# POLYBROMINATED DIPHENYL ETHERS IN FOOD FROM THE USA: TRENDS BY TIME AND LOCATION

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## Abstract

Our previous market basket surveys have described the levels of polybrominated diphenyl ethers (PBDEs) in food samples collected in the USA. To extend our previous findings, a convenience sample of thirty matched food samples were collected from three geographically disparate locations in the USA: Los Angeles, California; Dallas, Texas; and Albany, New York. Total PBDE levels were not statistically significantly different among the three locations. When comparing this study to our previous market basket surveys, total PBDE levels in meat were non-significantly higher in the new study and levels in dairy products were statistically significantly higher. These findings suggest that PBDE levels in some foods commonly consumed in the USA may be increasing despite declining commercial use of PBDEs.

## Introduction

PBDEs are commonly used flame retardants found in a variety of consumer products including mattresses, carpet linings, and electronics[1]. Exposure to PBDEs has been linked to deleterious health outcomes in both toxicological and epidemiological studies. In rodent studies, PBDE exposure has been linked to reproductive disorders including decreased sperm count and delayed onset of puberty, fetal malformations, behavioral changes, and endocrine disruption[2-7]. Recent, but limited, epidemiological studies have found PBDE exposure to be associated with reproductive and developmental disorders including cryptorchidism in newborn boys and decreased chest circumference, length, and weight at birth[8, 9]. Additionally, PBDE exposure in adult males has been associated with hormone disruption as well as decreased sperm count and testes size[10, 11]. Levels of PBDEs in human blood, adipose tissue, and milk have been shown to be 10-100 times higher in the United States than in Europe, China, and Japan[12-18].

Routes of PBDE exposure include ingestion of PBDE contaminated food, inhalation or ingestion of PBDE contaminated dust, and inhalation of PBDE contaminated air[19]. Market basket surveys conducted by us and others have described PBDE contamination of food worldwide[20-25]. Typically, levels of PBDEs are highest in food of animal origin, although the highest exposure levels in food of vegetable origin overlap with the lowest levels found in food of animal origin[26]. Among foods of animal origin, our previous market basket surveys have found the highest levels in fish, followed by meat, and then dairy products[24]. However, given American dietary intake patterns, typically the largest amount of dietary exposure to PBDEs was estimated to be from meat consumption.

Data presented in this abstract expands on our and other's market basket surveys to determine PBDE levels in food in the U.S. and worldwide. We collected matched samples of food from three locations across the United States to determine whether a difference in measured PBDE concentrations exists between regions.

Additionally, our previously calculated PBDE intake model was updated using the newly measured values to give a new estimation of American PBDE dietary intake by age and gender.

## Materials and Methods

*Sample Collection*

Thirty matched food samples were collected in May and June 2008 as part of a market basket survey from multiple supermarkets in three US cities: Albany, New York; Dallas, Texas; and Los Angeles, California. A total of 90 food samples were collected. Foods purchased as part of this market basket survey were similar or identical to those previously sampled and reported in our earlier studies for purposes of comparison. Samples were frozen and shipped on dry ice to the Wadsworth Center at the New York State Department of Health in Albany, New York.

*Chemical Analyses*

Methods of chemical analysis were adapted from methods previously described, with some modifications to also include PBDE congeners 183, 203, and 209 in the analysis[14, 27].

*Statistical Analyses*

Analysis of variance (ANOVA) was used to determine if PBDE levels detected in food varied by either collection location (this study) or by time collected (this study vs. previous study). A two-way ANOVA was run with location and food type as factors to determine if total PBDEs measured varied by location. To determine if a difference exists in total PBDE levels measured in different food groups (meat, fish, dairy) over time, two way ANOVAs were run separately for each food group with time collected (previous study or this study) and food type as factors. For each of the statistical analyses, non-detect levels were estimated as one half of the detection limit. However, analyzing the data using non-detect levels estimated as zero did not change any of the calculated significance levels.

