studies. It is noted that the congener profile for seafood items in these European regions do not reflect BDE-153...
as the predominant congener, which is a similar observation by Ohta and colleagues (2002) from Japan. The source of BDE-153 is uncertain at this time given the lack of association with seafood.

Correlation analysis revealed that BDE-153 had no correlation to BDE-47. BDE-153 is considered to be a minor component of the pentaBDE and octaBDE commercial products (La Guardia et al., 2006). However, the estimated half-life of BDE-153 is three times longer than that of the BDE-47 congener (6.5 years vs. 1.8 years, respectively) (Geyer et al., 2004), which suggests that over time, BDE-153 will become the predominant congener in the body. The apparent increase in the predominance of BDE-153 in human milk samples from New Hampshire is suggestive of a shift in congener patterns, which could be related to when our samples were collected (2005–2006) relative to other studies (e.g. 2004–2005) (Wu et al., 2007). It is also noted in recent work by Zhu and colleagues (2009) that BDE-153 was a predominant congener and accounted for 26% of the total PBDE concentrations in Chinese human milk samples.

4.2. BDE-153 and age

Our finding of an association between age and BDE-153 levels is the first human milk biomonitoring study to document this positive relationship in 22–40 year old women, which may be an indicator of the long half-life of this congener paired with the physiological demands of lactation on the body. However, we noted that the association between age and BDE-153 weakened when we accounted for changes in body composition due to weight changes. Sjödin and colleagues (2008) reported a linear decrease and positive quadratic trend with age and serum concentrations of BDE-153 in human milk samples from New Hampshire is suggestive of a shift in congener patterns, which could be related to when our samples were collected (2005–2006) relative to other studies (e.g. 2004–2005) (Wu et al., 2007). It is also noted in recent work by Zhu and colleagues (2009) that BDE-153 was a predominant congener and accounted for 26% of the total PBDE concentrations in Chinese human milk samples.

4.3. PBDE levels and exposure sources

We found an association between the home model and BDE-153. It is unclear from our results whether or not the household items themselves (e.g. curtains/valences) or the dust they collect are the sources of the exposure, or if there was an association between household dust concentrations and PBDE sources (e.g. furniture, curtains, electronics) as recently described by Allen et al. (2008). We were not able to make this distinction because we did not sample house dust from the homes of our participants nor did we measure bromine concentrations in consumer products.

We were surprised with the limited findings between PBDE levels and dietary intake given the work by Wu and colleagues (2007) who reported positive associations between PBDE levels in human milk and the consumption of dairy products and meat pre-pregnancy. Our limited findings may be attributed to the study's small sample size and our choice of timing for collecting dietary information (two 3-month periods). We selected the two time periods because pregnancy and the postpartum period can cause changes in dietary behavior (Devine et al., 2000; George et al., 2005). Perhaps if we expanded our time frame for assessing dietary intake (e.g. throughout the entire pregnancy), we may have encountered different results because the half-lives of many congeners are on the magnitude of years. However, we did find a small, but significant, positive association between BDE-153 levels and saturated fat intake during the 3-month postpartum period. This finding was not seen during the last trimester. The significant finding during the postpartum period may reflect the metabolic demands that are placed on the body by the process of lactation. BDE-153 is present in foods that contain saturated fat (e.g. meat, meat fat, dairy) based on the findings from market basket surveys in the US (Schecter et al., 2004; Huwe and Larsen, 2005; Schecter et al., 2010), however, it is not a predominant congener in these food samples. In addition, Fraser and colleagues (2009) recently reported that poultry fat intake and red meat fat intake (i.e. saturated fat) were significant determinants of serum levels of PBDE congeners including BDE-153. Fraser et al. (2009) also noted that there was
a stronger association with red meat fat intake and BDE-153 as compared to other congeners.

To our knowledge, this is the first study to find a significant, negative relationship with food consumption and PBDE levels in human milk. The association between fruit consumption and PBDE (Σ and BDE-47) levels during the last trimester suggests that fruit may contribute less to PBDE body burden. Our finding with fruit consumption corresponds with vegetarian and vegan trends that were observed by Fraser et al. (2009) and Schecter et al. (2006), respectively, where serum levels of PBDEs were lower in those participants who abstained from consuming foods of animal origin. However, we were surprised with our lack of findings between vegetable consumption and PBDE levels.

