HBM4EU E-waste study : assessing persistent organic pollutants in blood, silicone wristbands, and settled dust among E-waste recycling workers in Europe

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Abstract

E-waste recycling is an increasingly important activity that contributes to reducing the burden of end-of-life electronic and electrical apparatus and allows for the EU´s transition to a circular economy. This study investigated the exposure levels of selected persistent organic pollutants (POPs) in workers from e-waste recycling facilities across Europe.

The concentrations of seven polychlorinated biphenyls (PCBs) and eight polybrominated diphenyl ethers (PBDEs) congeners were measured by GC-MS. Workers were categorized into five groups based on the type of e-waste handled and two control groups. Generalized linear models were used to assess the determinants of exposure levels among workers. POPs levels were also assessed in dust and silicone wristbands (SWB) and compared with serum.

Four PCB congeners (CB 118, 138, 153, and 180) were frequently detected in serum regardless of worker’s category. With the exception of CB 118, all tested PCBs were significantly higher in workers compared to the control group. Controls working in the same company as occupationally exposed (Within control group), also displayed higher levels of serum CB 180 than non-industrial controls with no known exposures to these chemicals (Outwith controls) ($p < 0.05$). BDE 209 was the most prevalent POP in settled dust (16 µg/g) and SWB (220 ng/WB). Spearman correlation revealed moderate to strong positive correlations between SWB and dust. Increased age and the number of years smoked cigarettes were key determinants for workers exposure. Estimated daily intake through dust ingestion revealed that ΣPCB was higher for both the 50th (0.03 ng/kg bw/day) and 95th (0.09 ng/kg bw/day) percentile exposure scenarios compared to values reported for the general population.

This study is one of the first to address the occupational exposure to PCBs and PBDEs in Europe among e-waste workers through biomonitoring combined with analysis of settled dust and SWB. Our findings suggest that e-waste workers may face elevated PCB exposure and that appropriate exposure assessments are needed to establish effective mitigation strategies.

Keywords: e-waste; occupational exposure; biomonitoring; PCBs; PBDEs; Europe

1 Introduction

Electronic waste, or e-waste, refers to discarded electrical or electronic devices that are no longer usable (Cucchiella et al., 2015; Robinson, 2009). E-waste can contain a wide range of materials, including inorganic substances such as toxic metals and metalloids (e.g., lead, mercury, cadmium, zinc and organic materials (e.g., plastics), and organic compounds (e.g., polybrominated diphenyl ethers (PBDEs) and organophosphate flame retardants (PFRs), dioxins/furans (PCDD/Fs), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and phthalates (Akcil et al., 2015; Han et al., 2010; Kaya, 2016; Leung et al., 2008; Li et al., 2014; Liu et al., 2009; Luo et al., 2011; Wong et al., 2007; Wu et al., 2019). The increasing production and disposal of e-waste has become a major environmental concern, as improper recycling or disposal of e-waste can lead to the release of hazardous substances into the environment and pose a threat to human health (Awasthi et al., 2016; Rautela et al., 2021). The European Union (EU) has taken steps to address the issue of e-waste by implementing several regulations and initiatives aimed at promoting the recycling and disposal of e-waste. In 2002, the EU adopted the Waste Electrical and Electronic Equipment (WEEE) Directive, which requires EU member states to establish systems for the collection, treatment, and recovery of e-waste (Cucchiella et al., 2015; Shittu et al., 2021). The directive also sets targets for e-waste that must be collected and recycled each year.

In addition to the WEEE Directive, the EU has also implemented the Restriction of Hazardous Substances (RoHS) Directive, which limits the use of certain hazardous substances in electrical and electronic equipment ("EUR-Lex - 32002L0095 - EN," n.d.; Honda, 2012). The RoHS Directive aims to reduce the environmental impact of e-waste by reducing the number of hazardous substances used in electronic device production. These initiatives are in line with the EU's broader Circular Economy Action Plan, which seeks to promote sustainable practices throughout the product life cycle (*A new Circular Economy Action Plan*, 2020). By encouraging the design of electronic devices with fewer hazardous components and fostering a more circular approach to resource use, the Circular Economy Action Plan further supports the EU's efforts to tackle e-waste and foster a greener, more sustainable economy.

While the regulation of e-waste components is gaining momentum in the EU, concerns persist regarding the potential risks faced by workers in these facilities due to exposure to hazardous chemicals. This study specifically focuses on persistent organic pollutants (POPs), including PCBs and PBDEs, which were previously used as flame retardants in electronics (Abafe and Martincigh, 2015a; Die et al., 2019; Tue et al., 2013; Wang et al., 2016; Zhang et al., 2019). These substances are now restricted under the Stockholm Convention but can still be present in some of the e-waste components (Lallas, 2001). It is important to highlight that a significant knowledge gap exists in the literature concerning the exposure of e-waste workers to PCBs and PBDEs in the EU. As of now, there are very few existing studies that specifically address this aspect.

PCBs and PBDEs are exceptionally stable chemicals known for their remarkable persistence in the environment. They possess lipophilic properties, resist metabolic breakdown, and tend to bioaccumulate in organisms, particularly in fatty tissues. The adverse effects of PCBs and PBDEs are well-documented, and they have been linked to a range of negative health effects, including impacts on the endocrine and reproductive systems, as well as the development of cancer (Alharbi et al., 2018; Awasthi et al., 2016; Parvez et al., 2021; Qing Li et al., 2006; Shi et al., 2019). Occupational exposure to PCB and PBDE technical mixtures, specifically Aroclor 1254 and Penta-BDE, has been linked to disruption in the thyroid function and endocrine-disrupting effects on sex hormones (Eguchi et al., 2015, 2014; Zheng et al., 2017). The diverse composition of electronic components exposes workers dismantling electronics to a range of potentially harmful substances, including metals. Research by Li et al. demonstrated a negative correlation between blood cerium levels and global DNA methylation in populations living near e-waste recycling facilities (Li et al., 2020). Alabi et al. further highlighted the mutagenic and genotoxic potential of pollutants, particularly polycyclic aromatic hydrocarbons (PAHs) and metals, generated during e-waste processing (Alabi et al., 2012). These findings highlight the potential risks of occupational exposure to these POPs and emphasize the need to characterize exposure to identify effective measures to minimize exposure and protect Worker´s health in the e-waste recycling industry. While declining concentrations of legacy pollutants, such as PCBs, have been observed in marine and freshwater biota due to efforts following the Stockholm Convention, the ongoing emissions and exposure to POPs remain significant, particularly from e-waste recycling and historical applications where these chemicals are still in use, such as PBDEs in old furniture or electronics and PCBs in open applications (Hung et al., 2016; Li et al., 2023; Rigét et al., 2019; White et al., 2021).

The objective of this study was to evaluate the exposure levels of workers to POPs in various e-waste recycling facilities across Europe. Therefore, a comparative assessment was conducted to assess the exposure levels of workers involved in the processing of various types of e-waste categories. Considering the potential health risks linked to POPs and the extended exposure of workers engaged in work-related tasks within these environments, it is crucial to determine the chemical levels in e-waste recycling facility workers and compare them with baseline levels in the general population.

2 Materials and methods

2.1 Study population

The target population for this study was previously described by Scheepers et al. (Scheepers et al., 2021). Briefly, using cross-sectional design, e-waste workers and control population were recruited from fourteen companies in six countries located in the European region including Belgium, Finland, Latvia, Luxembourg, The Netherlands and Portugal (Table 1). For some participating countries, e.g., the UK and Poland, blood samples were either not collected or unavailable for PCB and PBDE analysis. The recruited companies were engaged in various e-waste processing activities, including sorting, dismantling, shredding and pre-processing of metal and non-metal components. Subject eligibility was determined using the inclusion criteria, which stipulated that an e-waste worker was qualified to participate in the study if they were involved in 1) sorting e-waste from household or industrial sources either manually or in a semi-automatic way, 2) dismantling electronic components, 3) shredding and pre-processing, 4) recycling of electronic components to recover precious metals or obtain granulated plastic for further re-use or resale (Supplementary Information Figure SI-1. The study population included both males and females aged between 18 and 64 years (Table 1).

