



Measurement of polychlorinated biphenyls with hand wipes and matched serum collected from Chinese E-waste dismantling workers: Exposure estimates and implications

Chen Zhao^{a,c,1}, Ji-Fang-Tong Li^{a,b,1}, Xing-Hong Li^{a,b,*}, Meng-Qi Dong^{a,b}, Yuan-Yuan Li^{a,b}, Zhan-Fen Qin^{a,b}

^a State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, PO Box 2871, 18 Shuangqing Road, Haidian District, Beijing 100085, PR China

^b University of Chinese Academy of Sciences, 19A Yuquan Road, Shijingshan District, Beijing 100049, PR China

^c College of Earth Sciences, Guilin University of Technology, Guilin 541006, PR China

HIGHLIGHTS

- Hand wipes were used to understand dermal/hand-to-mouth exposure to PCBs for e-waste workers.
- Dermal absorption of low-chlorinated CB congeners was non-negligible for e-waste workers.
- There were insignificant associations found between serum PCBs and thyroid hormones.

GRAPHICAL ABSTRACT



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ABSTRACT

To date, dermal/hand-to-mouth exposure to chemicals in the e-waste recycling environment has not been sufficiently understood, and the importance of dermal absorption of chemicals in e-waste dismantling workers remains controversial. In this study, we utilized hand wipes and matched sera to characterize dermal/hand-to-mouth exposure to PCBs for e-waste dismantling workers, and potential effects on thyroid hormones were also assessed. PCB loadings in hand wipes varied from 0.829–265 ng wipe⁻¹ (11.3–2850 ng m⁻² wipe⁻¹), with 37.2 ng wipe⁻¹ (432 ng m⁻² wipe⁻¹) as the median value. Serum concentrations of PCBs ranged from 32.3–3410 ng g⁻¹ lipid weight (lw) with 364 ng g⁻¹ lw as the median value. Between wipes and sera, lower-chlorinated congeners (e.g. CB-28, -66, -74, -99, -105 and -118) showed significant associations ($p < 0.01$), but higher-chlorinated congeners (e.g. CB-138, -153, -156, -170, and -180) did not. These lower-chlorinated CBs were the major contributors to estimated dermal/hand-to-mouth average daily doses (ADDs) and the hazard index (HI). Correspondingly, their estimated contributions to serum levels by dermal absorption were also significant, with the contribution of CB-28 being as high as 21.4%. As a consequence, dermal absorption of some low-chlorinated congeners was a non-negligible route for e-waste dismantling workers. Although insignificant association was shown between serum PCBs and thyroid hormones, the potential health risk should be of concern due to the high levels of PCBs observed in workers' sera.

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* Corresponding author at: State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, PO Box 2871, 18 Shuangqing Road, Haidian District, Beijing 100085, PR China.

E-mail address: lxhzpb@cees.ac.cn (X.-H. Li).

¹ Authors have the equal contribution.

1. Introduction

Various chemicals exist in the e-waste recycling environment, which are released from the chemical-containing products into the surrounding environment (e.g. air, dust and various surfaces) during the recycling processes (Ni et al., 2010; Song and Li, 2014). The e-waste recycling industry is especially flourishing in underdeveloped/developing countries, and large numbers of residents are engaged in e-waste recycling industries each year in these countries (Grant et al., 2013; Orisakwe et al., 2019; Song and Li, 2014; Wath et al., 2011). Because of poverty and/or the lack of self-protection awareness, e-waste workers in underdeveloped/developing countries generally do not employ the proper protective measures when they work, and are highly exposed groups to various e-waste-relating pollutants (Wang et al., 2016). Polychlorinated biphenyls (PCBs), with 209 congeners, are a group of chemicals typically observed in the e-waste recycling environment, and e-waste dismantling workers are highly exposed to PCBs (Song and Li, 2014). Due to the environmental persistence, bioaccumulation and potential toxic effects of PCBs, the health risks to e-waste workers caused by PCB exposure merit long-term and sustained attention (Grant et al., 2013; Qu et al., 2014).

PCBs can gain entry into the human body by inhalation, dermal absorption, dietary and inadvertent hand-to-mouth ingestion. Because research on the exposure pathways can help formulate appropriate and effective strategies to reduce the human health risk, efforts have been made to investigate the exposure pathways of PCBs for e-waste dismantling workers (Liu et al., 2019; Song and Li, 2014; Tue et al., 2013; Wang et al., 2016; Xing et al., 2009; Xing et al., 2011; Xing et al., 2010). To date, the general consensus suggests that dietary ingestion and inhalation constitute the main routes of exposure, while both dermal absorption by contact with dust/airborne particles and hand-to-mouth dust ingestion tend to be considered negligible (Liu et al., 2019; Song and Li, 2014; Tue et al., 2013; Wang et al., 2016; Xing et al., 2009; Xing et al., 2011; Xing et al., 2010). However, more recent reports have indicated that the previous dermal exposure assessments associated only with dust (Tue et al., 2013; Wang et al., 2016; Xing et al., 2011) and airborne particles (Liu et al., 2019) might not give comprehensive results for e-waste workers, and the exposure dose might be underestimated due to the contribution from other sources such as contact with e-waste articles and absorption from e-waste aerosol. For example, Wu et al. (2016) suggested the importance of dermal uptake of airborne flame retardants from human exposure to e-waste combustion fumes. In their review, Ma et al. (2021) reported that it might be a significant omission if direct dermal contact with e-waste articles is not considered when performing dermal exposure assessments. Also, a contrasting opinion was presented in a recent biomonitoring study, in which dermal absorption was inferred to be a significant contributor to body CB burden based on information derived from serum monitoring of e-waste recycling workers (Kaife et al., 2020). Overall, these results indicate that dermal exposure to PCBs for e-waste workers is not yet sufficiently understood, and that opinions on the importance of PCB dermal exposure are not unanimous.

