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Levels of polychlorinated dibenzo-p-dioxins, dibenzofurans (PCDD/Fs) and biphenyls (PCBs) in blood of informal e-waste recycling workers from Agbogbloshie, Ghana, and controls

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ABSTRACT

The formation and environmental release of highly toxic organohalogen compounds associated with informal recycling of waste electric and electronic equipment (e-waste) is a growing problem at e-waste dumps/recycling sites (EWRSs) in many developing countries worldwide. We chose a cross-sectional study design to measure the internal exposure to polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) as well as polychlorinated biphenyls (PCBs) of individuals working on one of the largest EWRSs of Africa, located at Agbogbloshie, Accra, Ghana, and in controls from a suburb of Accra without direct exposure to EWRS activities. In whole blood samples of 21 age matched male exposed individuals (mean age: 24.7 years, SD 6.0) and 21 male controls (mean age: 24.4 years, SD 5.7) 17 PCDD/F congeners were determined. Moreover three indicator PCB congeners (#138, #153 and #180) were measured in blood of 39 exposed (mean age: 27.5 years, SD 11.7) and 19 non-exposed (mean age: 26.8 years, SD 9.7) patients. Besides a health examination, biometric and demographic data, residential and occupational history, occupational exposures and working conditions were recorded using a standardized questionnaire. In the exposed group, median PCDD/F-concentrations were 6.18 pg/g lipid base WHO_{2005}-TEq (range: 2.1–42.7) and significantly higher compared to the control group with 4.60 pg/g lipid base WHO_{2005}-TEq (range: 1.6–11.6). Concentrations were different for 2,3,7,8-TetraCDD, three HexaCDD and all 10 PCDF congeners, indicating a combustion pattern. Using a multivariate regression analysis exposure to EWRS activities was the most important determinant for PCDD/F exposure. Median PCB levels for the indicator congeners #138, #153 and #180 were 0.011, 0.019 and 0.008 μg/l whole blood (ranges: 0.002–0.18, 0.003–0.16, 0.002–0.078) in the exposed group and, surprisingly, significantly higher in the controls (0.037, 0.062 and 0.022; ranges: 0.005–0.46, 0.010–0.46, 0.004–0.21). In a multivariate regression approach e-waste related activities had no positive influence on internal PCB exposure, but rather the time living in Accra. The internal PCB exposure is in particular notable for a country where PCBs have historically never been produced or used. The impact of EWRS activities on organohalogen compound exposure of individuals working at and living in the surroundings of the Agbogbloshie EWRS, and the surprisingly high PCB exposure of people living in Accra not involved in e-waste activities require further investigation.

1. Introduction

Formation of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) by thermal processes was first identified and quantified in emissions from municipal waste incinerators in 1977 (Olie et al., 1977). Intense research followed and it rapidly became obvious that PCDD/Fs and other halogenated (aromatic) compounds are formed in all combustion processes involving organic carbon, oxygen and halogens, especially in the presence of metals like copper working as catalysts (Fiedler, 1998). In most industrialized countries, significant emission sources, including cable incinerator plants, were identified and emissions were reduced both by technical and legislative measures at the end of the last century. As a result, a continuous reduction of the internal exposure to PCDD/Fs was observed in Germany and many other industrialized countries (Wittsiepe et al., 2000; Ulaszewska et al., 2011; Consonni et al., 2012).

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With the increasing production of home appliances and consumer electronics devices, such as refrigerators, washing machines, air conditioners, lighting equipment, TVs/monitors, computers, printers, photocopiers, fax machines, cell/smart phones, game consoles and batteries, in combination with the downward trend in prices and their ever shorter life span, a growing uncontrolled stream of those products at their end-of-life-time developed.

The wastes from electric and electronic equipment (WEEE or e-waste) contain relatively high amounts of valuable materials which are highly integrated into each other and therefore are difficult to separate. E-waste has been exported (sometimes illegally) as "second-hand-goods" by developed countries and informal e-waste dumps/recycling sites (EWRSs) were set up since the middle of the 1990s in developing countries in Africa and Asia, e.g. Ghana (Accra), Nigeria (Lagos), China (Guiyu in Shantou, Guangdong Province), India (Delhi) and Pakistan (Karachi) (Kuper and Hojsik, 2008; Robinson, 2009; Chi et al., 2011; Schluep et al., 2012). Informal recycling processes used can be different at EWRSs, but often include steps of manual dismantling, acid leaching, heating and wanton burning (Brigden et al., 2005, 2008; Jian et al., 2014).

The e-waste material itself can contain halogenated persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs) used as dielectric or flame-retardant plasticizers and brominated flame retardants (BFRs) like polybrominated diphenyl ethers (PBDEs), biphenyls (PBBs) and others. Moreover, the open burning of e-waste, that contain halogens, e.g. BFR containing or polyvinyl chloride (PVC) coated copper cables, provide optimal conditions for de novo formation of halogenated aromatic compounds, such as PCDD/Fs, its brominated equivalents PBDD/Fs, mixed brominated/chlorinated homologies (PXDD/Fs) and other dioxin-related compounds (DRCs) (Weber and Kuch, 2003; Hedman et al., 2005; Gullett et al., 2007; Duan et al., 2011; Hibbert and Ogunseitan, 2014). Due to uncontrolled combustion and thermal processing of e-waste, these POPs have been found in the air, bottom ash, dust, soil, water and sediment samples from EWRSs worldwide, partly in tremendous high concentrations (Li et al., 2007; Wong et al., 2007; Brigden et al., 2008; Liu et al., 2008; Ma et al., 2009a, 2009b; Wen et al., 2009, 2011; Ni et al., 2010; Tue et al., 2010b; Zhang et al., 2012b; Chan and Wong, 2013; Hu et al., 2013; Labunska et al., 2013; Ren et al., 2013; Wang et al., 2013; Hosoda et al., 2014; Xiao et al., 2014). The share of annual mass of e-waste derived PCDD/F in China alone is estimated to be in the range of several kilograms (Ni et al., 2010).