The control population comprised of individuals residing in the same geographical area as the e-waste workers but were not involved in e-waste processing or in activities with known occupational exposure. Moreover, the control group was subdivided into two categories, based on the occupation of the subjects: those who worked in the same industry as the individuals that were occupationally exposed to e-waste processing (referred to as Within controls), and those who worked in industries not associated with e-waste processing, such as healthcare, technology, research and development, and agriculture, among others (referred to as Outwith controls). The selection of Outwith controls was limited to only three countries: Latvia, Luxembourg, and Portugal.

Likewise, workers were also stratified into five subcategories (‘White goods’, ‘Brown goods’, ‘Batteries’, ‘Metals and Plastics’, and ‘Miscellaneous’) based on the activities performed as part of their duties (Table 1).

Additional information about the participants were obtained from three standardized questionnaires collected during sampling, covering risk management measures (RMMs), specific work tasks, relevant co-exposures from other sources outside of work, and availability and use of respiratory protective equipment (RPE).

Table 1. Characteristics of the study population: Mean and (standard deviation) values. The ‘N’ column displays the number of individuals sampled, including females (F) and males (M) in parentheses.

Main category	**Subcategory**	**Subcategory description**	**N (female/male)**	**Age (Y, SD)**	**BMI (kg/m², SD)**	**Shift duration (h, SD)**	**Length of employment (Y, SD)**
Control	Outwith controls	Non-industrial controls: Geographically matched individuals without known exposures to the studied chemicals.	30 (10/20)	39.4 (9.4)	26.0 (3.5)	7.9 (1.0)	2.1 (3.9)
Control	Within controls	Industrial controls: Non-e-waste processing personnel at the same facilities, without expected occupational exposure to the studied chemicals.	33 (15/18)	41.5 (10.8)	26.3 (5.4)	8.0 (0.2)	7.2 (7.6)
Worker	Batteries	Individuals involved in lead battery recycling	26 (2/24)	48.8 (8.4)	28.4 (3.8)	8.0 (0.0)	7.0 (7.9)
Worker	Brown goods	Individuals involved in processing electrical appliances such as televisions, radios, computers, DVD players, lights, etc.	18 (2/16)	38.6 (11.1)	26.3 (4.2)	8.1 (0.2)	8.4 (8.1)
Worker	Metals and plastics	Individuals involved in recycling metals and plastics	21 (2/19)	43.2 (10.4)	27.5 (5.3)	8.4 (1.2)	7.7 (8.3)
Worker	White goods	Individuals involved in processing domestic appliances such as heaters, washing machines, refrigerators, and dryers	35 (2/33)	43.0 (9.2)	26.9 (5.1)	8.0 (0.1)	6.5 (5.9)
Worker	Miscellaneous	Individuals involved in recycling miscellaneous e-waste products	6 (1/5)	42.7 (12.3)	28.2 (4.4)	7.8 (0.9)	2.3 (2.5)

2.2 Sample collection

Collection of serum samples of workers was performed in 2021 as part of the initial HBM4EU E-waste study (Scheepers et al., 2021). Peripheral blood collection of exposed and non-exposed volunteers was performed following signed informed consent according to WHO guidelines (World Health Organization, 2010). For the exposure assessment of POPs, whole blood samples were collected in trace elements blood containers (n = 169). Following centrifugation for 10 min at 2000 g, serum of at least 2 mL was transferred to polypropylene tubes that were washed prior with hexane and rinsed with purified water. Samples were immediately cooled to +4°C at the sampling location and were then shipped to the University of Antwerp, Belgium, for POP analysis.

Settled dust samples were collected from e-waste processing plants according to the protocol described by Scheepers et al., designed to evaluate exposure to metals in home environments (Loh et al., 2016; Scheepers et al., 2021). Briefly, country and e-waste plant matched settled dust samples (n = 52) were collected towards the end of a given work shift using a University Products Museum Vac

Vacuum with dial suction control (Adams, MA, USA). HEPA filter bags were pre-weighed, and at least 4 g of dust was collected from marked 1 m² areas on bare floors. If the quantity was insufficient, additional areas were included. Samples were stored at room temperature until transport to the Laboratoire National de Santé (LNS), Luxembourg.

Silicone wristband (SWB) samples, adult-size (202 mm L × 12 mm W × 2 mm T, weight 5.33 g, SD 0.10 g), were collected from all participating countries except for Finland. Briefly, silicone wristbands were first subjected to cleaning with 125 mL 1:1 mixture of ethyl acetate and hexane (v/v) for 30 min, followed by a second cleaning with 125 mL 1:1 mixture of ethyl acetate and methanol (v/v) for 30 min in an overhead shaker. The cleaned wristbands were then dried using nitrogen at 40°C and stored individually in aluminum/LDPE zip lock bags at -20°C. During the workweek, participants wore the precleaned wristbands exclusively during their working activities on the wrist of their dominant hand, starting from the first morning of the workweek. At the end of each workday, the wristbands were placed in their respective zip lock bags and stored overnight in a clean area. The collected wristband samples were kept cool or at room temperature (Aerts et al., 2018; Wang et al., 2019). For longer storage periods, the samples were kept at -20°C prior to transportation to the LNS. To establish background levels, one blank SWB for every ten SWB worn at the site was analyzed. These blank SWB were stored in their respective zip lock bags for the entire workweek.

2.3 Sample preparation

Extraction of PCBs and PBDEs from serum was accomplished following a protocol previously described by Dirtu et al (Dirtu et al., 2013). Briefly, 1 mL of serum was transferred to clean polypropylene conical tubes containing 50 µL of internal standard mix in iso-octane (^{13}C-HCB at 50 pg/µL from LGC Standards, the Netherlands, PCB-143 at 250 pg/µL from LGC Standards, BDE-103 and BDE-128 at 100 pg/µL from AccuStandard, the Netherlands, and ^{13}C-BDE-209 at 125 pg/µL from AccuStandard). Following addition of 500 µL of MilliQ water (PURELAB Flex system from Elga Veolia, Tienen, Belgium), 250 µL of formic acid (Merck, Darmstadt, Germany) and 5 mL of n-hexane/DCM/toluene mixture (4:4:0.5, v/v), samples were vortexed for one minute and sonicated for 20 min. Next, samples were centrifuged at 1600 g for 5 min and the upper layers were transferred to clean borosilicate glass tubes. The aqueous layers were re-extracted two more times with 4 mL of n-hexane/DCM/toluene mixture (4:4:0.5, v/v). The organic layers were combined and were evaporated to near dryness under gentle nitrogen stream and reconstituted in 500 µL hexane followed by vortexing for 1 min. Further sample clean-up was performed on silica solid phase extraction (SPE) cartridges (3 mL, 500 mg, Agilent Bond Elute SI) topped with 200 mg sulfuric acid impregnated silica (44%, w/w) and 75 mg sodium sulfate. Prior to loading, the silica SPE cartridges were first washed with 4 mL hexane. After loading, POPs were eluted with 6 mL hexane. Eluates were evaporated under gentle nitrogen stream to near dryness, then reconstituted in 50 µL iso-octane and 50 µL recovery standard containing PCB-207 at 50 pg/µL (LGC Standards) in iso-octane and transferred to autosampler vials containing inserts. For reliable quantification, standard calibration curves and quality controls were prepared and injected with each batch of samples.

