POLYBROMINATED DIPHENYL ETHERS, POLYCHLORINATED BIPHENYLS AND PERSISTENT PESTICIDES IN 7 AND 9 YEAR OLD CHILDREN AND THEIR MOTHERS IN THE CHAMACOS COHORT

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Introduction

Three commercial formulations of polybrominated diphenyl ethers (PBDEs), identified by their average bromine content, have been in commercial production since the 1970s. The ability of PBDEs to reduce flammability as well as stricter fire safety standards world-wide and increasing use of flammable materials in furniture were all factors that led to increased use of flame retardant chemicals over the last decades, including the PBDEs. Tetra- through hexaBDE congeners are present in common food items and almost universally detected in human biomonitoring studies in the United States. The Penta- and OctaBDE technical mixtures were voluntarily withdrawn from the US market in 2004; while commercial use of DecaBDE ended in 2013.

The objectives of the current study were to: (1) expand the current limited information on the association of PBDE serum concentration and age in young children; (2) compare patterns in PBDE exposure with other persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and persistent pesticides which are often present in indoor dust at concentrations orders-of-magnitude lower than PBDEs; (4) explore whether dilution in a larger body size and/or elimination from the body are plausible explanations for decreasing concentrations of PBDEs and other POPs as the children age; (5) evaluate whether PBDE and other POPs are decreasing in concentration since the phase out of PentaBDE in 2004; and (6) compare the POP concentrations in the longitudinal birth cohort study of low income Hispanic children and their mothers in California with concentrations in a national sample.[1]

Materials and Methods

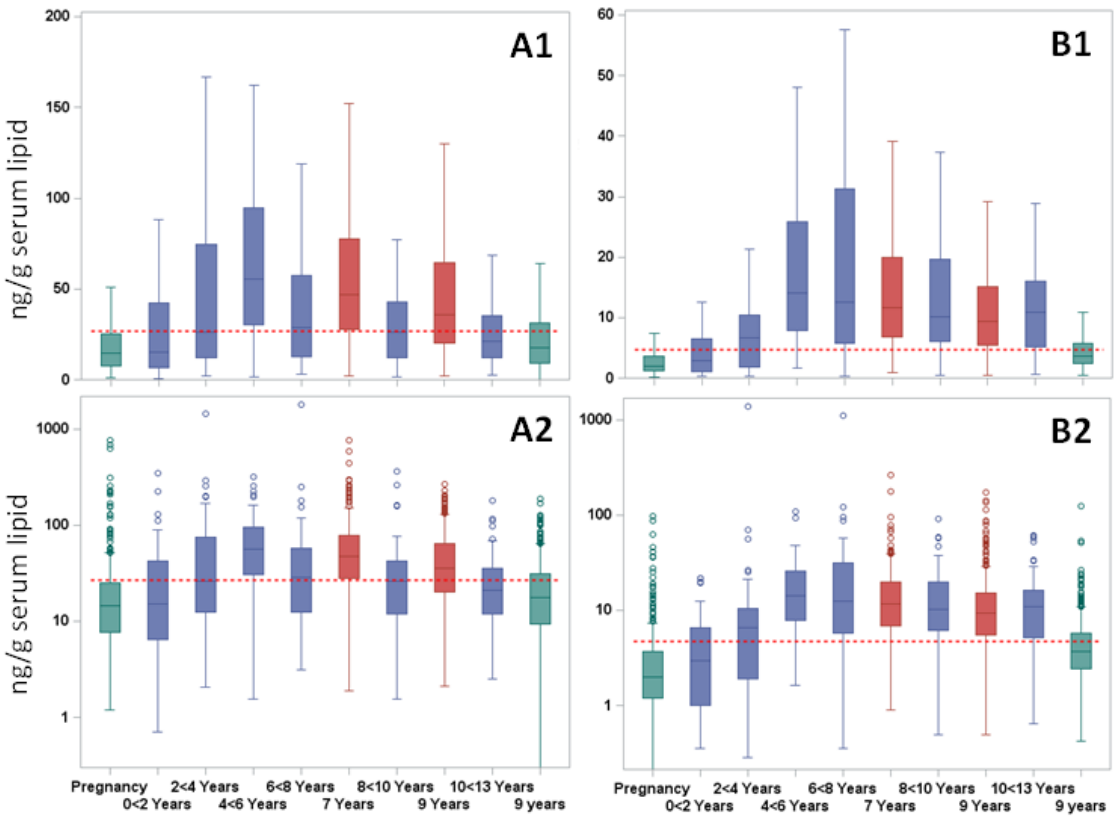
Study Population. Details of the Center for the Health Assessment of Mothers and Children of Salinas (CHAMACOS) recruitment and follow-up have been published elsewhere.[2,3] Study protocols were approved by the University of California, Berkeley and the Centers for Disease

