VARIATION IN SEVERAL CLASSES OF POPs (PCDD/Fs, PCBs, PBDEs, HBCDs) FROM A SINGLE WOMAN WITHIN AND BETWEEN TWO LACTATIONS

John Jake Ryan, Bruce C. Wainman[1], Dorothea F. Rawn, Wing F. Sun, Ivana Kosarac and Amy Sadler

Food Research Division, Health Products and Food Branch, Health Canada, Ottawa, ON K1A 0K9, CANADA and [1]Education Services, McMaster University, Hamilton, ON L8N 4A9, CANADA

Introduction

It has been well established that all humans contain measurable amounts of POPs in their body as evidenced by concentrations in human milk, blood and other tissues [1-3]. Most of this work has been carried on a cross-sectional basis with sampling taking place at one period of time. However, there have been a number of studies of the change of the body burdens of POPs in people over time from the same individuals in a longitudinal setting[4-7]. Some of this work has used measurements in milk of lactating women to show a decrease or elimination of dioxins, furans, PCBs and other organohalogen compounds. However there have been few reports on the changes in human milk for a wide number and classes of POPs in more than one nursing period over a relatively extended period of time.

We were able to obtain multiple samples and quantities of human milk from an individual over a period of more than two years involving two separate pregnancies and nursing periods. Some of these milk samples have been analysed for a wide spectrum of POPs including dioxins, furans, PCBs, PBDEs, HBCDs, and several organochlorine (OCs) pesticides. In these milks, we report here the changes and variation of these multiple compounds over this period and give our interpretation of these changes with respect to the body burden of the mother and to earlier literature reports.

Methods

Milk Sampling

The donor was born in June 1974, was a life time non smoker, exercised often, during the lactation period weighed about 65 kg, height 1.7 m, and had little or no weight change over the study time except for approximately 20 kg of weight gain and loss within the pregnancies. Her diet was considered normal consuming mostly commercial foods with no special nutritional requirements. The children were two daughters, both of whom were of normal weight and gestational age and both the mother and the two children were healthy during the nursing period. After a first birth date of April 30, 2003, over a time period of 84 days between June 11 and September 2, 2003, numerous human milks were collected by machine pumping in the first nursing period (primipara) which lasted about 10 months in total. A little over two years later, the second child was born on August 5, 2005 and during the second nursing period which lasted about 8 months, milk samples were taken between August 13 and November 11, 2005 over a period of 87 days. From the numerous aliquots available, six samples from each nursing period were selected for extensive analysis based on the first and last sampling day and, as far as possible, equal spacing among samples in the intervening period. Comparative human milks were obtained in 2002-3 (n=27) and 2005 (n=34) from the same regional area of Ontario as part of ongoing studies of chemicals in human milk[8]. Approval for sample collection from the mothers and determination of POPs was obtained from the research ethics boards (REB) of both Health Canada and McMaster University.

Analyte Determination

Analysis of the milks for each class of organohalogen i.e. PCDDs, PCDFs, PCBs, PBDEs, HBCDs, and organochlorine pesticides (OCs), was carried out by adding appropriate carbon-13 isotopes. Sample preparation involved solvent extraction, lipid removal with strong acid, followed by purification and fractionation on Florisil and carbon. Determination was by gas chromatography-mass spectrometry (GC-MS) in the high resolution electron impact mode except for HBCD where liquid chromatography (LC)-MS was used for identification and quantification [8-10]. Milk values are expressed on a milk lipid basis- the latter content was determined gravimetrically after organic solvent extraction. The average lipid content for the multiple donor samples was

3.0 +/- 0.6 percent fat. TCDD toxic equivalents (TEQ) for all dioxins (n=7), furans (n=10) and dioxin-like PCBs (n=12) are calculated using 2005 TEFs of the World Health Organization. Each set of analytes has an associated quality control (QC) milk sample which is analysed with unknown samples for comparative purposes. These QC milk samples form a data base which not only controls and confirms the performance of the analytical system but also allows a useful estimation of the variation due to the analytical method when comparing differences among contaminant values. Health Canada also participates regularly in the Norway Institute of Public Health interlaboratory studies for food and human milk.

Results and Discussion

The human milk samples showed the typical pattern for PCDDs and PCDFs (dioxins increasing with degree of chlorination; furans less variation) in the ppt range and for total PCBs (domination of congeners 138, 153, 180 and lesser amounts of 99, 118, 170, 187) at the ppb level. Due mainly to the lower TEFs for the mono *ortho* PCBs, the TEQ values expressed with the newer 2005 TEFs are somewhat lower than the case using the former TEF values of 1998. As expected, the PBDEs consist of, in decreasing order of concentration, congeners 47, 99, 100, 153, 154, 28, 183, and 85 which collectively form more than 97 % of all detectable PBDE congeners.

The data set for all analytes and for both nursing periods are shown in the table along with comparative data from two sub cohorts sampled in the same time frame. The single donor values are expressed as means and standard deviation and the comparative cohort numbers as median and range - the latter method used due to the skewed distribution of most environmental contaminants. Examination of the observational numbers **within each of the two nursing periods** shows almost no change in concentration on a lipid basis for any class of chemical over a period of about three months. The standard deviations (SD) of the mean values are all the same order as the SD of the quality control milk samples i.e. the same as the analytical variation of the method which is typically 10-30 % depending on the analyte. Examination of the data **between the two nursing periods** indicates a slight decrease of about 20-30 % from the first to the second nursing period for most classes of compounds (dioxins, furans, PCBs, OCs) except for the BFRs (PBDEs and HBCDs) which remain relatively the same. Figure 1 shows the values for PBDEs and PCDD/F TEQ as a function of time for both nursing periods. In addition the concentration levels for all analytes are within the range of values of the comparative cohorts[8] which were sampled from the same area and year of collection and are considered to be normal or background values.