Dust samples were weighed and sieved with a Retsch AS 200 digit vibratory sieve shaker to analyze the fine fraction (< 63 µm) only. Among these, 9 samples had insufficient particles smaller than 63 µm for analysis, leading to the selection of the fraction smaller than 2 mm. Following sieving, 100 mg of the fine fraction dust sample was weighed in an accelerated solvent extraction (ASE) cell filled with diatomaceous earth (Thermo Fisher, Merelbeke, Belgium) and subjected to accelerated solvent extraction with n-hexane/acetone (90:10, v/v). The extracted samples were concentrated at 40°C under a N2 flow. Subsequently, the extract volume was adjusted to 1 mL by transferring it to a 1-mL graduated amber flask, using ethyl acetate. An aliquot of 50 µL, spiked with 2.5 ng of $^{13}C_{12}$-BDE 99, 25 ng of $^{13}C_{12}$-BDE 209, and 5 ng of $^{13}C_{12}$-CB 28 (LGC), was transferred to a 2-mL amber glass vial with a glass insert for injection into the GC-MS system. For reliable quantification, each batch of samples included standard calibration curves and quality controls, incorporating a certified reference material solution (Sigma-Aldrich, Overijse, Belgium) and NIST SRM house dust (LGC). When necessary, dust extracts were appropriately diluted to ensure they fell within the linear range.

The SWBs were extracted following the method described by Kile et al (Kile et al., 2016) with some modifications. Subsequently, the resulting extracts underwent a clean-up process based on the protocol described by (Butt et al., 2016). In brief, SWB samples were pre-rinsed with water and isopropanol, cut into five pieces, and placed in an amber 60 mL glass vial. After addition of internal standard, extraction was performed twice with 25 mL ethyl acetate. The resulting extracts were combined, and 50 µL of n-dodecane was added before evaporation under a nitrogen stream. Ethyl acetate (450 µL) was added to obtain 500 µL of the final extract. To reduce interferences, the obtained extract was submitted to two successive solid-phase extractions. The first clean-up was achieved on Oasis HLB cartridges conditioned with 5 mL of dichloromethane, 5 mL of methanol and 5 mL of water successively. After loading of a 50 µL aliquot of final extract, the cartridges were washed with 5 mL of water and elution of the compounds was achieved with 10 mL ethyl acetate: dichloromethane (50:50, v/v). After evaporation to dryness at 40°C under nitrogen stream, the eluate was resuspended in 1 mL of n-hexane submitted to Sep-Pak Silica SPE. The second pass-through clean-up involved loading the sample extract onto n-hexane-conditioned cartridges and collecting the fraction that resulted from the sample loading and elution of PCBs and PBDEs using 10 mL of n-hexane. After addition of 50 µL of n-dodecane, the extract was evaporated to reduce the volume to approximately 50 µL and then transferred to a 2-mL amber glass vial with glass insert, before analysis with GC-ECNI-MS.

To reliably determine levels of PCBs and PBDEs, matrix-matched calibration curves from 1 to 1000 ng/WB (10-10000 ng/WB for BDE209) and quality controls (certified reference material solution spiked on blank SWB) were prepared and injected for each batch of samples. Control SWB, spiked with a known mixture of PCBs/PBDEs at a concentration of 25 ng/WB, were also analyzed every 20 samples. Measured recoveries fell within the range of 80-120%. If required, SWB extracts were appropriately diluted to ensure they fell within the linear range.

2.4 Instrumental analysis

Target analysis of PCBs in serum was performed using an Agilent 6890 Gas Chromatograph (GC) (Palo Alto, CA, USA) interfaced to an Agilent 5973 Series Mass Selective Detector (MSD) operated in electron capture negative ionization (ECNI) mode with methane as reagent gas and helium as carrier gas (Ali et al., 2012). PBDE analysis in serum was performed using an Agilent 7890A Series GC coupled to an Agilent 7000 Series Triple Quadrupole MS operating in ECNI mode. Analysis of PCBs and PBDEs from settled dust and silicone wristbands was performed using an Agilent 6890N gas chromatograph equipped with a DB-XLB column (15 m, 0.25 mm I.D., 0.10 µm film thickness) and an ALS 7683 autosampler, coupled to an Agilent 5975 inertXL MSD operating in SIM mode with electronic impact (EI) and methane-induced negative chemical ionization (NCI). The detailed methodology is provided in the Supplementary Information (Tables SI-1, SI-2 and SI-3).

The implementation of Internal Quality Assurance (QA) and Quality Control (QC) for serum analysis in our study involved the routine analysis of procedural blanks with a minimum of two blanks for every batch of 24 samples and matrix-matched spiked reference samples with at least one reference sample per batch (Table SI-4). Solvent blanks and spiked samples were consistently injected throughout the instrumental run. The analysis of procedural blank concentrations was conducted to detect any potential background contamination originating from the laboratory environment, and these blank values were then subtracted from the analyte concentrations in the samples. The Limits of Quantification (LOQs) were determined as 3xSD of the blank measurements. Our participation in inter-laboratory comparison exercises, such as the Arctic Monitoring and Assessment Program for Persistent Organic Pollutants in Human Serum (AMAP) served to ensure the external quality control of our study.

2.5 Calculation of estimated daily intake (EDI)

The EDIs of PCBs and PBDEs from e-waste workers were calculated using the average levels of pollutants measured in settled dust from their respective e-waste processing facilities, considering multiple measurements if taken from the same plant.

The EDI through dust ingestion was calculated based on established methodologies from previous publications (Ait Bamai et al., 2016; Christia et al., 2021). This resulted in a unique EDI value for each e-waste worker expressed in ng/kg bw/day.

$$EDI_{Ingestion} = (C_{dust} \times IngR \times FR \times Ba) / BW,$$

where *Cdust* represents the concentrations of quantified compounds in ng/g. *IngR* corresponds to dust ingestion rates of 20 and 50 mg/day for adults in the 50th and 95th percentile exposure scenarios, respectively (USEPA, 2017a). *FR* denotes the fraction of time spent at the workplace, assuming an 8-h workday, expressed as 8/24 (Klepeis et al., 2001). *BW* represents the body weight of the e-waste workers in kg, obtained from the questionnaire. Ba (theoretical bioaccessibility) values of PCB and PBDE congeners were estimated using their respective LogKow values obtained from the CompTox Chemicals Dashboard (Williams et al., 2017). These theoretical bioaccessibilities were calculated according to the method described by Christia et al (Christia et al., 2021).

Dermal exposure was assessed using the following equation:

$$EDI_{Dermal} = (C_{dust} \times BSA \times SAS \times AF \times IEF) / BW \times 1000,$$

where *Cdust* represents the concentration of pollutants in ng/g dry wt, BSA stands for the body surface area in cm^2/day, which was set to 2430 cm^2/day (Hammel et al., 2023), SAS is the soil adhered to skin in mg/cm^2, which was 0.01 mg/cm^2 (Hammel et al., 2023), AF was the fraction of analyte absorbed in the skin which was 0.03 for PBDEs (Johnson-Restrepo and Kannan, 2009) and 0.14 for PCBs (Mayes et al., 2002), IEF was the indoor exposure fraction (hours spent over a day in an indoor environment), which was 8/24 and finally BW is the body weight of the participant expressed in kg.

2.6 Data processing and statistical analysis

Analytes with concentrations below the LOQ were substituted with LOQ/2 (medium bound approach). The normality of concentration distributions in serum, SWB, and settled dust samples was assessed through various methods, including visual observation of histograms, numerical evaluation of skewness and kurtosis values, and Shapiro-Wilk tests. The results revealed positively skewed distributions for all matrices. Despite attempting log transformation, the Shapiro–Wilk test indicated a departure from normal distribution. Consequently, non-parametric tests were employed for analysis. Compounds with DF > 50% were used for statistical analysis.

The differences between worker and control group in main analyte categories were evaluated using the Mann-Whitney U test, while subcategory differences were assessed through Dunn's test for multiple pairwise comparisons. Statistical p-values from these comparisons were controlled using Benjamini-Hochberg correction. Spearman's correlation test was used to determine the strength and direction of the association between the levels of POPs measured in SWB, serum, and settled dust among the workers. Generalized Linear Models (GLMs) were employed to examine the impact of various factors, including RMMs, specific work tasks, co-exposures to sources outside of work, and the availability and use of RPE on the exposure levels to CB 138, 153, and 180. Prior to model fitting, the outcome variables were square root transformed to reduce skewness and kurtosis.

