Characteristics Associated with Elevated PBDE Serum Levels: Findings from a US Cohort of Frequent and Infrequent Consumers of Sport-caught Fish

Henry A. Anderson,[1] Pamela Imm, [1] Lynda Knobeloch, [1] Mary Turyk,[2] Victoria Persky[2], John Mathew[3] and Carol Buelow[3]

[1]Wisconsin Department of Health and Family Services, 1 W. Wilson Street, Room 150, Madison, WI, 53703, [2]Division of Epidemiology and Biostatistics, School of Public Health, University of Illinois at Chicago, 1601 W. Taylor Street, Chicago, IL 60612, [3]Wisconsin State Laboratory of Hygiene, 2601 Agriculture Drive, Madison, WI 53718.

Introduction:

Polybrominated diphenyl ethers (PBDEs) have been used as flame retardants for several decades in the United States. Their incorporation into mattress coverings, carpeting, upholstery fabric, clothing and plastics have greatly reduced fire-related deaths. However, their widespread use and resistance to degradation have resulted in global contamination. PBDEs have a tendency to bioaccumulate in the food chain and have recently been detected in a variety of foods including fish, meat, poultry and dairy products. However, in contrast to PCBs and DDE, diet and consumption of sport caught fish are not thought to be the major route of human BPDE exposure (Schecter et al., 2006, Morland et al., 2005). PBDEs have also become common indoor contaminants and are present in household dust and continue to be present in a wide variety of consumer products (Schecter et al., 2005). Human exposure to these chemicals is ubiquitous, yet poorly understood. While most US residents have detectable levels of PBDEs in their bloodstreams, concentrations vary greatly from person to person. This study was designed is to assess PBDE exposure among a well-characterized cohort of adult residents of the Great Lakes Basin and to evaluate associations with potential exposure risk factors.

Materials and Methods:

A cohort of 4,206 frequent and infrequent consumers of Great Lakes fish established during the early 1990s was re-contacted during 2004 and 2005. This cohort has been described previously (Hanrahan et al. 1999). Members were invited to participate in a follow-up study that involved collection of serum samples and completion of a self-administered survey that requested information about recent fish consumption and diet, employment history, lifestyle characteristics, and medication use. Data is currently available from 404 individuals who volunteered to participate in the follow-up study.

Serum samples were extracted three times with a hexane\ethyl ether mix. The extract was then concentrated to approximately 2 ml and fractionated using Florisil and silica-gel columns. The final extract was concentrated to 0.5 ml. For PCBs and DDE the concentrated sample was injected onto a gas chromatograph equipped with an electron-capture detector. PCBs were analyzed on a 60 meter DB-5 capillary column, with confirmation on a DB-1 column. PCB congeners included in this method are quantitated using the "Mullin 1994" mixture of Aroclors. DDE was analyzed on a 60 meter DB-1 column. For PBDEs the concentrated sample was injected onto a gas chromatograph-mass spectrometer operating in the negative ion mode. PBDE congeners were analyzed on a 30-meter DB-5HT capillary column.

Venous blood samples and surveys were collected from 281 men and 123 women. Serum was analyzed for 9 PBDE congeners, as well as DDE and 30 PCB congeners. Results for BDE 154 were not used because it did not separate completely from PBB 153 (polybrominated biphenyl 153). Geometric mean DDE, PCB, and PBDE levels in this cohort were 284, 224, and 27 ng/g lipid, respectively, and with maximum values of 3964, 4309, and 1359 ng/g lipid (non-detected congeners=0). PBDE congener 47 (2,2',4,4' tetra-BDE) was detected in 97% of the serum samples, while congeners, 99 (2,2',4,4',5 penta-BDE), 100 (2,2',4,4',6 penta-BDE), and 153 (2,2',4,4',5,5' hexa-BDE) were detected in 59, 34, 33 and 9% of the samples, respectively.

Relationships of PBDEs with demographics and exposures were examined using nonparametric statistical methodology. PBDE serum levels were expressed as an ordinal variable (individuals in the highest and lowest 10% of lipid-adjusted PBDE levels, Table 1) or as a continuous variable (Table 2).

Results and Discussion

In the ordinal analysis, those with higher PBDE levels were significantly older, had lower family income, higher PCB and DDE levels, more years of sport fish consumption, spent more hours/day outdoors, and were more likely to sleep on a water bed (Table 1). However, recent dietary patterns, including sport fish consumption, did not differ significantly by group.

In the continuous variable analysis, PBDE levels were positively associated with age, PCBs, DDE, years consuming sport fish, shellfish meals in the last month, and hours spent outdoors, and negatively associated with income and population density in zip code of residence (Table 2). PBDEs did not differ by gender, but PCBs and DDE were higher in male participants ($p<0.05$). PBDEs were lower in participants who lost 10 or more pounds in the last year, while PCBs and DDE were lower in participants who gained 10 or more pounds in the last year ($p<0.05$). PCBs were significantly higher in participants who used fish oil dietary supplements ($p<0.05$).

A multivariate model reflecting exposures independently associated with log sum PBDEs was created using stepwise selection procedures, with a p-value of 0.20 required for entry into the model and a p-value of 0.08 required for retention in the final model. In the final model log PBDEs were positively associated with log DDE, serum lipids, shellfish meals, pork meals, and hours on the computer, and negatively associated with income, and egg meals (Table 3). R-square for the model was 0.13. Identical results were obtained using a backwards selection procedure.

