ORGANOHALOGEN POLLUTANTS IN HUMAN SAMPLES OF THE CZECH POPULATION

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Introduction

Since the 70´s of the last century, numerous surveys on the levels, distributions and time trends of persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins and furans (PCDDs/Fs) or organochlorine pesticides (OCPs) occurring in various environmental compartments were initiated in many countries. In the recent two decades, an increasing attention has been focused also on brominated flame retardants (BFRs), a group of "emerging" halogenated POPs that, similarly to chlorinated POPs, tend to accumulate in lipid rich tissue across the food web. Human population is exposed to these man-made chemicals primarily via diet; fatty fish, meat and dairy products being the main sources. Inhalation is another route of the human exposure, because BFRs may occur in the ambient air and/or the indoor environment typically associated with solid dust particles [1,2].

The Czech population body burden of halogenated POPs is of a great concern due to incautious handling of hazardous chemicals in the past time. A monitoring of human matrices, including polybrominated diphenyl ethers (PBDEs) has been employed for this purpose in the recent decade.

The main aim of the presented study was to investigate levels and profiles of (i) BFRs (PBDEs) and (ii) "classical" POPs occurring in adipose tissue and in human breast milk of Czech population. To our knowledge, this is the first study in this field. The results of this study compared PBDE levels in human matrices of general Czech population with similar studies conducted recently in other countries.

Materials and Method

Breast milk.

Samples of breast milk were obtained from 56 Czech women living in Olomouc region (located in the north-east part of the Czech Republic) during the autumn 2007 in the co-operation with the Gynaecological-maternity Clinic, Faculty Hospital in Olomouc. The age of mothers participating in this study ranged from 21 to 45 years (mean and median 29 year). The breast milk was expressed manually into the glass bottles and stored at 10°C until analysis according to WHO methodology [3]. The analytical procedure used for analysis of human breast milk samples was described in detail in our earlier study [4,5].

Human adipose Tissue.

Adipose fat tissue samples for determination of selected POPs were collected from patients (n=98) who underwent a tumescent liposuction for aesthetic reasons. The mean age of the group was 35.5 years with a range of 17–60 years. The analytical procedure used for analysis of adipose tissue samples has been described in detail in our earlier study [6]; therefore, there is only a brief summary of procedure steps. Approximately 5 g of adipose fat tissue sample was homogenized with anhydrous sodium sulphate (20 g) and extracted in a Soxhlet apparatus for 8h using a hexane:dichlormethane mixture (1:1, *v/v*, 150 ml). The extract was rotary evaporated at 40°C and residues were weighted for a lipid determination. An aliquot of isolated fat (cca 750 mg) was dissolved in 10 ml of an internal standard (PCB 112) solution (cyclohexane:ethylacetate, 1:1, *v/v*). Sample extracts were then purified on a Bio Beads S-X3 column using cyclohexane:ethylacetate (1:1, *v/v*) as a mobile phase. A fraction corresponding to a elution volume of 14–30 ml was collected.

Results and discussions

The first data on the occurrence of BFRs in Czech humans employing breast milk as a bioindicator matrix were reported by Kazda et al. in 2004 [6], however, the information on the other major POPs, such as PCBs and DDTS was not provided.

An overview of the most abundant PBDE and PCB congeners together with OCP levels in both types of examined matrices collected in the Czech Republic within presented study is shown in Table I. The results clearly document ubiquitous occurrence of PBDEs emission sources in the environment of general Czech population.

