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Current data on the background burden to the persistent organochlorine pollutants HCB, p,p′-DDE as well as PCB 138, PCB 153 and PCB 180 in plasma of the general population in Germany☆

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ABSTRACT

Despite their long-term ban, persistent organochlorine compounds like hexachlorobenzene (HCB), p,p′-dichlorodiphenylethylene (DDE) as well as polychlorinated biphenyls (PCBs) are still of environmental concern. For the evaluation of potential occupational or environmental exposures to these substances, it is essential to know the current background burden of the general population. As representative and up-to-date information is missing for Germany, we have analysed a large dataset generated in studies on potential exposure to lower chlorinated PCBs to fill this gap for the levels of HCB, DDE as well as PCB 138, PCB 153 and PCB 180.

We have investigated $n = 2750$ plasma samples of persons of the general population living in North Rhine-Westfalia and Hesse aged 6–65 years and sampled between September 2010 and March 2014. For evaluation of the age-dependent accumulation in the general population we have generated seven age groups in the collective. Our laboratory used a validated and quality controlled procedure using GC/MS for quantification of the organochlorine compounds in plasma (LOQ: 0.01 μg/L). The median (95th percentile) levels for $\sum$ PCB 138 + PCB 153 + PCB 180 were 0.14 (0.73); 0.30 (0.82); 0.38 (0.88); 0.50 (1.14); 0.92; 1.58 (3.54) and 2.41 (4.82) μg/L plasma in the age groups 6–10 years ($n = 102$), 11–17 years ($n = 499$), 18–25 years ($n = 157$), 26–35 years ($n = 710$), 36–45 years ($n = 400$), 46–55 years ($n = 525$) and 56–65 years ($n = 357$), respectively. Similarly, the median (95th percentile) levels of p,p′-DDE were 0.18 (1.24); 0.18 (0.74); 0.24 (0.85); 0.30 (1.20); 0.45 (1.74); 0.64 (3.25) and 0.94 (4.7) μg/L plasma. Finally, the median (95th percentile) of HCB in plasma in these age groups was 0.05 (0.10); 0.06 (0.11); 0.08 (0.15); 0.08 (0.15); 0.11 (0.22); 0.14 (0.42) and 0.20 (0.68) μg/L plasma. Our results prove an overall substantial reduction in the body burden to organochlorine compounds in Germany compared to earlier studies. However, 15% and 3.6% of the examined collective exceeded the HBM-I- and HBM-II-values for PCBs established by the German Human Biomonitoring Commission. Due to a large sample size and a collection period from 2010 to 2014, our data might be suitable for the evaluation of additional exposures to these POPs and may serve as reference values.

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Introduction

Organochlorine pollutants like hexachlorobenzene (HCB), the insecticide dichlorodiphenyl-trichloroethane (DDT) (or its persistent metabolite p,p′-dichlorodiphenyl-dichloroethylene (DDE)) as well as the whole group of polychlorinated biphenyls (PCB) belong to the so called "dirty dozen", substances that are characterised by their high lipophilicity and high bioaccumulative potential. The production and use of these substances has been banned worldwide in the framework of the Stockholm convention, which became effective in 2004 (Stockholm convention secretariat). The resistance of these substances to microbial degradation as well as animal metabolism has led to their accumulation in the food chain and global distribution, which finally resulted in an age-dependent body burden in humans that is mirrored by their blood levels.

HCB is a fungicide that has been widely used in seed treatment. The use of HCB as a pesticide has been banned in Germany since 1981. The use of the insecticide DDT has been banned in Western Germany as early as in 1972, while its use continued

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in Eastern Germany until 1991. This altered usage pattern has resulted in regional differences in the body burden of the persistent DDT-metabolite DDE with higher blood levels in persons originating from the Eastern part of Germany as revealed in the last German Environmental Survey GerES III (German Human Biomonitoring Commission, 2003). The International Agency for research on cancer (IARC) has classified HCB and DDT as potential human carcinogens (class 2B) (IARC, 1991, 2001). Several epidemiological studies have shown endocrine as well as reproductive effects of HCB and DDE in humans (Chevrier et al., 2008, Longnecker et al., 2007). Most recently, a negative association between lung function and plasma DDE levels was found in a large Canadian health survey (Ye et al., 2015).