*Intake Model*

The intake model used for estimating dietary PBDE intake was calculated using the same method previously described[24]. Standard errors of intake were calculated as the square root of the sum of squared standard errors of mean PBDE level for each food type weighted by the squared food intake amounts.

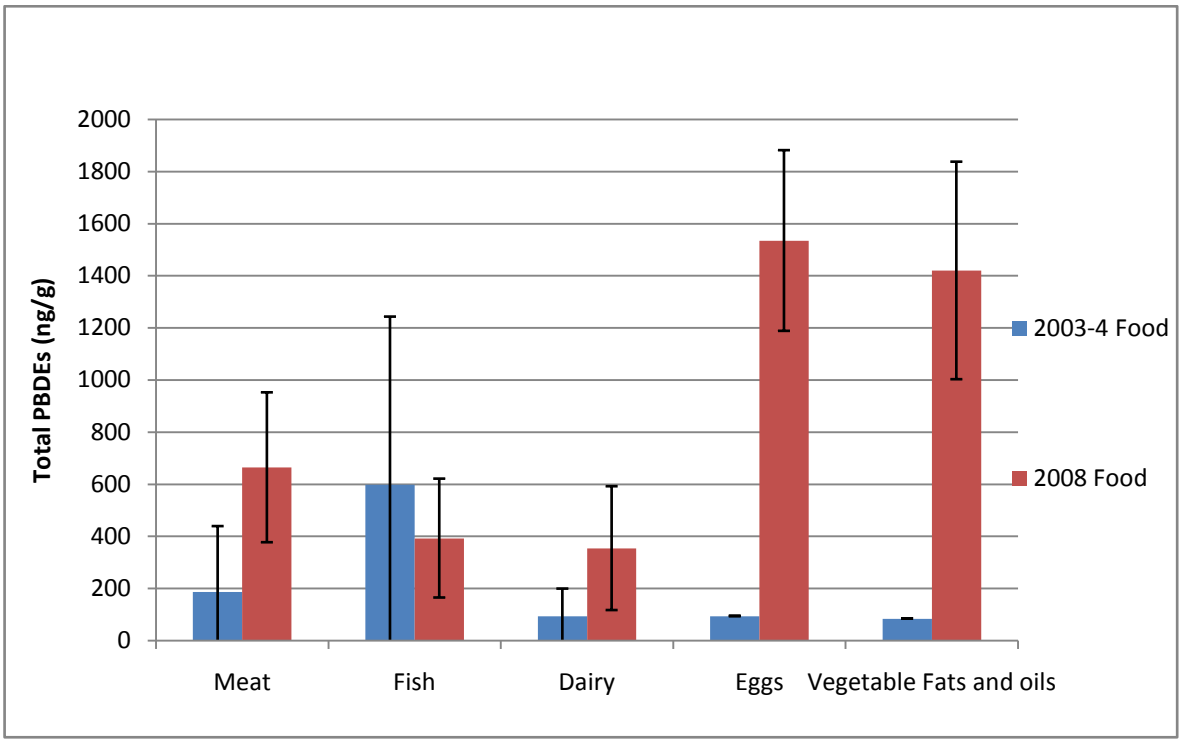
## Results and Discussion

No statistically significant difference in total PBDE levels in food from the three locations was detected ($F(2,29) = 1.43$, $p = 0.25$). Additionally, there was no statistically significant difference in total PBDE levels measured in this study compared to our previous market basket surveys[24]. However, as shown in Figure 1, there were some differences observed in total PBDE levels measured in individual food groups. In the previous market basket survey, median total PBDE levels in fish, 616 pg/g wet weight (ww), were higher than in meat, 190 pg/g ww[24]. In this study, the opposite was observed, with median total PBDE levels in fish, 457 pg/g ww, being lower than median total PBDE levels in meat, 719 pg/g ww. There was no statistically significant difference in total PBDE levels measured in either meat ($F(1,8) = 1.89$, $p = 0.18$) or fish ($F(1,7)=3.34$, $p = 0.08$) over time.