The hand-wipe approach is one of the sampling techniques used to evaluate exposure by dermal absorption and hand-to-mouth ingestion, which has been successfully applied to various organic flame retardants (e.g. phosphorus flame retardants, legacy and novel brominated/chlorinated flame retardants) in non-e-waste recycling environments (e.g. homes and offices) (Brouwer et al., 2000; Gong et al., 2014; Hoffman et al., 2015; Kang et al., 2012; Liu et al., 2018; Liu et al., 2017; Stapleton et al., 2012; Tay et al., 2018; Watkins et al., 2011). However, the application of the hand-wipe approach has not been extensively developed in the e-waste recycling environment. To our knowledge, only two studies involved the preliminary application of hand wipes for e-waste workers. One study investigated potential factors that might affect the collection of brominated/organophosphate flame retardants by hand wipes (Beaucham et al., 2019). Another collected 10 hand-

wipe samples to compare the contamination levels of some legacy organic contaminants (including PCBs) in hair samples, and high loadings of contaminants were found in the hand wipe samples, with a median PCB value of 23.3 ng wipe⁻¹ (Qiao et al., 2019). As described above, the importance of dermal absorption of polychlorinated biphenyls (PCBs) in e-waste dismantling workers remains controversial to date. Since the hand-wipe approach is a personal measurement of pollutants (Watkins et al., 2011), chemicals in hand wipes could have a close association with the internal doses if dermal exposure is an important route (Hoffman et al., 2015; Stapleton et al., 2012; Watkins et al., 2011). Therefore, hand-wipe measurements combined with human biomonitoring may be helpful in identifying the importance of human dermal exposure and/or hand-to-mouth uptake of PCBs.

Human exposure to pollutants derived from e-waste is likely to be associated with a series of adverse human health effects (Grant et al., 2013; Song and Li, 2014). The current literature suggests that exposure to certain congeners of PCBs may potentially be associated with an increased risk of thyroid cancer (Alsen et al., 2021). However, the reported effects of PCBs on thyroid hormones in adults are not consistent (Salay and Garabrant, 2009), and thus more studies are required to elucidate the disruptive effects of PCBs on thyroid hormone regulation.

In this study, hand wipes and matched serum samples were collected from e-waste dismantling workers. The objectives of this study were (1) to gain comprehensive knowledge on dermal/hand-to-mouth exposure to PCBs for e-waste dismantling workers by the hand-wipe sampling approach; (2) to identify the importance of dermal/hand-to-mouth exposure in combination with serum measurements; (3) to explore the relationship between serum and thyroid hormones (THs) and the potential health effects associated with PCBs.

2. Materials and methods

2.1. Sample collection

Taizhou city, located on the southeast coast of China, is one of the well-known e-waste recycling areas in China. Many people are employed to deal with e-wastes for recycling of metals or electronic components. In 2018, we recruited 76 workers who had been engaged in e-waste dismantling for at least one year to collect serum samples and hand-wipe samples. The volunteers came from two adjacent e-waste factories licensed by the Chinese government to operate e-waste recycling businesses. All volunteers who provided serum and hand-wipe samples were asked to perform similar tasks, primarily manually dismantling e-waste products and/or sorting out useful electronic components from fragmented e-waste. In the end, 34 and 42 qualified volunteers were recruited successfully in two factories.

Venous blood was drawn before breakfast, and hand wipes were collected after 4 h of work. The fasting blood samples (approximately 10 mL) were collected and centrifuged to separate the sera. The sampling method for hand wipes mainly followed that previously described (Gong et al., 2014). Briefly, the participants' two hands were wiped with four-layer sterile gauze pads (7.5 cm × 7.5 cm) infused with isopropyl alcohol, each hand was wiped four times, each time with an unused layer of the gauze pad. All hand-wipe samples were collected by a single investigator to avoid variabilities due to differences between investigators. Four field blank samples were also collected, two containing isopropyl alcohol only and the other two containing un-used gauze pads infused with isopropyl alcohol. The institutional review board of the Research Center for Eco-Environmental Sciences approved the study protocol prior to collection of samples, and all donors also gave their written informed consent to participate. The demographic information for these workers, including the gender, age, body weight and height, duration of e-waste recycling employment, and residential time in Taizhou were recorded by questionnaires. Three serum thyroid hormones (THs), total cholesterol and triglycerides were determined in a local hospital. The personal information of participants, total cholesterol,

triglycerides, levels of serum thyroid hormones (THs) and lipid contents are listed in Table SI-1 of the Supplementary information.

2.2. Sample pretreatment and chemical analysis

Thirty-two CB congeners commonly found in food and human tissues were determined in this study. The pretreatment of samples mainly followed procedures described in the previously published literature with minor modification (Cao et al., 2019a; Lv et al., 2015). Briefly, hand-wipe samples were spiked with surrogate standards and then extracted by the Soxhlet extraction method. The extracts were purified by a multi-layer chromatography column. Serum samples (2.0 mL) were firstly denatured with HCl and isopropanol, and then ultrasonically extracted with a mixture of methyl tert-butyl ether and hexane with three repeats. Next, the combined extracts were concentrated and purified by the same process as that of the hand-wipe samples mentioned above. Gas chromatography coupled with low resolution mass spectrometry (Agilent 6890 GC/5973 LRMS) was used for determining the target compounds. Detailed information on the chemicals and materials, sample extraction and clean-up, instrumental analysis including quality control and assurance is provided in the Supplementary information.

