

POLYBROMINATED DIPHENYL ETHERS AND HEXABROMOCYCLODODECANE IN THE UNITED STATES: LEVELS IN HUMAN BLOOD AND MILK, FOOD, AIR, FAST FOOD AND ENVIRONMENTAL SAMPLES

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Introduction

Polybrominated diphenyl ethers (PBDEs) have been reported at increasingly high levels in the U.S. population. To determine the past and present levels in humans and various sources of exposure, a series of studies were conducted measuring 10-12 PBDE congeners. Studies of human tissue included older and more recent blood samples, blood from vegans, partitioning between blood and milk of the same individuals, and fetal tissue. Exposure studies included assessment of levels in office and home environmental samples, in a broad range of foods, and in comparisons of cooked and uncooked foods. This paper presents findings from these studies. It includes indoor and outdoor air levels of PBDEs, PBDE levels in fast food from the USA and Germany, and hexabromocyclododecane in milk and blood.

Materials and Methods

Human milk and blood specimens were collected first in Texas and then elsewhere in the USA. Food was purchased from three large supermarket chains at stores in Dallas, Texas. Fast food was collected in the USA and to a limited extent in Germany at common fast food company outlets. Samples were collected in chemically cleaned containers and, in the case of biological specimens, were frozen and shipped frozen to the analytical laboratory on dry ice. Hexane wipe sampling of computer and computer monitor surfaces was performed for office electrical equipment. Household vacuum sweepings were collected from home vacuum sweepers and shipped in their original bags to the laboratory. Household drier lint was placed in plastic containers and shipped at room temperature. Air sampling was done in a manner to collect particulate and vapor phases separately. Ten to twelve PBDE congeners were measured by gas chromatography-mass spectrometry. The analytical methods for PBDEs have been previously described.^{1, 2}, and so have those for HBCDD.³ The milk hexabromocyclododecane congener specific analysis was performed at Health Canada and the blood analysis at ERGO Laboratory.

Results and Discussion

Previous work showed USA human milk PBDE levels to be the highest in the world, approximately 10-30 fold higher than reported in Europe.⁴ Similar high blood PBDE values from blood collected in 2003 were seen when compared to that collected in 1973 but far lower levels in dioxins, dibenzofurans and PCBs were detected in the 2003 specimens.⁵ We also reported high levels of PBDEs in USA household vacuum sweepings and in wipe samples from office computers and computer monitors.⁶

We report blood to milk partitioning of PBDEs shows ratios of close to or sometimes below 1.0 for the less brominated and hence smaller congeners whereas BDE 209, fully brominated with 10 bromines, had a blood/milk ratio higher than 1.0 and a range of 1.7– 132. This large range points out difficulty in calculations of blood/milk ratios for BDE 209. Vegan data shows a trend of lowered PBDE body burden proportional to the length of time without animal fat.

We show in Figure 1 data based on our new extended U.S market basket food survey with estimated PBDE dietary intake, in Figure 2 the effect of cooking on PBDE levels in some foods, in Figure 3 levels of PBDEs in fetal liver, and in Figure 4 levels in lint from German and USA household driers.

In our expanded market basket survey of PBDEs in USA food, fish has the highest level of contamination followed by meat and dairy products. Yet, since meat is consumed at far higher levels than fish in the USA, meat is the primary source of intake of dietary PBDEs in this country. These data continue to show high levels of PBDEs in USA biological specimens and also in some environmental samples. Documentation of lower amounts of PBDEs when fat is removed by broiling is similar to findings previously reported that PCBs and dioxins can be decreased by cooking food in such a way that lipid is decreased. The finding of fetal contamination with PBDEs although at a level lower than found in adults documents trans-placental transfer in humans from mother to fetus. Fetal PBDE levels in this study were not related to gestational age. Finding of higher levels of PBDEs in USA household drier lint compared to German levels continues the pattern previously reported with USA levels higher than European levels. The levels of PBDE in USA lint are lower than in USA vacuum sweepings. The origin of PBDEs in lint is not clear at the present time. A wide variation in levels is seen in biological and environmental samples. BDE 209 is noted in all types of specimens, biological and environmental, but appears especially high in environmental samples. Fast food, as expected, contained considerable amounts of PBDEs.

