PBDEs in U.S. milk, blood, and food, and temporal trends for PBDEs, PCDDs, and PCBs in US blood

Arnold Schecter[1], Olaf Päpke [2], John Jake Ryan[3], Robin Rosen[4], KC Tung[1], Marian Pavuk[5], Daniele Staskal[6], Linda Birnbaum[7], Hong Trong Quynh[8], John D. Constable[9]

[1]University of Texas School of Public Health at Dallas, Dallas, Texas USA
[2]ERGO Research Laboratory, Hamburg, Germany
[3]Health Canada, Ottawa, Ontario, Canada
[4]Southwestern Medical Center, Dallas, Texas, USA
[5]SpecPro, San Antonio, USA
[6]Curriculum in Toxicology, UNC, Chapel Hill, NC, USA
[7]EPA, Office of Research and Development, NHEERL, RTP, NC, USA
[8]Oncology Center, Ha Noi, Viet Nam
[9]Harvard Medical School, Boston, USA

Introduction

Recent findings document elevated PBDE flame retardant in human tissues in the USA and lower levels in Europe with increasing levels in both areas over the last decades[1-4]. Levels of PBDEs have also been reported in limited food surveys, and especially in fish[5,6]. We report here on findings of PBDEs in samples of recently collected USA blood, milk, and a market basket survey of food as well as two samples of human milk from Vietnam, a developing country. Levels of PBDEs in samples from U.S. nursing mothers milk are 10 – 100 times higher than European milks. Milk intake estimates for the USA and Germany are presented for nursing infants. Archived blood from Dallas, Texas from 1973 was analyzed for PBDEs, PCDD/Fs and PCBs and compared with blood from the same location collected in 2003. Almost no PBDE was found in 1973 US blood whereas current blood levels are similar to the elevated U.S. milk levels. Dioxins, dibenzofurans and PCBs were relatively high in the 1973 samples and much lower in the 2003 blood samples. Food PBDE levels are highest in fish, followed by meat and then dairy products in our survey. Levels were found to be extremely low in the two Vietnamese samples. All samples analyzed to date were positive for some levels of PBDEs with the exception of the 1973 archived US serum.

Materials and Methods

Human milk: Milk samples were collected from nursing mothers in Texas, USA in 2002 and individually analyzed for the presence of thirteen PBDE congeners.

Blood Samples: Blood was collected from individuals in two different states. Thirty-nine individual blood samples were collected recently and analyzed. Pooled blood (N=100) from a 1973 Dallas Texas archive was analyzed and compared to a pool, N=100, from the same area. Milk from Vietnam was collected in northern Vietnam from two nursing mothers in Hanoi.

Food Samples: Food samples for the market basket survey were purchased from three Dallas, Texas supermarket stores and analyzed for PBDEs.
All analyses employed techniques described elsewhere[7-9]. PBDEs are reported in pg/g or parts per trillion (ppt), wet weight (w.w.) in food. In blood and milk, PBDEs are reported in ng/g or parts per billion (ppb) lipid. PCDD/F/PCBs are reported in ppt in blood.

Results and Discussion
Table 1 provides our estimates of daily PBDE consumption in a German or US infant nursing for one year, showing 55.2 ng/day for the German[10] and 1,774 ng/day for the USA[4] infant. Table 2 shows levels of PBDEs, PCDDs. PCDFs and coplanar and mono-ortho PCBs in 1973 and 2003 blood serum from Dallas, Texas. PBDEs were very low, 0.77 ppb lipid, (ND: Not detected, using half the detection limit for purpose of calculation) in 1973 and very much higher (62 ppb) in 2003 blood. For dioxins, dibenzofurans and PCBs the opposite was the case with very high levels in 1973 compared to 2003 with total TEQ[11] of 150 ppt in 1973 and 25.3 ppt in 2003.

Figure 1 shows PBDE levels found in our human milk and blood analyses by percentiles and range; from about 5 ng/g, or ppb in blood and milk to 351 (median 31) and 419 ppb (median 34) for blood and milk, respectively. Figures 2-4 present our finding for PBDE levels in fish with the highest levels, range 8.5 to 3,078 and median of 1,725 ppt wet weight, to meat with levels from not detected to 1,373 ppt ww, and dairy products with the lowest levels, from 0.9-679, median 31.5 ppt, ww. Salmon contained the highest levels of PBDEs of all foods tested.