The total PBDE concentrations (sum of BDEs 28, 47, 49, 66, 85, 99, 100, 153, 154 and 183) ranged between 0.2 and 54.3 ng/g lipid (mean 4.4 ng/g lipid) in adipose tissue and between 0.23 and 20.51 ng/g lipid (mean 3.53 ng/g lipid) in breast milk. BDE congeners 47 (tetrabromo-), 153 (hexabromo-) and 183 (heprabromo-) were predominant and accounted up to 80% of the total PBDEs. Other relatively abundant representatives of this group were congeners BDE 99 and 100 (both pentabromo-). BDEs 28, 49, 66, 85 and 154 were detected only in several samples, mostly close to the limit of quantification

As compared to PBDEs, the levels of OCs (PCBs and OCPs) were up to 2-3 orders of magnitude higher, obviously, due to a relatively longer history of their intensive use in a wide range of areas. Although PCBs, DDT and several other persistent OCs were banned many decades ago, and, in spite of their decline witnessed in the Czech Republic alike worldwide, these POPs are still persisting in very high quantities in the environmental compartments. Not surprisingly, their transfer into the food chain still continues, what was documented also in this study. The OCs pattern found in Czech breast milk was: PCBs > DDTs > HCB > HCHs.

The mean values for the sum of seven indicator PCB congeners (No. 28, 52, 101, 118, 138, 153 and 180) in breast milk and adipose tissue were 592 and 625 ng/g lipid, respectively. The following PCBs profile was found: PCB-153 > PCB-138 > PCB-180 > PCB-118 > PCB-101 ~ PCB-52 ~ PCB-28. The dominating PCB congeners No. 153, 138 and 180 contributed together to the total PCBs content by almost 80%

Regarding to other chlorinated POPs, p,p´-DDE was their major representative in all analyzed samples. The contamination input in the food chain obviously occurred many years ago since the parent compound, p,p´-DDT, was significantly lower. Other OCPs like HCB (hexachlorobenezene) and β-HCH (hexachlorocyclohexane), were apparently in all samples at levels above LOQs (see Table 1).

Interestingly, PBDE profile in human adipose tissue found in our present study was not identical to that one observed in breast milk samples. While in the latter matrix BDE 47 was the most abundant congener followed by BDE 153, opposite ratio of these major PBDEs was found in adipose tissue samples (see Fig. 1). Biotransformation and accumulation kinetic properties of individual PBDE congeners after human exposure may contribute to these differences. As shown in Fig. 1, a similar trend was observed in a recent Japanese study [7], while this was not reported in a study conducted in Sweden, which documented, in both human milk and adipose tissue, the levels of BDE 47 to be approximately 4-times higher than BDE 153. The use of Penta-BDE technical mixture with a majority of BDE 47 was probably dominating in this country [8]. In this Scandinavian country this difference could be attributed to different uses of these products or a different in the diet of individuals. On the other hand, no significant differences were found by a comparison PCB profiles in both examined matrices.

Generally, based on available studies, PBDE levels in breast milk and adipose tissue samples collected within this study were comparable to those reported in other European countries but considerably lower (by almost one order of magnitude) than results from the United States, probably due to a less extensive use of this group of BFRs in common goods and products. The PBDEs content measured in this study was comparable with data reported in similar samples collected in Spain, Sweden, Belgium and Japan, whilst slightly lower than in the United States [9-18]. The levels of PCBs were similar to those found in other European countries. While no age dependency was found for PBDEs, an increase of PCB and OCP levels with age was observed. Different exposure routes of donors were documented by the absence of the relationship between PCBs and OCPs.

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Table 1: Levels of PBDEs, PCBs and OCPs determined in breast milk and adipose tissue collected in the Czech Republic during the year 2007 (ng/g lipid)

	Breast milk (n=56)			**Adipose tissue (n=98)**		
Analyte	**Mean**	**RSD (%)**	**Median**	**Mean**	**RSD (%)**	**Median**
BDE 47	1.52	88	1.11	1.1	157	0.7
BDE 99	0.73	114	0.53	0.5	182	0.2
BDE 100	0.30	123	0.17	0.5	166	0.3
BDE 153	0.54	146	0.21	1.3	158	1.0
BDE 154	0.14	123	0.11	0.2	181	0.1
BDE 183	0.32	164	0.15	0.7	116	0.4
∑ 10 PBDEs	3.23	96	2.06	4.4	139	3.1
PCB 118	17.84	139	13.43	17.7	58	14.3
PCB 138	143.61	68	119.84	121.6	57	110.1
PCB 153	229.81	61	202.76	233.6	64	219.8
PCB 180	192.98	79	166.67	245.0	64	230.7
∑ PCBs	591.63	67	497.81	625.5	60	595.0
HCB	40.78	54	34.72	120.4	111	72.1
β-HCH	11.28	62	8.31	23.9	81	17.5
p,p´-DDE	178.95	68	148.81	582.5	73	478.8
p,p´-DDD	4.26	45	3.94	1.7	90	1.3
p,p´-DDT	22.04	79	16.64	24.9	72	20.4
∑ DDTs	220.14	59	195.07	615.6	71	509.7

