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Polybrominated diphenyl ether exposure to electronics recycling workers – a follow up study

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Abstract

Workers at an electronics recycling plant have previously been shown to have elevated serum levels of polybrominated diphenyl ethers (PBDEs) compared to referents without occupational PBDE exposure. Subsequent structural changes and industrial hygiene measures at the plant were applied to improve the work environment. The present study aims to assess the impact of these work environment changes on the occupational exposure to PBDEs.

Blood were drawn from the workers and analyzed at two different laboratories, and serum concentrations of several PBDE congeners were determined by GC/MS or GC/HRMS. Cross-sectional studies were performed prior to (in 1997; $N = 19$) and after (in 2000; $N = 27$) workplace improvements. Longitudinal studies were performed on twelve of the workers that were sampled at both occasions.

Even though the amount of processed goods had doubled in 2000 as compared to 1997, there was a significant decrease in the serum levels of BDE-183 and BDE-209. For BDE-209 the levels observed in year 2000 were even lower than in referents with no occupational exposure. In contrast to the decrease of higher brominated diphenyl ethers, the concentrations of BDE-47 did not significantly change. For BDE-153, the cross-sectional study indicated no change, whereas the longitudinal follow up indicated a significant increase.

This study shows that the industrial hygiene improvements clearly reduced the occupational exposure to BDE-183 and BDE-209 at the plant. Still, the levels of hexa- to nonaBDEs but not BDE-209 were elevated, compared to referents with no occupational exposure.

Keywords: PBDE; BFR; Brominated flame retardants; Occupational exposure; Blood levels; Analysis

1. Background

An increasing number of electric and electronic devices for household or professional use are present in the technosphere and, finally, approach their end of life. To promote sustainability it is important to optimize the recycling of these products. Printed circuit boards hold valuable metals as copper, silver and gold and the cabinets are made of polymers that either can be recycled as such or used for energy production (Imai et al., 2003; Drohmann et al., 2004). It is however important that added chemicals do not interfere in the recycling processes, leading to poor quality of down stream products or to occupational exposure to personnel handling the goods. Brominated flame retardants (BFRs) are used in ABS-plastics, high-impact polystyrene, epoxy resins, and rubber (WHO, 1995, 1997), and may thus appear in electrical appliance consumer products, such as printed circuit boards, computer housing, cables and TV-sets (WHO, 1997). Work related exposure to BFRs such as tetrabromobisphenol A, and in particular polybrominated diphenyl ethers (PBDEs), have

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been reported from several electronics dismantling facilities in Scandinavia (Sjödin et al., 1999; Hovander et al., 2001; Thomsen et al., 2001). The toxicity of BFRs and PBDEs was recently summarised by Birnbaum and Staskal (2004) and Gill et al. (2004). PBDEs have been shown to have toxic effects, such as endocrine effects (Vos et al., 2003) and developmental neurotoxicity (Eriksson et al., 2001; Viberg et al., 2003a,b, 2005).

In 1999 a first survey was published, confirming that workers dismantling discarded electronics at an electronics recycling plant in Southern Sweden were exposed to PBDEs (Sjödin et al., 1999). These results were based on analysis of serum samples drawn from the workers in 1997. The exposure to BFRs at this plant was also described by indoor air measurements at the facility (Sjödin et al., 2001). In response to these findings, industrial hygiene measures to reduce PBDE exposure were taken by the company. The primary aim of the present study was to evaluate if the work place improvements had reduced the personal PBDE exposures (i.e. serum concentrations) on group level as well as on individual level.

2. Material and methods

2.1. The plant

Manual dismantling of electronic goods such as personal computers, printers, television sets and radios is performed at the plant. Discarded electronics goods are stored inside the facility until further processing. Trucks are used for transport of goods within the facility, which has an open lay out (Fig. 1). The discarded electronics are dismantled at different work stations using air pressure driven tools, components are separated, and hazardous components are removed. All plastics, such as computer cabinets, are separated further into bromine containing and non-bromine containing plastics, using X-ray technology. All plastic fractions are separately ground to pieces in a shredder for volume reduction. The plastic material is packed and transported to other industries, either for recycling of the plastics, or for incineration.