A statistically significant difference was noted when comparing the total PBDE levels measured in dairy based food products in this study and the previous market basket survey[24]. In this study, the minimum total PBDE

level measured in dairy foods, 40 pg/g ww, exceeded the median level measured in the previous market basket survey, 37 pg/g ww[24]. When comparing matched samples, total PBDE levels in dairy were significantly higher in the current study than the previous study ($F(1,5) = 13.46$, $p < 0.01$).

Figure 1. Median total PBDEs (with 2x standard errors) in food collected in 2003-4 and 2008 by food group




Median total PBDE levels measured in eggs in this study were 1630 pg/g ww, with all three samples measuring above 1000 pg/g ww. In the previous study, a pool of six eggs had a total PBDE level of 85 pg/g ww[24]. This is, however, a non-statistically significant difference, most likely due to the limited sample size. Total PBDE levels measured in margarine in this study far exceeded the one sample of margarine measured in the previous study, with a median of 1485 pg/g ww in this study compared to 88 pg/g ww in the previous study[24]. The median total PBDE level measured in vegetable based oils (either olive oil or a blend of olive and canola oil) was 1630 pg/g ww, compared to 9.4 pg/g ww in canola oil and 34.2 pg/g ww in olive oil, measured in a study of vegetable based foods collected in 2006[26].

Figure 2 compares total PBDE levels measured in various food groups in samples taken recently from three different countries, Spain, Belgium, and the U.S.[21, 25]. In this figure, the U.S. data is a combination of our current and previous market basket surveys. The Spanish and Belgian studies measured only one egg sample each, so standard errors could not be calculated.

Figure 3 shows the previously calculated estimation of dietary intake of PBDEs from various food groups per kilogram of bodyweight per day broken down by age and gender[24]. Figure 4 describes the newly calculated estimated total dietary intake of PBDEs from different food groups for the U.S. population broken down in the same manner as in Figure 3. The estimated total dietary intake of PBDEs for young children in this study is 6.5 ng/kg/day compared to 2.6 ng/kg/day calculated previously[24]. In this study, for all groups, meat provided the largest portion of PBDE intake. The youngest individuals had the highest amount of PBDE intake per kilogram

body weight. PBDE intake per kilogram of body weight decreased as age increased. After meat, dairy based foods and eggs are the greatest sources of PBDE intake for Americans.

Figure 2. Median total PBDEs (with 2x standard errors) in food collected in various countries

