Levels of Polybrominated Diphenyl Ethers in Milk from Italian Women Living in Rome and Venice

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Introduction

Increasing concern on the potential effects on neurobehavioral development in children caused by perinatal exposure to polybrominated diphenyl ethers (PBDEs) has prompted a number of regulatory actions aimed to reduce PBDE maternal body burden. In fact, in spite of a still incomplete congener-specific PBDE toxicological characterization, many experimental studies consistently reported neurotoxic effects following neonatal exposure to these compounds.^{1,2}

Breast milk monitoring studies on PBDEs are of primary importance to carry out an adequate risk assessment at the actual levels of exposure, since it has been shown that levels in breast milk reflect maternal body burden during pregnancy and are a dose-metric of prenatal exposure to PBDEs. Additional exposure occurs during breast-feeding. Although in humans it is difficult to distinguish between exposure of offspring by transplacental or by breast milk transfer, both human and animal data from a different species indicate that accumulation of highly persistent chemicals ingested *via* milk far exceeds the contribution from maternal-foetal transfer.^{2,3}

In order to characterize the current levels of infant exposure due to breast feeding in Italy, we analyzed milk from mothers from the general population of Rome and Venice and its surroundings. In this latter case, an assessment of PBDE levels was carried out as a function of maternal fish consumption, remarkably higher than the national mean value in some subgroups of the local population.

Materials and methods

Sampling. Lactating mothers from Rome (one group/pool: 10 donors) were enrolled in the period from January 2000 to July 2001. Mothers from Venice and its surroundings (three

ORGANOHALOGEN COMPOUNDS - Volume 66 (2004)

groups/pools: LC, 10 donors; MC, 13 donors; HC, six donors) were enrolled in the period from April 1998 to October 2000. Age distribution of mothers enrolled in all groups was 21–40 years. Except for an excess fish consumption of the HC group, dietary habits of all subjects were substantially representative of the mean Italian diet. Mothers from Venice and its surroundings were grouped according to their consumption of fish and molluscs: Pools LC, MC, and HC are associated with respectively low, medium, and (remarkably) high fish consumptions. Mothers from the Rome area had an average fish consumption.

Analysis. Milk pools from each group were formed by adding the same quantity of milk from each donor. Samples were added with ¹³C-labelled standards and subjected to liquid-liquid extraction with ethyl ether and *n*-hexane. Clean-up was carried out by a multi-step procedure as described by Päpke et al.⁴ HRMS-HRGC(SIM) was utilized for quantification of PBDE congeners 28, 47, 66, 85, 99, 100, 138, 153, 154, and 183.

Results and discussion

The cumulative PBDE concentration in the milk pool from Rome was of 4.1 ng/g, lipid base (*lb*). Levels in the three pools from Venice (LC, MC, and HC), characterized by increasing levels of fish consumption, were respectively of 2.8, 2.5, and 1.6 ng/g *lb*. Congener-specific data are shown in Table 1 and Figure 1. As observed in most studies on human milk,⁵⁻¹⁰ in all our pools PBDE congeners 47, 99, 100, and 153 are the most abundant, their sum accounting for over 80 % of total concentration. In the pools from Venice and its surroundings, the cumulative PBDE levels appear to decrease with increasing fish consumption. Contrary to that, H₆BDE 153 relative contribution progressively increases, to become the predominant congener in the group at the highest level of fish consumption: this results in a contamination profile different from what is usually observed in human milk, with T₄BDE 47 standing out for its relative abundance.

The combined concentrations of the three most abundant PBDE congeners 47, 99, and 153 range between 1.3 and 3.3 ng/g *lb*: these values match those recently reported for some European countries (Table 2 and Figure 2), spanning from 1.5 to 5.3 ng/g *lb*^{5,6} and on average in the order of 3 ng/g *lb*.⁷⁻¹⁰

The outcome of the analysis of edible benthonic mollusks (clams and *cardium*) collected in the Venice lagoon during milk sampling showed a congener profile with a strong predominance of T₄BDE 47 and P₅BDE 99, with H₆BDE 153 levels approximately 15–20 % of the T₄BDE 47 concentration (Figure 3). The biological differences between the two organisms might account for the large diversity in body burdens (40 and 15 ng/g *lb*, respectively): however, it should be pointed out that clams, grown in a lagoon area under a greater industrial impact, exhibit a PBDE body burden much higher than that measured in *cardia*, the latter being obtained from a fishing zone. Although a correlation between PBDE levels in lagoon biota and the body burden of local people was beyond the scope of our study, it is nevertheless of interest to highlight the presence of PBDEs in low trophic niches at levels comparable with those of several PCB congeners.

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Table 1. PBDE congener concentrations and distribution in human milk samples (ng/g lb) from Venice and Rome.

| PBDE | Venice (LC) | Venice (MC) | Venice (HC) | Rome |
|------------------------|-------------|-------------|-------------|--------|
| T ₃ BDE-17 | 0.0038 | 0.0042 | < 0.002 | 0.0039 |
| T ₃ BDE-28 | 0.065 | 0.064 | 0.036 | 0.082 |
| $T_4BDE-47$ | 1.5 | 0.90 | 0.55 | 1.9 |
| T ₄ BDE-66 | 0.015 | 0.037 | < 0.006 | 0.019 |
| P ₅ BDE-85 | 0.035 | 0.045 | 0.018 | 0.074 |
| P ₅ BDE-99 | 0.41 | 0.51 | 0.14 | 0.97 |
| P ₅ BDE-100 | 0.28 | 0.19 | 0.15 | 0.48 |
| H ₆ BDE-138 | < 0.01 | 0.020 | < 0.01 | 0.013 |
| H ₆ BDE-153 | 0.41 | 0.47 | 0.60 | 0.47 |
| H ₆ BDE-154 | 0.025 | 0.047 | 0.020 | 0.070 |
| H ₇ BDE-183 | 0.061 | 0.19 | 0.050 | 0.092 |
| $\Sigma PBDE$ | 2.8 | 2.5 | 1.6 | 4.1 |



Table 2. PBDE congener concentrations and distribution in human milk samples (ng/g lb) from some European countries. Values rounded off to two figures.

| Country | Year | 47 | 99 | 100 | 153 | 154 | S PBDE | Reference |
|-----------------|-----------|------|------|------|------|--------|---------------|-----------|
| Belgium | 2000-2001 | 1.7 | 0.35 | 0.17 | 0.43 | 0.12 | 2.8 | 8 |
| Finland | 1996 | 0.85 | 0.35 | | 0.29 | | 1.5 | 5 |
| Sweden | 2000 | 1.2 | 0.21 | 0.14 | 0.32 | 0.02 | 1.8 | 10 |
| The Netherlands | 1998 | 1.2 | 0.37 | 0.31 | 0.95 | < 0.08 | 2.9 | 7 |
| UK | 2001-2003 | 3.0 | 0.9 | 0.6 | 1.4 | 0.5 | 6.6 | 6 |





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ORGANOHALOGEN COMPOUNDS - Volume 66 (2004)