These data suggest US body burden of PBDEs are the highest in the world with all persons testing positive for PBDEs at the present time. Milk from the US is 10-100 folds higher than European PBDE levels. Two milk PBDE analyses from Hanoi, Vietnam are 0.5 and 0.45 ppb, lipid. Food levels are higher than found in other countries (Spain and Japan) and highest in fish, followed by meat and then milk. Wild fish have sometimes been reported at even higher levels of contamination[12-14].

Further research is needed to adequately determine levels and routes of PBDE intake in humans. Perhaps more important is the need for toxicity data, both in humans and in animal models, in order to estimate the potential for adverse health impacts.

Acknowledgments
We wish to thank all volunteers and also funding agencies including the CS Fund, Warsh-Mott Legacy, the Albert Kundstadter Family Foundation, and the Samuel Rubin Foundation which generously provided funds for this work. Partial funding provided by the NHEERL-DESE Training in Environmental Sciences Research, EPA CT 826513. This abstract does not necessarily reflect EPA policy.

	Germany	*USA*
Consumed milk/day (ml)	**800**	**800**
Lipid content of milk (%)	**3**	**3**
Body weight of infant (kg)	**5**	**5**
Concentration (ng/g milk fat)	**2.3**	**74**
Total PBDE intake ng/day	**55.2**	**1,774**
PBDE intake/day in ng/kg BW	**11**	**355**

Table 1. Estimates of PBDE consumption in a German or US infant nursing for one year

Serum pool	**PBDEs (ppb** lipid)	**PCDD/Fs (ppt** lipid)		**Coplanar and mono ortho PCBs** (**ppt** lipid)		**Total TEQ**
		Measured	TEQ	Measured	TEQ	
1973	**0.77**	**4,108**	**85**	**211,483**	**65**	**150**
2003*	**62**	**529**	**16**	**26,427**	**9.3**	**25.3**

Table 2. Measured levels of PBDEs, PCDD/Fs, and PCBs in pooled human serum, 1973 and 2003 (*N=100).

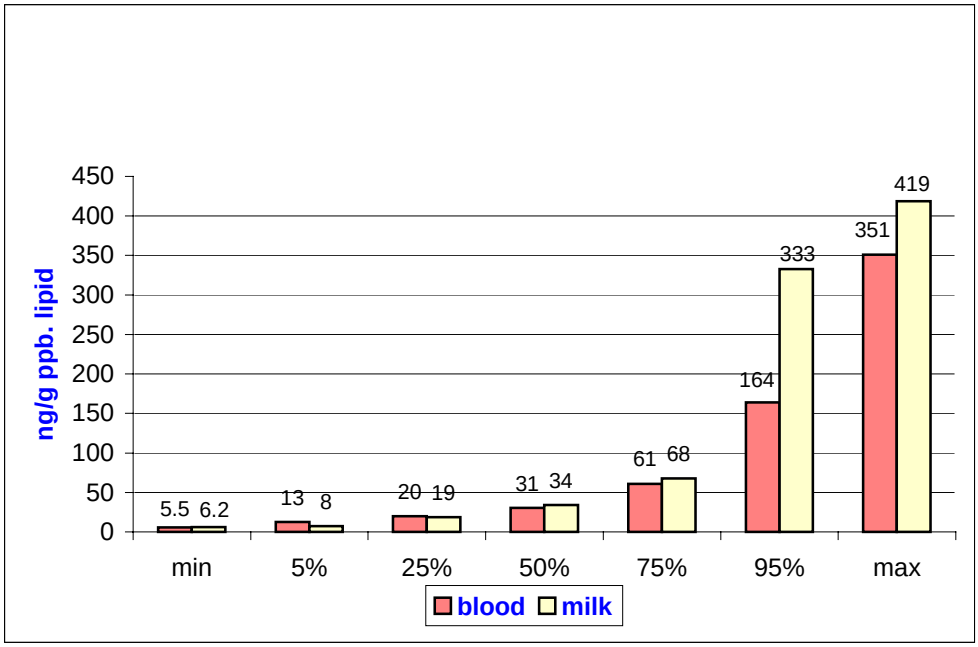

Figure 1. PBDE levels in U.S. human milk (N=52) and blood (N=29). 2003. (ng/g or ppb, lipid)