Independent variables were selected from the basic questionnaires based on the results from univariate analyses such as Spearman correlations, variance inflation factors, and previous literature findings (Coakley et al., 2018; Schecter et al., 2018; Singh et al., 2019; Wang et al., 2012; Wannomai et al., 2021; Yu et al., 2020) while considering multicollinearity. A detailed description of the selection criteria employed is given in the Supplementary Information. Potential outliers were investigated based on the Cook's distance, where samples with Cook's Di distance over 0.5 deemed to be highly influential and considered for removal from the dataset used for the GLM.

Data processing and plot generation were conducted using the Python programming language. Significance level of $p < 0.05$ was considered statistically significant for all statistical analyses.

3 Results and discussion

3.1 Basic description of serum PCB and PBDE concentrations

The prevalence of exposure to POPs among the study participants was evaluated by examining the detection frequencies (DFs, %) in serum, along with concentrations (Table 2). Among the 16 measured POPs, four analytes were quantifiable in over 50 % of all samples, regardless of main category specific stratification, namely CB 118 (65% and 58%), CB 138 (89% and 91%), CB 153 (89% and 91%), CB 180 (89% and 91%) (Table 2).

CB 153 exhibited the highest median concentration among POPs with levels of 86 and 126 ng/L in controls and workers, respectively. This was followed by CB 180 (54 and 95 ng/L), CB 138 (43 and 57 ng/L), and CB 118 (12 and 13 ng/L). (Table 2). The absence of detectable levels of lower chlorinated congeners containing four or fewer chlorine substituents, such as CB 52, could be attributed to high LOQs (Table SI-1).

These PCB congeners exhibited consistently high detection frequencies in numerous studies conducted across Europe indicating their ubiquitous nature. In a study from Catalonia, Junqué et al. reported detection frequencies of 67% for CB 153 and 73% for CB 180 in serum samples from pregnant mothers during the first trimester (Junqué et al., 2020). Additionally, Haug et al. reported these congeners to be the major contaminants in the adult population of the HELIX project, which recruited participants from six birth cohorts across Europe (Haug et al., 2018). CB 28 and 52 were not detectable in any of the analyzed samples. Despite exhibiting higher concentrations in certain samples, none of the measured PBDE congeners were detectable in at least 50% of the samples. BDE 209, for instance, had a detection frequency of 9% among workers and 11% among controls (Table 2).

Table 2. Serum detection frequencies (DF) and concentrations of PCBs and PBDEs in E-waste workers and controls (ng/L).

	Controls (n=63)				Workers (n=106)			
	DF (%)	**p25**	**p50**	**p75**	**DF (%)**	**p25**	**p50**	**p75**
CB 28	0	< LOQ	< LOQ	< LOQ	0	< LOQ	< LOQ	< LOQ
CB 52	0	< LOQ	< LOQ	< LOQ	0	< LOQ	< LOQ	< LOQ
CB 101	21	< LOQ	< LOQ	< LOQ	34	< LOQ	< LOQ	14
CB 118	65	< LOQ	12	19	58	< LOQ	13	29
CB 138	89	29	43	83	91	40	57	121
CB 153	89	56	86	167	91	79	126	269
CB 180	89	27	54	107	91	51	95	175
BDE 28	21	< LOQ	< LOQ	< LOQ	14	< LOQ	< LOQ	< LOQ
BDE 47	13	< LOQ	< LOQ	< LOQ	13	< LOQ	< LOQ	< LOQ
BDE 99	30	< LOQ	< LOQ	3	26	< LOQ	< LOQ	2
BDE 100	32	< LOQ	< LOQ	5	26	< LOQ	< LOQ	3
BDE 153	35	< LOQ	< LOQ	4	37	< LOQ	< LOQ	5
BDE 154	24	< LOQ	< LOQ	2	37	< LOQ	< LOQ	3
BDE 183	0	< LOQ	< LOQ	< LOQ	0	< LOQ	< LOQ	< LOQ
BDE 209	11	< LOQ	< LOQ	< LOQ	9	< LOQ	< LOQ	< LOQ

3.2 Associations between serum PCB levels and worker categories

Analysis of main category and subcategory-specific differences revealed significantly higher serum concentrations of all PCBs with DF > 50% except for CB 118 in the worker population compared to the control group ($p < 0.01$) (Figure 1A). A similar pattern was observed in a study by Yang et al., who compared POP exposure profiles in control-matched electronic waste recycling sites in Northern China (Yang et al., 2013). Their findings suggest similar exposures to CB 118 among both local residents and e-waste dismantling workers, regardless of occupation (Yang et al., 2013). The observed differences in CB 138, 153 and 180 concentrations could be attributed to country-specific factors due to specific regulations in place or used production processes. However, upon further evaluation, the influence of country as a confounding factor is observed only for CB 180, as indicated by the Mann-Whitney U test (data not shown).

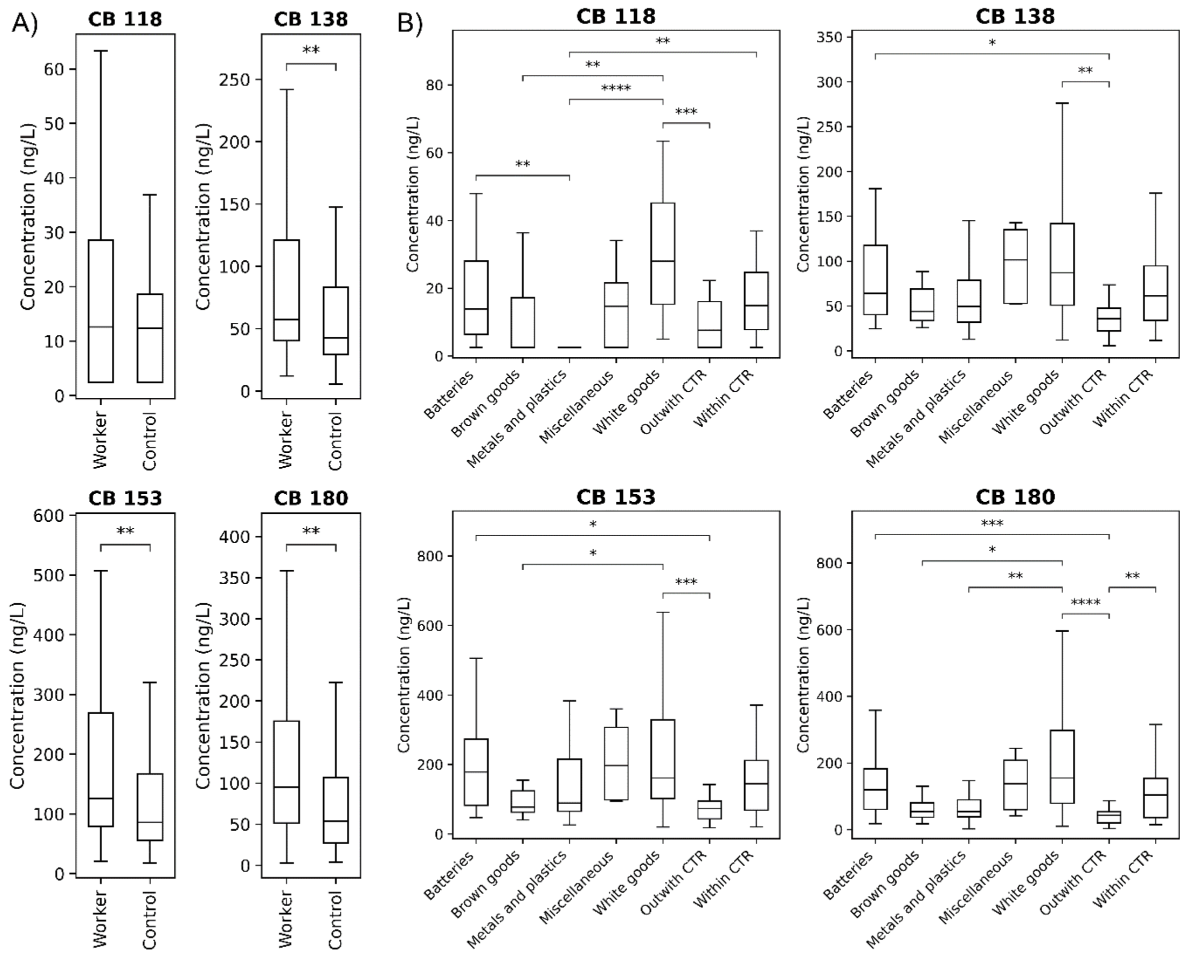

*Figure 1. Main and subgroup specific distribution of POPs with DF over 50%: A) Main group specific distribution of POPs B) Subgroup specific distribution of POPs. The plots depict a 90% interval, spanning from the 5th to the 95th percentiles, with the horizontal line symbolizing the 50th percentile. Subcategory differences were assessed via multiple pairwise comparisons using Dunn's test, followed by Benjamini-Hochberg correction. P-value annotation legend: *: p < 0.05, **: p < 0.01. Tabulated version can be found in Table SI-5 and SI-6.*