Our population was initially chosen for study of PCB and DDE exposure effects because Great Lakes sport fish consumption was high. Our findings suggest that in such a population sport fish consumption may also be contributing to PBDE exposure and the positive correlation of PBDE with PCB and DDE raises concerns of cumulative exposure. Because of the dietary habits of our population, the effects of sport fish consumption on PBDEs may obscure identification of effects of exposure to PBDEs from the wide variety of consumer products in residential and work environments including textiles, plastics and foam components likely to contain PBDEs. These findings confirm the environmental persistence and bioaccumulative properties of this class of chemicals and provide new clues regarding sources of exposure to brominated fire retardants.

Table 1. Comparison of PBDE exposure groups: Mean or Percentage of Category

	PBDE ng/g lipid (LOD=0)			
	>100	**7.4-100**	**<7.4**	**P-value ***
Number of participants	40	323	41	
% males	75%	70%	58%	0.22
Age in years	60.8	57.7	54.0	0.004
Income <$55,000/year	73.5%	50.3%	50.0%	0.02
State of residence: Illinois	14.8%	74.1%	11.1%	0.24
Michigan	8.6%	85.9%	5.5%	
Ohio	12.2%	76.2%	11.6%	
Wisconsin	6.1%	80.5%	13.4%	
Body mass index, kg/m^2	29.1	29.8	28.1	0.21
% with weight loss of >10 lb in last year	15.0%	27.5%	36.6%	0.09
Serum lipids, mg/dL	767	718	668	0.16
% taking anti-lipid medications	27.5%	30.7%	22.0%	0.49
PCB Level, ng/g lipid	548	412	228	0.005
DDE Level, ng/g lipid	585	427	221	0.0001
% Recent fish oil supplement use	22.5%	11.5%	14.6%	0.13
Sportfish ingestion years	43.4	34.5	27.0	0.001
Shellfish meals in past month	1.6	1.5	1.1	0.34
Commercial fish meals in past month	2.4	3.2	3.0	0.30
Sportfish meals in past month	1.7	2.0	1.2	0.15
Total fish/shellfish meals in past month	5.7	6.7	5.3	0.51
Meat/poultry servings/week	9.9	10.5	10.3	0.60
Dairy servings/week	13.9	15.8	16.6	0.41
Hours outdoors/day	5.3	4.2	3.4	0.02
Hours indoors/day	14.6	16.3	17.5	0.02
Hours in a car/day	1.7	1.7	1.8	0.66
Hours in a boat/day	2.4	1.5	1.1	0.06
Hours TV viewing/day	2.7	2.9	2.4	0.14
Hours using a computer/day	1.9	2.2	1.9	0.38
Number of new cars owned 10 yr	4.0	3.9	4.3	0.69
% Who sleep on a water bed	15.8%	5.0%	0%	0.006
% who use tobacco products	21.1 %	15.4%	12.8%	0.58

*P-values for continuous and categorical variables from Wilcoxon rank sum tests and Chi-square tests, respectively.

Table 2. Associations of PBDEs, PCBs, and DDE with Demographic and Exposures in 404 Participants

	Sum PBDEs	Sum PCBs	DDE
Age in years	0.14*	0.50*	0.46*
Income	-0.13*	-0.07	-0.09
Population density in zip code	-0.10*	-0.08	-0.05
PCB Level, ng/g lipid	0.17*	-	0.70*
DDE Level, ng/g lipid	0.24*	0.70*	-
Sportfish ingestion years	0.15*	0.54*	0.38*
Shellfish meals in past month	0.13*	0.08	0.02
Hours outdoors/day	0.12*	0.26*	0.13*

Associations between PBDEs, BDE 209, PCBs, and DDD and exposures from Spearman correlation coefficients. *P-value<0.05

Table 3. Independent Predictors of log PBDE Serum Levels (ng/g lipid): Backward Regression Analysis in 316 Participants with Complete Data, R-square for Model=0.13

	Parameter Estimate	Standard Error	p-value
Income	-0.07	0.025	0.008
Log DDE Level, ng/g lipid	0.29	0.07	<0.0001
Shellfish meals in past month	0.08	0.03	0.01
Pork meals/week	0.06	0.03	0.07
Egg servings/week	-0.11	0.03	0.005
Hours using a computer/day	0.05	0.02	0.01
Serum Lipids	0.0005	0.0003	0.07

Acknowledgement: This research was funded by the US Environmental Protection Agency, Grant No RD-83025401-1. The research and researchers were supported in part by the Center for Disease Control and Prevention Training Program Grant #1 T01 CD000189-01.

References

Hanrahan LP, Falk C, Anderson HA, Draheim L, Kanarek MS, Olson J. 1999. Environ Res Sec A 80:S26-S37.

Morland KB. Landrigan PJ, Sjodin A, Gobeille AK, Jones RS, McGahee EE, Needham LL, Patterson DG JR. 2005. Environ Health Perspect 113:1689-92.

Schecter A, Papke O, Joseph JE, Tung KC. 2005. J Toxicol Environ Health A 68(7):501-513.

Schecter A, Papke O, Harris TR, Tung KC, Musumba A, Olson J, Birnbaum L. 2006. Environ Health Perspect 114:1515-20.