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Original article

# Decabromodiphenyl ether in breast milk collected from Saudi mothers

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ABSTRACT

*Objectives:* This study presents for the first-time temporal changes of decabromodiphenyl ether (deca-BDE) in Saudi human breast milk samples.
*Methods:* Seventy-five samples were taken, and then the extracted and cleaned samples were put via solid-phase extraction (SPE) before being analyzed with gas chromatography-mass spectrometry.
*Results:* In this research, women eat more meat (69%), followed by eggs (50%), and milk (36%). The bulk of donors (44%) and eggs (33%) eat fish and eggs twice a week. The most of the moms who took part had finished higher education (68%). BDE-209, -28, -138, and -208 were the predominant congeners. BDE-209 was detected in all analyzed samples (median concentration 0.19 ng/g lw) suggesting recent exposure of mothers to deca-BDE formulations. The levels of BDE-209 (the most Carcinogenic agent type) were the highest in breast milk were higher (2.5–8 Fold) the levels of other BDES. Higher rates of egg consumption were positively associated with higher breast milk levels of BDE-28 (P = 0.01).
*Conclusions:* Increased exposure to BDE-209 raises health concerns since its breakdown may result in lower brominated congeners and/or other products, which are more toxic than the parent compound. Furthermore, Eggs could be a way for these congeners to get into the bodies of mothers. Also, our results suggest that mothers in Saudi Arabia get BDE-28 from sources other than their jobs, like eating eggs. More research needs to be done to find out if chickens raised in Saudi Arabia have eggs with a lot of deca-BDE.


## 1. Introduction

Polybrominated diphenyl ethers (PBDEs) are chemical pollutants. They are added & during the manufacture of electronic equipment such as Computers and mobile phones and also in the interior parts of cars and airplanes. These pollutants leach into the environment by volatilization and are known to have a high bioaccumulation capacity through the food chain (Babichuk et al., 2022; Zhao et al., 2022). PBDEs are known to cause maternal toxicity at and are detected in breast milk; with potential fetal toxicity (Hites, 2004). Toxicity may affect serval organs such as the thyroid, liver, kidney, and nervous Systems. These pollutants are also potentially Carcinogenic (Sun et al., 2013). The level of exposure to PBDEs are expected to be affected by two main factors: the close proximity to products containing PBDEs and their level in these products.

PBDEs have 209 congeners and are distributed into three marketable products (penta-, octa- and deca-BDE) depending on the number of bromine atoms in their chemical structure (Birnbaum and Cohen Hubal, 2006). The penta-BDE formulation. primarily consists of BDE-47 and -99; the octa-BDE formulation consists of a mixture of *hexa*-to nona-BDEs, while the deca-formulation comprises 98% BDE-209 and various nona-BDEs (Miller et al., 2012). Usage of the penta-BDE formulation has been banned in many countries worldwide since 2004 but the deca-formulation continued to be used until 2013 (Darnerud et al., 2015; Guo et al., 2016).

Deca-BDE is used in conjunction with antimony trioxide as a flame retardant in the housings of electronic equipment (Morf et al., 2005). Based on animal studies, the possible health effects of decaBDE in humans involve the liver, thyroid, reproductive/de-

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velopmental effects, and neurological effects (Agency. January, 2006). Deca-BDE is classified as a possible human carcinogen by EPA. In the environment, Dcca-BDE can be debrominated to fewer bromine atoms products which may have higher (Zhang et al., 2017). In May 2009, commercial octa-BDE was added to the Stockholm Convention as it meets the criteria for the so-called persistent organic pollutants. There are currently no restrictions on the manufacture or use of PBDEs in Asian countries. Breast milk and house dust have been indicated as the possible main exposure pathways for PBDEs ingested or inhaled by breastfed infants after delivery (Jones-Otazo et al., 2005).

The aim of the current study is to investigate the levels deca-BDE in breast milks of Saudi lactating mothers for the first time in Saudi Arabia.

## 2. Materials and method

### 2.1. Patients and sample collection

In this perspective study, ethical approval was obtained from institutional review board of King Khalid University Hospital, Riyadh (ref, No. 21/0044/IRB). Inclusion Criteria were: lactating mothers with overall normal health condition and who are willing to participate in the study. Exclusion criteria were mothers with underlying health problems and those who were not willing to participate. Included 75 mothers were fully informed with the nature and objectives of the study and signed a consent form. A self-reported questionnaire regarding age, height, weight, diet, residence, and parity was used. About 100 ml breast milk were collected in a clean glass. Samples were immediately Ferzan (at −80 °C).

