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High levels of PAH-metabolites in urine of e-waste recycling workers from Agbogbloshie, Ghana



Torsten Feldt^{a,b,*}, Julius N. Fobil^c, Jürgen Wittsiepe^d, Michael Wilhelm^d, Holger Till^e, Alexander Zoufaly^f, Gerd Burchard^{a,f}, Thomas Göen^g

^a Clinical Research Unit, Bernhard Nocht Institute for Tropical Medicine, Bernhard-Nocht Str. 74, 20359 Hamburg, Germany

^b Department of Gastroenterology, Hepatology and Infectious Diseases, University Hospital Düsseldorf, Moorenstr. 5, 40225 Düsseldorf, Germany

^c Department of Biological, Environmental & Occupational Health Sciences, School of Public Health, University of Ghana, P.O. Box LG13, Legon, Ghana

^d Department of Hygiene, Social and Environmental Medicine, Ruhr-University Bochum, Universitätsstr. 150, 44801 Bochum, Germany

^e GIZ – Regional Coordination Unit for HIV & TB (GIZ-ReCHT), 32 Cantonment Crescent, Cantonments, Accra, Ghana

^f Department of Medicine, Section Infectious Diseases and Tropical Medicine, University Medical Center Hamburg-Eppendorf, Martinistr. 52, 20246 Hamburg, Germany

^g Institute and Outpatient Clinic of Occupational, Social and Environmental Medicine, University of Erlangen-Nuremberg, Schillerstr. 25/29, 91054 Erlangen, Germany

HIGHLIGHTS

- Urinary PAH levels were assessed in e-waste recycling workers and controls in Ghana.
- The PAH exposure of the general population was higher than in developed countries.
- Informal e-waste recycling was associated with increased individual PAH exposure.
- Respiratory symptoms were frequent in persons involved in e-waste recycling.

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ABSTRACT

The informal recycling of electronic waste (e-waste) is an emerging source of environmental pollution in Africa. Among other toxins, polycyclic aromatic hydrocarbons (PAHs) are a major health concern for exposed individuals. In a cross-sectional study, the levels of PAH metabolites in the urine of individuals working on one of the largest e-waste recycling sites of Africa, and in controls from a suburb of Accra without direct exposure to e-waste recycling activities, were investigated. Socioeconomic data, basic health data and urine samples were collected from 72 exposed individuals and 40 controls. In the urine samples, concentrations of the hydroxylated PAH metabolites (OH-PAH) 1-hydroxyphenanthrene (1-OH-phenanthrene), the sum of 2- and 9-hydroxyphenanthrene (2-/9-OH-phenanthrene), 3-hydroxyphenanthrene (3-OH-phenanthrene), 4-hydroxyphenanthrene (4-OH-phenanthrene) and 1-hydroxypyrene (1-OH-pyrene), as well as cotinine and creatinine, were determined. In the exposed group, median urinary concentrations were 0.85 µg/g creatinine for 1-OH-phenanthrene, 0.54 µg/g creatinine for 2-/9-OH-phenanthrene, 0.99 µg/g creatinine for 3-OH-phenanthrene, 0.22 µg/g creatinine for 4-OH-phenanthrene, and 1.33 µg/g creatinine for 1-OH-pyrene, all being significantly higher compared to the control group (0.55, 0.37, 0.63, 0.11 and 0.54 µg/g creatinine, respectively). Using a multivariate linear regression analysis including sex, cotinine and tobacco smoking as covariates, exposure to e-waste recycling activities was the most important determinant for PAH exposure. On physical examination, pathological findings were rare, but about two thirds of exposed individuals complained about cough, and one quarter about chest pain. In conclusion, we observed significantly higher urinary PAH metabolite concentrations in individuals who were exposed to e-waste recycling compared to controls who were not exposed to e-waste recycling activities. The impact of e-waste recycling on exposure to environmental toxins and health of individuals living in the surroundings of e-waste recycling sites warrant further investigation.

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* Corresponding author at: Clinical Research Unit, Bernhard Nocht Institute for Tropical Medicine, Bernhard-Nocht Str. 74, 20359 Hamburg, Germany. Tel.: +49 211 8108721; fax: +49 211 8116294.

E-mail addresses: feldt@bni-hamburg.de, torsten.feldt@med.uni-duesseldorf.de (T. Feldt), jfobil@ug.edu.gh (J.N. Fobil), wilhelm@hygiene.rub.de (M. Wilhelm), holger.till@giz.de (H. Till), gerd.burchard@bni-hamburg.de (G. Burchard), thomas.goen@ipasum.med.uni-erlangen.de (T. Göen).