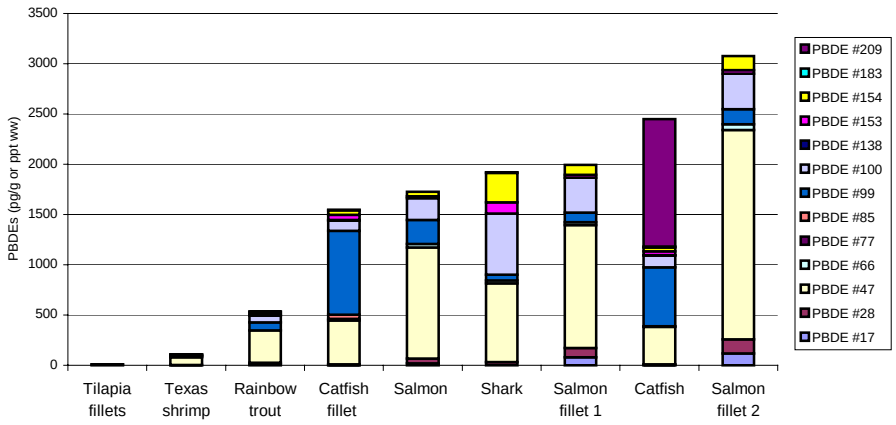

Figure 2. PBDE congener levels and profiles of individual samples of fish products purchased in USA. (pg/g or ppt, ww)

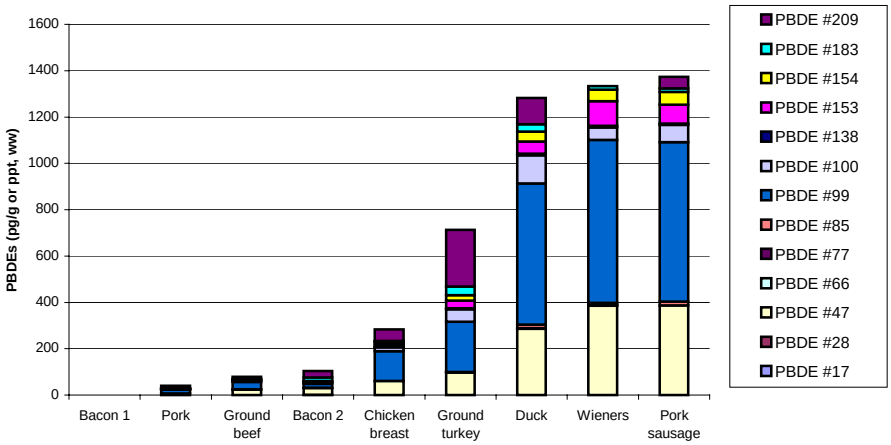

Figure 3. PBDE congener levels and profiles of individual samples of meat products purchased in USA. (pg/g or ppt, ww)