At EWRSs direct involvement in open burning without protective gear, and environmental contamination with PCDD/Fs and DRCs may result in accumulation in humans, but only very few data on internal human exposure exist (Yang et al., 2013; Tue et al., 2014). Typical pathways are direct soil/dust ingestion, inhalation of fumes and consumption of contaminated local food, especially fish. Data from Chinese, Indian and Vietnamese EWRSs including human health risk assessments have been summarized recently (Sepulveda et al., 2010; Tsydenova and Bengtsson, 2011; Zhang et al., 2012c; Chan and Wong, 2013; Tue et al., 2013; Song and Li, 2014).

At the Agbogbloshie EWRS near Accra, Ghana, the technical processes used seem to be more outmoded compared to Asian sites. Workers, often children and adolescents, working 10–12 h per day without any form of protective gear are exposed to frequent burns, cuts, and continuous inhalation of highly contaminated fumes (Brigden et al., 2008; Akormedi et al., 2013; Sthiannopkao and Wong, 2013; Jian et al., 2014). To our knowledge this is the first study on internal exposure to PCDD/Fs and PCB at an African EWRS and the first study on blood levels of PCDD/F of directly exposed EWRS workers worldwide.

In this paper, we report levels of PCDD/Fs and PCB in blood samples of workers from the Agbogbloshie EWRS in Accra, Ghana, and of controls from the surrounding area without exposure to e-waste recycling.

2. Materials and methods

2.1. Study design

We conducted a cross-sectional study at the Agbogbloshie EWRS, one of the largest EWRS in Africa, located in Accra, the capital of Ghana. About 40,000 people live in this area under the most deplorable environmental conditions and largely represent a migrant population from northern parts of Ghana.

For comparison, a second group of individuals was recruited in Kwabenya North, a suburb of Accra, which is located about 25 km north of the city center of Accra. Compared to Agbogbloshie, the environment and ambient air in Kwabenya North are relatively pristine. The population in Kwabenya North consisted predominantly of migrants who are transitioning from rural areas to an urban fringe area, mostly unskilled and of low socio-economic status, similar to the migrant group working on the e-waste dumpsite.

Details of the recruitment, study procedures and sample collection have been described in detail before (Feldt et al., 2014). A total of 75 exposed and 40 unexposed individuals were recruited in October 2011. Due to technical complexity and high costs, PCDD/F analysis was conducted in those patients with the highest risk of exposure and in age-matched controls. Among patients with a sufficient blood sample volume of at least 30 ml, participants who were directly involved in the burning of e-waste materials, who worked at the site for at least one year, and who also lived at the EWRS, were selected for PCDD/F analysis. For 21 participants of the exposed group who met these criteria, 21 participants from the control group were chosen by best-matching age (see Table 1a). After this selection, analysis of PCB was conducted in all patients with sufficient sample volume being available for testing (39 exposed subjects and 19 controls) (see Table 1b).

2.2. Laboratory analysis

The method for determination of PCDD/Fs in blood samples was described in detail before (Wittsiepe et al., 2007). In brief, the method includes: (a) extraction of blood fat with organic solvents, (b) multiple-column chromatography cleanup using modified silica gels and activated charcoal, and (c) instrumental determination with capillary gas chromatography and high-resolution mass spectrometry. Toxicity equivalent concentrations were calculated according to the WHO 2005 model (Van den Berg et al., 2006). All concentrations were adjusted on a lipid base.

Determination of PCB was performed using a recently developed method featuring on-line solid-phase extraction and large volume injection gas chromatographic high resolution mass spectrometry (Wittsiepe et al., 2014). Concentrations were calculated on a whole blood volume base. Because most values of the lower chlorinated indicator PCBs (#28, #52, #101) were below LOD or in the range of blank samples, these data were not included here.

2.3. Statistical analysis

For statistical calculations values below the limit of detection (LOD) were set to 1/2 LOD. Statistical analysis was performed using STATISTICA software system (version 10 & 12.5, Statsoft, Inc., Tulsa, www.statsoft.com). The Mann–Whitney U test was used to compare differences of between POP concentrations in the exposed and control group. In addition, we used the t-test for the same purpose with log-transformed POP concentrations.

To investigate associations between e-waste related exposure and internal POP concentrations, and to control for confounding factors, multivariate regression models were used. The covariates *years working/living at site*, *years living in Accra in general*, *age*, and *exposed/non-exposed to e-waste recycling processes* were included. The variables describing the time working/living at the EWRS and in Accra were together and

Table 1
Participants' characteristics.

Parameter	Total	Controls	Exposed	P
a) PCDD/F study group				
Number of participants	42	21	21	
Age [years]	24.5 (SD 5.8)	24.4 (SD 5.7)	24.7 (SD 6.0)	0.85
Female gender	0	0		
Weight [kg]	67.0 (SD 6.1)	66.0 (SD 5.5)	68.0 (SD 6.6)	0.30
Height [cm]	169 (SD 6.3)	168 (SD 5.1)	171 (SD 7.2)	0.13
Body mass index [kg/m^2]	23.5 (SD 2.0)	23.5 (SD 2.0)	23.4 (SD 1.9)	0.87
Working/living at site	6.4 (SD 6.1)	8.4 (SD 7.4)	4.3 (SD 3.4)	0.013
Activities within recycling process[a]				
Burning of materials			21 (100%)	
Collection			14 (67%)	
Dismantling			18 (86%)	
Ash/wire collection after burning			17 (81%)	
Lead smelting			6 (29%)	
b) PCB study group				
Number of participants	58	19	39	
Age [years]	27.3 (SD 11.0)	26.8 (SD 9.7)	27.5 (SD 11.7)	0.40
Female gender	12 (21%)	5 (26%)	7 (18%)	
Weight [kg]	67.1 (SD 11.0)	66.7 (SD 11.3)	67.2 (SD 11.0)	0.86
Height [cm]	167 (SD 8.8)	167 (SD 8.5)	167 (SD 9.0)	0.78
Body mass index [kg/m^2]	24.1 (SD 3.5)	24.0 (SD 4.8)	24.1 (SD 2.8)	0.004
Working/living at site	5.6 (SD 5.5)	7.3 (SD 7.3)	4.8 (SD 4.3)	0.005
Activities within recycling process[a]				
Burning of materials			14 (36%)	
Collection			24 (62%)	
Dismantling			16 (41%)	
Ash/wire collection after burning			16 (41%)	
Lead smelting			4 (39%)	