¹ Present address: Department of Gastroenterology, Hepatology and Infectious Diseases, University Hospital Düsseldorf, Moorenstr. 5, 40225 Düsseldorf, Germany.

1. Introduction

It is estimated that 20–50 million tons of electronic waste (e-waste) accrue annually worldwide, mainly in industrialized countries (UNEP, 2005; Wong et al., 2007). A high proportion of electronic products generated in developed countries end-up as obsolete or semi-spent materials in resource-poor countries (Robinson, 2009; Schmidt, 2006). Informal recycling of e-waste in West Africa is an emerging problem due to the rapidly growing use of electric and electronic equipment (EEE) in Africa, and particularly due to imports from industrialized countries. West Africa serves as the major trading route of used EEE into the African continent, with Ghana and Nigeria being the main import hubs. In Ghana alone, it is estimated that 215,000 tons of new and used EEE were imported, and 129,000 tons of e-waste were generated in 2009 (E-Waste Africa Programme, 2011).

The recycling methods include melting of electronic boards on open fires in order to recover metals and valuable chips, burning cable wires to extract copper, and finally open burning of residual valueless materials. Most of the processes are also performed by children and adolescents particularly. This mode of recycling results in the emission of a multitude of toxins in high concentrations, exposing e-waste workers and communities.

Besides a number of heavy metals and a variety of other organic toxins like polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), it was shown that polycyclic aromatic hydrocarbons (PAHs) represent an important emission of informal e-waste recycling (Asante et al., 2012; Brigden et al., 2008; Ma et al., 2009; J. Wang et al., 2012; Wang et al., 2010; Yu et al., 2006).

Unprotected workers at these sites are exposed to PAH-containing dust and gaseous fumes via inhalation, ingestion and dermal contact. It was also shown, that PAH contamination extends to the surroundings of e-waste recycling areas (Tang et al., 2010; Y. Wang et al., 2012). Those PAH emissions represent a health hazard for e-waste workers and residents living on or near e-waste recycling areas, since several PAH metabolites have been recognized as carcinogenic (IARC, 2012). The association between high PAH exposure and the risk of lung cancer has been well established (Mastrangelo et al., 1996), and e-waste recycling activities in southern China have been associated to an increased inhalation cancer risk (J. Wang et al., 2012).

Most available data on environmental pollution associated to e-waste recycling originate from China and India. Compared to those countries, being commonly associated to e-waste recycling, the technical processes used in Ghana seem to be more primitive (Bridgen et al., 2008). To our knowledge, no data on PAH exposure associated to e-waste recycling activities has been reported from Africa, where e-waste recycling activities are an emerging environmental problem.

In this paper, we report PAH-metabolite levels in the urine of e-waste workers at one of the largest e-waste dumps in Africa, and in controls from the periphery of this site without exposure to e-waste recycling. The assessment of the individual PAH exposure was conducted by the determination of 1-hydroxypyrene and several monohydroxyphenanthrenes in urine, which were proved to be reliable biomonitoring parameters for human PAH exposure in several former studies (Heudorf and Angerer, 2001; Kuusimäki et al., 2004; Hemat et al., 2012).

2. Material and methods

2.1. Study areas

A cross-sectional study was conducted at the Agbogbloshie e-waste dumpsite, one of the largest informal recycling sites for e-waste in Africa, located in Accra, the capital of Ghana. About 40,000 people live and work in this highly polluted environment, permanently laden with dense smokes from the burning of e-wastes. Used electrical equipment, such as obsolete computers, refrigerators and old television sets, are manually

dismantled at numerous small workshops, and plastic materials, including coated wires and cables, are burned using scattered open fires in order to retrieve valuable metals. Individuals recruited at Kwabenya North, a suburb of Accra which is located about 25 km north of the city center of Accra, served as control group. No e-waste recycling site or heavy industry is located in the surroundings of the control site, economic activities in the area comprise mostly of commerce and a surface stone quarry. The control site at the very periphery of Accra was chosen in order to minimize a possible exposure to emissions from the e-waste site.