The shredder, which in 1997 was placed indoors in the dismantling hall, was the main contaminating source of PBDEs in the factory (Sjödin et al., 2001). Hence, when major industrial hygiene improvements were made at the plant, the shredder was placed outside the building (Fig. 1). In 1997 there was no ventilation in the factory, and the temperature and quality of air was regulated by opening the doors and windows. For improvement a specific process-ventilation system was installed, forcing the airflow from ceiling to floor in order to remove particles and dust from the air. The flow rates of this process-ventilation were 7845 l/s to the factory and 8280 l/s going out. In 1997, only brooms were used for cleaning of the floor. The cleaning routines were then improved. An industrial vacuum cleaner operating on wet floors was installed and the work benches and work stations surfaces were wept with wet rags. In November 1999 the changes were fully implemented.

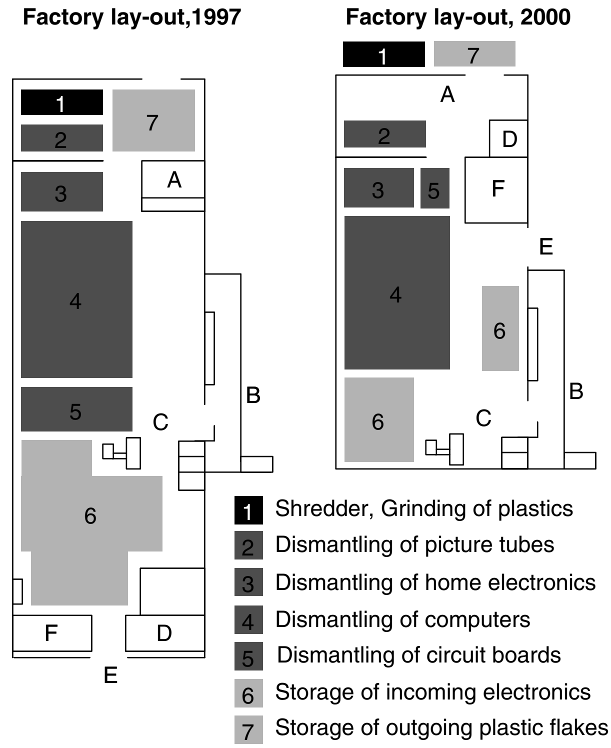


Fig. 1. Schematic figure of the electronics recycling factory plant lay-out, in 1997 (left) and 2000 (right). The figure is showing the structural changes made within the plant between the years. The major change made was moving the shredder (the main contamination source) outside the plant. A process-ventilation system was installed at the dismantling stations and the storage area was reduced. The letters given in the figure are; Storage of hazardous waste (A); loading dock (B); sorting and weighing of incoming electronics (C); locker room and showers (D); main entrance (E); office (F).

During the same time period as the industrial hygiene improvements were implemented the volume of dismantled and recycled electronics at the factory increased markedly, and was doubled between 1997 and 2000. In 1997 the factory processed 1900 tons of discarded electronics, in comparison to 3800 tons in 2000, which corresponds to 75 and 195 tons of brominated plastic materials, 1997 and 2000, respectively. Even though the workforce was increased, the productivity of the workers was higher in 2000 than in 1997. The processed electronics comprise of discarded goods, with different age, from households and industry in Sweden The relative amount of bromine containing plastics being dismantled did not change. In the time period of 1997–2000, the relative bromine content of the plastic fraction was 3–4%. BFR-specific information is not available.