Control and Prevention (CDC) Committees for Protection of Human Subjects. No personally identifiable information on the research subjects were made available to CDC researchers.

Measurements of POPs. The analytical methodology has been published elsewhere.[4] Analytical data were corrected by subtracting the median blank value. Concentrations below the LOD were substituted with the LOD/√2.

Statistical Methods. Statistical evaluation was limited to PBDE and PCB congeners having a greater than 60% detection frequency overall in the study (except maternal 26 weeks) and the pesticide *p,p'*-DDE having a 100% detection rate. Changes in child serum concentration from age seven to nine years of age and their association with increasing body weight were assessed in mixed effects models using SPSS (IBM, Armonk, NY).

Figure 1. Concentrations (ng/g lipid) of A BDE-47 and (B) BDE-153 on a (1) linear and (2) log10 scale by age of the child (7 and 9 years) given in red and their mothers at week 26 of pregnancy and at age 9 of their children in green. A cohort of Texas children [5] stratified by 2 year age groups between <2 and >10 years are included as a comparison in blue. Median and interquartile range (IQR) concentration (box) and outlier range (bars) [Outlier range is the 25th percentile – 1.5 IQR and 75th percentile + 1.5 IQR]. The red horizontal dashed line represents the National Health and Nutrition Examination Survey (NHANES) estimate for the age category 12-19 years of age for the survey years 2003/04.[1]



Results and Discussion

POP levels in the CHAMACOS children were generally comparable to a convenience sampling of Texas children[5] with slightly higher concentration of BDE-47 and similar median concentration of BDE-153 (Figure 1). The maternal BDE-47 concentration at 9-year collection were likewise comparable with the oldest group of Texas children (10 to <13 years) while the BDE-153 concentration is about half that of the Texas children (Figure 1). However, in the Texas children we see a sharp increase in concentration from the youngest age group (<2 years) to an apex concentration at 4 <6 years and 6 < 8 years for BDE-47 and BDE-153, respectively, which then declined at ages >8 years. Sjodin et al (2014) speculated that this increase up to age 4-6 years was due to the hand-to-mouth behavior of young children exposing them to residential dust which is known to contain high concentration of PBDEs.[6-9] Subsequent declining concentration may be caused by elimination of PBDEs and/or dilution in a growing body as the child ages and hand-to-mouth behaviors end or decrease. The current longitudinal study provides an opportunity to investigate these possibilities to a greater extent than what was possible in the Sjodin et al. 2014 publication.[5]

Table 1. Percentage change between age seven and nine by year and kilogram of weight gain for selected PBDEs, PCBs, DDT and DDE for participants with repeated measurements only.