2.3. Data analysis

All statistical analyses were performed with Excel (2016), OriginPro8 SRO software (www.OriginLab.com) and the SPSS 23.0 software package (SPSS Inc., Chicago, IL, USA), and statistical significance was set at the p -value <0.05 level. Compounds detected in >60% samples were included in the statistical analysis. For samples below the method detection limits (MDLs), PCB loadings/levels were calculated by $MDL / \sqrt{2}$ when descriptive statistics were conducted. The one-sample Kolmogorov-Smirnov Test was used to test the normality of the distribution of continuous variables. Spearman correlation analysis was employed to examine the relationships between log-transformed CB concentrations in serum and/or hand-wipe samples as well as log-transformed CB concentrations and log-transformed TH levels in serum. Principal component analysis was performed only for CB congeners simultaneously detected in serum and hand-wipe samples.

The estimation models included hand surface areas, average daily doses (ADDs) by dermal and/or hand-to-mouth uptakes based on CB loadings/levels in hand wipes, body fat content and body mass index, internal doses obtained by dermal and/or hand-to-mouth uptakes and their corresponding contributions to serum levels as well as corresponding health risks, as set out in Eqs. (1) to (9) of the Supplementary information.

3. Result and discussion

3.1. PCBs in serum

3.1.1. Levels

Residual levels of PCB congeners in sera of workers are displayed in Table 1. Twelve of the thirty-two CB congeners with frequency >60% were reported, and the median value of total concentrations of all CB congeners (\sum PCB) was 364 ng g⁻¹ lw. The median serum level for the general population living in Taizhou was 26.2 ng g⁻¹ lw (Lv et al., 2015), which is over 10-fold lower than the value obtained in this study. Furthermore, the body burden of PCBs in Taizhou inhabitants related to e-waste recycling in 2006 was reported (Zhao et al., 2010). The median value of five congeners (CB-105,-118,-138,-153,-180) in 2006 was 118 ng g⁻¹ lw, which was approximately 2-fold lower than the present result (203 ng g⁻¹ lw for the same five congeners). The comparison suggested that people associated with e-waste dismantling activities in Taizhou have been continuously exposed to high levels of PCBs over the past decade.

Table 1
Descriptive statistics for PCBs measured in serum samples (n = 76).

Compound	N	Serum level (ng g ⁻¹ lw)				
		Min.	Max.	Mean	SD	Median
CB-28	69	<MDL ^a	284	65.5	63.9	40.3
CB-60	49	<MDL	66.0	64.2	71.5	43.9
CB-66	51	<MDL	204	31.0	40.5	17.6
CB-74	54	<MDL	368	13.4	17.4	6.11
CB-99	59	<MDL	289	35.0	42.4	23.2
CB-105	64	<MDL	221	101	105	62.3
CB-118	67	<MDL	636	35.7	37.6	20.3
CB-138	66	<MDL	636	83.6	94.6	67.3
CB-153	70	<MDL	707	60.6	83.9	43.9
CB-156	49	<MDL	120	13.9	18.9	8.46
CB-170	46	<MDL	117	22.9	28.3	14.5
CB-180	56	<MDL	155	15.3	20.2	9.20
\sum PCB		32.3	3410	541	535	364

^a MDLs: method detection limits.

3.1.2. Profiles

The profile of CB congeners in serum can be seen in Fig. 1. For workers, the most abundant groups were penta-CBs and hexa-CBs, followed by tetra-CBs, \leq tri-CBs and hepta-CBs. For convenient description, we defined low-chlorinated CBs here as congeners containing less than or equal to 5 chlorine atoms, and high-chlorinated CBs as congeners containing more than 5 chlorine atoms. Compared to the relative abundance of six reported CB congeners in the general population (Lv et al., 2015), a shift towards lower-chlorinated congeners (e.g. CB-28 and CB-118) could be seen for e-waste workers, as shown in Fig. SI-1. E-waste recycling workers in Ghana also tended to accumulate lower-chlorinated PCBs (Kaifre et al., 2020) when compared to the reference group. This indicated the elevated accumulation of low-chlorinated CBs in the workers, which was likely associated with e-waste dismantling activities.

In order to reduce human health risk from exposure, the sources of chemicals in e-waste workers are of particular concern. For PCBs, it is well-known that the high-chlorinated CBs (e.g. hexa- and hepta-CB) accumulated in humans are generally related to dietary ingestion (Lv et al., 2015; Xing et al., 2010), and elevated low-chlorinated CB congeners in e-waste workers are likely related with inhalation exposure (Xing et al., 2009), whereas the dermal/hand-to-mouth uptake via

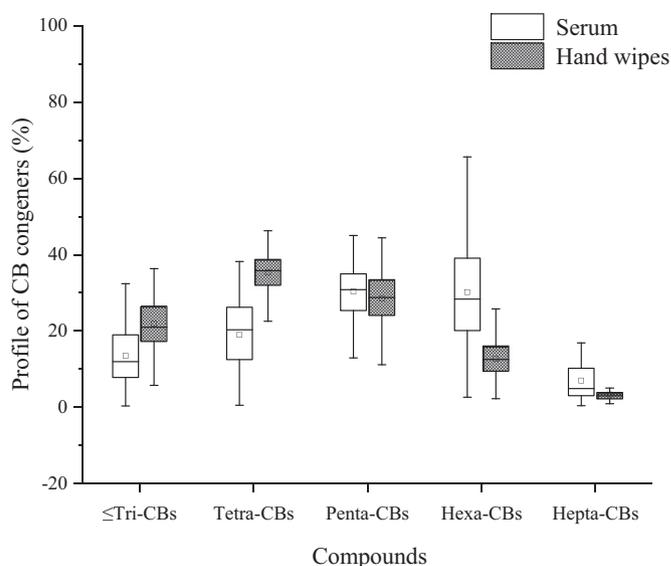


Fig. 1. Box plot (minimum, 25% quartile, median, 75% quartile, and maximum; the squared rectangle represents the mean) (outliers are not shown) of PCB profiles in serum and hand wipes from e-waste dismantling workers in Taizhou, China.