### 2.2. Samples analysis

PBDEs Extraction and clean up from breastmilk was don done according to (Pajewska-Szmyt et al., 2019). The procedure allowed us to obtain recovery values between 96.46% and 119.98% with acceptable relative standard deviations (3.36–12.71%). The method was validated using parameters such as linearity, limit of detection and quantification (LOQ), intra-day precision, and reproducibility. PBDEs analysis were obtained using GC–MS instrument. Compound quantification based on the signals in the mass chromatograms and comparisons with internal standards. Duplicate samples, blanks and spiked samples were processed and analysed with each batch of samples. Quality assurance were maintained throughout the course of the project using standard quality assurance procedures and documentation.

### 2.3. Sample size calculation

Sample size calculation was done using Raosoft online to specify the number of subjects needed with an error margin to meet the desired confidence level. In order to obtain a confidence level of 95% and a 3.15% margin of error, a minimum sample size of 100 would enable us to achieve the study objectives.

### 2.4. Statistical analysis

Statistical analysis was applying to calculate minimum, maximum, mean, standard deviation and confidence levels. Data were presented as mean ± SD and median (Q1-Q3) Non-parametric tests were used to investigate these values according to the characteristics of the participants. Most of the analyses (96.2%) were above LOQ, values below the LOQ were treated as ½ of these limits in calculating the means. A P-value less than 0.05 was considered significant. Further analysis was done using multiple linear regression with a significant p value.

## 3. Results and discussions

### 3.1. Maternal characteristics

Mothers' demographic data (age, parity, employment, smoking, diet, etc.) is shown in Table 1. The majority (about 90%) of mothers were over 26 years old with average age of 31.4 years. almost all residence was in Riyadh. People who live in cities may be more likely to be exposed to flame retardants at home or at work, which could lead to higher PBDEs levels. Majority of mothers (about 90%) had a BMI less than 30 with average 25.6 ± 3.0 kg/m$^2$. About 90% of mothers were primiparous. In this study, the mothers ate more

**Table 1**
Patients Characteristics.

| Characteristic | Mean or N | SD or % |
|---|---|---|
| Age | 31.4 | 4.4 |
| 21–25 | 6 | 8.00% |
| 26–30 | 27 | 36.00% |
| >30 | 42 | 56.00% |
| Mean BMI, kg/m2 (SD) | 25.6 | 3 |
| <25 | 27 | 37.00% |
| 25-29.9 | 41 | 56.20% |
| 30-34.9 | 3 | 4.10% |
| 35-39.9 | 2 | 2.70% |
| Urban residence | 74 | 98.00% |
| ≤5 | 1 | 1.3 |
| >5 | 74 | 98.70% |
| Rural | 1 | 1.30% |
| ≤5 | - | |
| >5 | 1 | 100.00% |
| Education | | |
| ≤High school | 24 | 32.00% |
| College | 8 | 10.70% |
| ≥Bachelor's degree | 43 | 57.30% |
| Birth order | | |
| First | 68 | 90.70% |
| Second or higher | 7 | 9.30% |
| Parity | | |
| Primiparous | 68 | 90.70% |
| Multiparous | 7 | 9.30% |
| Diet (before pregnancy) | | |
| Omnivore | 74 | 98.70% |
| Vegetarian | 0 | 0 |
| Weekly Egg Consumption | | |
| Never | 0 | 0 |
| Sometimes | 4 | 5.6 |
| Once a week | 3 | 4.2 |
| Twice a week | 24 | 33.3 |
| More than twice a week, but not every day | 38 | 50.7 |
| Every day | 3 | 4 |
| Weekly Fish Consumption | | |
| Never | 6 | 8 |
| Sometimes | 16 | 21.3 |
| Once a week | 19 | 25.3 |
| Twice a week | 33 | 44 |
| More than twice a week, but not every day | 1 | 1.3 |
| Every day | 0 | 0 |
| Weekly Meat Consumption | | |
| Never | 0 | 0 |
| Sometimes | 0 | 0 |
| Once a week | 1 | 2.7 |
| Twice a week | 4 | 5.3 |
| More than twice a week, but not every day | 52 | 69.3 |
| Every day | 18 | 24 |
| Weekly Milk Consumption | | |
| Never | 0 | 0 |
| Sometimes | 2 | 2.7 |
| Once a week | 0 | 0 |
| Twice a week | 2 | 2.7 |
| More than twice a week, but not every day | 27 | 36 |
| Every day | 44 | 58.7 |

meat (69%), then eggs (50%), and then milk (36%). Most donors ate fish (44%) and eggs (33%) two times a week. Most of the mothers who took part had graduated from college (68%).