Figure 1: Comparison of PBDE and PCB profiles in both types of examined matrices

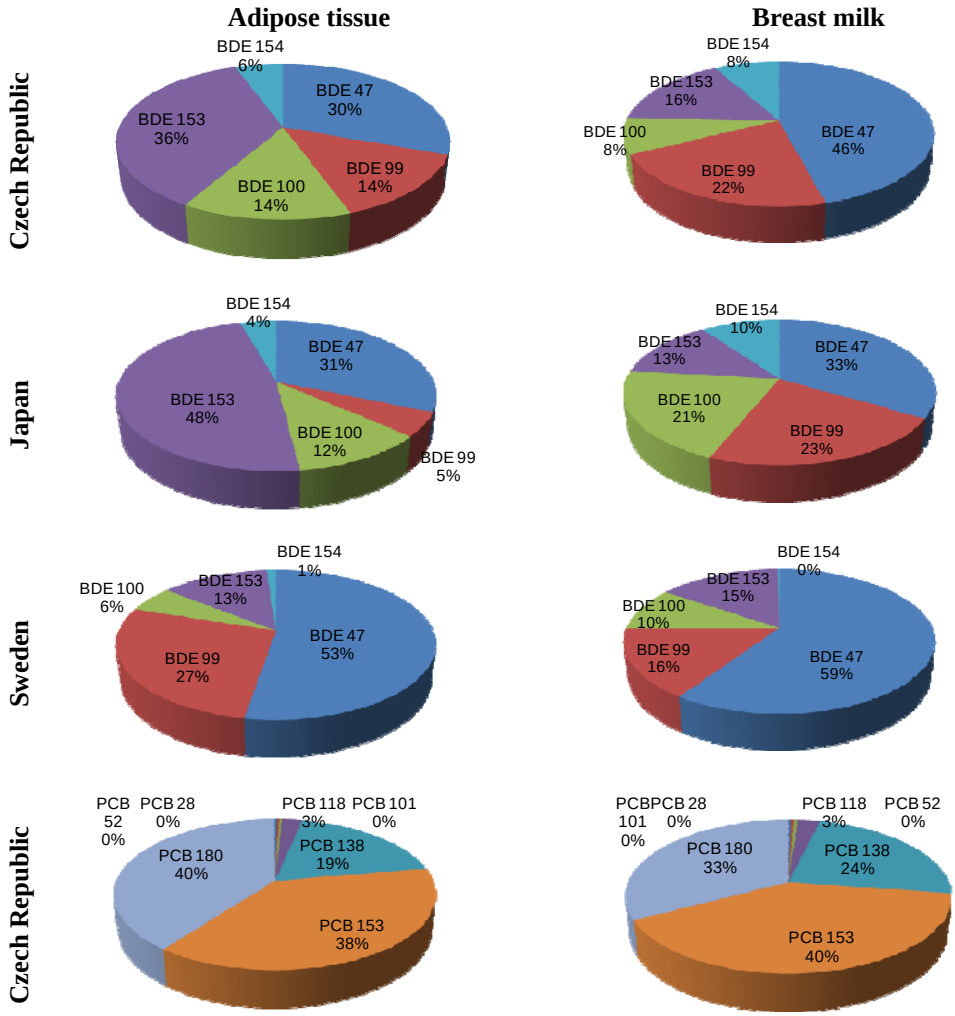

Figure 2: Comparison of PBDE levels in various countries; (A) breast milk and (B) human adipose tissue