contact with dust had been assessed as negligible in an early study (Xing et al., 2011). However, a more recent study (Wang et al., 2016) showed an increase of 2–3 orders of magnitude in the dermal daily exposure dose by contact with dust for e-waste workers in Taizhou compared with the early result (Xing et al., 2011). In fact, dust-related dermal exposure could not provide comprehensive results of dermal exposure, because adsorption of aerosol and contact with e-waste articles and contaminated surfaces could also lead to dermal exposure (Ma et al., 2021; Weschler and Nazaroff, 2012; Wu et al., 2016). Furthermore, Kaifie et al. (2020) inferred that the elevated plasma levels of less-chlorinated CBs in manual dismantling workers were most likely associated with dermal exposure. These clues implied that the importance of dermal uptake of PCBs by e-waste workers might have changed, and its contribution to internal doses needed to be evaluated.

Overall, e-waste workers in Taizhou are still at a high exposure level. The dermal exposure to PCBs and the related human health risks deserve attention.

3.2. PCBs in hand wipes

3.2.1. Loading

Descriptive statistics for PCB loadings in hand-wipe samples are presented in Table 2. Twenty-four of the thirty-two CB congeners were able to be detected in hand-wipe samples. Detections of PCBs in hand wipes indicated that these compounds could be present on the skin of the hand and that human exposure to PCBs might occur by dermal absorption and hand-to-mouth ingestion. The mass loading of total CBs varied from 0.829 ng wipe⁻¹ (or sample⁻¹, 4 h per wipe) to 265 ng wipe⁻¹, with 37.2 ng wipe⁻¹ as the median value. Considering that CB loading in hand-wipe samples might be affected by the hand surface area of a specific individual, CB mass was further normalized to hand surface area estimated by Eq. (1) in the Supplementary information. Consequently, the hand-area-normalized loading of total CBs was in the range of 11.3–2850 ng m⁻² wipe⁻¹ with a median value of 432 ng m⁻² wipe⁻¹.

Frederiksen et al. (2020) reported that the median value of CB mass loading in hand wipes was 113 ng wipe⁻¹ and 4.6 ng wipe⁻¹ for residents living in contaminated high-rise apartment buildings and a reference group in Denmark, respectively. The mass loading in this study

(37.2 ng wipe⁻¹) was lower than that in the exposure group, but far higher than that in the reference group in Denmark. Furthermore, the current data were somewhat higher than the observed loading (23.3 ng sample⁻¹) in hand wipes of e-waste workers in another e-waste recycling region in China (Qiao et al., 2019). Recently, Cao et al. (2019b) studied the CB loading in human forehead wipes from students and college staff in a central region of China. The median value of total CB loading during haze days was 1300 ng m⁻², which appeared to be higher than the value obtained in the current study. However, when compared to the congeners simultaneously determined in two studies, it was found that the loadings of most of CB congeners (excluding CB-37, CB-60 and CB-52) in the current study were close to or higher than the values reported in Cao et al.'s (2019b) study. In addition, Frederiksen et al.'s (2020) study showed that a relatively short dermal exposure time (1–2 years) might lead to a relatively rapid increase in serum concentration levels of low-chlorinated congeners, while e-waste workers participating in this study had exposure durations varying from 1 to 30 years. These comparisons indicated that the amount of PCBs attached to the e-waste workers' hand skin was not trivial. E-waste workers were continuously and chronically exposed to CBs at close to haze levels, and the dermal exposure of e-waste workers to PCBs might have an influence on the internal doses.

3.2.2. Profile

In this study, the most abundant congener in hand-wipe samples was CB-28, followed by four congeners (CB-44, CB-70, CB-118 and CB-101) with similar loadings. The relative abundance of CB congeners was different from that reported by Frederiksen et al. (2020), in which the five most abundant congeners were CB-52, CB-31, CB-44, CB-28 and CB-18. This difference was also found in the report of Cao et al. (2019a), where the five most important congeners were CB-37, CB-60, CB-7, CB-206 and CB-4. These differences might be related to the different sources/processes in the studies. In the study of Frederiksen et al. (2020), PCBs in hand-wipe samples mainly came from the release of CB-containing building materials, differing from the release from the e-wastes in the study. In the study of Cao et al. (2019a), PCBs were associated with haze days, and more likely came from the air since the samples were collected from the forehead skin. In the current study, wipes were taken from e-waste workers' hands, which were frequently

Table 2
Descriptive statistics for PCB loading measured in hand-wipe samples (n = 76).