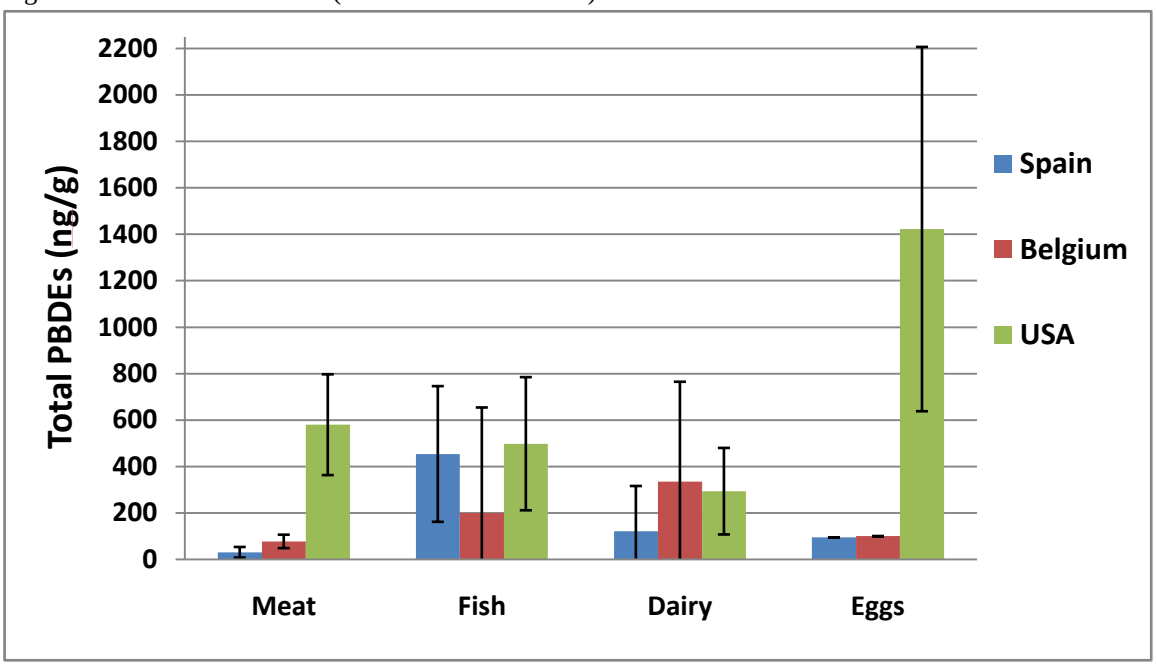



Figure 3. Estimated total dietary PBDE intake by food group, age, and gender (2003-04)

