ORGANOHALOGEN BODY BURDENS IN CALIFORNIA WOMEN

Myrto Petreas[1], Evan Rogers[2,3], Guomao Zhao[2,3], Gayle Windham[4], Rajiv Bhatia[5], M. Judith Charles[2]

[1]Hazardous Materials Laboratory, Department of Toxic Substances Control, Berkeley CA 94704
[2]Department of Environmental Toxicology, University of California, Davis CA 95616
[3] Public Health Institute, Berkeley CA 94704
[4]Environmental Health Investigations Branch, Department of Health Services, Oakland, CA 94612
[5]San Francisco Department of Public Health, San Francisco, CA, 94102, USA

Introduction

Persistent organic pollutants enter the natural environment via a multitude of pathways. Body burdens reflect cumulative exposures to such chemicals and can be used to assess temporal and spatial trends. Body burdens of organochlorine compounds (dioxins, PCBs, OCPs) are declining in most of the industrialized countries as a result of source reduction measures. Polybrominated diphenyl ethers (PBDEs), on the other hand, show increasing trends and measurements in California populations appear to be very high[1,2]. In this report we compare similarities and differences in organohalogen body burdens between immigrant and US-born groups in California assembled from different studies.

Materials and Methods

Immigrant Population

Between 1997 and 1999 serum was collected from a group of 50 women (19-40 years old) participating in a reproductive outcome study[3]. The women were born in SE Asia (primarily Laos) and had immigrated to the San Francisco Bay Area within the previous 2-20 years (mean=14.4 yrs, sd=4.5 yrs). The Laotian women were selected because of suspected higher organochlorine pesticide body burdens from SE Asia and, potentially, from consumption of fish caught in the San Francisco Bay. Half the women only had an elementary school level education and most (84%) had a prior birth.

Comparison populations

A group of 53 women, residents of the San Francisco Bay Area, and participating as controls in a breast cancer study, was used for comparison[1,2]. These women were 28-65 years old; most (83 %) were born in the USA; 8.5 % were of Hispanic and 2 % were of Asian origin, while the majority (83 %) were Non-Hispanic White and 2 % were African American. Over 95 % had college education, and 70% had at least one prior birth. Breast adipose tissue was collected during biopsy or breast surgery between 1996-98.

For a historic comparison population, archived serum was obtained from a case-control study of genital anomalies in male offspring, nested in the Child Health and Development Study (CHDS) cohort. The CHDS target population included all women who became pregnant and used services at selected Kaiser Foundation clinics and hospitals in the San Francisco Bay Area during the years 1959-1967. Women in the nested study ranged in age from 14-45 (median=26); 74 % had one or more prior pregnancies; and 69 % had graduated from high school. Only 3.9 % of the women were Asian and 3.2 % Hispanic, while 65 % were White and 23 % African American. Over 77% were US-born; 9.6 %

foreign-born (of whom 2 % born in Asia); and 14.8 % were of unknown birthplace. Serum from a subset of 50 women was used in this comparison.

Analytical methods

Samples were kept frozen below -20 °C until analysis. Serum was thawed, surrogate standards were added, proteins denatured with acetic acid and samples were extracted with hexane/dichloromethane, cleaned up through a glass column custom-packed with Florisil, eluted with hexane/dichloromethane, concentrated, and recovery standards were added. Analysis was performed by dual GC-ECD equipped with DB-XLB and Rtx-5MS columns. Elution orders and analyte identification were confirmed by GC/MS. Total lipids were calculated from total cholesterol and triglycerides as described by Phillips[4]. Results were reported as ng/g lipid.

Adipose samples were homogenized and ^{13}C labelled internal standards added. Samples were extracted with 1:1 hexane:dichloromethane, cleaned up with GPC and acid silica, concentrated, and recovery standards added. OCPs and PBDEs were analysed by LRMS by using ECNI (Finnigan 4510), with a DB5ms column (60m, 0.25 mm ID, 0.25 um film thickness). Methane was used as the reagent gas; the ion source pressure was 0.6 Torr and the ion source temperature was 100 °C. The electron energy was typically 70eV and the electron current was kept at 0.3 mA. PCBs were analysed by isotope dilution HRMS (Finnigan MAT 90), with a DB5ms column (60m, 0.25 mm ID, 0.25 um film thickness). Perfluorokerosene (PFK) was used to establish the lock masses and selected ions characteristic of the analytes were monitored. Lipid content of the adipose samples was determined gravimetrically in an aliquot of the extract and results were reported as ng/g lipid.