Data presented as means (standard deviation) or number (proportion of the respective population). P-values were calculated using unpaired, 2-sided t-test.
[a] More than one activity possible.

optionally used in the model. We used the log10-transformed concentration values as dependent variables to ensure normal distribution of residuals. The variables *years working/living at EWRS*, *years living in Accra in general* and *age* were log2-transformed, so that their associations with POP-levels were estimated for a doubling in the natural scale. Multivariate regression analysis was computed by the SAS software (version 9.3, SAS Institute, Cary, NC, USA) procedure GENMOD, which fits a generalized linear model to the data by maximum likelihood estimation. The results of these regression analyses are expressed as adjusted regression coefficients (β) together with their 95% CIs and Wald p-values.

2.4. Ethical considerations

The study was conducted in accordance with the ethical principles of the Declaration of Helsinki. Ethical clearance was obtained from the institutional review board of the Noguchi Memorial Institute for Medical Research, University of Ghana, Accra.

3. Results

Analysis of PCDD/Fs were conducted on 21 exposed and 21 unexposed age-matched participants, and of PCB in 39 exposed and 19 unexposed participants, as described above. Biometric, socio-demographic and medical parameters of participants, as well as comparison of the data, have been published in detail before (Feldt et al., 2014). Basic biometric data and information about activities within the recycling process are given in Table 1. Exposed and unexposed participants were not different in age, weight, height and body mass index (BMI).

Descriptive statistical parameters (arithmetic mean, geometric mean, 50th (median) and 95th percentile) of the blood fat content and of PCDD/F blood levels on a lipid base of the exposed individuals and controls as well as P-values of the Mann–Whitney U-test and t-test (applied to logarithmized values) of testing for differences are given in Table 2. Corresponding data for PCB on a whole blood volume base are given in Table 3.

Concentrations of 2,3,7,8-TetraCDD (#D48), 1,2,3,4,7,8-HexaCDD (#D66), 1,2,3,6,7,8-HexaCDD (#D67) and 1,2,3,7,8,9-HexaCDD (#D70) and all 2378-chlorosubstituted PCDF congeners were significantly higher in exposed individuals compared to non-exposed ones. Looking at the geometric mean or median values of the sum of PCDF [pg/g lipid base] the concentrations of the exposed group were about four times higher than those of the controls, and the P95 value was as much as about seven times higher. Median concentrations of 1,2,3,4,7,8-HexaCDF (#F118), 1,2,3,6,7,8-HexaCDF (#F121) and 2,3,4,6,7,8-HexaCDF (#F130) were 2.3, 3.0 and 4.3 times higher; those for 1,2,3,4,6,7,8-HeptaCDF (#F131) and 1,2,3,4,7,8,9-HeptaCDF (#F134) were 5.8 and 5.0 times higher (exposed group vs. controls). The shift in the congener pattern of the PCDD/F exposure between the exposed individuals and controls is shown in Fig. 1 as box-and-whisker plots presenting minima, 25th, 50th, 75th percentiles and maxima. For all congeners (except OctaCDD) and TEq, median and maximum concentrations were higher in the exposed group. It is very evident that the most clear-cut differences in internal exposure were found for PCDF.

For the PCB congeners #138, #153 and #180 [μg/l whole blood], we also found differences between EWRS workers and controls; but here, on a group basis, the concentrations found in controls were about three times higher based on geometric means or medians.

Results of the multivariate regression approach are shown in Table 4. The variables describing the exposure time (*working/living at site* and *living in Accra*) were strongly correlated. Especially in the PCB control group, some persons were living for a long time in Accra, but for a shorter time at the control site. We included both variables optionally in the models for PCDD/Fs and PCB. For PCDD/F concentrations, positive associations were observed between the exposure and time working/living at the site with strongest effects for higher chlorinated PCDF. The age of the participants, which is normally one of the most influencing

Table 2
Descriptive statistical parameters of blood fat content and PCDD/F blood levels [pg/g lipid base] in exposed individuals and controls.