Previous animal studies showed a hepatotoxic and hepatocarcinogenic potential of HCB in rodents. On that basis and exposure guidance values from different organisations, Aylward et al. (2010) have previously derived biomonitoring equivalents for HCB in serum with cancer-risk specific doses of 800–1500 ng/g lipid in serum (app. 6–11 μg/L serum) for the highest target risk level (corresponding to a risk of 1.00 E−04).

PCBs are technical mixtures consisting of up to 150 PCB congeners (out of 209 possible congeners) which differ in the position and number of chlorine atoms (Frame et al., 1996). As a result of their high stability, these technical mixtures have been extensively used in transformers, capacitors, as plasticisers, flame retardants or as additives for sealant materials during construction of buildings. The production and use of PCBs is legally banned in Germany since 1989, although the production already stopped some years earlier. As for HCB and DDE, food is meanwhile the main source of exposure for PCBs in the general population.

The IARC has recently classified PCB as a human carcinogen (class 1) (Lauby-Secretan et al., 2013). There are numerous epidemiological studies that showed neurological, immunological or endocrine effects of PCB-exposure on humans (Harper et al., 1995; Heilmann et al., 2010; Grandjean et al., 2001; Lee et al., 2006; Winneke et al., 1998). For human biomonitoring, the blood levels of 6 "indicator" congeners are usually quantified, three of them showed especially long biological half-lives (PCB 138, PCB 153 and PCB 180).

As all these organochlorine pollutants are banned in Germany at least for more than 25 years now, the exposure level and in consequence the body burden in the general population has strongly declined over the years and present human exposures seemed to be improbable. However, the irregular handling or removal of contaminated industrial waste might still lead to environmental contamination and potential exposures of workers or residents, as recent cases for PCBs in Germany (Schettgen et al., 2012a) or most recently for HCB in Austria (http://kaernten.orf.at/news/stories/2683507/) showed. Moreover, due to the long biological half-lives of these compounds, even former (occupational) exposures that were a long time ago might still be elucidated by human biomonitoring. For a comprehensive evaluation of such potential exposures, it is an essential prerequisite to know the background burden of the general population to the compounds in question.

However, the background burden to organochlorine pollutants in the adult German population has not been assessed since the German Environmental Survey (GerES III) in 1997–1999 (German Human Biomonitoring Commission, 2003). These reference values are outdated and cannot be used to evaluate present exposures. Previous studies investigating these compounds either used plasma samples that were collected several years ago (Schettgen et al., 2011) and/or were limited by a small number of samples investigated with inappropriate age distribution (Fromme et al., 2015).

In order to fill this gap about the current background burden to these organochlorine compounds, we used a large dataset from studies conducted in the last 3.5 years evaluating elevated exposures to lower chlorinated PCBs in the indoor environment. As previous investigations showed, indoor exposures to lower chlorinated PCBs do not lead to elevated blood levels of the less volatile higher chlorinated PCBs (≥6 chlorine atoms) (Gabrio et al., 2000; Liebl et al., 2004; Herrick et al., 2011; Schettgen et al., 2012b; Meyer et al., 2013). Therefore, our data might give a good impression on the current body burden to organochlorine compounds in Germany.

Materials and methods

Study groups

We have obtained plasma samples from 2755 persons (1470 female, 1285 male) aged between 6 and 65 years. All persons resided in North Rhine-Westfalia or Hesse (Western Germany) and participated in human biomonitoring studies to evaluate a possible additional exposure to the volatile lower chlorinated PCBs, e.g. by the stay in a contaminated building (for example schools, universities, company buildings, etc.).

In all cases, an exposure to lower chlorinated PCBs due to the former use of PCB-containing building materials (e.g. caulk or ceiling tiles) was verified by air monitoring in these buildings, which partially exceeded the preventive threshold value of 300 ng/m^3 air (sum of PCBs × 5). This gave reason to conduct human biomonitoring studies for the persons working in or attending these buildings in order to evaluate the internal exposure to these lower chlorinated PCBs. These human biomonitoring studies were offered to all persons concerned by the employer or responsible authorities and planned and conducted by the supervising occupational physician. Altogether the plasma samples were collected at 7 different locations spread over North Rhine-Westfalia and Hesse. As these studies were solely planned for the evaluation of indoor exposure to lower chlorinated PCBs, detailed anamnestic informations (like dietary habits, BMI, number of children, etc.) have not been acquired.