2.2. Subjects and sampling

In June 1997 nineteen out of 27 employees participated (Sjödin et al., 1999). Of these, 13 employees were sampled once again in June–July, 2000, together with 14 workers

Table 1
Study groups of electronics dismantlers

Year of sampling	Laboratory A		Laboratory B	
	Blue collar	White collar	Blue collar	White collar
1997 only	6	0	–	–
1997 and 2000	9	1	3	0
2000 only	2	1	5	6

All blood samples from year 1997 were analyzed at Laboratory A, whereas the samples from year 2000 were analyzed either at Laboratory A or at Laboratory B.

not previously sampled (Table 1), out of a workforce of 47 persons. There were four female participants in 1997, and 10 in year 2000. The personnel are divided into blue collar and white collar workers in the present study to identify dismantling and office workers, respectively. The blue collar workers operate at individual workstations, dismantling discarded electronics by hand, using pneumatic driven tools. All dismantling is performed in one large open space within the factory (Fig. 1). The dismantling hall is also used partly for storage of incoming goods. White collar workers perform their office work tasks within the same factory building, but the offices are separated from the large dismantling hall by walls. There was no or little change of work tasks between 1997 and 2000 for blue collar workers participating in both samplings. Informed consent was obtained from all subjects and the study was approved by the Ethics Committee, Lund University, Sweden (LU 227-97).

Blood (10 g) was drawn from a cubital vein into evacuated plain tubes (Vacutainer, Rutherford NJ, USA) on an ordinary work day. The serum was centrifuged, transferred to dark coloured acetone-washed glass bottles, frozen and kept at −20 °C until start of the chemical analysis. The samples from 1997 have previously been analyzed at Lab A (Stockholm University). No serum was available for reanalysis. The samples from 2000 were divided in two groups, and sent to two laboratories, A and B (ERGO Forschungsgesellschaft mbH), respectively. For workers participating in the longitudinal study, 9 out of 12 samples were analyzed at Lab A. In blue collar workers whose samples were analyzed at Lab A, the median age was 47 years (range 31–57), and the median employment time was 5 years (range 1–8). The corresponding figures for Lab B were for median age 49 years (range 34–53), and for median employment time 2 years (range 0.2–8).

For comparisons with a non-occupationally exposed population, we used a male referent group of abattoir workers with little or no computer experience from southern Sweden, sampled in year 2000 and analyzed by Lab A (Thuresson et al., 2005a).

2.3. Chemical analysis

The sample preparation and analysis procedure were similar between calendar-years and laboratories. All serum samples (5 g) were extracted by liquid/liquid extraction between serum and organic solvents, and all cleanup was performed using sulfuric acid in order to remove lipids from the samples. Analysis of serum samples from 1997 was performed at Lab A as described by Sjödin et al. (1999). Serum samples from 2000 were analyzed either at Lab A as described by Thuresson et al. (2005a) or at Lab B as described by Päpke et al. (2004). Lab A used GC–MS, with the MS being run in the electron capture negative ionisation (ECNI) mode and selective ion monitoring (SIM) of the bromide ions: *m*/*z*: 79 and 81 (Buser, 1986). Lab B used HRMS with ^{13}C-labeled reference standards for quantification. All PBDE concentrations reported have been adjusted to gravimetric determinations of lipid weight (l.w.), and are expressed as pmol/g l.w. Performance of extraction, cleanup and analysis for methods used by both Lab A and B have been reported previously (Sjödin et al., 1999; Hovander et al., 2000; Päpke et al., 2004; Thuresson et al., 2005a).

As samples drawn from the workers in the year of 2000 were analyzed at two different laboratories, a possibility for interlaboratory comparison appeared, and we planned for split sample analyzes. However, when all analyzes at Lab A and B had been performed and the sample codes were broken, it was realised that there had been a misunderstanding of the sampling procedure at the plant, and all that remained for any kind of interlaboratory comparison was samples from five individuals sampled twice, 4 weeks apart, and analyzed at the two Labs, but not true split samples. BDE-209 was not quantified in any of these five samples at Lab B. BDE-47 was quantified only in one sample, BDE-183 and BDE-99 in four samples, and BDE-99 and BDE-153 in all five samples. In contrast, these congeners were quantified in all samples at Lab A. Thus, it was not meaningful to compare laboratory results for BDE-47 and BDE-209. The correlations between Lab A and Lab B were good or fair for BDE-99 ($r_s = 0.90$), BDE-100 ($r_s = 0.89$), BDE-153 ($r_s = 0.90$), and BDE-183 ($r_s = 1.0$); all reaching statistical significance. The ratio Lab A/Lab B (median) were 1.8 for BDE-99, 1.4 for BDE-100, 1.2 for BDE-153, and 1.1 for BDE-183. Thus, even if a formal split sample analysis is lacking, it seems reasonable to collapse data from Lab A and Lab B for BDE-153 and BDE-183.