PCDD/F [pg/g lipid base]	Controls (N = 21)				Exposed (N = 21)				MWU	t-Test (log)
	AM	GM	P50	P95	AM	GM	P50	P95	P*	P*
Fat content [mg/g whole blood]	3.54	3.37	3.41	5.13	3.17	3.00	3.07	5.02	0.222	0.290
2,3,7,8-TetraCDD (#D48)	0.66	0.53	0.65	1.3	1.31	0.99	0.9	3.0	**0.0260**	**0.0099**
1,2,3,7,8-PentaCDD (#D54)	1.40	1.22	1.3	2.7	3.44	2.05	1.9	12	0.0994	0.0530
1,2,3,4,7,8-HexaCDD (#D66)	0.87	0.64	0.63	1.4	1.81	1.23	1.0	5.6	**0.0099**	**0.0106**
1,2,3,6,7,8-HexaCDD (#D67)	2.72	2.42	2.4	4.4	5.96	4.45	3.5	18	**0.0052**	**0.0034**
1,2,3,7,8,9-HexaCDD (#D70)	1.91	1.30	1.4	4.1	3.48	2.41	2.4	12	**0.0228**	**0.0273**
1,2,3,4,6,7,8-HeptaCDD (#D73)	13.6	10.7	9.1	40	16.3	13.7	13	38	0.0944	0.203
OctaCDD (#D75)	105	80.7	73	270	92.2	77.7	75	250	0.950	0.845
2,3,7,8-TetraCDF (#F83)	0.65	0.44	0.55	1.7	2.23	1.52	1.4	5.2	**0.0004**	<**0.0001**
1,2,3,7,8-PentaCDF (#F94)	0.68	0.50	0.48	1.6	2.08	1.54	1.2	4.9	**0.0005**	<**0.0001**
2,3,4,7,8-PentaCDF (#F114)	3.59	3.10	3.2	7.3	10.7	6.51	4.9	33	**0.0048**	**0.0085**
1,2,3,4,7,8-HexaCDF (#F118)	2.17	1.94	2.1	3.5	8.24	5.70	4.9	27	<**0.0001**	<**0.0001**
1,2,3,6,7,8-HexaCDF (#F121)	1.91	1.68	1.8	3.5	9.70	6.33	5.4	33	<**0.0001**	<**0.0001**
2,3,4,6,7,8-HexaCDF (#F130)	0.79	0.66	0.65	1.6	4.09	2.97	2.8	12	<**0.0001**	<**0.0001**
1,2,3,7,8,9-HexaCDF (#F124)	0.35	0.24	0.215	1.0	0.63	0.48	0.50	1.4	**0.0142**	**0.0155**
1,2,3,4,6,7,8-HeptaCDF (#F131)	3.50	2.47	2.6	8.3	21.4	14.9	15	74	<**0.0001**	<**0.0001**
1,2,3,4,7,8,9-HeptaCDF (#F134)	0.59	0.36	0.28	1.7	2.44	1.40	1.4	8.0	**0.0004**	<**0.0001**
OctaCDF (#F135)	3.20	1.08	0.85	6.1	4.91	2.80	2.8	8.6	**0.0024**	**0.0082**
Sum P(4–8)CDD	127	100	89.5	337	125	105	97.2	343	0.450	0.794
Sum P(4–8)CDF	15.8	11.5	11.5	31.5	67.1	48.0	39.4	210	<**0.0001**	<**0.0001**
Sum P(4–8)CDD/F	143	114	103	419	192	158	155	569	0.0645	0.104
WHO_{2005}-TEq (PCDD/F)	4.52	4.02	4.60	7.37	12.1	8.56	6.18	36.3	**0.0020**	**0.0008**

AM = arithmetic mean; GM = geometric mean; P50 = 50th percentile (median), P95 = 95th percentile.
* P-values were calculated using Mann–Whitney U-test or t-test of logarithmized values (values ≤ 0.05 are indicated in bold).

parameters had a weaker influence with a tendency towards negative correlation. In Fig. 2, this context is visualized. The highest PCDD/F levels were found in young men (19–28 years old) working at the EWRS for 2–10 years. In this subgroup indicated by the year-annotations, a nearly linear relationship between exposure time and internal PCDD/F-exposure with an increase of the internal PCDD/F exposure of about 5 pg/g lipid base WHO_{2005}-TEq per year was observed.

PCB concentrations correlated positively with the PCDD/F levels within the group of the EWRS workers. But highest internal PCB exposure was found for individuals in the control group. Here we found a significant influence of the time the participants were living at Accra.

4. Discussion

In developed countries, a wide variety of anthropogenic sources have historically contributed to environmental PCDD/F background levels, e.g. thermal processes, medical, municipal and hazardous waste combustion processes and special industrial sources like secondary copper smelting, chlorine bleaching of pulp and paper, production of chlorinated compounds as well as chlorinated products itself. Environmental releases of PCDD/Fs have declined substantially and continuously in the last decades in most developed countries due to regulatory activities, improved emission controls, technical changes in the industrial processes and the closing of a number of facilities. This has also been confirmed and tracked by numerous biomonitoring and human biomonitoring studies (e.g. Wittsiepe et al., 2000; Lakind et al., 2009; Fang et al., 2013). In contrast, environmental PCDD/F background-levels have been so far low in developing countries, resulting in a low internal exposure (Consonni et al., 2012). This also applies to PCBs, because they have typically not been produced or used in developing countries.

Environmental pollution from EWRS and individual exposure of unprotected workers constitute an emerging problem in developing countries. During EWRS activities, PCDD/Fs are formed by burning of chlorine containing materials; and PCBs are part of e-waste itself, being used as dielectric or flame-retardant plasticizers. We investigated the levels of both contaminants in blood of individuals working in Agbogbloshie, one of the largest EWRS in Africa, and in controls from a suburb of Accra without direct exposure to e-waste. To our knowledge, this is the first study on internal exposure to PCDD/Fs and PCB at an African EWRS and the first study on blood levels of PCDD/Fs of directly exposed EWRS workers worldwide.

The above-mentioned time trend in human PCDD/F and PCB background levels and the strong influence of participants age on internal exposure levels have to be considered for comparison with literature data. In Table 5a we compare 95th percentile (P95) lipid based concentration values of our study with the most recent results on background exposure levels in Germany (Fromme et al., 2015), a recent study from South Africa (Pieters and Focant, 2014) and the comprehensive review of Consonni et al. (2012) for blood concentrations in general (non-exposed) populations of 161 studies from all continents (except of Africa and Central/South America) published in 1989–2010. The P95-concentration

Table 3
Descriptive statistical parameters of PCB blood levels [μg/l whole blood] in exposed individuals and controls.

[μg/l whole blood]	Controls (N = 19)				Exposed (N = 39)				MWU	t-Test (log)
	AM	GM	P50	P95	AM	GM	P50	P95	P*	P*
2,2′,3,4,4′,5′-HexaCB (#138)	0.086	0.035	0.037	0.46	0.025	0.012	0.011	0.087	**0.0121**	**0.0043**
2,2′,4,4′,5,5′-HexaCB (#153)	0.103	0.054	0.062	0.46	0.036	0.019	0.019	0.14	**0.0054**	**0.0026**
2,2′,3,4,4′,5,5′-HeptaCB (#180)	0.057	0.031	0.022	0.21	0.021	0.012	0.008	0.066	**0.0066**	**0.0034**

AM = arithmetic mean; GM = geometric mean; P50 = 50th percentile (median), P95 = 95th percentile.
* P-values were calculated using Mann–Whitney U-test or t-test of logarithmized values (values ≤ 0.05 are indicated in bold).