Compound	N	Hand-wipes (ng wipe ⁻¹)					Hand-wipes (ng m ⁻² wipe ⁻¹)				
		Min.	Max.	Mean	SD	Median	Min.	Max.	Mean	SD	Median
CB-8	75	<MDL	10.8	1.77	2.27	0.693	<MDL	121	20.0	25.6	8.41
CB-28	72	<MDL	52.1	10.8	11.1	6.58	<MDL	561	123	125	75.2
CB-37	63	<MDL	14.0	3.40	3.09	2.28	<MDL	151	38.5	35.0	27.1
CB-44	72	<MDL	18.3	4.98	4.51	3.23	<MDL	205	57.0	51.9	38.3
CB-49	70	<MDL	12.0	2.82	2.75	1.70	<MDL	129	32.1	31.5	20.5
CB-52	71	<MDL	14.0	3.94	3.53	2.18	<MDL	171	44.8	41.1	26.1
CB-66	73	<MDL	18.2	4.03	4.13	2.45	<MDL	196	45.9	47.2	28.1
CB-70	74	<MDL	21.6	5.08	4.97	3.37	<MDL	232	58.0	56.8	37.4
CB-74	72	<MDL	10.7	2.62	2.34	1.79	<MDL	116	29.8	26.7	20.2
CB-77	55	<MDL	3.19	0.976	0.862	0.624	<MDL	38.7	11.1	10.0	6.42
CB-82	60	<MDL	2.70	0.825	0.682	0.641	<MDL	30.3	9.40	7.72	6.92
CB-87	74	<MDL	11.7	2.69	2.59	1.70	<MDL	128	30.8	29.3	20.3
CB-99	75	<MDL	7.05	1.81	1.57	1.27	<MDL	80.5	20.7	17.9	14.1
CB-101	75	<MDL	18.0	4.38	3.95	2.95	<MDL	205	50.3	45.2	36.9
CB-105	75	<MDL	9.90	2.43	2.10	1.61	<MDL	113	27.9	23.9	19.8
CB-118	76	0.525	20.2	4.82	4.28	3.33	6.32	230	55.3	48.8	38.0
CB-128	70	<MDL	4.14	1.08	0.935	0.729	<MDL	44.5	12.4	10.6	8.40
CB-138	72	<MDL	10.4	2.71	2.42	1.84	<MDL	119	31.1	27.4	21.6
CB-153	76	0.115	10.7	2.49	2.27	1.64	1.56	122	28.6	25.8	19.3
CB-156	68	<MDL	2.88	0.858	0.651	0.632	<MDL	32.8	9.85	7.42	6.81
CB-170	62	<MDL	3.30	0.812	0.657	0.609	<MDL	35.4	9.30	7.49	7.35
CB-180	67	<MDL	2.30	0.691	0.544	0.557	<MDL	25.7	7.93	6.23	5.59
CB-183	48	<MDL	0.750	0.273	0.182	0.220	<MDL	8.56	3.13	2.11	2.43
CB-187	48	<MDL	0.809	0.271	0.194	0.211	<MDL	9.56	3.10	2.24	2.34
∑ PCB		0.829	265	61.2	59.3	37.2	11.3	2850	699	676	432

exposed to multiple sources including air and dust, PCB-containing products and contaminated surfaces, and so on. In addition to air-to-skin transfer, other source-skin transfer processes might be contributors to on-hand PCBs in this study. Regardless of PCB sources, a common feature could be extracted from these studies, which was that low-chlorinated PCBs accumulated more in the skin wipes. In the study, the profile of PCB homologous groups in hand-wipe samples is displayed in Fig. 1. Tetra-CBs, penta-CBs and \leq tri-CBs constituted the largest group, followed by hexa-CBs and hepta-CBs. In total, approximately 86% of the total loading was made up of \leq penta-CBs. Low-chlorinated CB congeners made up the majority of the total loading, which could also be found in the reports of Cao et al. (2019a) and Frederiksen et al. (2020). In short, lower-chlorinated congeners (e.g. di- to penta-CBs) adhered more readily to human skin than higher-chlorinated congeners (e.g. hexa- to hepta-CBs), and dermal/hand-to-mouth exposure to PCBs for e-waste workers might be more related to the low-chlorinated CBs.

3.2.3. Estimation of average daily dose based on CB loadings in hand wipes

Average daily doses (ADDs, $\text{pg kg}^{-1} \text{d}^{-1}$) via dermal and hand-to-mouth uptakes were estimated based on Eqs. (2) and (3) in the Supplementary information, respectively. The median values of the estimated ADDs for PCBs are listed in Table SI-3. The ADD for $\sum \text{PCB}_{16}$ was the sum of the individual ADDs listed from di- to penta-CBs (from CB-8 to CB-118), and that for $\sum \text{PCB}$ was the sum of all individual ADDs listed.

The ADD of $\sum \text{PCB}$ was $4960 \text{ pg kg}^{-1} \text{d}^{-1}$ by dermal absorption and $402 \text{ pg kg}^{-1} \text{d}^{-1}$ by hand-to-mouth uptake, respectively. For each specific congener, ADD varied from 4.27 to $974 \text{ pg kg}^{-1} \text{d}^{-1}$ by dermal absorption, and ranged from 0.311 to $145 \text{ pg kg}^{-1} \text{d}^{-1}$ by hand-to-mouth uptake, respectively. Overall, dermal absorption was 1–2 orders of magnitude larger than hand-to-mouth uptake.

Whether by dermal absorption or hand-to-mouth uptake, the estimated ADDs showed wide variations among individual congeners. Generally, the ADDs for low-chlorinated CB congeners (Table SI-3) were larger than those for high-chlorinated CBs. For example, the dermal ADDs for CB-28, CB-101 and CB-44 ranked the top three, at $974 \text{ pg kg}^{-1} \text{d}^{-1}$, $463 \text{ pg kg}^{-1} \text{d}^{-1}$ and $406 \text{ pg kg}^{-1} \text{d}^{-1}$, respectively. For some high-chlorinated CBs such as CB-183 and CB-187, the ADDs were only several $\text{pg kg}^{-1} \text{d}^{-1}$. The ADDs of CB congeners by hand-to-mouth uptake showed similar rules to dermal ADDs. In total, the ADD of low-chlorinated CB congeners (di- to penta-CBs, $\sum \text{PCB}_{16}$) via dermal/hand-to-mouth uptake could account for about 93% total dermal ADD and 86% total hand-to-mouth ADD, respectively (Table SI-3). Therefore, the major contributors to ADDs via dermal/hand-to-mouth uptake were low-chlorinated CB congeners.