Results and Discussion

Fig.1 shows levels of major OCPs and PCBs in the serum of the Laotian immigrants to California and the two mostly US-born groups. The Laotians' PCB levels are the lowest of all three groups. The Laotian samples show high DDT and DDE levels, but not as high as the archived serum of the mostly US-born group from the 1960s. Additionally, a significant decline in DDT, DDE and â-HCH was observed between the mostly US-born participants from the 1960s to the 1990s. Levels of the other OCPs and PCB cannot be easily differentiated; sample matrix (partitioning in serum vs. adipose) and age differences between the older adipose group and the two younger serum groups may obscure any differences.

Serum samples were originally scheduled for PCB and OCP analyses[3], with the PBDEs added on as a secondary objective. Only 1 mL of serum was available for analysis, limiting our ability to detect low levels. Therefore, only PBDE #47 could be assessed in serum without interference from the blank and the limit of quantitation for PBDE #47 in serum was much higher (10 ng/g fat) than in adipose (<0.5 ng/g fat). With this limitation, PBDE #47 could be measured in the serum of only 24 of the 50 Laotian immigrants. PBDE #47 was not measurable in any of the archived serum from the 1960s. It is noteworthy that the immigrant women acquired PBDE body burdens equivalent to (or even greater than) those of the contemporary US-born, while their PCB body burdens remained quite lower. This may reflect different half-lives of contaminants and the recent introduction of PBDEs in the environment.

Table 1 shows concentrations of PBDE-47 in the three groups of California women in relation to data from a composite sample of human milk collected in 2000 from the US[5]. The levels of PBDE #47 in the Laotian samples are shown both including (n=50) and excluding (n=24) the samples below the limit of quantitation (~10 ng/g lipid). When the adipose samples were divided into a "younger" and "older" group based on the median age (48), the difference in PBDE #47 between these two age groups was statistically significant ($p<0.05$)[2]. Although no demographic information was provided, the

Table 1. Levels of PBDE#47 (ng/g fat) and demographic characteristics of the three California groups in relation to a composite human milk sample from the USA.

Group	N	PBDE#47 (ng/g fat)			Age (yrs)		US-born
		mean	median	range	median	range	%
Serum from Laotian Immigrants							
All, including <10 ng/g fat	50	50.6	10	<10-511	32.0	19-40	0
Only >10ng/g fat	24	94.6	49.7	20-511	35.5	20-40	0
Adipose from Breast Cancer Controls							
All	32	28.9	16.5	5.2-196	48.0	28-62	88
Younger than 48 yr	16	42.1	23	7.1-196	43.0	28-48	100
Older than 48 yr	16	15.8	13	5.2- 42.3	51.5	48-62	75
Serum from 1960s							
	50	—	—	<10	26.0	14-45	77
US human milk composite (from Papke et al, 2000)	1	120	—	—	lactating		—

composite milk sample was presumably obtained from young (lactating) women, consistent with the observation of higher PBDE levels in younger women.[1,2,6]

The Laotian group shows a distinctly different pattern of organohalogen body burdens, reflecting past exposures and differences in half-lives of the various analytes. Their profiles are dominated by DDT exposures with minimal PCB contribution. On the other hand, their PBDE #47 levels appear to have risen to levels experienced by native-born residents of the same area. It is important to generate more data on various sub-populations (immigrant, low SES, different diet and lifestyle) in order to protect public health for all California residents.

References

1. MX Petreas, J She, J Winkler, P Visita, M McKinney, P Reynolds, D Smith, D Gilliss, S Hurley, S Jeffrey, E Mahone. Organohalogen Compounds, 48:17-21, 2000
2. J She, M Petreas, J Winkler, P Visita, M McKinney, D Kopec. *Chemosphere*, 46:697-707, 2002
3. G Windham, P Mitchell, D Lee, B Lasley. Epidemiology 12:S58, 2001
4. Phillips, DL, JL Prickle, VW Burse, JT Bernert, O Henderson, LL Needham. Arch Envir Contam Toxicol. 18: 495-500, 1989
5. O Papke, L Bathe, A Bergman, P Furst, D Meironyte Guvenius, T Herrmann, K Noren. Organohalogen Compounds, 52: 197-200, 2001
6. M Petreas, J She, FR Brown, J Winkler, P Visita, C Li, D Chand, J Dhaliwal, E Rogers, G Zhao, MJ Charles. Submitted to Dioxin 2002.

FIG.1 MAJOR OCPs and PCBs IN CALIFORNIA WOMEN