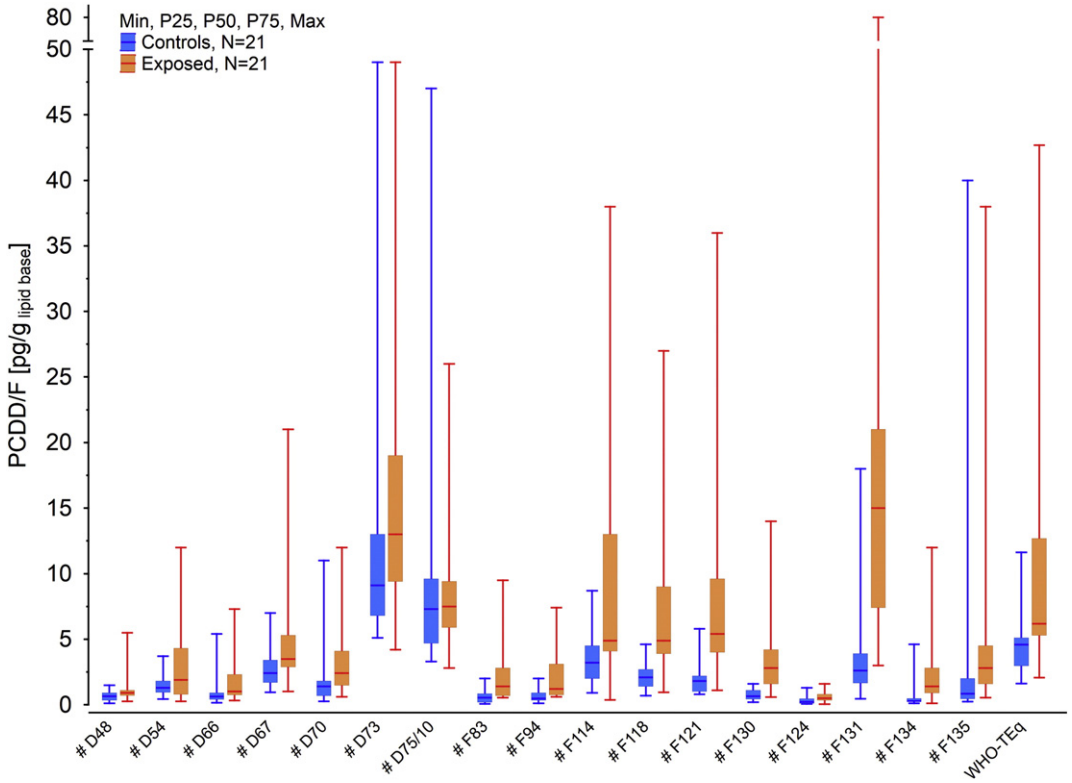

Fig. 1. Distribution (minimum, 25., 50., 75. percentile and maximum as box-and-whisker-plot) of internal exposure for different PCDD/F congeners (abbreviations explained in Table 2) and WHO_{2005}-TEq [pg/g lipid base] of workers from the Agbogbloshie EWRS and of the control group.

Table 4
Multivariate linear regression analysis of PCDD/F (a), PCB and organochlorine pesticide (b) concentrations and exposure to e-waste recycling activities, working/living time at the site (a) or time living in Accra (b) and age.