### 3.2. Levels and congener profiles of Deca-BDE

The median (Q1-Q3) and average (SD) concentrations of congeners in the breast milk samples are shown in Table 2. The repeated-measures ANOVA results indicated that the congener concentrations were same across the three samples. Among PBDEs, BDE-209, -28, -138, and -208 were the predominant congeners. The levels of the deca BDE-209 (The most Carcinogenic agent type) were the highest in breast milk were higher (2.5–8 Fold) the levels of other PBDEs.

The level in our study is still lower to compound in other countries (Hites, 2004) but it is alarming that the Deca BDE-209 is the highest level any other types of PBDEs (Table 2), Other Countries showed that BDE -209 is found at a significantly lower concentration than other brominated PBDEs (Jakobsson et al., 2012). As Mentioned in the introduction, BDE–209 is carcinogenic compound. PBDEs are commonly used in electronics as flame retardants. USA, Canada, and several Europeans countries banned the Production of several BDE (Noren and Meironyte, 2000). This is not the care in other countries like china (Pajewska-Szmyt et al., 2019).

The predominance of BDE-209 in human milk was also found in previous study (Shi et al., 2013) and many other studies from China (Yang et al., 2016; Li et al., 2017). BDE-209 was also the most abundant congener in human milk from a few European countries, including Belgium and Sweden (Croes et al., 2012; Darnerud et al., 2015). After Penta-BDE and Octa-BDE were phased out, Deca- BDE has become the only commercial PBDEs produced and used in China in the past several years. Because BDE-209 is the main component in commercial Deca-BDE (N95%), we infer that the wide and heavy industrial usage of commercial Deca-BDE should be the primary source for the predominance of BDE-209 in this study.

Most of the electronics in Saudi Arabia are imported from China and that is another concern. A third Concern is the recent increase of involvement of females in industry in Saudi Arabia and hence the levels of BDEs are expected to rise over the next decade. All these concerns dictate further investigations in the future and our study results can be used as a base-line for future studies.

Increased exposure to BDE-209 raises health concerns since its breakdown may result in lower brominated congeners and/or other products, which are more toxic than the parent compound (Darnerud et al., 2015). In addition, higher brominated congeners less readily transfer from blood to breast milk compared to the lower brominated congeners (Mannetje et al., 2013; Thomsen et al., 2010). Therefore, the levels of BDE-209 in breast milk may underestimate the actual body burden of this compound, since serum levels may be higher than those in breast milk.

On the other hand, because the partitioning of BDE-209 between serum and human milk is quite low (serum/milk ratio N 10), the proportion of BDE-209 in human milk would be accordingly low (Inoue et al., 2006; Mannetje et al., 2012). Thus, we infer that the predominance of BDE-209 in human milk might suggest that the Saudi population is continuously exposed to elevated Deca-BDE levels.

**Table 2**
Mean and Median concentration of BDEs (ng/g lw).

| | Mean ± SD | Median (Q1 - Q3) | N < LOD |
|---|---|---|---|
| BDE-209 | 0.8 ± 0.5 | 0.9 (0.2–1.2) | 63 (84.0) |
| BDE-28 | 0.1 ± 0.1 | 0.1 (0.1–0.2) | 2 (2.7) |
| BDE-138 | 0.3 ± 0.1 | 0.2 (0.2–0.3) | 62 (82.7) |
| BDE-208 | 0.2 ± 0.1 | 0.2 (0.1–0.3) | 2 (2.7) |

Note: Data presented as mean ± SD and median (Q1-Q3); LOD indicates limit of detection.

### 3.3. Association of deca-BDEs levels with maternal characteristics and dietary habits

Table 3 shows the correlation between maternal features and maternal nutrition and deca-BDEs congers levels. There was no statistically significant correlation between maternal age and deca-BDEs congeners. In contrast to other persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), PBDEs have not been observed to accumulate with maternal age (Dimitriadou et al., 2016). This could be because PBDEs have only recently been made and used, which means that people are exposed to more of them than to other POPs whose production and use have been banned for a long time (Muller et al., 2016). Also, it was suggested that the lack of a link between deca-BDEs levels and age could be due to a behavioral cohort effect, in which younger women may be more likely to use consumer products with PBDEs or eat more foods with PBDEs. Also, it could be because there wasn't enough control over possible confounders like parity, diet, living habits, and educational level, which can hide the age correlation (Thomsen et al., 2010). None of the congeners were shown to be significantly associated with body mass index before pregnancy (Table 3).