A single plasma sample was collected from each participant by venepuncture and subsequential centrifugation between September 2010 and March 2014 and frozen at −20 °C until analysis. The studies were carried out according to the Helsinki Declaration recommendations, and all participants provided their written informed consent on the donation of blood samples for scientific purposes. In a descriptional questionnaire, the participants were asked about possible previous occupational contact to PCBs. 5 male persons stating possible previous contact to PCBs due to electrical works were excluded from the following statistics. Due to the age-dependent accumulation of organochlorine compounds in fatty tissues we have generated 7 age-groups of the whole collective:

Group 1: children aged between 6 and 10 years (n = 102; 48 f, 54 m; median age: 9 years)
Group 2: adolescents aged between 11 and 17 years (n = 499; 282 f, 217 m; median age: 14 years)
Group 3: young adults aged between 18 and 25 years (n = 157; 100 f, 57 m; median age: 23 years)
Group 4: adults aged between 26 and 35 years (n = 710; 359 f, 351 m; median age: 29 years)
Group 5: adults aged between 36 and 45 years (n = 400; 204 f, 196 m; median age: 41 years)
Group 6: adults aged between 46 and 55 years (n = 525; 297 f, 228 m; median age: 51 years)
Group 7: adults aged between 56 and 65 years (n = 357; 180 f, 177 m; median age: 59 years)

Analysis of organochlorine compounds in plasma

In order to determine the internal exposure to organochlorine compounds, the serum samples were analysed for HCB, p,p′-DDE and 21 different PCB congeners using a slightly modified method approved by the Deutsche Forschungsgemeinschaft (DFG) described previously (Schulte et al., 1993; Schettgen et al., 2011).

Shortly, 2 ml of the plasma sample were deproteinised using formic acid. The organochlorine compounds were then extracted with n-hexane (containing PCB 54 as well as isotopically labelled PCBs as internal standards), cleaned up on a silica gel column and analysed by GC/EI-MS in Selected Ion Monitoring-Mode (SIM). The limit of quantification was determined to be 0.01 μg/L plasma for all analytes investigated. For quality control purposes, bovine serum was spiked with all analytes at a concentration of 0.4 μg/L and included in every analytical series. The between-day imprecision in this period of time (August 2010–April 2014, $n = 356$) has been determined to be in the range of 5.9–7.4% for all analytes.

Additionally, a reagent blank was included in every analytical series. Due to an accurate preparation of reagents and glassware, no organochlorines could be detected in these reagent blanks. Accuracy of our results is guaranteed over the whole study period by biannual successful participation in a round robin for the determination of HCB, DDE as well as the indicator-PCBs in plasma in the environmental concentration range organised in Germany (www.g-equas.de).

Statistical analysis

All examinations were conducted with SPSS.21 (IBM, 2013). Data distribution was investigated by Kolmogorov–Smirnov test and skewness and kurtosis were observed. In addition Levene's test for equality of variances and Kolmogorov–Smirnov test for distribution of residuals of the dependent variables was conducted. Due to their results and to the left censored and right skewed distribution pattern of PCB data nonparametric procedures were elected for further investigations. The distribution of single congeners for the age classes was examined by Kruskal–Wallis-test. Mann–Whitney-*U*-test was conducted to detect differences in the distribution of dependent variables between men and women. Even to detect significant differences in PCB levels between bordering age classes Mann–Whitney-*U*-test was chosen. The limit of statistical significance was set at $p < 0.05$.

The figures were prepared using Microcal™ Origin® Version 6.0.

Results

The results for the levels of HCB, p,p′-DDE as well as for the higher chlorinated PCBs PCB 138, PCB 153 and PCB 180 in the different age groups are given in Table 1. Overall, HCB ($n = 2747$) and DDE ($n = 2748$) were quantifiable in 99.9% of all samples. PCB 180 was quantified in 99.8% ($n = 2745$), whereas PCB 153 and PCB 138 were quantified in every sample. Values below LOQ were assigned half of the LOQ (0.005 μg/L plasma) for statistical purposes.

Overall, we found the expected age-dependent increase in plasma organochlorine levels for all analytes investigated. However, the differences in plasma levels between group 1 (age 6–10 years) and group 2 (age 11–17 years) were no longer significant for HCB and DDE (Mann–Whitney-*U*-test, $p = 0.434$ and $p = 0.982$, respectively). We attribute this to the decreasing environmental exposure in the last 30 years, as probably already the parents of these children and adolescents were born after the ban of both substances.