Subgroup specific stratification revealed that individuals in the White goods category had significantly higher concentrations of serum PCBs compared to the Outwith controls for all four congeners. On the other hand, Battery workers generally exhibited elevated levels of serum PCBs compared to the Outwith controls, except for CB 118 (Figure 1B). Although these findings imply increased PCB exposure among these subpopulations, the composition of the Outwith control group could have impacted the observed differences. Notably, the recruitment of Outwith controls was limited to Latvia, Luxembourg, and Portugal. This restricted geographical representation could have introduced sampling bias, potentially affecting the findings.

Interestingly, CB 180 also showed differences within the control groups. The Within control group exhibited slightly higher levels of CB 180 compared to the Outwith controls, although to a lesser extent than the worker population. This finding indicates that the Within control group may experience additional exposure either due to their proximity to the industrial working environment or variations in their geographical representation. This mirrors a similar finding from the HBM4EU chromates study, where a distinction between control groups was also seen, involving 'bystander' exposure (Santonen et al., 2022). The HBM4EU chromates study found that apart from workers directly exposed to Cr(VI) in plating, welding, or other surface treatment activities, some office workers recruited as controls from the same companies might have experienced indirect exposure to Cr(VI), as inferred from higher urinary Cr(VI) levels observed in the Within control group. Overall, these findings indicate that further considerations are required to study the potential exposure of bystanders, and it should be integrated into the health surveillance programs of e-waste companies.

3.3 GLM to assess the effects of potential exposure sources and effectiveness of mitigation

To evaluate potential connections between RMMs, specific work tasks, relevant co-exposures from other sources outside of work, availability and use of RPEs and serum PCB levels, GLMs were employed. The outcome variables included contaminants where subcategory stratified DF exceeded 50%. The selection criteria regarding the variables and evaluation of model performances are described in the Supplementary Information (Figure SI-2, 3 and 4).

The findings from the GLMs revealed a notable positive association between age and serum PCB concentration. Age was treated as a dichotomous variable, with workers younger than 40 years serving as the reference. The corresponding estimated coefficients (β) and confidence intervals (CI) were 0.32 (95% CI [0.19, 0.46], $p < 0.01$), 0.35 (95% CI [0.23, 0.48], $p < 0.01$) and 0.42 (95% CI [0.27, 0.58], $p < 0.01$) for CB 138, 153 and 180, respectively (Figure 2). This indicates that workers older than 40 years exhibited 1.37, 1.41, and 1.52 ng/L higher concentrations of CB 138, CB 153, and CB 180 compared to workers younger than 40 years.

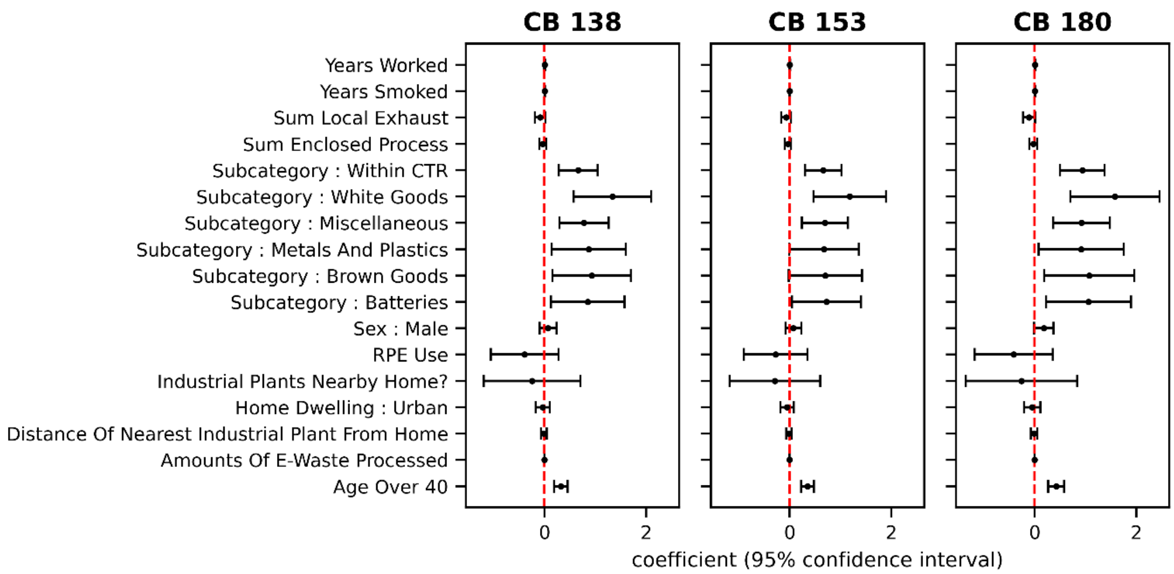

Figure 2. Comparative forest plots of the coefficients of the three GLM models: Model coefficients for explanatory variables with A) CB 138, B) CB 153, and C) CB 180 as endogenous variable. The points represent the coefficients of corresponding predictors and the error bars correspond to the 95% confidence interval. Exogenous variables such as subcategory ("Outwith Control"' as reference category), sex ("female" as reference category), industrial plants nearby home ("no" as reference category), home dwelling ("rural" as reference category), age (category denoted to lower than 40 years, as reference), use of RPE ("not used" as reference category) and e-waste processing involving work in enclosed environments ("no" as reference category) were dummy-encoded. Detailed information regarding the results of the GLM can be found in Table SI-7.

The importance of age as a determinant of PCB exposure, such as CB 138, 153 and 180 has been reported by numerous studies (Coakley et al., 2018; Singh et al., 2019; Yu et al., 2020). Additionally, the number of years smoked cigarette was also positively correlated for CB 153 and CB 180 (CB 153: $\beta = 0.01$, 95% CI [0, 0.01], $p < 0.05$; CB 180: $\beta = 0.01$, 95% CI [0, 0.01], $p < 0.05$)). This implies an annual increase of 1 ng/L of serum PCB levels among cigarette smokers, assuming other factors remain unchanged. While several studies have investigated the association between smoking and POP levels in humans, the findings have been inconsistent, warranting further research to elucidate a clearer relationship (Fierens et al., 2005; Jönsson et al., 2005; Moon et al., 2017).

Our findings suggested that participation in the e-waste processing was a strong determinant for PCBs. Subcategory stratification also emerged as an important factor for PCB levels, with all subcategories, except for Brown goods and Metals and plastics for CB 153, exhibiting higher coefficients compared to the Outwith controls as a reference. For example, the odds ratio of e-waste workers involved in processing Miscellaneous e-waste having higher serum PCB concentrations than Outwith controls ranged between 4.7 ($\beta = 0.78$, 95% CI [0.29, 1.26], $p<0.01$) and 6.3 ($\beta = 0.92$, 95% CI [0.37, 1.48], $p<0.01$) for PCB 138 and PCB 180, respectively. This increased for individuals belonging to White goods, from 14.6 ($\beta = 1.34$, 95% CI [0.58, 2.10], $p<0.01$) to 23.6 ($\beta = 1.58$, 95% CI [0.70, 2.46], $p<0.01$]). Interaction effects of age with main and subcategories were non-significant (data not shown). This underpins the importance of implementing suitable risk management measures and comprehensive workplace regulations to effectively mitigate exposure to PCBs in e-waste recycling facilities.