4.4. Strengths and limitations of our research

There were several strengths to our study. First, the collection of serial human milk samples enabled us to evaluate changes in PBDE levels over time. Our results indicate no significant difference in mean PBDE levels from months one, two, and three, which was similar to findings by LaKind et al. (2009) and Hooper et al. (2007). LaKind and colleagues (2009) found no consistent pattern in the increase or decrease of the chemicals tested (including PBDEs) in human milk samples from months one to three in their 10 participants. Hooper and colleagues (2007) found that the 0–28 d after birth group was not different from the 29 to 56 d after birth group. Additionally, over the long-term, Hooper and colleagues (2007) found that depuration of PBDEs by lactation is slow in primiparae, averaging 1–3% per month or a 12–18% decrease after 6 months. Consequently, it appears that one human milk sample during the first 3-months postpartum would estimate PBDE levels.

An additional strength to our study was a standardized milk collection protocol and the collection of a complete expression, which provided information on these parameters and their influence on lipid-adjusted PBDE levels in human milk. There appears to be a gap in the biomonitoring literature evaluating if the time of day and/or quantity of milk collected (e.g. complete expression vs. foremilk expression) influences lipid-adjusted PBDE levels in human milk. It is noted that collection amounts vary in human milk biomonitoring studies and most likely collection times. For example, studies document collecting anywhere from 30 mL to 100 mL of human milk from participants without documentation on what time of day the sample was collected. However, median ΣPBDE and BDE-47 levels are similar between Boston, Massachusetts (Wu et al., 2007) and our work suggesting that a standardized timing protocol for milk collection and a complete expression may not be necessary. This gap in knowledge could be answered in future human milk biomonitoring research of PBDEs.

We also viewed our survey work as strengths to our study because this evaluated potential exposure sources. To our knowledge, this is the first study to inquire about year of purchase on items in the mother’s environment that are part of her day-to-day living, such as electronics, appliances, and cushioned furniture. We also standardized lifestyle and living environment questions from the Greater Boston PBDE Body Burden Project (Wu et al., 2007). Although we had a different population in our study, we had similar findings for questions such as smoking, occupational, and hobby exposures.

The use of a validated food frequency questionnaire was a strength to our study; we were confident with the accuracy of the instrument used to assess dietary intake based upon its validation for the Women’s Health Initiative (Patterson et al., 1999). In addition, we obtained a comprehensive analysis of 125 nutrients, and quantified the frequency and amount of food consumption for 27 selected questions, which provided a substantial amount of dietary information – an essential aspect given that diet is considered to be an important source of exposure.

In general, collecting dietary information can pose a challenge in obtaining accurate information due to prevarication bias, the reliance on memory, and the estimation of portion sizes. As noted by Willet (1990), food frequency questionnaires have their drawbacks including restrictions in collecting information with fixed food lists, which we encountered as we were unable to distinguish between kinds of dairy fat consumed because some participants included multiple types. Because Wu and colleagues (2007) found an association between ΣPBDE levels in human milk and consumption of dairy fat, this would have been an important aspect to evaluate from our data.

Our findings are limited due to the small sample size and our specific population of nursing mothers, therefore our results may not be applicable to the general population. As noted from previous research, infants ingest more PBDEs compared to adults (Jones-Otazo et al., 2005; Schecter et al., 2006), and males are 2.1 times more likely to be above the 95th percentile for BDE-153 as compared to females (Sjödin et al., 2008).

5. Conclusions and future directions

This was the first study in the state of New Hampshire to document and evaluate PBDE levels in nursing mothers over time. Our results indicate that PBDE levels from New Hampshire are within the range that has been reported in the US, and that levels are stable during the first 3-months of lactation. Our findings revealed a higher predominance pattern with BDE-153 compared to other studies, which may be reflective of its long half-life. Our data indicates that PBDE levels are influenced by diet and the home environment, contributing to the limited body of knowledge on exposure sources of PBDEs. The negative association between fruit consumption and PBDE (Σ and BDE-47) levels during the last trimester was surprising, and suggests that fruit may contribute less to PBDE levels in the body.

In the future, it will be important to document time-trends of PBDEs given the phase-out of pentaBDE and octaBDE commercial products and to further evaluate geographical trends in the US. Lastly, further research should assess for potential health consequences associated with PBDE body burden and the nursing infant, with a focus on the toxicology and long half-life congeners BDE-153.

Funding sources

New Hampshire Agricultural Experiment Station Grant H-485; University of New Hampshire President’s Research Fund for Excellence; and the American Dietetic Association Foundation. This is Scientific Contribution Number 2430 from the New Hampshire Agricultural Experiment Station.

Acknowledgements

Enormous thanks to: Kristin McDonald, Jean Picard, Margaret Lorentzsen, Shanna Miller, Kathy Kendall-Tackett, Aviva Meyer, Sarah Phillips, the New Hampshire Breastfeeding Task Force, Seacoast area lactation consultants, and our participants.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2010.06.017.
References