a) PCDD/F [log10, pg/g lipid base]	Exposure vs. no exposure		Working/living at site [log2, years]		Age [log2, years]	
	β (95% CI)	P*	β (95% CI)	P*	β (95% CI)	P*
2,3,7,8-TetraCDD (#D48)	0.328 (0.141–0.514)	**0.0006**	0.07 (0.005–0.135)	**0.0346**	−0.247 (−0.546–0.053)	0.1061
1,2,3,7,8-PentaCDD (#D54)	0.279 (0.062–0.496)	**0.0118**	0.07 (−0.006–0.145)	0.0714	−0.093 (−0.442–0.256)	0.6002
1,2,3,4,7,8-HexaCDD (#D66)	0.341 (0.148–0.534)	**0.0005**	0.075 (0.007–0.142)	**0.0296**	−0.283 (−0.594–0.027)	0.0736
1,2,3,6,7,8-HexaCDD (#D67)	0.323 (0.174–0.472)	<**0.0001**	0.075 (0.022–0.127)	**0.0050**	−0.275 (−0.515 to −0.035)	**0.0248**
1,2,3,7,8,9-HexaCDD (#D70)	0.325 (0.112–0.537)	**0.0028**	0.073 (−0.001–0.147)	0.0540	−0.365 (−0.707 to −0.023)	**0.0365**
1,2,3,4,6,7,8-HeptaCDD (#D73)	0.162 (0.017–0.307)	**0.0283**	0.069 (0.019–0.120)	**0.0070**	−0.296 (−0.529 to −0.063)	**0.0127**
OctaCDD (#D75)	0.024 (−0.134–0.181)	0.7701	0.051 (−0.004–0.106)	0.0673	−0.188 (−0.441–0.065)	0.1457
2,3,7,8-TetraCDF (#F83)	0.598 (0.364–0.833)	<**0.0001**	0.081 (−0.001–0.163)	0.0520	−0.163 (−0.540–0.214)	0.3970
1,2,3,7,8-PentaCDF (#F94)	0.494 (0.285–0.704)	<**0.0001**	−0.001 (−0.074–0.072)	0.9701	−0.325 (−0.662–0.012)	0.0584
2,3,4,7,8-PentaCDF (#F114)	0.399 (0.189–0.608)	**0.0002**	0.101 (0.028–0.174)	**0.0070**	−0.267 (−0.603–0.071)	0.1212
1,2,3,4,7,8-HexaCDF (#F118)	0.544 (0.389–0.699)	<**0.0001**	0.094 (0.040–0.148)	**0.0007**	−0.444 (−0.693 to −0.194)	**0.0005**
1,2,3,6,7,8-HexaCDF (#F121)	0.650 (0.484–0.816)	<**0.0001**	0.094 (0.036–0.152)	**0.0014**	−0.421 (−0.687 to −0.154)	**0.0020**
2,3,4,6,7,8-HexaCDF (#F130)	0.716 (0.550–0.882)	<**0.0001**	0.078 (0.020–0.136)	**0.0084**	−0.456 (−0.723 to −0.189)	**0.0008**
1,2,3,7,8,9-HexaCDF (#F124)	0.346 (0.146–0.546)	**0.0007**	0.062 (−0.008–0.132)	0.0812	−0.547 (−0.869 to −0.225)	**0.0009**
1,2,3,4,6,7,8-HeptaCDF (#F131)	0.854 (0.661–1.046)	<**0.0001**	0.092 (0.025–0.159)	**0.0072**	−0.468 (−0.777 to −0.158)	**0.0030**
1,2,3,4,7,8,9-HeptaCDF (#F134)	0.617 (0.371–0.862)	<**0.0001**	0.020 (−0.065–0.106)	0.6417	−0.579 (−0.974 to −0.183)	**0.0041**
OctaCDF (#F135)	0.515 (0.261–0.769)	<**0.0001**	0.131 (0.043–0.22)	**0.0037**	−0.561 (−0.969 to −0.152)	**0.0071**
Sum P(4–8)CDD	0.067 (−0.083–0.218)	0.3809	0.059 (0.006–0.111)	**0.0281**	−0.212 (−0.454–0.030)	0.0861
Sum P(4–8)CDF	0.705 (0.534–0.876)	<**0.0001**	0.105 (0.045–0.165)	**0.0006**	−0.503 (−0.778 to −0.228)	**0.0003**
Sum P(4–8)CDD/F	0.193 (0.047–0.340)	**0.0099**	0.069 (0.017–0.120)	**0.0086**	−0.297 (−0.533 to −0.061)	**0.0137**
WHO_{2005}-TEq (PCDD/F)	0.391 (0.231–0.551)	<**0.0001**	0.081 (0.025–0.136)	**0.0047**	−0.267 (−0.524 to −0.009)	**0.0424**

b) PCB [log10, µg/l whole blood]	Exposure vs. no exposure		Living at Accra [log2, years]		Age [log2, years]	
	β (95% CI)	P*	β (95% CI)	P*	β (95% CI)	P*
2,2′,3,4,4′,5′-HexaCB (#138)	−0.117 (−0.383–0.149)	0.3896	0.225 (0.117–0.333)	<**0.0001**	−0.042 (−0.270–0.188)	0.7227
2,2′,4,4′,5,5′-HexaCB (#153)	−0.148 (−0.371–0.074)	0.1914	0.156 (0.069–0.243)	**0.0004**	0.046 (−0.163–0.254)	0.6687
2,2′,3,4,4′,5,5′-HeptaCB (#180)	−0.251 (−0.470 to −0.032)	**0.0245**	0.160 (0.085–0.234)	<**0.0001**	0.186 (−0.021–0.393)	0.0786

* P-values ≤ 0.05 are indicated in bold.

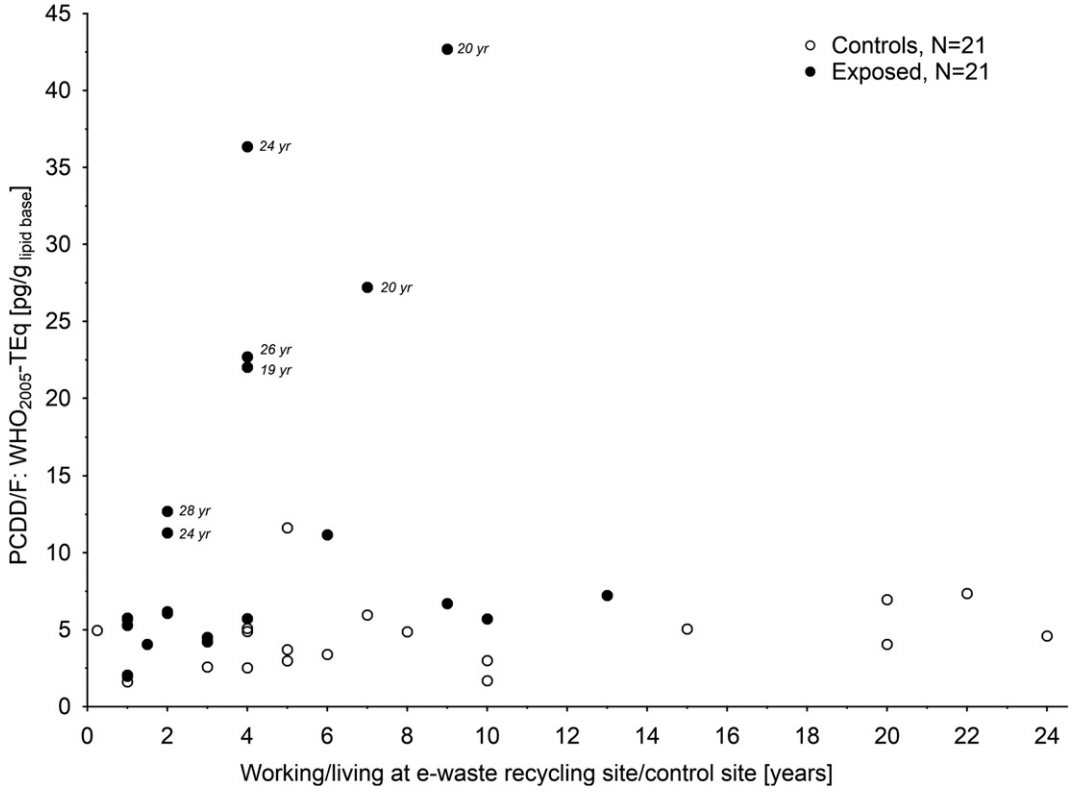

Fig. 2. Scatterplot of internal PCDD/F-exposure (WHO_{2005}-TEq (PCDD/F) [pg/g lipid base]) against time of working years at the Agbogbloshie EWRS or living years at the control site.