Factors pertaining to RMMs, such as RPE use and number of exhaust units available during e-waste processing, as well as other factors, such as sex, type of home location, and distance of nearest industrial plants to home did not exhibit statistically significant associations with PCB exposure. However, RPE use and availability of exhaust units showed decreasing trends of exposure levels of all three CBs with having partly borderline significance ($p<0.2$). The assessment of RPE use involved self-reported questionnaires that asked workers to indicate whether they used RPE during work activities. Sum of local exhaust units was assessed by e-waste facilities. Thus, taking into consideration the underestimation of the use of RPE and local exhaust units, our finding suggests that the use of RPE and local exhaust ventilation is one of the most effective means of reducing exposure to PCBs.

Although our study has identified several key factors that contribute to PCB exposure, there could be other sources of exposure that were not accounted for in our analysis. Dietary habits, for example, were found to be one of the major contributors to elevated POP concentrations (Aerts et al., 2019; Chen et al., 2018; Grešner et al., 2021). Especially, a higher consumption of food products with higher fat content such as milk, fish, liver, and eggs, has consistently been found to be a significant determinant of POP levels (Jeon et al., 2021; Zahira et al., 2021). Although our study did not provide enough evidence to establish a direct link between residency and POP concentrations, previous research has shown that geographical factors, such as coastal or inland locations, and rural or urban settings, can impact PCB concentration in breast milk (Shen et al., 2012; Zhang et al., 2016, 2011; Zhu et al., 2022). For instance, a study in Zhejiang Province gathered breast milk samples from the general maternal population and found that levels of 18 PCB congeners found in urban samples were nearly twice as high as those in rural samples (Shen et al., 2012). Additionally, Zhang et al. reported significantly higher PCB concentrations in breast milk from coastal regions compared to inland regions ($p < 0.01$) (Zhang et al., 2011). Therefore, further research is needed to fully understand the complexity of PCB exposure and to identify additional predictors.

3.4 POP levels in settled dust and wristbands

The concentrations and detection frequencies of PCBs and PBDEs were also investigated in the settled dust and SWB samples obtained from recycling factories and e-waste workers, respectively (Table 3). Among the measured POPs, the dominant contaminant was BDE 209 in both settled dust (8 µg/g, 77%) and SWB (220 ng/WB, 94%). In settled dust, BDE 209 was followed by BDE 183 (0.45 µg/g, 70%), CB 28 (0.14 µg/g, 58%), and CB 52 (0.11 µg/g, 56%). On the other hand, SWB samples exhibited elevated levels of CB 101 (8.1 ng/WB, 61%), CB 138 (5.3 ng/WB, 86%, and CB 118 (4.5 ng/WB, 90%).

BDE 209 has consistently been identified as one of the most prominent pollutants in dust samples collected at e-waste recycling facilities (Abafe and Martincigh, 2015b; He et al., 2017; Takahashi et al., 2017; Yang et al., 2013). While our study identified lower chlorinated PCBs as important contributors to settled dust POP levels, similar to Hong et al., it is important to note that some studies, such as the one conducted by Takahashi et al. 2017, have reported different concentration distributions for PCB congeners. They found that penta-PCBs were the major contributors to settled dust contaminants in floor dusts from Vietnamese end-of-life vehicle (ELV)-processing households, as well as in informal e-waste recycling sites and open dumpsites in India (Chakraborty et al., 2018; Hong et al., 2018; Takahashi et al., 2017). Specifically, Takahashi attributed the elevated levels of penta-PCBs to recycling activities related to old electrical capacitors and transformers, containing specific PCB technical mixtures. These variable findings highlight the potential influence of different environmental contexts and sources of contamination in different regions.

Table 3. Levels of PCBs and PBDEs in SWB (ng/WB) and settled dust (µg/g). (N.M stands for "Not Measured")

	SWB (n=79) in ng/WB				Settled dust (n=43) in µg/g			
	DF (%)	p25	p50	p75	DF (%)	p25	p50	p75
CB 28	N.M.	N.M.	N.M.	N.M.	58	< LOQ	0.1	1.4
CB 52	N.M.	N.M.	N.M.	N.M.	56	< LOQ	0.1	0.9
CB 101	61	< LOQ	8.1	19.6	54	< LOQ	0.1	0.5
CB 118	90	2.0	4.6	11.4	54	< LOQ	0.1	0.5
CB 138	86	1.9	5.3	14.8	42	< LOQ	< LOQ	0.3
CB 153	87	1.5	3.5	8.3	44	< LOQ	< LOQ	0.4
CB 180	53	< LOQ	1.1	3.4	35	< LOQ	< LOQ	0.1
BDE 28	15	< LOQ	< LOQ	< LOQ	16	< LOQ	< LOQ	< LOQ
BDE 47	63	< LOQ	2.9	13.6	61	< LOQ	0.2	0.5
BDE 99	37	< LOQ	< LOQ	16.2	72	< LOQ	0.3	0.5
BDE 100	34	< LOQ	< LOQ	2.4	37	< LOQ	< LOQ	0.1
BDE 153	47	< LOQ	< LOQ	6.5	61	< LOQ	0.1	0.4
BDE 154	25	< LOQ	< LOQ	0.8	30	< LOQ	< LOQ	0.1
BDE 183	66	< LOQ	2.1	7.2	70	< LOQ	0.5	0.8
BDE 209	94	54.4	220.6	830.5	77	1.5	8.1	24.6

While SWBs have been widely used as passive samplers to measure levels of various pollutants in the general population and occupational settings, their application for assessing occupational exposure to environmental pollutants in e-waste recycling is still relatively novel (Hammel et al., 2018; Romanak et al., 2019; Samon et al., 2022; S. Wang et al., 2020; Yin et al., 2023). Notably, studies by Wang et al. and Matsukami et al. have used SWBs to assess the exposure of e-waste dismantlers and recycling workers to organophosphate esters and halogenated flame retardants in Dhaka, Bangladesh, and Vietnam, respectively (Matsukami et al., 2022; Y. Wang et al., 2020). In our study, the detection frequency of PBDEs in SWBs matched the trend observed in Bui Dau Village, Vietnam, an e-waste recycling area where valuable metals and plastics are extracted from discarded electronic devices (Matsukami et al., 2022). BDE 209, 183, and 47 were the most frequently detected PBDEs in both settings. Similar patterns have been observed among office building occupants in the USA, UK, China, and India, with BDE 209 being the most abundant contaminant followed by penta-BDEs (Young et al., 2021). The higher levels of BDE 209 compared to other PBDEs are attributed to the recent restrictions on deca-BDE, which, for example, was only banned in 2019 in the UK while penta-BDEs were banned in 2004. In contrast, Nguyen et al. reported significantly higher detection frequencies of PBDEs in SWB samples among e-waste recycling workers in Québec, Canada, even though the wristbands in their study were only worn for an average sampling time of 8 hours compared to our study, where they were employed for a full workweek of exposure (Nguyen et al., 2020). The differences in detection frequencies may be related to the post-deployment wash procedure used in our study, which may have removed some of the target analytes that are likely particle bound at room temperature.