Analyte	N	Change per Year			Change per kilogram weight gain		
		% Change	95% CI	p-value	% Change	95% CI	p-value
PBDEs							
BDE-28	228	-9.6	(-14.4,-4.4)	<0.001	-0.4	(-1.2,0.5)	0.44
BDE-47	228	-8.3	(-13.0,-3.4)	<0.001	-0.4	(-1.3,0.5)	0.36
BDE-85	228	-4.2	(-9.3,1.1)	0.12	-0.9	(-1.8,0.0)	0.04
BDE-99	228	-11.6	(-16.7,-6.2)	<0.001	-0.4	(-1.4,0.6)	0.43
BDE-100	228	-10.0	(-14.0,-5.9)	<0.001	-0.7	(-1.5,0.2)	0.11
BDE-153	228	2.3	(-1.6,6.4)	0.25	-2.6	(-3.3,-1.9)	<0.001
BDE-154	228	-13.4	(-17.7,-8.9)	<0.001	-0.8	(-1.7,0.0)	0.06
PCBs							
PCB-74	210	-3.7	(-6.7,-0.7)	0.02	-0.5	(-0.9,-0.1)	0.02
PCB-99	224	-3.2	(-6.1,-0.1)	0.04	-0.5	(-0.9,-0.1)	0.01
PCB-118	209	-2.1	(-5.3,1.2)	0.21	-0.7	(-1.1,-0.3)	<0.001
PCB-138/158	224	-3.0	(-6.0,0.0)	0.05	-0.6	(-0.9,-0.2)	<0.001
PCB-153	224	-2.9	(-5.9,0.2)	0.07	-0.6	(-1.0,-0.2)	<0.001
PCB-180	224	-3.2	(-6.1,-0.1)	0.04	-0.6	(-0.9,-0.2)	0.01
Pesticides							
DDE	228	-1.7	(-6.7,3.6)	0.53	-2.4	(-3.4,-1.4)	<0.001
DDT	228	-7.5	(-12.1,-2.6)	<0.001	-1.0	(-2.0,-0.1)	0.04

Abbreviations: BDE-28, 2,4,4'-tribromodiphenyl ether; BDE-47, 2,2',4,4'-tetraBDE; BDE-85, 2,2',3,4,4'-pentaBDE; BDE-99, 2,2',4,4',5-pentaBDE; BDE-100, 2,2',4,4',6-pentaBDE; 2,2',4,4',5,5'-hexaBDE; BDE-154, 2,2',4,4',5,6'-hexaBDE; CB-74, 2,4,4',5-tetrachlorobiphenyl; CB-99, 2,2',4,4',5-pentaCB; CB-118, 2,3',4,4',5-pentaCB; CB-138/158, 2,2',3,4,4',5'-hexaCB and 2,3,3',4,4',6-hexaCB; CB-153, 2,2',4,4',5,5'-hexaCB; CB-180, 2,2',3,4,4',5,5'-heptaCB; and p,p'-DDE, 2,2-Bis(4-chlorophenyl)-1,1-dichloroethene

In mixed effects models accounting for weight gain as the children aged from seven to nine years, resulted in an annual decrease (-8.3 to -13.4%) in tri- to hexaBDEs concentrations

($p<0.001$), except for BDE-85 and BDE-153., These two congeners were not associated with year of serum collection and instead showed an -0.9 to -2.6% decrease per kilogram (kg) of weight gain during the study period ($p<0.05$, Table 1). In the case of tetra- to heptachlorobiphenyls we observed -0.5 to -0.7% decrease in serum concentration per kg weight gain ($p<0.05$) and -3.0 to -3.7% decrease in serum concentration per year of aging ($p<0.05$) except CB-118 and CB-153 which were not associated with date of serum draw. p,p'-DDE decreased -2.4% per kg of weight gain between the two sampling points ($p<0.001$, Table 1).

Overall, these findings suggest that dilution in a larger body size as the children ages may play an important role in explaining reductions in body burden in the case of traditional POPs such as PCBs and p,p'-DDE. By contrast, in the case of PBDEs, such reductions would mainly be explained by lower exposures in more recent years as illustrated by decreased PBDE serum concentrations, possibly amplified by the presumed shorter biological half-life of PBDEs than of traditional POPs.

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