

POLYBROMINATED FLAMES RETARDANTS

HIGH LEVELS OF PBDES IN 5 % OF 220 BLOOD SAMPLES FROM THE SWEDISH POPULATION

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Introduction

Brominated flame retardants i.e. PBDEs are found in the low ng/g range in the general population in Northern Europe¹⁻³. It is generally assumed that the levels in the general population are a result of exposure through food. Elevated levels have been reported for workers in computer recycling facilities^{4,5}. Recent data shows that human samples from Northern America contain higher levels than the values reported from Europe⁶⁻⁸. In the here presented study blood from a cohort of 220 persons from Sweden were analysed for PBDEs (Tetra-Hexa). This specific cohort existed of mother and son pairs of a control group and a group of which the sons were diagnosed with testicle cancer. In addition to the here presented PBDE data all samples were analysed for 35 individual PCB congeners, DDE, chlordanes and HxCBz. Results concerning the epidemiology of this study will be presented elsewhere, here we discuss our findings of high levels of PBDEs in 5% of the samples in both the control and case group.

Material and Methods

Blood samples were collected throughout Sweden but mostly in the Örebro region of both mothers and sons of which the sons were diagnosed with testicle cancer. In addition an age matched control pair was selected. For each person 20 ml blood was taken and frozen until analysis. Of the blood samples 10 ml was mixed with ethanol and applied to a hydromatrix column after the internal standard ¹³C PBDE #77 was added. After elution of the blood lipids from this column with a mixture of isopropanol/hexane and removing water from the concentrated extract with sodium sulphate, the amount of blood lipids was established gravimetrically. Removal of the lipids was achieved by using a multiple layer silica column consisting of H₂SO₄-silica, neutral silica and KOH silica. The extraction and clean up method was adapted from Pöpke et al.^{9,10}. 2 µl of cleaned sample extract concentrated in 50 µl tetradecane was analysed by GC/MS. Two different ionisation methods were used. Electron impact monitoring the two most abundant masses of the bromine molecular cluster for screening of the samples and NCI ionisation monitoring masses 79 and 81 to achieve lower detection limits. The first 35 blood samples were only run in the EI mode due to a different spiking protocol including ¹³C PBDE #47, #99 and #153. A laboratory blank sample was analysed with each set of 10 samples, blank values were in all cases lower than 10 % of the values measured in the samples.

Results and discussion

The results of the analysis are summarised in Table 1 and graphically displayed in Figure 1. In addition the levels of three computer recycling employees analysed by the same method are given in Table 1. In total 220 blood samples were analysed within this study consisting of mother/son case pairs

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and mother/son control pairs. In the large majority of the blood samples levels of around 5 ng/g lipids were found. The distribution of the samples is shown in Figure 1, where the frequency is plotted against the levels measured. Figure 1 almost resembles a normal distribution between 1.0 and 13 ng/g lipids with a mean value of 4.9 ng/g lipids for the sum of tetra through hexa PBDE isomers. This is in accordance with other studies in Europe where levels between 2 and 7 ng/g (lw) are reported. This is somewhat lower than levels reported for computer recycling employees from our own studies and the literature^{4,5,11}. What is surprising however is that in 10 of the samples extreme high levels of PBDEs were found, with one extreme exceeding 1000 ng/g (lw). The high levels were both found in the case group and the control group. What is even more surprisingly is that two mother/son pairs could be identified (case 42, control 145) with high levels. In both mother/son pair the mother showed the highest levels of PBDEs. The same pattern was seen in both the mother and son in these two mother/son pairs.

Table 1. Levels of PBDE ng/g lipids in case and controls of the testicle cancer study and their mothers and the levels in computer recycling employees from ref¹¹

| Sample Information | NCI Data | | | | EI Data | | |
|----------------------------------|------------|------------|------------|------------|---------------|----------------|----------------|
| | # 47 | # 99 | # 153 | Sum PBDE | # 47 | # 99 | # 153 |
| Case 4 na | na | na | na | 10 | < 15 | < 20 | |
| Case 35na | na | na | na | 53 | < 15 | < 20 | |
| Case 42178 | 73 | 22 | 272 | 192 | 48 | < 40 | |
| Mother Case 42 | 716 | 353 | 153 | 1221 | 740 | 313 | 156 |
| Mother Case 119 | 177 | 325 | 35 | 537 | 192 | 323 | 22 |
| Mother Case 37 | na | na | na | na | 21 | < 15 | < 20 |
| Control 153 | 28 | 6 | 6 | 40 | < 11 | < 30 | < 40 |
| Control 145 | 35 | 12 | 4 | 52 | 21 | < 15 | < 20 |
| Mother Control 145 | 478 | 156 | 52 | 686 | na | na | na |
| Mother Control 117 | 106 | 19 | 9 | 134 | 99 | 22 | < 20 |
| Mean (n =7) | 245 | 135 | 40 | 420 | 166 | 176 | 89 |
| Rest of samples (n = 136) | 2.2 | 1.1 | 1.6 | 4.9 | < 5 | < 10 | < 20 |
| Computer Recylcer 1 (n = 7)4.1 | | 3.6 | 2.0 | 11 | | | |
| Computer Recylcer 2 (n = 7)3.2 | | 3.1 | 2.5 | 10 | | | |
| Computer Recylcer 3 (n = 7)5.3 | | 4.5 | 1.2 | 12 | | | |
| Mean | 4.2 | 3.7 | 1.9 | 11 | | | |