3.5 Correlation analysis in serum, settled dust and SWB

Table 4 reports the results of Spearman's correlation analysis between levels of targeted PCBs and PBDEs in serum, settled dust, and SWB samples among workers. While no positive correlations were found between serum and settled dust, or serum and wristband concentrations, CB 153 exhibited weak but significant negative associations. A Spearman correlation of -0.26 ($p < 0.01$) was observed between serum CB 153 levels and settled dust concentrations, while a correlation of -0.24 ($p < 0.01$) was found between serum CB 153 levels and wristband CB 153 levels. Human exposure to PCBs occurs through multiple pathways, including inhalation, ingestion, dermal contact, and dietary intake (Nguyen et al., 2019). The lack of correlation between PCB levels in serum and settled dust could be attributed to the predominant source of exposure to PCBs, which is through dietary intake or the potential effectiveness of RPE used by the workers (Covaci et al., 2008; Whitehead et al., 2015). In addition, results could be attributed to the difference in metabolism between PCB congeners, where less-chlorinated congeners are generally metabolized more readily than highly chlorinated ones (hexa-CBs), leading to shorter retention times in the body (Grimm et al., 2015; Hopf et al., 2013; Othman et al., 2022). In contrast, the inhalation of PBDE contaminated dust is a more significant pathway for PBDE exposure (Frederiksen et al., 2009; Kim et al., 2016). Also, highly lipophilic PCBs tend to accumulate in adipose tissues, which were not assessed in this study.

While serum pollutant levels were generally not correlated with either settled dust or SWB measurements, SWB demonstrated moderate to strong correlations with settled dust concentrations. It is essential to recognize that the SWB and dust samples were collected during a one-week period, while the serum samples could potentially represent exposure over a substantially longer duration. Consequently, the serum POPs levels may not accurately represent the workers' current exposure levels.

*Table 4. Spearman Correlation for POP concentration in different matrices. p-value annotation legend: *: 0.001 < p <= 0.01; **: 0.0001 < p <= 0.001. For values represented by ND, no correlation coefficients or p-values could be calculated.*

	Serum and settled dust (n=94)		**Serum and SWB (n=70)**		**SWB and settled dust (n=79)**	
	Corr. Coeff.	**p-value**	**Corr. Coeff.**	**p-value**	**Corr. Coeff.**	**p-value**
CB 28	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
CB 52	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
CB 101	N.D.	N.D.	N.D.	N.D.	0.42	**
CB 118	-0.03	0.79	-0.04	0.74	0.42	**
CB 138	-0.12	0.23	-0.06	0.62	0.69	**
CB 153	-0.26	*	-0.24	*	0.60	**
CB 180	-0.15	0.15	-0.14	0.23	0.71	**
BDE 28	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
BDE 47	N.D.	N.D.	N.D.	N.D.	0.57	**
BDE 99	N.D.	N.D.	N.D.	N.D.	0.65	**
BDE 100	N.D.	N.D.	N.D.	N.D.	0.63	**
BDE 153	N.D.	N.D.	N.D.	N.D.	0.50	**
BDE 154	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
BDE 183	N.D.	N.D.	N.D.	N.D.	0.63	**
BDE 209	N.D.	N.D.	N.D.	N.D.	0.58	**

However, it is important to acknowledge that while our study employed a one-week sampling period for SWBs, other studies utilizing shorter sampling durations have reported correlations between POP concentrations in wristbands and serum. This highlights the potential for POP exposure assessment even with shorter sampling intervals. Recent studies, for example, have shown that the accumulation of contaminants in SWBs is not solely time-dependent. For instance, Frederiksen et al. demonstrated a plateauing of several tri-chlorinated PCBs on silicone wristbands by day 31 (Frederiksen et al., 2022). However, the tendency for a curvilinear shape diminished with higher chlorination levels. This suggests that the accumulation rate of certain contaminants may slow over time, potentially influencing the correlation with serum levels.

Furthermore, Nguyen et al. and Wang et al. have shown that high detection rates for PBDEs and PCBs can be achieved with SWBs worn for a single workday or a full day (Nguyen et al., 2020; Y. Wang et al., 2020). This indicates that accumulation can occur in a relatively short time frame. Additionally, Nguyen et al. observed a significant and positive correlation between log-transformed BDE-209 in blood plasma and wristband after a single 8hr workday of sampling(Nguyen et al., 2020). This finding suggests that for certain contaminants, a strong correlation between serum and SWB levels can be established even over short periods.

Although there is limited research on the effectiveness and suitability of SWBs in estimating serum PCB levels, a study conducted by Hammel discovered a significant and positive correlation between SWB levels of BDE-47, -99, -100, and -153 and corresponding serum biomarkers (Hammel et al., 2018) indicating the potential applicability of such monitoring devices for assessing PBDE levels in occupational settings. The strong correlations between SWB and settled dust suggest the use of SWBs as a potentially valuable tool for assessing individual workers' POP exposure in occupational settings.

On the other hand, the strong positive correlations observed between SWB and dust, as well as the varying degrees of correlation between SWB and serum and dust and serum, may potentially be attributed to several factors. Adherence to established PPE protocols during work activities, particularly correctly placing SWB over PPE, could explain the stronger positive correlation observed between SWB and dust measurements. This practice enables SWBs to directly capture dust particles on the PPE surface, thereby providing a more accurate reflection of workplace exposure. The effectiveness of RPE could account for the weaker correlations between SWB/serum and dust/serum. By mitigating inhalation exposure, which could be a significant contributor to serum PCB levels in occupational environments, RPE might limit the direct association between environmental exposure markers like SWB or dust and serum levels. It is also important to consider that dietary intake significantly influences serum PCB levels that could potentially diminish correlations with environmental exposure markers such as SWB and dust.

3.6 Intake estimation of PCBs and PBDEs through settled dust

Exposure to PCBs and PBDEs can occur through multiple pathways, including inhalation, dermal absorption, dust ingestion and dietary intake. Considering reports that propose dermal contact and dust ingestion as primary exposure routes for occupational e-waste workers, it is crucial to assess exposure to PCBs and PBDEs by evaluating estimated daily intake values through dust ingestion and dermal absorption of dust (Doan et al., 2022; Hammel et al., 2023; Muenhor et al., 2010; Nguyen et al., 2019; Ohajinwa et al., 2019a, 2019b; Tue et al., 2013). As depicted in Table 5, dust ingestion estimates were calculated considering the 50th and 95th percentile exposure scenarios and subsequently compared with previously reported values regarding PCB/PBDE exposure in the general population. The EDI values via dust ingestion with percentiles (p25, p50, and p75) for both scenarios, along with the corresponding reference doses (RfD) for each congener and EDI values via dermal absorption of dust, are provided in Table SI-8.

Our findings indicated that e-waste workers exhibited the highest median EDIs (0.03 and 0.09 ng/kg bw/day) for both the 50th and 95th percentile exposure scenarios for ΣPCB measured in dust, surpassing most of the values reported in selected countries evaluating PCB exposure among the general public with the exception of values reported from US and Canada (Table 5) (Harrad et al., 2009). By contrast, our findings indicate that occupational exposure to PBDEs (ΣPBDE) via dust ingestion is comparable to levels reported for the general population. Overall, even under the worst-case exposure scenario, the total EDI values remained below the available RfD values for the e-waste workers, suggesting a low health risk (Table SI-8).

Table 5. Comparison of Median Estimated Daily Intake of PCBs and PBDEs via dust ingestion with other studies

Reference	Country	Sampling place	Year	Sample (n)	Class	50th percentile exposure scenario (ng/kg bw/day)	95th percentile exposure scenario (ng/kg bw/day)
This study	**six EU countries**	***e-waste factory***	***2023***	***103[a]***	***PCB***	***0.03***	***0.09***
(Dirtu and Covaci, 2010)	Romania	home	2010	18[b]	PCB	0.01	0.04
(Harrad et al., 2009)	Canada	home	2009	10[c]	PCB	0.07	0.19
(Harrad et al., 2009)	New-Zealand	home	2009	20[c]	PCB	0.01	0.03
(Harrad et al., 2009)	UK	home	2009	20[c]	PCB	0.01	0.03
(Harrad et al., 2009)	US	home	2009	20[c]	PCB	0.06	0.14
(Roosens et al., 2010a)	Belgium	university housing	2010	19[d]	PCB	0.00	0.01
(Coelho et al., 2016)	Portugal	home	2016	28[e]	PCB	NA	0.01
This study	**six EU countries**	***e-waste factory***	***2023***	***103[f]***	***PBDE***	***0.27***	***0.82***
(de la Torre et al., 2020)	three EU countries	home	2020	21[g]	PBDE	0.03	1.22
(D'Hollander et al., 2010)	Belgium	home, office	2010	53[h]	PBDE	0.01*	0.24*
(Hassan and Shoeib, 2015)	Egypt	home, car, workplace	2005	31[i]	PBDE	0.06	0.14
(Wilford et al., 2005)	Canada	home	2005	68[j]	PBDE	0.11	2.57
(Cunha et al., 2010)	Portugal	home, car	2010	20[k]	PBDE	0.06	1.28
(Fromme et al., 2009)	Germany	home	2009	34[l]	PBDE	0.03	0.13
(Dirtu and Covaci, 2010)	Romania	home	2010	18[m]	PBDE	0.43	1.08
(Coelho et al., 2016)	Portugal	home	2016	28[n]	PBDE	NA	0.49
(Roosens et al., 2010b)	Belgium	home	2010	53[o]	PBDE	0.01	0.04
(Brommer et al., 2012)	Germany	home, car	2012	23[p]	PBDE	0.04	0.3
(Pasecnaja et al., 2021)	Latvia	home	2021	34[q]	PBDE	1.24	3.1