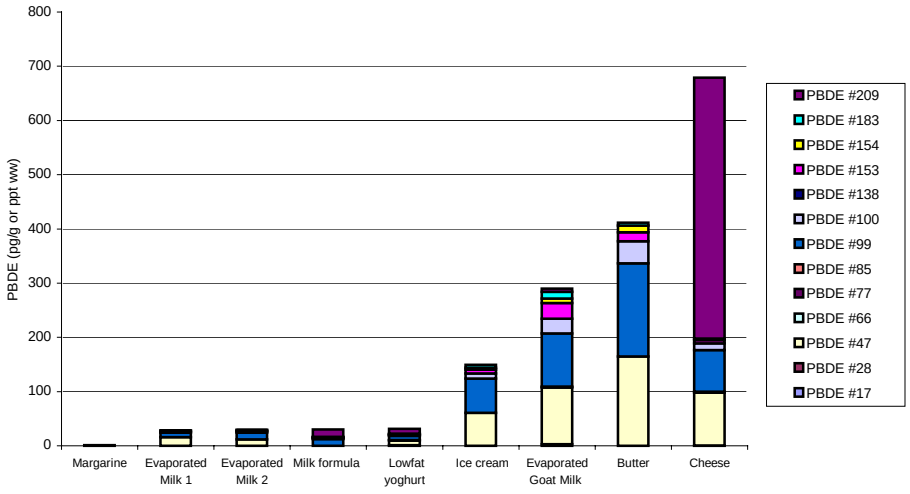

Figure 4. PBDE congener levels and profiles of individual samples of dairy meat products purchased in USA. (pg/g or ppt, ww)

References

1. Meironyté D, Norén K., Bergman Å. (1999) Analysis of polybrominated diphenyl ethers in Swedish human milk. A time-related trend study, 1972-1997. *J. Toxicol. Environ. Health A.* 58, 329-341.

2. Thomsen C, Lundanes E, Becher G. (2002) Brominated flame retardants in archived serum samples from Norway: A study on temporal trends and the role of age. *Environ. Sci. Technol.* 36, 1414-1418.

3. Sjödin A, Jones RS, Focant, JF, Lapeza C, Wang RY, McGahee EE, Zhang Y, Turner WE, Patterson DG. (2004) Retrospective time trend study of Polybrominated Diphenyl ether and Polybrominated and polychlorinated biphenyl levels in human serum from the United States. 2004. *Environ. Health Perspect.* 112: 654-658.

4. Schecter AJ.; Pavuk M, Päpke O, Ryan JJ, Birnbaum L, Rosen R. (2003) Polybrominated Diphenyl Ethers (PBDEs) in U.S. mother's milk. *Environ. Health Perspect.* 111:1723-1729.

5. Bocio A, Llobet JM, Domingo JL, Corbella J, Teixido A, Casas C. (2003) Polybrominated Diphenyl Ethers (PBDEs) in foodstuffs: Human exposure through the diet. *J. Agric. Food Chem.* 51: 3191-3195.

6. Ohta S, Ishizuka D, Nishimura H, Teruyuki N, Aozasa O, Shimidzu Y, Ochiai F, Kida T, Nishi M, Miyata H. (2002) Comparison of polybrominated diphenyl ethers in fish, vegetables, and meat and levels in human milk of nursing women in Japan. *Chemosphere.* 46:689-696.

7. Päpke O, Bathe L, Bergman Å, Fürst P, Meironyte-Guvenius D, Herrmann T,Norén K. (2001). Determination of PBDEs in human milk from the United States-Comparison of results from three laboratories. *Organohalogen Compd.* 52:197-200.

8. Schroeter-Kermani C, Hel D, Hermann T, Päpke O. (2000) The German environmental specimen bank- Application in trend monitoring of PBDEs in human blood. *Organohalogen Compd.* 47:49-52.

9. Päpke O, Ball M, Lis A, Scheunert K. (1989) PCDD/PCDF in whole blood samples of unexposed persons. *Chemosphere*. 1989;19:941-948.

10. Fürst P. (2001). Organochlorine pesticides, dioxins, PCBs and polybrominated biphenylethers in human milk from Germany in the course of time. *Organohalogen Compounds* 52:185-188

11. Van den Berg M, Birnbaum L, Bosveld AT, Brunstrom B, Cook P, Feeley M, et al. (1998) Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for human and wildlife. *Environ. Health Perepect.* 106(12): 775-792.

12. Manchester-Neesvic J, Valters K, Sonzogni W. (2001) Comparison of Polybrominated Diphenyl Ethers (PBDEs) and Polychlorinated Biphenyls (PCBs) in Lake Michigan Salmons. *Environ. Sci. Tech.* 35:1072-1077.

13. Johnson A, Olson N. (2001) Analysis and occurrence of polybrominated diphenyl ethers in Washington state freshwater fish. *Arch. Environ. Contam. Toxicol.* 41: 339-344.

14. Zennegg M, Kohler M, Gerecke AC, Schmid P. (2003) Polybrominated diphenyl ethers in whitefish from Swiss lakes and farmed rainbow trout. *Chemosphere.* 51:545-553.