The lack of distinctive variation or change within the nursing period is similar to that reported recently for several donors by Hooper et al.[11] for PBDEs and PCBs (1- 3 % decrease per month) over a wide concentration range and Sjödin et al.[12] for PBDEs, PCBs and OCs over nursing periods of about 120 and 80 days, respectively which is similar to the nursing period here. However our values differ somewhat from earlier studies[4-7] of dioxins and PCBs which showed mostly declines in lactation levels in a single nursing period. On the other hand, reports in the literature on milk levels between lactation periods are scarcer. Our data indicate a decline in most analytes except for the BFRs. This result is in agreement with the detailed study by Abraham et al.[6] on this topic and also consistent with the known difference in the POP content of human milk between primiparous and multiparous donors[4,7] even though the individuals are not the same in each group. In their work with OCs in human milk, Czaja et al.[13] suggested that the time interval between nursing and pregnancy may be a significant determinant of the milk concentration. The longer the interval between lactation periods the less the difference in the chemical content of the milk- a proposition which may be pertinent in the data reported here. Lastly, there are two other parameters which could affect these milk levels; 1) the known contemporary decline of many POPs in the tissues of humans from industrialized countries and, 2) the known increase in the human content of most POPs with age.

In summary, we find little or no variation in several classes of POPs for human milk from the same donor within two nursing periods each of three months. Between the two nursing periods however, the environmental values appear to decline somewhat for most classes except for the BFRs which remain relatively the same. Long term breast feeding does not appear to result in a large decrease in the body burden of persistent chemicals in the mother. However multiple pregnancies may accomplish this to some degree.

Table 1. PCDD/Fs and PCB TEQ, total PCBs, BFRs, and OCs in human milks over two time periods all values on milk lipid basis								
First Nursing period 2003 (primiparous) ; 84 days sampling								
no	Date	PCDD/F TEQ (n=17) ppt	PCB TEQ (n=11) ppt	Total PCBs (n=33) ppb	Total PBDEs (n=11) ppb	α-HBCD ppb	HCB ppb	*trans*-nonachlor ppb
1	June 11	6.0	1.7	36	9.1	0.21	5.5	9.6
2	July 1	6.1	1.6	35	9.4	0.27	6.8	8.7
3	July 8	5.0	1.6	40	9.7	0.29	7.6	7.2
4	July 21	5.9	1.5	30	8.8	0.23	5.9	8.0
5	Aug 22	7.0	1.8	37	9.4	0.18	7.0	9.3
6	Sept 2	6.7	1.8	37	12.7	0.28	6.3	9.0
	Mean	6.1	1.7	36	9.8	0.25	6.5	8.6
	SD	0.7	0.1	3.1	1.4	0.04	0.8	0.9
Second nursing period 2005 (multiparous); 87 days sampling								
1	Aug 8	5.8	1.2	27	14.1	0.28	4.2	6.1
2	Aug 30	4.7	1.1	23	9.5	0.23	4.1	5.3
3	Sept 15	5.4	1.3	28	11.5	0.20	4.4	6.5
4	Oct 6	5.1	1.3	28	11.0	0.22	4.3	6.0
5	Oct 25	4.3	1.2	26	9.8	0.20	4.2	5.7
6	Nov 7	5.0	1.1	23	9.5	0.21	3.9	5.3
	Mean	5.0	1.2	26	10.9	0.22	4.2	5.8
	SD	0.5	0.1	2.4	1.8	0.03	0.15	0.5
General population samples for comparison								
1	2002-3 samples; n=27							
	Median	8.7	2.4	81	37	0.6	9.6	10.2
	Range	3.0-14.1	0.8-8.9	26-439	3.2-960	0.2-8.8	7.1-13	7.1-26
2	2005 samples; n==34							
	Median	6.6	2.2	63	20	0.4	8.3	10.2
	Range	0.6-29.1	0.3-10.9	10-213	3.7-580	0.1-28	1.0-31	1.9-43

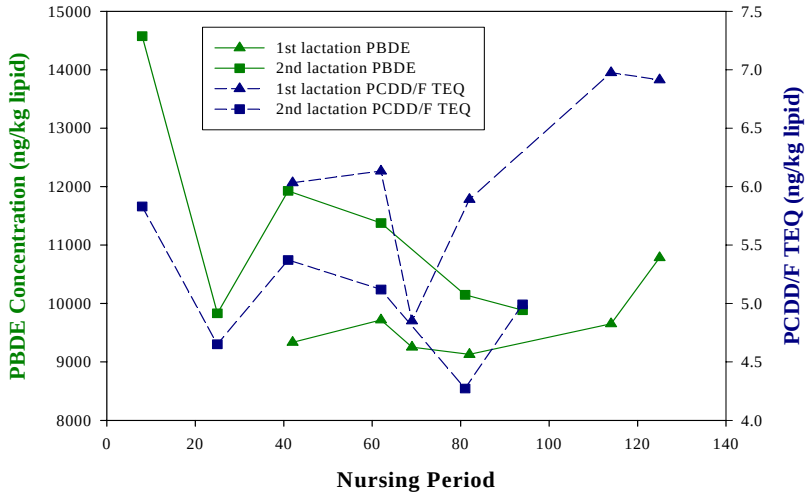

Figure 1: Comparison of PBDE and PCDD/F TEQ between two nursing periods.

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