The general population is mainly exposed to PBDEs through the food chain which results in background levels of 1-6 ng/g lipids. Occupational exposure has been identified for employees of computer recycling facilities. The extreme high level for the 10 samples presented here are a surprise and an indication of an unknown source for PBDEs. None of the persons showing the highest PBDE

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Histogram PBDE in Human Blood (n=143)

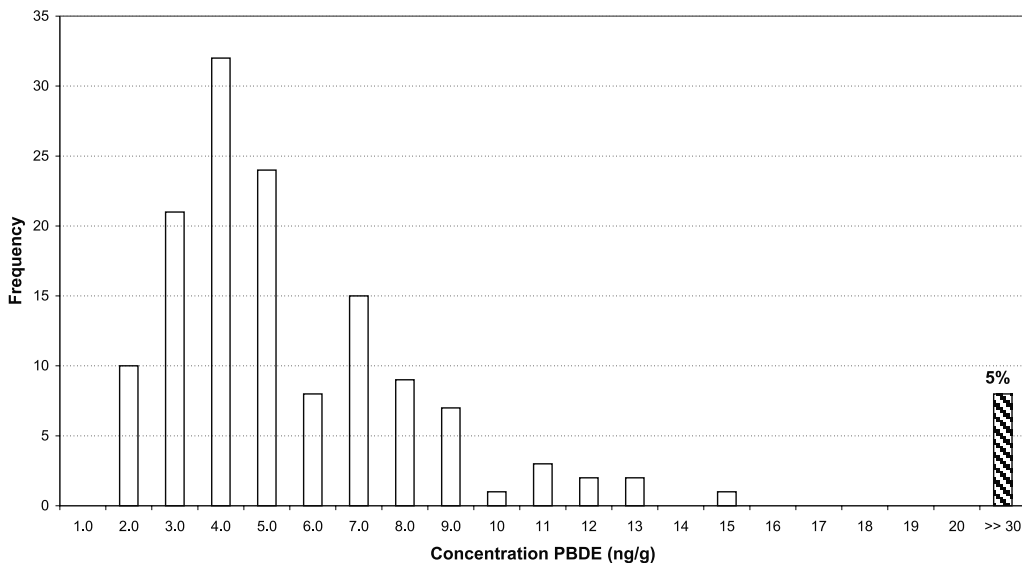


Figure 1. The distribution of 143 samples rerun using NCI to enhance the detection limit for PBDE, 5% of the samples showed levels of PBDE larger than 30 ng/g lipids

levels were working at a computer recycling facility or similar industries. Although the reason is still unknown, it is speculated that the high levels might be related to the home environment. This is strengthened by the fact that two mother/son pairs showed high levels of PBDE. In addition this might explain the higher levels found in several samples in the US where PBDEs are used to a larger extent in the home environment than in Europe.

To definitely exclude contamination during sampling of the blood and to follow up this study, new samples will be taken of the persons with the extreme high PBDE levels. A more thorough investigation of the work and home situation will also be part of this follow up study.

Acknowledgements

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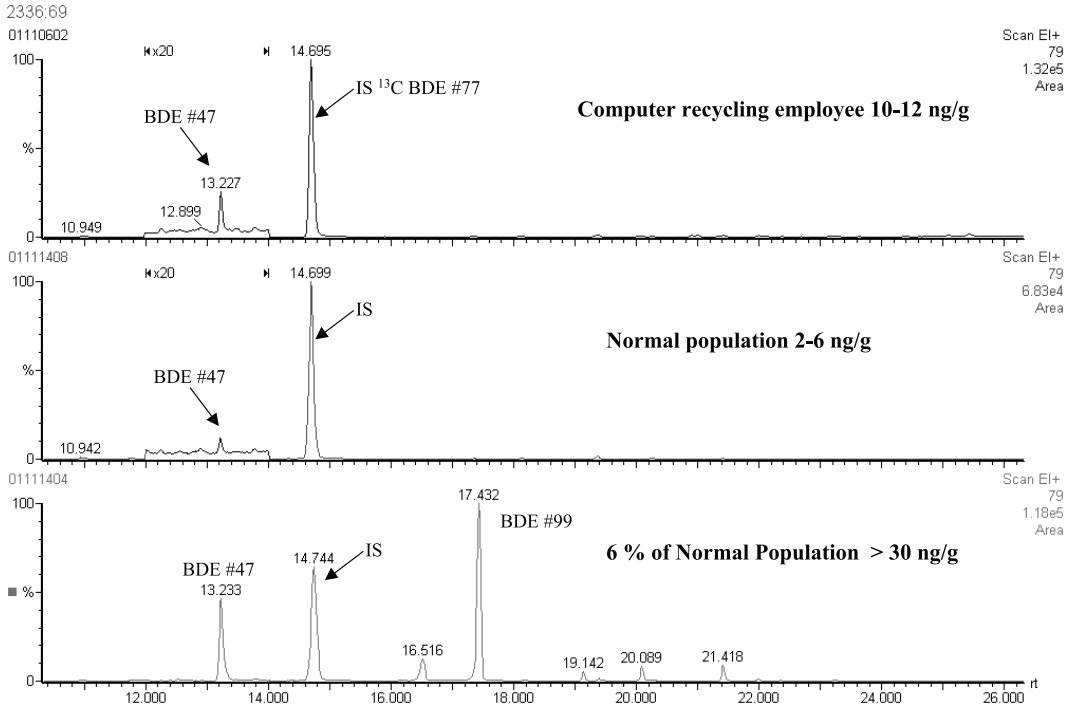


Figure 2. A NCI GC/MS chromatogram showing mass 79 of a 'normal' sample, a computer recycler and one of the extreme high level samples.

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