OctaBDEs and nonaBDEs were determined only at Lab A, in serum samples from 2000. BDE-154 was determined only at Lab B. The method used by Lab A did not permit quantification of BDE-154 since this compound co-eluted with 2,2′,4,4′,5,5′-hexabromobiphenyl with the MS technique applied.

3. Statistics

Non-parametric tests were used for group comparisons (Mann–Whitney *U*-test), paired observations (Wilcoxon sign rank test), and correlations (Spearman's rank correlation). In the calculations, values below the limit of quantification (LOQ) were set to half of the LOQ. A *p*-value of <0.05 was considered to be statistically significant.

4. Results

The samples from year 2000 were analyzed at two different laboratories, thus, the results are reported separately for each laboratory (Table 2). In blue collar workers the median and ranges from the two laboratories were rather similar, with a variation of less than a factor of 2 for BDE-99, -100, -153, and -183. The concentrations of PBDEs were not correlated with duration of employment as a blue collar worker. Thus, for comparisons over time in blue collar workers, we combined the two data sets.

The longitudinal analysis in twelve blue collar workers sampled both in 1997 and 2000 (Fig. 2) showed a significant decrease over time in the serum concentrations of the highest brominated diphenyl ethers, BDE-183 and BDE-209 , with a median decrease of 26% and 46%, respectively. On group level similar results were found. There was a significant decrease over time in the serum concentrations of BDE-183 and BDE-209, in the blue collar workforce (Fig. 3). In year 2000, the levels were almost half or a third of those previously observed for BDE-183 and BDE-209,

Table 2
Serum concentrations (pmol/g l.w.) of six PBDE-congeners in electronics dismantlers in year 2000

PBDE-congener	Laboratory A					Laboratory B				
	Blue collar ($n = 11$)		White collar ($n = 2$)		LOQ	Blue collar ($n = 8$)		White collar ($n = 6$)		LOQ
	Median	Range	Subject 1	Subject 2		Median	Range	Median	Range	
BDE-47	7.3	3.6–110	8.0	4.5	3	4[a]	<4–16	4[b]	–	4
BDE-99	3.0	1.1–23	2.1	1.0	1	3.1[c]	<0.7–6.6	0.7[d]	<0.7–1.2	0.7
BDE-100	2.7	1.1–18	2.2	1.5	1	1.9	0.57–4.6	1.0	0.60–2.3	0.6
BDE-153	7.9	4.6–19	4.3	12	4	5.5	3.0–17	2.1	0.90–5.6	0.9
BDE-183	4.4	1.0–9.6	0.5	2.0	1	7.3	2.4–12	1[e]	<1–1.7	1
BDE-209	2.0[f]	<1–5.4	1.9	5.4	1	3[b]	–	3[b,g]	–	3
l.w. (g)	0.034	0.021–0.047	0.024	0.026		0.041	0.016–0.048	0.034	0.024–0.057	
l.c. (%)	0.57	0.38–0.86	0.62	0.48		0.57	0.27–0.82	0.59	0.46–0.74	

Lipid weight (l.w.) and relative serum lipid content (l.c.), as well as limit of quantification (LOQ) are given. The samples were analyzed at two different laboratories.
[a] Four samples below LOQ.
[b] All samples below LOQ.
[c] Three samples below LOQ.
[d] Four samples below LOQ.
[e] Five samples below LOQ.
[f] One samples below LOQ.
[g] Four samples only.

