RECENT TRENDS IN LEVELS OF BROMINATED DIPHENYL ETHERS (BDES) IN HUMAN MILKS FROM CANADA

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Introduction

The pioneering work of Meironyté et al. 1999[1] showed that the concentrations of brominated diphenyl ethers (BDEs) in human milk in Sweden increased steadily from 1972 to 1997. Data on this topic from the same group[2] in the years 1998-2000 indicate that the levels have stabilized or reached a plateau in Sweden with typical values of total BDEs now being about 3-4 µg/kg (parts per billion; ppb) milk lipid. The changes in levels for BDEs in human milk from other European countries[3] have not been extensively studied. The presence and trends for BDEs in human milk in North America, where brominated flame retardants (BFRs) including BDEs are widely used, is more tentative. This is due to a lack of multiple BDE values from recently obtained human milks. We reported previously[4,5] on the BDE content of 72 individual human milks collected across Canada in 1992 and found median levels for total BDEs of 2.9 ppb lipid. It was also noted that there was a large individual variation of values among the data set caused by a small number of very high samples (mean more than 5 times the median). Other pooled Canadian milk samples from earlier years in 1982 and 1986 showed a low level of BDE content. Reports on the BDE content of milks from the United States using pooled but not individual samples by ourselves at Dioxin 2001 from a New York State composite in 1997 and by Päpke et al[6] from a single composite from two US cities showed even higher levels between 100 and 200 ppb. However, to date there are no North America data on the BDE content of human milk from recently obtained individual samples in order to estimate general exposure.

Current levels of BDEs in people and foods are lower than PCBs by at least an order of magnitude[4,5] but the toxicological data base for evaluating their safety and risk is not too extensive. It has been thought that BDEs are not as potent as PCBs although recent experiments on laboratory animals now cast doubt on that assumption[7]. We report here on the collection of individual Canadian human milks from 2001 and 2002 from several regions of Canada and the BDE content of those from the Vancouver area of western Canada. Comparison is made directly to early samples from the same area and to other countries.

Sampling and Methods

Sample Collection

Human milks have been obtained between 2001 and 2002 from a milk bank in Vancouver, in collaboration with the Alberta provincial health department in Edmonton, and through two community groups in Quebec. In most cases information on age and smoking status was available and, less frequently, on parity, weight and height of the donor. The amount of milk obtained has varied between 5 and 20 mL.

Analysis

Samples were analysed as reported previously[4]. In summary, this involved the addition of six isotopically labelled BDEs, extraction with a mixture of acetone-hexane and lipid removal with strong acid. Further purification was accomplished on columns of acid silica and Florisil with the BDEs eluting in the dioxin fraction from which it was separated by an active carbon column. Determination

was by gas chromatography with high resolution mass spectrometry in the electron impact mode. Quantification was by the isotope dilution method using a eight point calibration curve. Sample batches included a laboratory blank to estimate contribution from that source (which is significant for BDEs; the total in the blank is subtracted from the total in the sample) and a reference or repeat control sample for accuracy and precision. The laboratory has participated in both international interlaboratory studies sponsored by the flame retardant industry (BSEF)[8].