We studied further the potential relationship between maternal diet and deca-BDEs levels (Table 3). Ingestion of contaminated food, particularly that of animal origin such as fish, meat, milk, and eggs, has been demonstrated to be a primary source of human exposure to PBDEs (Matovu et al., 2019). In the current investigation, correlations between the intake of fish, meat, and milk and PBDEs were not significant. Similarly, minor and insignificant correlations between human milk PBDEs and fish diet were discovered (Campoy et al., 2001). Variability in the absorption and metabolism of the contaminants among individuals may account for these weak relationships. In addition, contributions from numerous sources of PBDEs, such as other dietary items and indoor and outdoor exposure, may potentially account for the absence of connection. According to prior research (Jones-Otazo et al., 2005; Wilford et al., 2005; Wu et al., 2007), food consumption is not the only main source of human exposure to BDEs; nonetheless, environmental scientists and epidemiologists may need to focus on alternate pathways such as dust for the biomonitoring of PBDEs.

On the other hand, our study found that higher rates of egg consumption were linked to higher levels of BDE-28 in breast milk ($P = 0.01$). This shows that eggs could be a major source of BDE-28 in the diet. Some authors have found high levels of PBDEs in fatty foods that come from animals, like eggs (Babalola and Adeyi, 2018). These results agree with those findings.

A recent study by (Polder et al., 2016) found that eggs from free-range chicken had high amounts of BDE-209 (0.2–311 ng/g lw). The authors said that the levels were caused by the birds' behavior of scavenging, which put them in contact with particle-bound contaminants (like BDE-209) that could be broken down into lower congeners (such as BDE-28). Eggs could be a way for these congeners to get into the bodies of mothers. Also, our results suggest that mothers in Saudi Arabia get BDE-28 from sources other than their jobs, like eating eggs. More research needs to be done to find out if chickens raised in Saudi Arabia have eggs with a lot of PBDEs.

Also, regression analysis showed that mothers who previously lived outside the kingdom also had significantly higher level of BDE-28 ($p = 0.03$). Living outside the kingdom increases the exposure to electronics and hence the result is not surprising.

One of the strengths of the study that it is the first study to report the levels of deca-BDEs in human breast milk samples in Saudi Arabia. The study also explored the association of maternal

**Table 3**
Correlation between BDE and participant characteristics.

| BDEs | Age | | BMI | | Residing Years | | Fish and Fish Products | | Meat and Poultry | | Milk and Milk products | | Eggs | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | R | P-value | R | P-value | R | P-value | R | P-value | R | P-value | R | P-value | R | P-value |
| BDE-209 | 0.18 | 0.57 | 0.16 | 0.63 | −0.16 | 0.61 | 0.18 | 0.57 | −0.03 | 0.92 | 0.39 | 0.21 | −0.17 | 0.6 |
| BDE-28 | 0.00 | 0.97 | 0.05 | 0.69 | −0.03 | 0.78 | −0.14 | 0.23 | 0.03 | 0.81 | −0.01 | 0.97 | −0.29 | 0.01 |
| BDE-138 | 0.30 | 0.32 | 0.02 | 0.96 | 0.07 | 0.82 | 0.03 | 0.93 | −0.45 | 0.12 | 0.09 | 0.77 | 0.36 | 0.23 |
| BDE-208 | 0.18 | 0.13 | −0.04 | 0.74 | 0.09 | 0.43 | 0.03 | 0.77 | 0.12 | 0.31 | 0.05 | 0.67 | −0.1 | 0.39 |

Note: Data presented as correlation coefficient (R); P-value < 0.05 considered significant.

PBDE levels with maternal age, BMI and diet in Riyadh city. A limitation of the study is that the number of individual participants recruited into the study (n = 75) is relatively small to allow generalisations be made. Secondly, in saudia, limited data on the levels of PBDEs in other matrices such as food items and other human samples is available. This limited comparison of the observed levels in breast milk with those from the possible sources of the pollutants. Another limitation of the study was that exposure factors such as indoor and outdoor dust contamination, which have been proven to be important sources of the pollutants have not been considered in the present study.

## 4. Conclusion

Our study is the first investigation of deca-BDE in the milk of Saudi lactating women. Among BDEs, BDE-209, -28, -138, and -208 were the predominant congeners. BDE-209 was detected in all analyzed samples (median concentration 0.19 ng/g lw) suggesting recent exposure of mothers to deca-BDE formulations. The levels of BDE -209 (the most Carcinogenic agent type) were the highest in breast milk were higher (2.5–8 Fold) the levels of other BDES. The levels were in the same range of data, or lower than those of mothers in other countries. Higher rates of egg consumption were positively associated with higher breast milk levels of BDE-28 (P = 0.01). Several concern are raised including consumption of eggs and mothers who previously lived outside Saudi Arabia. The results may be used as base-line for future studies Since a gradual rise of PBDEs levels is expected in our Country. More research needs to be done to find out if chickens raised in Saudi Arabia have eggs with a lot of deca-BDE.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jksus.2023.102622.

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