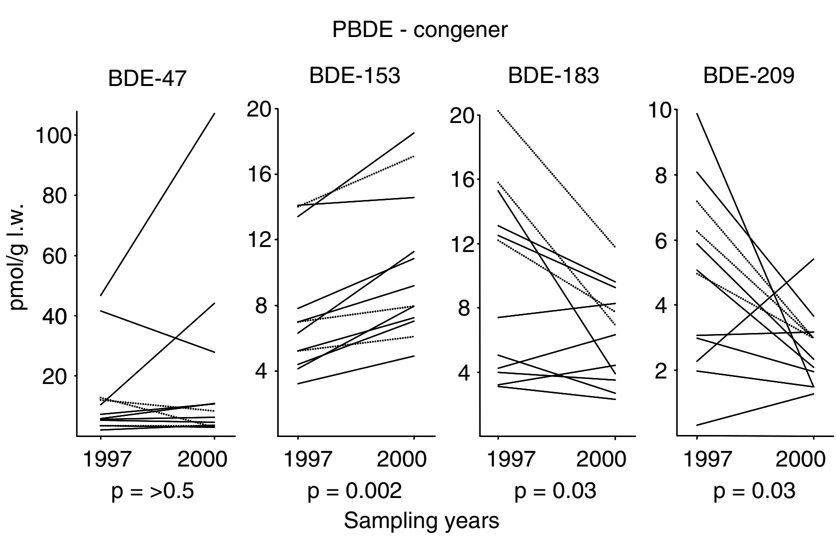
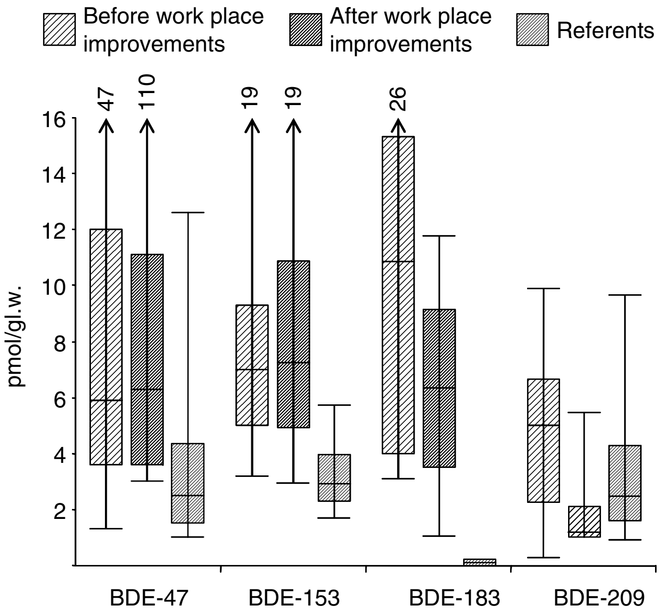


Fig. 2. Serum concentrations (pmol/g l.w.) of four PBDE-congeners in twelve blue collar electronics dismantlers. The subjects were sampled before and after industrial hygiene improvements in the workplace were implemented. Solid lines represent analyzes at Lab A in 1997 and 2000. Dotted lines represent analyzes at Lab A in 1997 and Lab B in 2000. The *p*-values given refer to Wilcoxon signed rank test for paired comparisons.



	BDE-47	BDE-153	BDE-183	BDE-209
A:	>0.25	>0.25	0.05	<0.001
B:	0.003	<0.001	<0.001	0.15
C:	0.008	<0.001	<0.001	0.002

Fig. 3. Serum concentrations (pmol/g l.w.) of four PBDE-congeners in electronics dismantlers and referents. The electronic dismantlers were sampled prior to (year 1997; $N = 19$, data from Sjödin et al. (1999)) and after industrial hygiene improvements were implemented in the workplace (year 2000; $N = 19$; present study, blue collar workers only). For comparison, serum concentrations in a male referent group of abattoir workers, working in a slaughterhouse, with no occupational exposure is also given ($N = 17$; data from Thuresson et al. (2005a)). The median is shown as a bar within each box (that represents 50% of the samples), and the whiskers represents the range of samples. *p*-Values for differences between groups refer to Mann–Whitney *U*-test. (A) Before versus after work place improvements; (B) before work place improvements versus referents; (C) after work place improvements versus referents.

respectively. In contrast, the concentrations of BDE-47 and BDE-153 in the workforce did not significantly differ between the years (Fig. 3). When performing intra-individual comparisons there was even a significant increase of BDE-153 levels between years (Fig. 2).