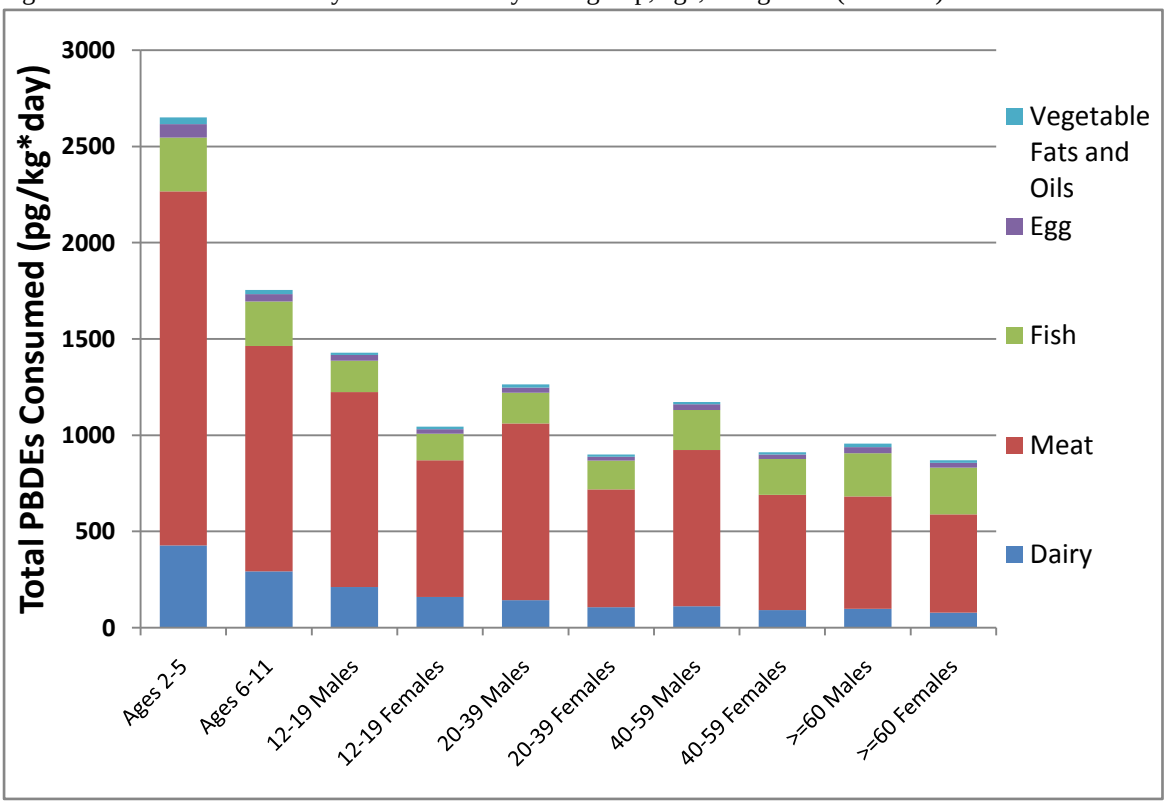
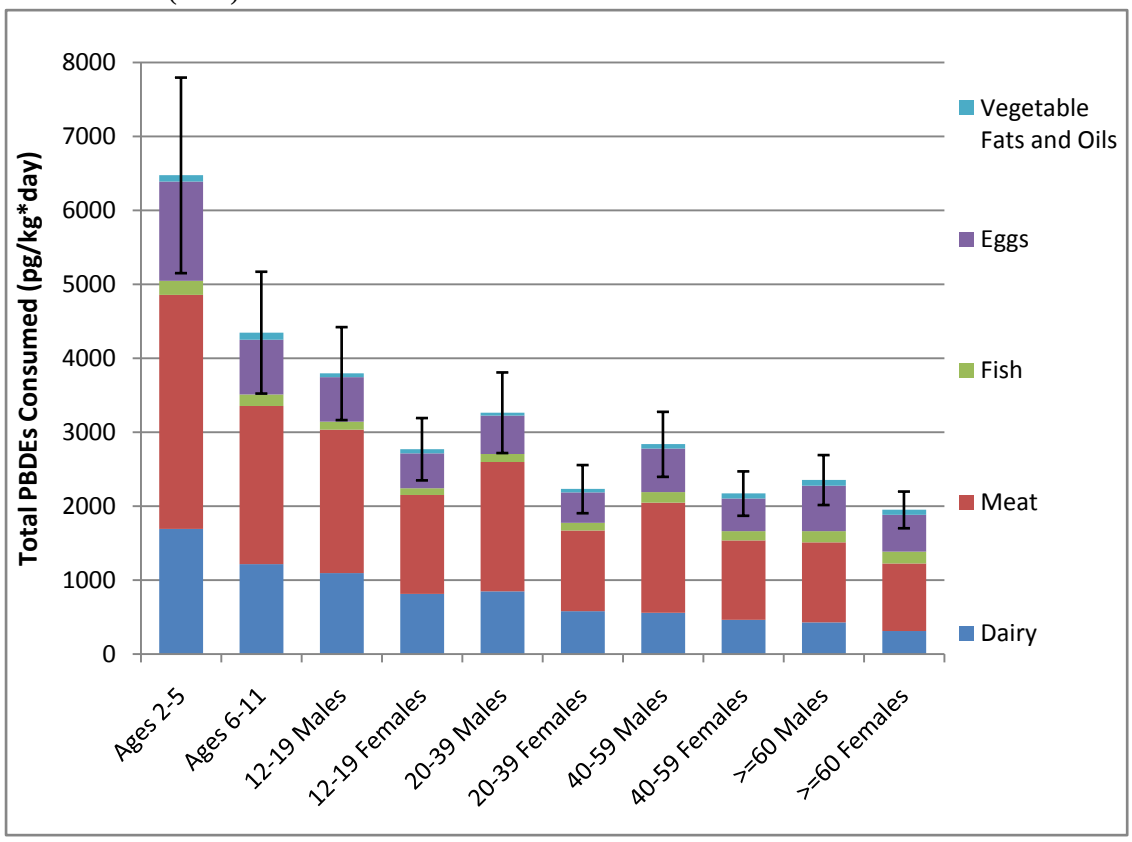


Figure 4. Estimated total dietary PBDE intake by food group, age and gender with error bars representing 1.96 standard errors (2008)




Data presented here suggest that PBDE levels in US food may not be decreasing, as would be expected due to declining use of PBDEs. In some commonly consumed food types, including eggs and cheese, PBDE levels may be increasing when compared to our previously conducted market basket surveys. However, these data do represent a small convenience sample of foods selected and cannot be considered a representative sampling of all US foods. There was considerable variation observed in levels of PBDEs measured, even in the same food types, which led to uncertainty in the intake estimations. Further research still needs to be conducted to determine representative levels of PBDEs in US foods as well as the effects of food packaging and shipping containers on PBDE levels. Additionally, more research needs to be done to describe the health effects of mixtures of PBDEs and other endocrine disrupting chemicals.

**Disclaimer:** This abstract does not reflect NIEHS/NIH policy.

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