2.2. Recruitment

Approaching e-waste workers and their community is a sensitive issue. Starting several weeks before recruitment, preparatory community entry activities were conducted at both sites in order to inform and familiarize the communities with the study team, purpose and procedures. Inclusion criteria comprised age ≥ 18 years and, for the exposed group, occupation in the e-waste recycling process or on the recycling site. Most workers were involved in several different activities within the recycling process and we did not select participants on the basis of performed activities. Exclusion criteria comprised evidence of severe anemia according to clinical examination for both groups and, for the control group, previous stay at, or residence ≤ 3 km from the e-waste recycling site. Recruitment was conducted during 3 days in October 2011, using a mobile study office installed in an open canopy.

2.3. Study procedures

Persons eligible for study participation were approached by the study team at both sites. The participants interrupted their work for the study procedures, which were completed within approximately one hour on the e-waste recycling site. The study objectives and procedures were explained in English and in the local languages. After giving written informed consent, participants were asked to fill a questionnaire with socio-demographic and medical information with the assistance of a field worker, who translated questions into the local language if necessary. A qualified study physician conducted a standardized interview, recorded biometric data, medical history and performed a short physical examination according to a standard protocol. Participants were asked to provide a urine sample. In addition, field blank samples were taken. All individuals willing to participate were included if the inclusion criteria were met, until the intended sample size of 40 exposed individuals and 40 controls with complete datasets and samples, comprising blood samples for the analysis of other toxins within a different study project, was reached. Due to a low success rate in blood sampling, a total of 75 exposed individuals had to be recruited in order to reach the intended sample size of 40 individuals with complete samples. Urine samples were frozen at -20 °C and shipped to Germany on dry ice after completion of the recruitment, where they were stored at -20 °C until analysis.

2.4. Laboratory analyses

2.4.1. PAH metabolites in urine

The determination of 1-, 2-, 3-, 4- and 9-hydroxyphenanthrenes (1-, 2-, 3-, 4- and 9-OH-phenanthrenes) as well as 1-hydroxypyrene (1-OH-pyrene) was carried out using a modified method proofed and published by the German Research Foundation (DFG) (Lintelmann, 1999). In brief, the hydroxylated PAHs were extracted from the urine after enzymatic hydrolysis by a solid phase extraction process, which was coupled online with high performance liquid chromatography (HPLC) and fluorescence detection (Hemat et al., 2012). The different hydroxylated PAHs were clearly separated by HPLC with the exception of 2- and 9-OH-phenanthrene, which co-elute from the analytical column, and were quantified as sum. The limits of quantification (LOQ) were

16 ng/L urine for 1-OH-phenanthrene, 4 ng/L for 2-/9-OH-phenanthrene, 5 ng/L for 3-OH-phenanthrene, 8 ng/L for 4-OH-phenanthrene and 12 ng/L for 1-OH-pyrene. Aliquots of quality control material prepared in human urine were analyzed in each series to verify the comparability of the analyses. Accuracy was proofed for 1-OH-pyrene by the successful participation in the proficiency test of the German External Quality Assessment Scheme (G-EQUAS) (Göen et al., 2012).

Urinary concentrations of PAH metabolites were expressed standard-ized by the relation to the urinary creatinine content ($\mu\text{g/g}$ creatinine), and as molar sum of phenantrenes and pyrene per mole creatinine ($\mu\text{mol/mol}$ creatinine).

2.4.2. Cotinine in urine

Cotinine in urine was determined using a gas chromatography mass spectrometry procedure proofed and published by the DFG working group (Müller, 2003). In brief, cotinine was extracted from the urine using dichloromethane and quantified after gas chromatographic separation by mass spectrometry in single ion monitoring mode (Eckert et al., 2011). Deuterated cotinine was added to the urine before sample preparation as an internal standard. Calibration was performed using standard solutions of cotinine which were prepared in non-smoker urine and were treated in the same manner as the samples. The LOQ of cotinine was 1 $\mu\text{g/L}$. Quality control was performed by analyzing aliquots of control material in each series and accuracy was proofed by the successful participation in G-EQUAS too. Urinary cotinine levels were used to estimate tobacco smoke exposure; concentrations of less than 50 $\mu\text{g/L}$ were interpreted as no recent active smoking.

2.4.3. Creatinine in urine and serum

Creatinine in urine was determined photometrical as picrate according to the Jaffé method (Larsen, 1972). Quality control was performed by analyzing aliquots of control material in each series and accuracy was proofed by the successful participation in G-EQUAS. Serum creatinine levels were determined in a commercial clinical laboratory with the compensated Jaffé method. Estimated glomerular filtration rate (eGFR) was calculated using the Cockcroft-Gault equation (Cockcroft and Gault, 1976).