OctaBDE and nonaBDE congeners were determined only at Lab A in 2000, with concentrations markedly higher than in the referent group (Table 3). BDE-85 was also determined by Lab A but only at low concentrations in comparison with other PBDE congeners, median 0.36 (range <0.1–2.4) pmol/g l.w. Conclusive quantifications of BDE-154 were only possible for Lab B, reporting median 0.17 (range <0.05–0.57) pmol/g l.w.

White collar workers sampled and analyzed in the year 2000 (Table 2) had as a group lower serum levels of all PBDEs, except BDE209, compared to blue collar workers investigated (both labs combined). When only analyzes performed at lab B for blue and white collar workers were compared, statistically significant results were found for BDE-153 and BDE-183.

5. Discussion

This study shows that the work related exposure to BDE-183 and BDE-209 at an electronics dismantling facility can be reduced by standard industrial hygiene measures like structural process planning, good ventilation, and good cleaning procedures. The observed decline of BDE-183 and BDE-209, which was observed both for individuals and on cross-sectional group level, is notable when considering the simultaneous doubling of the quantity of bromine containing goods at the plant. The composition of PBDEs in the processed electronics is not known, but the relative bromine content in the goods did not change between years. As the trend in PBDE use for electronics in Europe has been towards decaBDE during recent years (European Commission, 2002) it is even likely that the relative amount of decaBDE in the dismantled good should have increased between 1997 and 2000.

BDE-183 and BDE-209 have rather short apparent half-lives in humans – three months and 15 days, respectively (Thuresson et al., 2005b) – and are thus suitable biomarkers for evaluation of exposure six months after the full implementation of workplace improvements. It is notable that the BDE-209 concentrations among the workers in year 2000 did not exceed levels observed in male referent

Table 3
Serum concentrations (pmol/g l.w.) of octaBDEs and nonaBDEs in blue collar electronics dismantlers in year 2000

Study groups	octa-1	octa-2	octa-3	BDE-203	BDE-206	BDE-207	BDE-208
Electronics dismantlers ($n = 11$)	0.24 (<0.1[a]–0.47)	4.0 (1.2–8.6)	0.44 (<0.1[a]–1.0)	0.59 (0.24–1.4)	0.28 (<0.2[a]–0.73)	0.70 (0.43–1.4)	<0.1[a] (<0.1[a]–0.35)
<LOQ	1	–	1	–	3	–	7
Referents ($n = 17$)	0.074 (<0.04[a]–0.71)	<0.6[a] (<0.06[a]–0.71)	<0.086 (<0.05[a]–0.19)	<0.2[a] (<0.2[a]–0.49)	0.091 (<0.06[a]–9.7)	0.27 (<0.08[a]–0.65)	<0.08[a] (<0.08[a]–0.65)
<LOQ	5	14	4	16	6	2	13
p-Value[b]	<0.001	<0.001	<0.001	<0.001	0.015	<0.001	0.001

Median, range and the number of samples below limit of quantification (<LOQ) are shown. For comparison, serum levels in a referent group with no occupational exposure are given (data from Thuresson et al. (2005a). All octa- and nonaBDEs were quantified with the response factor for BDE-203.
[a] <LOQ.
[b] Mann–Whitney *U*-test.

workers without any known occupational exposure to PBDEs (Fig. 3). In contrast, the levels of BDE-183, which is considered to be the marker PBDE congener among electronics dismantlers (Sjödin et al., 1999; Hovander et al., 2001; Thomsen et al., 2001), and the levels of octa- and nonaBDEs were still markedly higher than the concentrations observed in the referent group.