Data on dermal ADDs of PCBs estimated by hand wipes were not available until 2019, mainly associated with two reports of Cao et al. (2019a, 2019b). One reported the ADD of whole-body dermal absorption of PCBs for the general population in China, which was estimated to be $6700 \text{ pg kg}^{-1} \text{d}^{-1}$ (Cao et al., 2019b). However, the permeability coefficient model was used to estimate the ADD, different from the model used in our study (relative absorption model). The estimation results from different models are not comparable because ADD estimated by the permeability coefficient model might be 1–2 orders of magnitude higher than that by the relative absorption model (Cao et al., 2019a). Therefore, if the relative absorption model had been used in the report of Cao et al. (2019b), dermal ADDs of PCBs in the study would likely be 1–2 orders of magnitude higher than those for the general population. In another report of Cao et al. (2019a), the ADD was estimated to be $14.2 \text{ ng kg}^{-1} \text{d}^{-1}$ during haze days, based on the measurement of PCBs on the forehead wipes. Notably, in the report of Cao et al. (2019a), the whole-body skin area was about 1.6 m^2 , exposure levels in covered areas were 20% of those on the forehead and exposure duration per day was 24 h. These parameters are at least 3.5-fold higher than the values used in this study, where exposed area (naked skin area) per day was 0.2215 m^2 , exposure duration per day was 10 h and covered

areas were not considered. Then, with similar estimation parameters, dermal ADDs of PCBs for e-waste workers might be close to or higher than those during haze days.

Regardless, this ADD result could further confirm that dermal exposure of e-waste workers to PCBs via dermal absorption was non-negligible. Dermal and/or hand-to-mouth uptakes of PCBs for e-waste workers were primarily associated with low-chlorinated CB congeners. Exposure to PCBs by dermal absorption was relatively more important compared to hand-to-mouth uptake.

3.3. Association between serum and hand-wipe samples

The published studies (Stapleton et al., 2012; Watkins et al., 2011) indicated that the importance of dermal exposure to chemicals could be supported by the significant relationships between hand-wipe loadings and serum levels. Therefore, the present study further investigated the relationship of PCBs between hand wipes and serum samples to examine the importance of dermal/hand-to-mouth exposure of e-waste workers to PCBs. Notably, only eleven CB congeners detected simultaneously in serum and hand-wipe samples were analyzed. Before correlation analysis, the CB loadings/levels in hand-wipe and serum samples were log-transformed due to the abnormal distribution of the dataset. The association results are listed in Table SI-2.

As shown in Table SI-2, the relationships highly depended on the individual congeners. High-chlorinated congeners such as CB-138, CB-153, CB-156, CB-170 and CB-180 showed no correlations between hand-wipe and serum samples, but significant associations were found for low-chlorinated congeners such as CB-28, CB-66, CB-74, CB-99, CB-105 and CB-118 ($p < 0.05$). This suggested that these low-chlorinated CBs in the hand-wipe samples could be linked to the body CB burden. For $\sum \text{PCB}_{11}$ (sum of eleven CB congeners detected simultaneously in serum and hand-wipe samples), significant associations between serum and hand-wipe samples were also found. Taking both CB-28 and CB-153 as representatives, the different associations between hand wipes and serum samples are plotted, respectively, in Fig. 2.

Principal component analysis (PCA) showed distinct clustering characteristics of CB congeners between serum and hand-wipe samples, as shown in Fig. SI-2. All CB congeners in hand-wipe samples clustered together, indicating that PCBs adhering to hand skin were from common sources. Compared to those in hand-wipe samples, CB congeners in serum samples were clearly more dispersed, which might indicate the presence of multiple sources of PCBs in residents living in e-waste recycling areas (Song and Li, 2014). PC1 could account for 57% of the variance, which was mainly related with CBs in hand-wipe samples and low-chlorinated CBs in serum samples. PC2 could account for 27%, which was mainly related to high-chlorinated CBs in serum samples. The PCA results clearly supported the supposition that low-chlorinated CB congeners (especially CB-28 and CB-66) in serum were more closely related to hand-wipe samples than high-chlorinated CB congeners.

Theoretically, chemicals in hand wipes may be likely to be from external microenvironments (e.g. air, dust, CB-containing products and contaminated surfaces) and/or internal exposure (redistributed to skin) (Frederiksen et al., 2020; Watkins et al., 2011). In this study, we first hypothesized that PCBs in hand wipes were primarily from circulatory redistribution to skin; thus, all congeners detected in hand wipes should be related to congeners in serum. Nevertheless, this was not the case for high-chlorinated CB congeners (e.g. CB-153 and CB-180). In addition, as shown in Tables 1 and 2, CB-60 was found frequently in the serum, but was less frequently detected in the hand wipes (detection frequency $< 60\%$), which also did not meet the hypothesis of blood-to-skin redistribution. Therefore, it appeared less likely that PCBs in hand-wipe samples were greatly affected by internal exposure, but instead were mainly from the external microenvironment.

In sum, since there were significant relationships for some CB congeners containing ≤ 5 chlorine atoms and $\sum \text{PCB}_{11}$ between hand-wipe