Results and Discussion

The BDE content of twenty milks collected in 2001-2002 from Vancouver, British Columbia is listed in table 1 by total BDEs (sum of the 7 main congeners usually containing more than 95 % of all BDEs). Figure 1 shows the means and medians for the seven individual congeners. From figure 1, the congener pattern of the median values is mostly uniform with BDE 47 dominating with lesser but significant amounts of congeners 99, 153, 100, 28, 154, and 183 in that order. However, the variation of the amounts of the individual congeners is large with many of the congeners showing a large difference between the mean and median values due to a few high skewed values. In particular, the total BDEs in sample 19 are much higher than the other samples and the pattern (relative amounts of the various congeners) is anomalous with BDE 153 rather than 47 dominating. This anomaly whereby certain milks have either an altered BDE pattern and/or much higher concentrations than most other samples collected at the same time from the general population has been noted in our previous work and does not seem to be shared by other POPs such as PCBs and dioxins. Descriptive statistics for these individual samples are listed in table 2 along with the nine samples from the same city which were collected in 1992. In addition, the pooled values from British Columbia and Canada are also shown for the years 1982 and 1986. Probably the most pertinent comparison is the individual milk results from Vancouver between 1992 and 2002 in which the BDE median values have increased about fifteen fold. Other results for pooled samples from the mid and early 1980s show a low level of presence of BDEs. Data from Sweden, both recent pooled samples[2] and individual analyses[9] taken in the late 1990s, are listed for comparative purposes and indicate lower levels than found in Canada.

In the use of human milk to monitor background exposure to and trends of BDEs, there is a problem of high individual sample values which appear to occur with BDEs and not with other POPs. Nevertheless, the present results on individual human milks samples from the Vancouver area in Canada show an increase of more than an order of magnitude for the BDEs in the space of about 10 years. We are in the process of analysing further individual milks from two other regions of Canada in order to obtain further data on these trends and the individual variation among donors.

Table 1. Total BDEs (sum of 7 congeners) in 20 human milks from Vancouver

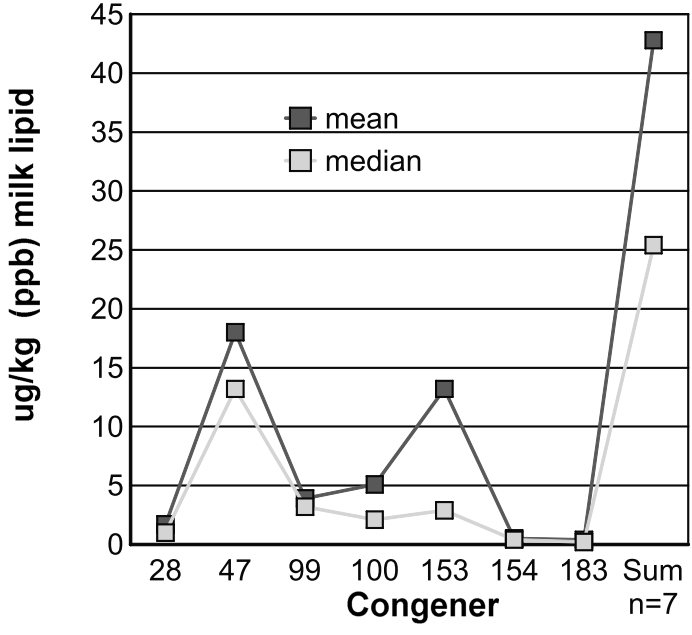
Sample No	1	2	3	4	5	6	7	8	9	10
µg/kg lipid	14.6	27.7	35.7	7.2	89.7	22.2	65.8	25.4	13.9	40.8
Sample No	11	12	13	14	15	16	17	18	19	20
µg/kg lipid	2.6	95.8	0.93	28.4	32.8	25.4	18.5	10.6	281.9	16.4

Table 2. Comparison of levels of total BDEs (?g/kg lipid) in human milks

	Individual Samples					Pooled Samples			
Year	2001-2	1992	1992	1996-1999	Year	1986	1986	1982	1998-2000
Place	Van[1]	Van	Can[2] ref 5	Swed[3] ref 9	Place	Van	Can	Can	Swed ref 2
Number	20	9	72	93	Number in pool	23	100	200	120
Mean	42.8	4.7	15.0	4.0					
Median	25.4	1.7	2.9	3.2	Value	0.7	0.6	0.2	3.4
Min	0.9	0.9	0.5	0.9					
Max	281.9	16.4	589	28.2					

1- Vancouver area, British Columbia, Canada; 2-Canada; 3- Sweden

Figure 1: Mean and median of 20 human milks



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