The indoor air at the plant contains BDE-47 and BDE-153 (Sjödin et al., 2001), and the workers still have elevated concentrations of this congener compared to the referent population (Fig. 3). There was no obvious reduction of BDE-47 and BDE-153 levels in the workers, neither on a group nor intra-individual basis (Figs. 2 and 3). These PBDE congeners have long estimated half-lives, 1.8 and 6.5 years, respectively (Geyer et al., 2004), and work-place exposure reductions will take time to be reflected in serum levels. This may, at least partly, explain the apparent lack of reduction of low-medium brominated PBDEs, in contrast to the reduction of the higher brominated congeners. Also, there seems to be a general trend that BDE-153 is taking over as the dominating PBDE congener in cohorts with non-occupational exposures, and BDE-153 is now the major PBDE congener present in human blood in populations from the Netherlands (Weiss et al., 2004), the Faroe Islands (Fängström et al., 2004) and Sweden (Thuresson et al., 2005a). Thus, increasing non-occupational exposure to BDE-153, which has been observed in a Swedish longitudinal study (Jakobsson et al., 2005) could tentatively, and in part, explain the finding of increasing levels of BDE-153 in the workers. A speculative explanation for increasing concentrations of BDE-153 may be metabolic formation from higher brominated diphenyl ethers present both in the work and home environments.

The indoor air at the electronics recycling plant also contained BDE-85 and BDE-154 (Sjödin et al., 2001). These congeners are present in the pentaBDE technical products, however, at lower concentrations than BDE-47, -99, -100 and -153 (Sjödin et al., 1998). This is also reflected in the serum PBDE pattern observed in the workers.

It has previously been shown that PBDE-exposure (in this case BDE-209) can differ depending on work task within the same factory facility (Thuresson et al., 2005a). This was also observed in our study. The white collar workers, working within the facility but in a separate area, clearly had lower levels of BDE-47, BDE-153 and -183 than the blue collar workers.

The analytical methods used at the different laboratories and between sampling occasions were similar, but not identical. The most obvious discrepancy between the two laboratories was the concentration data reported on BDE-209 for which the LOQ was different due to differences in the background (blank sample) concentrations. The low LOQ at Lab A is mainly dependent on the use of a Clean room for cleanup of the samples. The LOQ for BDE-47 was also higher at Lab B, most likely for the same reason. For BDE-153 and BDE-183 the concentrations in samples taken four weeks apart (i.e. considerably shorter than calculated half-lives of several years for BDE-153 (Geyer et al., 2004) and three months for BDE-183 (Thuresson et al., 2005b)), were very similar at Lab A and Lab B. Moreover, in the longitudinal analysis nine out of twelve samples from 2000 were analyzed at the same laboratory as in 1997, and the observations in the remaining three workers did not diverge from the rest (Fig. 2). Thus, there are no strong implications that our results have to a larger extent been biased by the analytical design. Extensive air sampling had not been performed before or after the workplace changes. Thus we could only use biomarkers for exposure, which have the advantage of capturing a larger time window of exposure, and integrating exposure from all occupational sources. As there is also environmental exposure to PBDEs, the use of a suitable referent population to estimate background levels, is crucial. It can be noted that we have found rather similar concentrations of BDE-47, -153, -183 and -209 as in the present referents (abattoir workers sampled 2000) in another group of Swedish males sampled 2001 (Jakobsson et al., 2005) – thus our choice of referent population is not invalidating the results.

6. Conclusions

Conventional industrial hygiene improvements in an electronics dismantling plant reduced the exposure to BDE-183 and BDE-209, despite a doubling of the quantity of bromine containing recycled goods between 1997 and 2000. For BDE-209 the serum levels in the workers did not exceed levels that are observed in the general population, whereas the levels of investigated hexa- to nonaBDEs still were markedly increased.

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