The hexabromocyclododecane levels are similar in the general population in the USA and Europe, unlike PBDEs where USA levels far exceed European levels.³ PBDE air levels in the USA samples were higher indoors than outdoors as has been found in other countries, except for outdoor air at an electronic recycling worksite in California with very high measured levels.⁷ We conclude air contributes a very small amount of total intake of PBDEs amount in most cases, although not always for individuals exposed to the highest reported levels in indoor or outdoor air.

References

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Figure 1: Daily PBDE dietary intake of U.S. population by age and food group (pg/kg body weight)

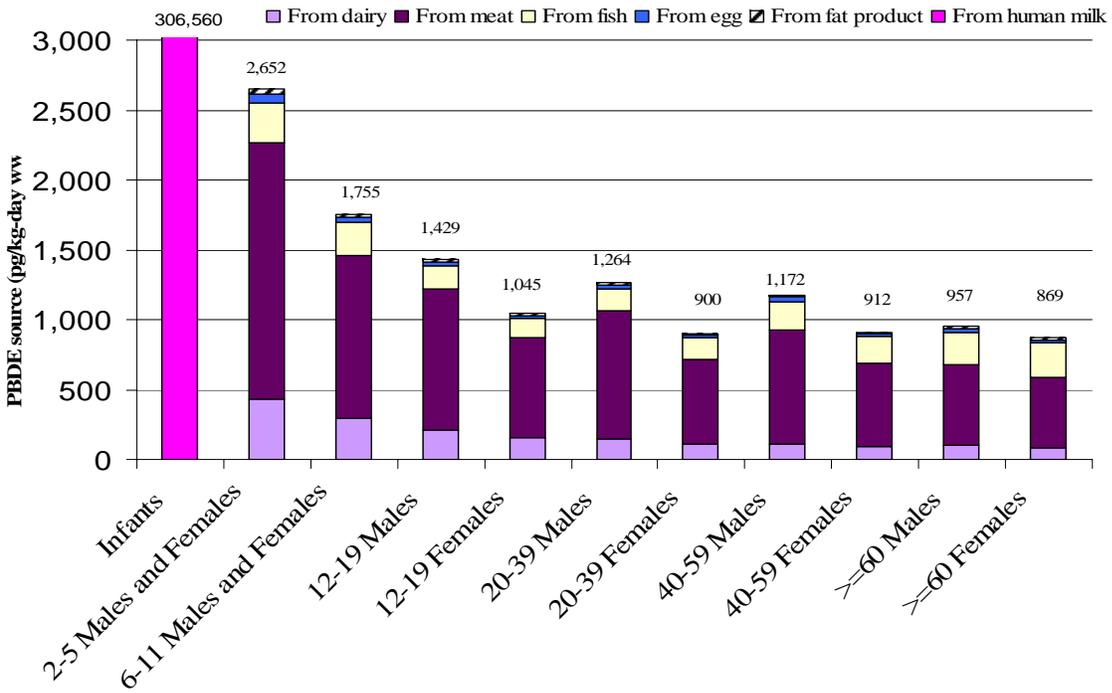


Figure 2: PBDE levels before and after cooking

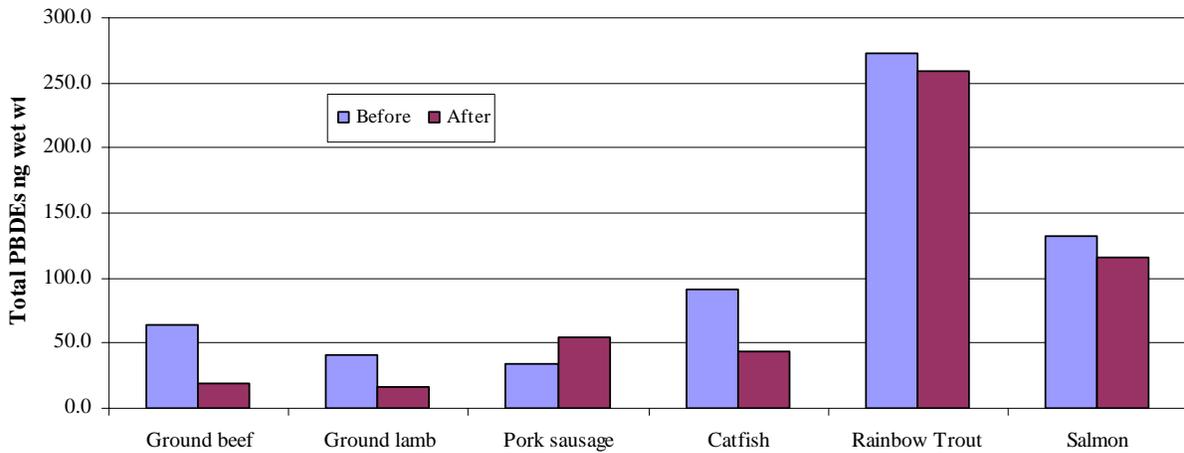


Figure 3: Fetal liver PBDE levels (ng/g or ppb lipid, half detection level. All BDE #77 and BDE #209 are not detected and presented at half detection level)

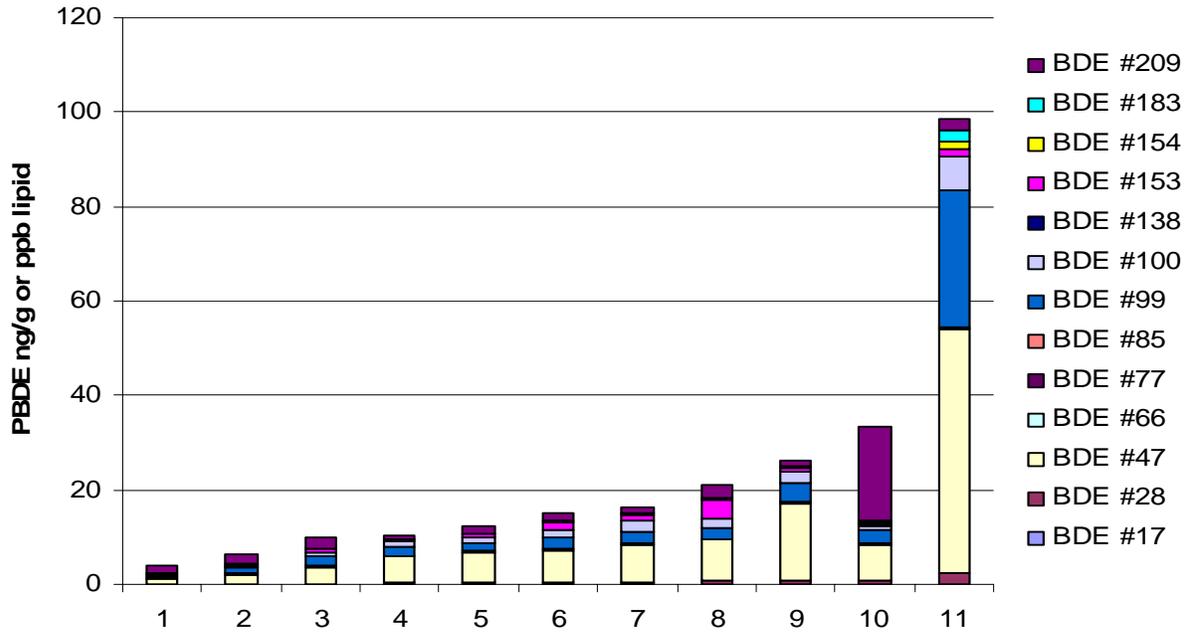


Figure 4: USA and German household drier lint (ng/g or ppb wet weight)