For all other organochlorine substances, the results indicate a significant increase of the body burden over age. This is illustrated exemplarily for the sum of higher chlorinated PCBs in plasma in Fig. 1 (see also Supplemental material). The plasma levels of the PCBs correlated excellently with each other, while there was no significant correlation between the levels of the other organochlorine compounds as shown in the scatter plots, probably indicating different exposure sources for these organochlorines (see Supplemental material).

Because the different body-fat distribution between man and woman (or different BMI) might have an influence on the body burden to these lipophilic compounds, we have investigated possible differences in the plasma levels of the different gender. The results for this comparison are shown in Table 2 for the complete study group. The age distribution for both groups was determined to be not significantly different (Mann–Whitney-*U*-test, $p = 0.21$).

As a result, the plasma levels of HCB have shown to be significantly higher in women compared to men (Mann–Whitney-*U*-test, $p < 0.001$). On the other hand, the levels of PCB 180 and the sum

Table 1
Plasma levels of organochlorine pollutants in the different age groups.

		6–10 years ($n = 102$)	10–17 years ($n = 499$)	18–25 years ($n = 157$)	26–35 years ($n = 710$)	36–45 years ($n = 400$)	46–55 years ($n = 525$)	56–65 years ($n = 357$)
HCB (μg/L plasma)	Median	0.05	0.06	0.08	0.08	0.11	0.14	0.20
	95th perc.	**0.10**	**0.11**	**0.15**	**0.15**	**0.22**	**0.42**	**0.68**
	Max. value	0.12	0.27	0.19	0.55	0.99	1.22	3.38
p,p′-DDE (μg/L plasma)	Median	0.18	0.18	0.24	0.30	0.45	0.64	0.94
	95th perc.	**1.24**	**0.74**	**0.85**	**1.20**	**1.74**	**3.25**	**4.7**
	Max. value	3.5	9.1	3.1	22.5	10.0	30.4	37.3
PCB 138 (μg/L plasma)	Median	0.05	0.09	0.12	0.15	0.24	0.39	0.56
	95th perc.	**0.22**	**0.23**	**0.25**	**0.33**	**0.53**	**0.93**	**1.26**
	Max. value	0.37	0.40	0.58	0.71	1.08	1.70	3.98
PCB 153 (μg/L plasma)	Median	0.06	0.13	0.17	0.21	0.37	0.63	0.92
	95th perc.	**0.32**	**0.35**	**0.38**	**0.49**	**0.79**	**1.41**	**1.94**
	Max. value	0.57	0.58	0.84	0.89	1.55	2.65	5.45
PCB 180 (μg/L plasma)	Median	0.03	0.07	0.10	0.14	0.29	0.57	0.87
	95th perc.	**0.19**	**0.25**	**0.29**	**0.34**	**0.65**	**1.23**	**1.87**
	Max. value	0.43	0.65	0.40	0.77	1.43	4.59	9.08
$\sum$ PCB 138 + 153 + 180	Median	0.14	0.30	0.38	0.50	0.92	1.58	2.41
	95th perc.	**0.73**	**0.82**	**0.88**	**1.14**	**1.95**	**3.54**	**4.82**
	Max. value	1.38	1.45	1.80	2.37	3.57	8.19	18.5
	$n >$ HBM-I	–	–	1 (0.6%)	2 (0.3%)	32 (8%)	198 (38%)	190 (53%)
	$n >$ HBM-II	–	–	–	–	2 (0.5%)	28 (5.3%)	68 (19%)

Table 2
Levels of POPs in plasma samples of female and male participants; significant differences.

		Females (n = 1470)	Males (n = 1280)	Significance (Mann–Whitney-U)
HCB (μg/L plasma)	Median	0.10	0.09	**p < 0.001**
	95th perc.	**0.40**	**0.26**	
	Max. value	2.46	3.38	
p,p′-DDE (μg/L plasma)	Median	0.37	0.35	p = 0.167
	95th perc.	**2.53**	**1.94**	
	Max. value	30.4	37.3	
PCB 138 (μg/L plasma)	Median	0.19	0.19	p = 0.215
	95th perc.	**0.80**	**0.82**	
	Max. value	3.98	2.54	
PCB 153 (μg/L plasma)	Median	0.29	0.29	p = 0.075
	95th perc.	**1.25**	**1.34**	
	Max. value	5.45	3.27	
PCB 180 (μg/L plasma)	Median	0.20	0.23	**p < 0.001**
	95th perc.	**1.06**	**1.25**	
	Max. value	9.1	4.1	
$\sum$ PCB 138 + 153 + 180	Median	0.68	0.71	**p = 0.021**
	95th perc.	**3.12**	**3.36**	
	Max. value	18.5	9.9	