Congeners included: a) CB 28, 52, 101, 118, 138, 153, 180 b) CB 118, 153, 138, 187,183, 156, 180, 170 c) CB 28, 31, 52, 101, 118, 138, 153, 180 d) CB 118, 138, 153, 180, 170 e) CB 28, 52, 101, 138, 153, 180 f) BDE 28, 47, 99, 100, 153, 154, 183, 209 g) BDE 28, 49, 71, 47, 100, 99, 85, 154, 153, 184, 183, 201, 204, 197, 203, 196, 208, 207, 206, 209 h) BDE 28, 47, 100, 99, 154, 153, 183, 197, 196, 203 i) BDE 47, 99, 100, 183, 209 j) BDE 17, 28, 47, 66, 100, 99, 85, 154, 153, 138, 183, 209 k) BDE 28, 49, 47, 66, 100, 99, 85, 154, 153, 183, 197, 203, 196, 207, 206, 209 l) BDE 99, 100, 153, 154, 183, 209 m) BDE 28, 47, 100, 99,154, 153, 183, 209 n) BDE 28, 47, 99, 100, 153, 153, 183, 209 o) BDE 47, 100, 99, 154 and 153 p) BDE 47, 99, 183, 209 q) BDE 17, 28, 47, 49, 99, 100, 138, 139, 153, 154, 155, 183, 209.

*Mean Estimated Daily Intake was taken. When the dust intake was described in ng/day, values were converted by dividing the amounts by 70 kg to obtain the corresponding EDI in ng/kg bw/day.

Dermal absorption at the 50th percentile concentration amounted to 0.01 and 0.03 ng/kg bw/day for ΣPCB and ΣPBDE, respectively. While absorption at the 95th percentile concentration showed values of 0.03 and 0.22 for ΣPCB and ΣPBDE, respectively (Table SI-8). These findings suggest that, in occupational settings, dermal absorption and dust ingestion contribute roughly equally to personal exposure to PCBs and PBDEs. This aligns with previous research conducted in metal shredding facilities in Wallonia, Belgium, which observed similar contributions of soil and dust ingestion to PCDD/F/dl-PCB exposure as dermal absorption (Doan et al., 2022).

Dermal exposure was the highest for BDE-209, with a median value of 0.02 ng/kg-bw/day and a 95th percentile of 0.21 ng/kg-bw/day. In contrast, PCB exposure was lower, with CB-28 exhibiting a median of 0.003 ng/kg-bw/day and a 95th percentile of 0.16 ng/kg-bw/day. Interestingly, our estimated dermal exposures based on dust levels suggest that European e-waste dismantlers have higher ΣPBDE exposure compared to Canadian e-waste workers, with median and 95th percentile levels of 0.03 ng/kg-bw/day and 0.22 ng/kg-bw/day, respectively, compared to 0.00199 ng/kg-bw/day in Canada (Nguyen et al., 2019). While dietary exposure was not assessed in this study, it remains a recognized pathway for PBDEs and PCBs in the general population (Muenhor et al., 2010). Additionally, emerging evidence suggests that inhalation of indoor air and ingestion of indoor dust are significant contributors to overall exposure (Nguyen et al., 2020).

4 Conclusions

Our study evaluated occupational exposure to PCBs and PBDEs among e-waste workers in Europe through the assessment of serum, settled dust and SWB samples. We observed statistically significant differences in the levels of specific PCB congeners, namely CBs 118, 138, 153, and 180, among workers and controls engaged in processing e-waste, especially white goods and batteries. Additionally, individuals who were not directly involved in e-waste processing, e.g., performing office work at the same recycling facilities, exhibited higher PCB levels compared to controls recruited outside the e-waste processing industry, suggesting elevated background exposures. Despite this, the relatively low RfD values for PCBs and PBDEs among e-waste workers suggest a low health risk associated with occupational exposure. Our findings highlight the significance of several factors in determining the exposure to PCBs among e-waste recycling workers such as age, type of e-waste processed and number of years smoked. Our findings indicate that effective reduction of PCB exposure levels in e-waste workers can be attained through the implementation of RPE and localized exhaust ventilation systems. PCBs and PBDEs exhibited moderate to strong correlations between SWB and settled dust measurements, however, no correlations between serum and SWB levels, as well as between serum and settled dust levels, could be achieved due to the low detection frequencies of several congeners in serum and SWB. The findings of this study provide valuable insights for policymakers and stakeholders seeking to establish effective regulations and promote sustainable e-waste management practices. By identifying specific pollutants of concern and understanding their prevalence in the e-waste recycling process, targeted strategies can be developed to address potential environmental and occupational health risks.

Funding sources

Adam Cseresznye acknowledges the Flemish Exposome Project (Flexigut - FFB200392) for his PhD fellowship. Paulien Cleys was supported by the Research Foundation – Flanders-Belgium (FWO) under Grant number 1S70820N, which provided her a PhD fellowship at the University of Antwerp. Yu Ait Bamai acknowledges a fellowship from the Japan Society for the Promotion of Science (JSPS) through the Fund for the Promotion of Joint International Research (Fostering Joint International Research (A), grant number 19KK0288). Giulia Poma was supported by the Exposome Centre of Excellence of the University of Antwerp (BOF grant, Antigoon database number 41222). This project has also received funding from the European Union's Horizon 2020 research and innovation program under grant agreement No 733032 (HBM4EU). In addition, the Finnish Work Environment fund participated in financing this work (grant number 200345).

Ethical approval

The study was conducted in accordance with regulations of the European Union and the current General Data Protection Regulation (GDPR). More information on the Legal and Ethics Policy and Material and associated Data Transfer Agreement is described elsewhere (“HBM4EU Legal and Ethics Policy Paper,” 2018; “Material and Associated Data Transfer Agreement,” 2020). The study was conducted in accordance with the Declaration of Helsinki. Study protocols have been approved by ethical review boards in each of the participating countries, with the approvals granted before recruiting the study participants. The ethical boards reviewing and approving the study are as follows: Portugal: Ethical Committee of the National Institute of Health Dr. Ricardo Jorge (Ethics Committee for Health, INSA), authorized on the 22nd September 2021) and Ethical Committee of the Lisbon School of Health Technology authorized on the 13th March 2020. Finland: Coordinating ethics committee, HUS Joint Authority, Helsinki, Finland. Decision number HUS/1357/2021, dated 2.6.2021. For Belgium (KU Leuven), the information on ethics approval is Reference number: S64321, Authorization date: 8th October 2020, Approved by: Ethics Committee Research UZ/KU Leuven. For the Netherlands, the CMO Regio Arnhem Nijmegen, approved on 20th January 2021 (Reference number: 2020–7089; National registry: NL67044.091.18). For Luxembourg, the National Research Ethics Committee, approved on the 4th August 2021, and the Ministry of Health approved on 17th November 2021 (Reference number: 83bx30c66). For Latvia, Rīgas Stradiņš University Research Ethics Committee, approved on 14th April 2021 (Reference number: 22–2/250/2021).

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