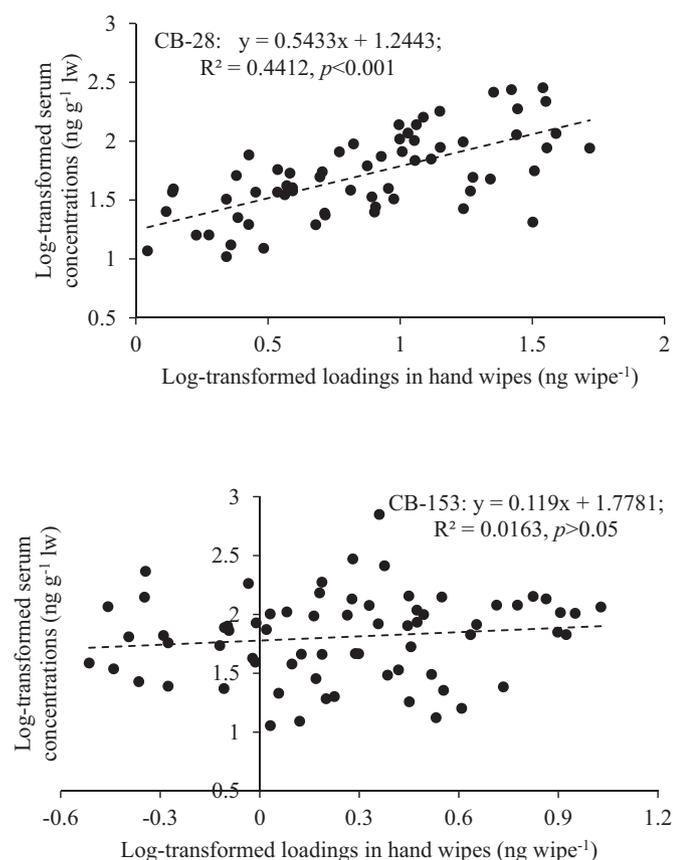


Fig. 2. Relationships of log-transformed loadings/levels of CB-28 and CB-153 between hand wipes and serum samples.

loadings and serum levels, dermal/hand-to-mouth exposure might be one of the missing links in workers' exposure to low-chlorinated CBs from external microenvironments.

3.4. Relative importance of dermal/hand-to-mouth route

3.4.1. Fraction of dermal/hand-to-mouth route among all exposure routes

The relative importance of dermal/hand-to-mouth uptake was evaluated by comparing the ADDs obtained in this study with the dietary/inhalation ADDs reported in published studies. However, ADD estimates for individual CB congeners were limited, so ADDs for \sum PCB were used for comparison. A previously estimate of ADD for e-waste workers was 1808 ng d⁻¹ by inhalation (1512 ng d⁻¹ from the gas phase and 296 ng d⁻¹ the particle phase) (Wang et al., 2016), which could be recalculated as 30,133 pg kg⁻¹ d⁻¹ when the human body weight of 60 kg was considered. Xing et al. (2010) estimated that the dietary ADD for residents in Taizhou was about 92,790 pg kg⁻¹ d⁻¹. Then, it was found that the dermal ADD obtained in the study approximately accounted for 4% of the combined ADDs of all pathways mentioned, while that for the hand-to-mouth route accounted only for 0.3%. Therefore, in terms of its relative proportion across all exposure pathways, the dermal/hand-to-mouth route did not appear to be dominant. However, the bioaccessibilities of PCBs were not considered in the inhalation estimation (Wang et al., 2016) and dietary estimation (Xing et al., 2010). Meanwhile, the dermal ADD in this study only focused on exposure at work and did not include exposure at home and outdoors. In addition, dermal absorption might also occur by contact with textiles (Cao et al., 2019a; Cao et al., 2019b; Liu et al., 2017; Weschler and Nazaroff, 2012), which was not included in our assessment estimates. Thus, the percentage contribution of dermal/hand-to-mouth uptake for e-waste workers in the current study might be underestimated.

3.4.2. Percentage contribution to body burden

The serum level could represent the overall intake of PCBs by all exposure routes, including dietary/dust hand-to-mouth ingestion, inhalation, and dermal absorption (Cherrie et al., 2006). According to Eqs. (4)–(6) in the Supplementary information, the estimated ADDs in this study could further be used to obtain the internal dose associated with dermal absorption (EID_{hw}^{da}) and hand-to-mouth uptake (EID_{hw}^{hm}). Based on Eq. (7) in the Supplementary information, their percentage contributions (Percentage_{hw}^{da} or Percentage_{hw}^{hm}) to internal dose (serum level) could be obtained. Notably, only eleven CB congeners simultaneously detected in hand-wipe and serum samples were considered, and these calculated values can be seen in Table SI-3.

The estimated EID from 11 individual congeners was 5.58 ng g⁻¹ lw for hand-to-mouth ingestion (EID_{hw}^{hm}) and 49.5 ng g⁻¹ lw for dermal absorption (EID_{hw}^{da}), accounting for 1.54% and 13.6% of the serum level of total PCBs (364 ng g⁻¹ lw), respectively. This 1.54% contribution indicated that hand-to-mouth ingestion only had a minor effect on overall body CB burden. Although the 13.6% contribution of the dermal route did not appear especially dominant, the total EID associated with dermal uptake (49.5 ng g⁻¹ lw) was more than the serum level (26.2 ng g⁻¹ lw) of the general population living in Taizhou (Lv et al., 2015).

For each specific congener, the EID associated with dermal exposure (EID_{hw}^{da}) ranged from 0.387 ng g⁻¹ lw (CB-170) to 13.7 ng g⁻¹ lw (CB-74), and that related with the hand-to-mouth ingestion (EID_{hw}^{hm}) was from 0.0350 ng g⁻¹ lw (CB-101) to 1.70 ng g⁻¹ lw (CB-28). Correspondingly, the percentage contribution (individual congeners in hand wipes *verse* in sera) by dermal absorption varied from 1.91% (CB-170) to 21.4% (CB-28), while the percentage contribution by hand-to-mouth intake ranged from 0.136% (CB-105) to 3.06% (CB-28). Notably, dermal exposure to some low-chlorinated congeners (e.g. CB-28, CB-66, and CB-74) could account for more than 15% of body burden, especially CB-28, which was up to 21.4%. The percentage contribution to body burden underlined the importance of dermal exposure to some low-chlorinated PCBs for e-waste workers.

In sum, it could be concluded from these results that dermal absorption was an important route of exposure to some low-chlorinated PCBs for e-waste workers.

3.5. Non-carcinogenic risk based on sera and hand wipes

3.5.1. Serum

For non-cancer effects of PCBs, the “critical concentration levels in humans” for total PCBs have been recommended as 700 ng g⁻¹ lw for women less than 45 years old and 1800 ng g⁻¹ lw for all others (AFSSA, 2010; Frederiksen et al., 2020). In this study, 28% of serum levels exceeded 700 ng g⁻¹ lw. Of these, one exceeded 1800 ng g⁻¹ lw, and 4 of the women <45y exceeded 700 ng g⁻¹ lw.