of PCB 138 + PCB 153 + PCB 180 were significantly higher in men (Mann–Whitney-U, $p < 0.01$ and $p < 0.001$, respectively), while all other levels did not show any gender-difference. The same gender difference for HCB and PCB 180 has already been reported as a result of the previous GerES III (German Human Biomonitoring Commission, 2003). However, the reason for this oppositional difference remains unclear. Although statistically significant in the whole group, the gender-variance in the levels is only marginal and we therefore resigned to present separate statistics in the different age groups.

Discussion

As expected, the data obtained in our study showed a serious decline in the internal body burden to organochlorine compounds in Germany compared to the last reference values published by the Human Biomonitoring Commission (German Human Biomonitoring Commission, 1999, 2003). The 95th percentiles for PCBs determined in the present study are only about 20–40% of these previous plasma reference values (German Human Biomonitoring Commission, 1999), illustrating impressively the reduced body burden to these contaminants and the positive effect of governmental regulation on environmental health. As anticipated, the reduction compared to the previous reference values is more pronounced in the younger age groups (<35 years) born after the ban of these substances.

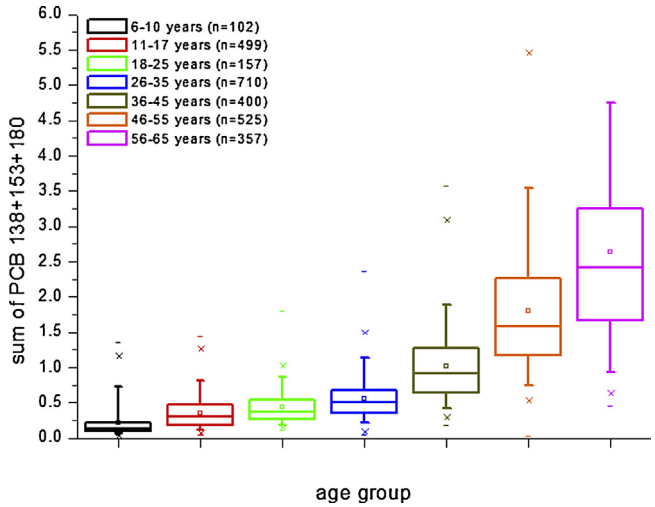

Fig. 1. Boxplots of the plasma levels for $\sum$ PCB 138 + 153 + 180 in the different age groups.

A comparison of the levels found in this study with several recent studies investigating these POPs in plasma or serum samples of the general population is given in Table 3. Compared to our previous study on plasma samples collected in 2003–2004 (Schettgen et al., 2011), the levels of all parameters have considerably declined, especially for HCB. However, it has to be considered that this previous study was suffering from limited sample size in each age group (n = 15). The median levels found in the present study are slightly lower than those of a previous Italian study with a comparatively higher median age for the sampled persons (Turci et al., 2010). In contrast, the median PCB-levels of a most recent German study in rural Bavaria are lower than those found in our present study, although the sampled collective showed a higher median age (Fromme et al., 2015).

While our present dataset also included data for altogether 21 PCB-congeners, we chose to present here only the data for the higher chlorinated indicator PCB 138, PCB 153 and PCB 180 for the following reasons. As our study participants were recruited in studies to evaluate possible additional exposures to lower chlorinated PCBs in public buildings, it would not be justified to use the data obtained for PCB 28, PCB 52 or PCB 101. The same is true for PCB 66, PCB 74, PCB 99 or the dioxin-like PCBs 105 and 118, as previous studies showed that indoor exposure in a public building might result in additional exposure to these congeners (Schettgen et al., 2012b; Meyer et al., 2013). Concerning higher-chlorinated dioxin-like PCB-congeners (e.g. PCB 156, PCB 167, PCB 189), they are usually detected at relatively small rates (25–82% of all samples) and would not give substantial new information without consideration of the other dl-PCBs.