Table 5
Comparison of 95th percentile values of a) PCDD/F blood levels [pg/g lipid base] and b) PCB blood levels [μg/l whole blood] from this study with data on background levels from other studies.

a) PCDD/F [pg/g lipid base]	This study		Fromme et al. (2015)[a]	Pieters and Focant (2014)[b]	Consonni et al. (2012)[c]
	Controls	Exposed			
2,3,7,8-TetraCDD (#D48)	1.3	3.0	1.87	1.1	4.9
1,2,3,7,8-PentaCDD (#D54)	2.7	12	5.92	3.5	12.2
1,2,3,4,7,8-HexaCDD (#D66)	1.4	5.6	4.15		
1,2,3,6,7,8-HexaCDD (#D67)	4.4	18	22.67	Σ_{Hx} 21.4	Σ_{Hx} 68.1
1,2,3,7,8,9-HexaCDD (#D70)	4.1	12	4.73		
1,2,3,4,6,7,8-HeptaCDD (#D73)	40	38	27.30	Σ_{Hp} 17.2	Σ_{Hp} 80.0
OctaCDD (#D75)	270	250	255.59	137.1	734.4
2,3,7,8-TetraCDF (#F83)	1.7	5.2	1.33	1.3	4.2
1,2,3,7,8-PentaCDF (#F94)	1.6	4.9	1.05	1.0	4.7
2,3,4,7,8-PentaCDF (#F114)	7.3	33	19.30	4.1	20.6
1,2,3,4,7,8-HexaCDF (#F118)	3.5	27	5.12	Σ_{Hx} 5.2	Σ_{Hx} 27.0
1,2,3,6,7,8-HexaCDF (#F121)	3.5	33	5.73		
2,3,4,6,7,8-HexaCDF (#F130)	1.6	12	2.55		
1,2,3,7,8,9-HexaCDF (#F124)	1.0	1.4	1.11		
1,2,3,4,6,7,8-HeptaCDF (#F131)	8.3	74	10.26	Σ_{Hp} 2.4	Σ_{Hp} 22.7
1,2,3,4,7,8,9-HeptaCDF (#F134)	1.7	8.0	0.87		
OctaCDF (#F135)	6.1	8.6	2.79	2.1	12.6
WHO_{2005}-TEq (PCDD/F)	7.37	36.34	17.9		

b) PCB [μg/l whole blood]	This study		Wittsiepe et al. (2008)[d]	Schettgen et al. (2014)[e]
	Controls	Exposed		
2,2′,3,4,4′,5′-HexaCB (#138)	0.46	0.087	0.39	0.13
2,2′,4,4′,5,5′-HexaCB (#153)	0.46	0.14	0.76	0.19
2,2′,3,4,4′,5,5′-HeptaCB (#180)	0.21	0.066	0.76	0.15

[a] Germany, time of examination 2013, 70 (37 female) participants, age 4–76 yrs (median: 42 yrs), individual samples.
[b] South Africa, time of examination 2010, 693 adults (448 female), age 37–84 yrs, 21 pool samples.
[c] Selected statistics (weighted by number of subjects) for blood concentrations in general (non-exposed) populations in 161 studies from 26 nations analyzing individual (I) or pooled (P) blood/plasma/serum samples published in 1989–2010.
[d] Germany, time of examination 2000–2002, 232 pregnant women, age 19–42 yrs (median: 32 yrs), individual samples.
[e] Germany, time of examination 2010–2013, 157 (100 female) participants, age 18–25 yrs, individual samples, values for plasma were multiplied by a factor of 0.5 to estimate whole blood levels.

values of the individual congeners of our control group is in the range or lower than the German or South African studies, except for PCDF, where the levels in controls exceeded the levels of the South African study by a factor of 2–4. The P95-values for all congeners are lower compared to the 1989–2010 multi-study values. The P95-PCDD-values of the exposed group are in the range or exceed the corresponding ranges of most actual studies from Germany and South Africa, whereas the P95-PCDF-data exceed the reported background ranges of all studies. While making this comparison it should not be forgotten, that the mean age of our participants is about 25 and well below the average age of subjects in the other studies.

In Table 5b we compare the P95-PCB-values on a whole blood volume basis of controls and the exposed group with data on background levels from Germany (Wittsiepe et al., 2008; Schettgen et al., 2014), where the data in the latter case were originally measured in plasma and transformed to corresponding estimated whole blood values by multiplication with a factor of 0.5. Compared to the data measured by our group in 2000–2002, both groups (exposed and non-exposed) in this study are in the same or below the concentration range of the 2000–2002 data. Considering the significant reduction in background levels during the last years, the comparison with the more recent data of Schettgen et al. (2014) shows that the levels of the controls of our study exceeded the German background levels by a factor of 1.4 to 3.5, whereas the P95 values of the exposed group are below these levels.