3.5.2. Hand wipes

The non-carcinogenic risk of individual CB congeners was assessed by hazard quotients (HQs), which were calculated based on Eq. (8) in the Supplementary information (EPA, 2011). In Eq. (8), RfD is the dermal/oral reference dose of an individual CB congener (20 ng kg⁻¹ d⁻¹) (Cao et al., 2019a). The hazard index (HI) method was used to assess the total non-carcinogenic risks from multiple congeners, which was equal to the sum of HQs for all individual congeners, as shown in Eq. (9) of the Supplementary information. As shown in Table SI-3, for e-waste workers in this study, the dermal HI was 0.25 for total PCBs and 0.23 for low-chlorinated congeners (di- to penta-CBs), respectively. For hand-to-mouth uptake, HI was 0.025 for total PCBs and 0.021 for low-chlorinated congeners (di- to penta-CBs), respectively. HIs for dermal exposure and/or hand-to-mouth were lower than the non-carcinogenic risk threshold value (HI: 1), but the dermal HI for both total PCBs and low-chlorinated congeners exceeded the potential risk level (HI ≥ 0.1).

In any case, the health risk assessment highlighted the potential non-carcinogenic risk for e-waste workers exposed to PCBs. The non-carcinogenic risk caused by dermal uptake of PCBs deserves attention.

3.6. Relationship between PCBs and THs/demographic information

The possible associations between PCB exposure and circulating levels of thyroid hormones (THs) were also determined in order to evaluate the biological effects of PCBs in humans. Both PCB levels and TH concentrations in serum were log-transformed before correlation analysis. Pearson correlation analysis showed an insignificant association between all listed CB congeners (detected frequency > 60%) and FT₄/TSH. Although FT₃ showed a significant positive association with both CB-105 ($p = 0.013$) and CB-153 ($p = 0.048$), and a marginal association with CB-138 ($p = 0.054$), these relationships disappeared (partial correlation analysis) after controlling for age, BMI and gender. The insignificant relationship observed in this study was consistent with the conclusion reached in the review of Salay and Garabrant (2009), where PCBs generally showed no clear association with FT₃/FT₄/TSH in adults.

Notably, a previous study of ours (Lv et al., 2015) revealed a significant negative association between PCB exposure and TSH levels in pregnant women residing in Wenling (the same region as this study), which was inconsistent with the result observed in this study. We speculated that the inconsistent results might be related to the different exposure characteristics of the study populations. First, since potential TH effects related with PCBs might be concentration-dependent, the 10-fold higher level of CB (364 ng g⁻¹ lw for total PCBs) in this study than that in pregnant women in the previous study (26.2 ng g⁻¹ lw for total PCBs) might result in the results between the two studies being incomparable. Secondly, in addition to PCBs, the e-waste dismantling workers may have been exposed to other potential thyroid-disrupting chemicals. If these potential TH interferents were not separated and removed, the results may be invalid. Clearly, more studies are required to elucidate the relationship between PCB exposure and thyroid homeostasis in the future.

The demographic information for workers, including age, height, body weight, BMI, body fat, gender, duration of e-waste recycling employment and residential time, is listed in Table SI-1. Relationships were examined between PCBs in serum/hand-wipe samples and demographic information, but no relationships were found between PCB loadings/levels and worker variables.

3.7. Limitations

Our research aims to investigate dermal/hand-oral exposure to PCBs in e-waste dismantling workers using a hand-wipe sampling approach. There exist some limitations to the results in this study. First, the consistency of PCB exposure levels during the same period has not been assessed. A single hand-wipe sampling does not adequately represent average PCB exposure over a period of time. Second, a recent study suggested that many factors might affect the removal efficiencies of chemicals, such as the wipe material, the number of sequential wipes, and lipophilicity of the chemicals (Beaucham et al., 2019). In this study, the hand-wipe approach was performed according to a previous work (Gong et al., 2014). The hand-wipe efficiencies were not assessed, which might result in bias when characterizing the extent of PCB exposure. In addition, several research studies (Cao et al., 2019b; Gong et al., 2014; Liu et al., 2017; Weschler and Nazaroff, 2008) indicated that chemical levels might vary in different body locations and that dermal absorption might also occur through contact with textiles. Therefore, estimating ADD based on hand wipes alone might result in certain uncertainties. Finally, the absence of a control group also contributed to the limitations in this study.

4. Conclusion

This study identified the characteristics of dermal/hand-to-mouth exposure to PCBs actually attached to the skin surface of e-waste dismantling workers and provided evidence of the importance of dermal exposure to low-chlorinated CBs for workers. Human health outcomes vary with exposure pathway (Weschler and Nazaroff, 2012), so the dermal exposure of e-waste workers requires specific attention to the toxic effects and health risks of low-chlorinated CB congeners. The PCB evaluation practice in this study can be used as a reference case for assessing dermal/hand-to-mouth exposure to other toxic chemicals present in the e-waste dismantling environment. Despite the insignificant relationship found between PCB exposure and expression of THs, the results could help in understanding the potential health effects of PCBs for e-waste workers.

CRedit authorship contribution statement

Chen Zhao: Investigation, Data curation, Formal analysis, Validation, Writing – original draft. **Ji-Fang-Tong Li:** Investigation, Methodology, Formal analysis, Validation. **Xing-Hong Li:** Conceptualization, Methodology, Formal analysis, Supervision, Resources, Funding acquisition, Project administration, Writing – review & editing. **Meng-Qi Dong:** Investigation, Formal analysis, Data curation. **Yuan-Yuan Li:** Investigation, Data curation. **Zhan-Fen Qin:** Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.149444>.

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