The German Human Biomonitoring Commission has recommended health-related exposure limits, HBM (human biomonitoring) values for PCBs. They represent threshold levels below which no adverse effects on health are expectable (HBM-I) and above which relevant health effects might occur (HBM-II) (Angerer et al., 2011). For PCBs, these values were derived for children and women in childbearing age to protect these high risk groups from possible neurological or immunological effects connected with PCBs. These HBM-values for PCBs were evaluated based on the plasma levels of

Table 3
Overview on the results of several recent studies on the background burden in plasma of the general population to POPs.

Country	Year of samples	Number of samples		Age (years)	HCB (μg/L plasma)	p,p′-DDE (μg/L plasma)	PCB 138 (μg/L plasma)	PCB 153 (μg/L plasma)	PCB 180 (μg/L plasma)	Ref.
Germany	2010–2014	$N = 2750$	Median	33	0.09	0.36	0.19	0.29	0.21	This study
			Range	6–65	<0.01–3.38	<0.01–37.3	0.01–3.98	<0.01–5.45	<0.01–9.08	
Germany	2013	$N = 70$	Median	42	n.a.	n.a.	0.082	0.152	0.153	Fromme et al. (2015)
			Range	4–76	n.a.	n.a.	0.621[b]	1.138[b]	1.079[b]	
Italy (Pavia)	2010	$N = 59$	Median	41.7[a]	0.145	0.27	0.4	0.56	0.47	Turci et al. (2010)
			Range	n.a.	0.1–0.4	<0.05–2.04	<0.05–1.1	<0.05–2.1	<0.05–1.55	
Italy (Novafeltria)	2010	$N = 36$	Median	42.3[a]	0.265	0.685	0.22	0.56	0.30	
			Range	n.a.	0.05–2.77	<0.05–2.25	0.11–1.64	0.05–3.42	0.05–0.95	
Germany	2003–2004	$N = 105$	Median	35	0.27	0.60	0.33	0.46	0.39	Schettgen et al. (2011)
			Range	5–84	0.04–4.1	0.06–14.5	0.04–1.79	0.03–2.51	0.02–2.59	

[a] Mean age.
[b] Max. value.

the sum of (PCB 138 + PCB 153 + PCB 180), multiplied by a factor of 2, and were 3.5 μg/L plasma as HBM-I-value and 7 μg/L plasma as HBM-II-value. This HBM-I-value was reached and sometimes even exceeded in several study participants, but mostly from persons of older age groups (>40 years). 423 participants had values exceeding the HBM-I-value, $n = 13$ of them were women of childbearing age (<45 years). The HBM-II-value for PCBs was exceeded by $n = 98$ participants, of whom two persons were women of childbearing age. More detailed information is given in Table 1. This shows impressively, that PCBs may still pose a possible health risk to the general population and that – albeit strongly declining levels over the last 30 years – organochlorine compounds are still of environmental concern.

Our study has strengths and limitations. The large sample size allows an age and sex group stratified description of results. Since we used nonparametric statistics, tests for group differences will not be influenced by inhomogeneity of variances and outliers. On the other hand, our study is regionally limited to some cities in North Rhine-Westfalia and Hesse. Possible German-wide regional differences or food consumption patterns (e.g. fish consumption) are not reflected by these values. Moreover, a collection of detailed anamnestic data of our participants was not part of the study protocol, so that important informations for data evaluation are lacking (BMI, consumption of fish or fatty food, number of children, lactation habits, etc.). With respect to the plasma levels of DDE, it would also have been advantageous to know the origin of the participants (Western/Eastern Germany) or possible migration background.

This study describes the most current available dataset on plasma organochlorine levels in Germany at present. As long as new representative data are not generated within a novel nationwide Environmental Survey, we would propose to currently use these data for the evaluation of possible environmental or (former) occupational exposures to these contaminants.

Conclusion

In conclusion, we have conducted the most current and largest study on human biomonitoring of persistent organic pollutants in Germany so far. The results of our study revealed a strong decline in the plasma levels of persistent organochlorine compounds in Germany investigated in 7 age groups from 6 to 65 years of age. Nevertheless, the high number of persons exceeding the HBM-I and HBM-II-values for PCBs clearly shows that PCBs might still pose an environmental health risk.

Although the collective was regionally restricted and therefore not fully representative for the general population in Germany, we propose the data from our study to be used as reference values for the evaluation of potential exposures to HCB, p,p′-DDT or higher chlorinated PCBs.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ijheh.2015.02.006.

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