The reason for the higher PCB levels in the control group which clearly exceeds background levels of industrialized countries like Germany is not clear. However, a general increase in PCB sources in Africa has been reviewed by Gioia et al. (2014) recently. Besides direct exposure by EWRS activities, which we do not observe in our study, secondary exposure from EWRS sites, bioaccumulated in the food chain, e.g. by consumption of contaminated fish, exposure by leakage and wrongly disposed transformers/capacitors or biomass burning might be the reason for the elevated PCB levels. Asante et al. (2011) found an increase in internal exposure to PCB via human milk analysis in Accra from 2004 to 2009 and significantly higher PCB exposure in more industrialized areas of Ghana. Luzardo et al. (2014) determined the serum PCB levels of recent immigrants from Sub-Saharan countries to the Canary Islands. For 106 Ghanaian immigrants they measured median levels of 0.67 μg/l for the sum of marker PCBs #28, 52, 101, 118, 138, 153, 180 (range 0.005–1.44; values for serum were multiplied by a factor of 0.5 to estimate whole blood levels). Compared to this study, values of the controls in our study were in a similar range or slightly lower. In general, they found significant associations between the country-specific implementation of information and communication technologies and the import volume of second-hand electronic equipment into the countries and the PCB levels of the immigrants. Further studies are necessary to explore the main route(s) of human exposure to PCB in Ghana and to find the reasons for these general observations and to explain our findings.

A cross sectional study design is not suitable to establish a causal relationship between internal exposure and EWRS activities. We found statistically significant differences between the age-matched cohorts of exposed and controls. In a multivariate statistical analysis, we found a clear impact of the exposure source and exposure time on the internal PCDD/F exposure. Furthermore, for a subgroup of workers, a nearly linear relationship between the time working at the Agbogbloshie EWRS and their PCDD/F blood levels was observed. The negative effect of age on PCDD/F-levels observed in this model might have been be a result of the age structure of the EWRS workers, where the youngest people are often performing the dirtiest jobs with the highest exposure risks.

This is particularly alarming, since prenatal and early-life exposure to PCDD/F has been associated to have negative effects on intrauterine and childhood growth, neurodevelopment, respiratory and immune function, in addition to carcinogenic potential (Lee et al., 2007; Burns et al., 2011; Gascon et al., 2013; Kishi et al., 2013; Ten Tusscher et al., 2014).

Only very few data on environmental PCDD/F levels from Ghana and the Agbogbloshie EWRS are available. Brigden et al. (2008) reports relatively low PCDD/F levels of 31 pg TEq/g in ash contaminated soil at the Agbogbloshie market and much higher levels of about 1 ng TEq/g in sediments from the adjacent shallow lagoon. Both samples show a similar congener profile, consistent with combustion residues from other EWRS sites. The 2,3,7,8-congener profile found at EWRS is characteristic: PCDD concentrations increase with the grade of chlorination and for PCDF the highest levels were found for 1,2,3,4,6,7,8-HeptaCDF, and Penta/HexaCDF (Ma et al., 2008; Zhu et al., 2008). The congener's different bioavailability and half-life times in humans lead to a typical internal exposure pattern, as it has been found in blood samples of other hot spots like the surrounding of a cable pyrolysis plant (Wuthe et al., 1992).

There are only few studies investigating the influence of EWRS activities on the internal PCDD/F exposure. Chan et al. (2007) measured PCDD/F-levels in 5 human milk samples of women at the Taizhou (TZ) EWRS (Zhejiang Province, China) and a control site. The levels at TZ were 21.0 ± 13.8 pg/g lipid base WHO_{1998}-TEq and about 2-fold higher (but statistically not significant) compared to controls. The PCDD/F pattern in the milk sample of TZ was similar to that found in blood of our EWRS workers with the concentrations being in a comparable range. At two Vietnamese EWRSs, Tue et al. (2014) found higher PCDF (but not PCDD or dioxin-like PCB on a TEq-basis) levels in 9 milk samples of exposed women compared to a control group. All concentrations were considerably lower than in this study.

We observed no association between PCB exposure and direct EWRS activities. A human biomonitoring study conducted at the Bangalore EWRS (South India, 25 participants), including a control group from Chidambaram near the coast (20 participants), gave similar results (Eguchi et al., 2012). Although the levels were lower (median: PCB #138: 7.9, PCB #153: 3.4, PCB #180: 2.8 pg/g serum wet weight), again a significant direct influence of EWRS activities on internal PCB exposure was not observed. Their results indicate consumption of fish as the major PCB pathway. The same results were already found by Tue et al. (2010a) in human milk samples from three EWRS sites in Vietnam (20 participants), and by Bi et al. (2007) in human blood samples of 26 inhabitants and by Xing et al. (2009) in 19 human milk samples of Guiyu (Shantou, Guangdong Province, China): EWRS activities had no direct influence on the internal PCB exposure. On the other hand, at the Tianjin EWRS in Northern China, Yang et al. (2013) found statistically significant higher PCB serum levels in 35 exposed people (17 dismantling workers and 18 residents) compared to 21 controls, but dismantling workers had no higher PCB than the residents.

At Agbogbloshie and most other EWRS sites, the workers have a low living standard and a low fish consumption can be assumed. These circumstances might be the reason, that in most case–control-studies the internal PCB exposure of the controls is often in the same range or even higher (as in our study) compared to the exposed ones.

5. Conclusions

Overall, our data indicate a direct exposure of workers at the Agbogbloshie ERWS, Accra, Ghana, to PCDD and especially to PCDF as combustion products. PCDD/F blood levels were related to the time participants were working at the EWRS. We found a relatively high PCB exposure in the control group, which might be caused by nutritional factors, for example higher consumption of contaminated fish. Often, and in Agbogbloshie quite naturally, children and adolescents are performing the most tedious jobs at the EWRS. They were not included in our study due to ethical considerations, but it can be assumed, that exposure levels of children are higher compared to adults (Shen et al., 2010; Xu et al., 2014).

It is estimated that more than 49 million tons of e-waste were generated worldwide in 2012, increasing to 65.4 million tons in 2017 (Robinson, 2009; StEP initiative — Solving the e-waste problem,

2014). But, although the global e-waste problem in general and the import to and informal recycling in developing countries have been recognized for some years now, its size and complexity are increasing at a much faster rate than the efficacy of strategies to contain it (Zhang et al., 2012a; Li et al., 2013; Man et al., 2013; Premalatha et al., 2014). As shown in this study, human exposure of toxic substances contained in or formed by informal recycling of e-waste is one important aspect to be addressed.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2015.03.008.

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