2.5. Statistical analysis

Statistical analysis was performed with STATA v12.1 software (STATA Corp LP, Texas, USA). A Mann–Whitney U test was used to compare PAH urine concentrations between the study groups. We used unpaired two-sided t-test to compare continuous normally distributed biometric variables, and chi-square test to compare categorical variables between the study groups. A linear regression analysis, including the logarithm of urinary PAH metabolite concentrations as dependent variable and exposure to e-waste, sex, and tobacco smoking (vs. no current tobacco smoking, defined as urinary cotinine levels $<50 \mu\text{g/L}$) as independent variables, was performed. All tests were 2-sided and p-values of less than 0.05 were considered statistically significant.

2.6. 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

3.1. Biometric, socio-demographic and medical parameters

We recruited 75 participants working at the Agbogbloshie e-waste recycling site (exposed group), and 42 controls. Biometric, socio-demographic and medical parameters of participants are shown in

Table 1. Overall mean age was 25.8 years, and 18% were female. None of the women reported a current pregnancy. The exposed participants worked at the Agbogbloshie site since 3.9 years. The vast majority (90.7%) of exposed participants also resided at the recycling site, and almost all (98.6%) reported that they moved to Accra in order to find work. Most exposed participants were involved in several activities within the e-waste recycling process, 36% were involved in burning of materials, 61% in the collection of e-waste, 48% in dismantling, 45% in ash collection and 44% in other activities on the recycling area. Exposed and control groups did not differ in age (26.1 vs. 25.2 years, $p = 0.59$) and gender distribution (female gender 17.3 vs. 19%, $p = 0.82$). Most common occupations of non-exposed individuals were driver (31.0%), quarry worker (14.3%) or trader (11.9%). Compared to controls, exposed participants were more likely to lack formal education (33.8% vs. 2.4%, $p = 0.001$), to be Muslims (88% vs. 7.1%, $p < 0.001$) and to be married (44% vs. 19.5%, $p = 0.02$). More exposed participants reported that they usually take their meals in informal restaurants (chop bars) compared to controls (94.7% vs. 69%, $p < 0.001$). There was no significant difference in the number of meals comprising fish (5.4 vs. 6.7, $p = 0.182$) or meat (6.6 vs. 6.2, $p = 0.657$) weekly between exposed participants and the control group. Exposed participants and controls were not different in height, weight and body mass index (BMI), but exposed participants had a significantly lower diastolic (74.7 vs. 78.6 mm Hg, $p = 0.007$) and systolic (119.5 vs. 125.1 mm Hg, $p = 0.03$) blood pressure. When being asked about medical complaints in the past 4 weeks before recruitment, exposed participants reported significantly more cough (64% vs. 9.5%, $p < 0.001$), chest pain (25.3% vs. 0%, $p < 0.001$), and dizziness or vertigo (16% vs. 0%, $p = 0.006$) compared to the control group. They also took pharmaceutical herbal preparations more frequently (51.4% vs. 20%, $p = 0.001$). On clinical examination, abnormalities were rarely found; only one exposed participant had skin changes (data not shown). Serum creatinine concentrations and glomerular filtration rate (GFR) were within the normal range in all study participants except in one control participant. There was no significant difference in serum creatinine concentration or eGFR between the study groups.

3.2. PAH, creatinine and cotinine urine concentrations

Urine samples were obtained from 72 exposed and 40 control participants. PAH metabolite concentrations were calculated on creatinine base in order to adjust for hydration status of participants. The concentrations of PAH metabolites, creatinine and cotinine, and comparisons between the groups are summarized in Table 2. The concentrations of all measured PAH metabolites were significantly higher in exposed individuals compared to non-exposed individuals. The urine concentration of 1-OH-phenanthrene was 0.85 vs. 0.55 $\mu\text{g/g}$ creatinine ($p < 0.001$), of 2-/9-OH-phenanthrene 0.55 vs. 0.37 $\mu\text{g/g}$ creatinine ($p = 0.005$), of 3-OH-phenanthrene 0.99 vs. 0.63 $\mu\text{g/g}$ creatinine ($p < 0.001$), of 4-OH-phenanthrene 0.22 vs. 0.11 $\mu\text{g/g}$ creatinine ($p < 0.001$), and of 1-OH-pyrene 1.33 vs. 0.54 $\mu\text{g/g}$ creatinine ($p < 0.001$).

The molar sum of phenantrenes was 1.57 vs. 1.08 $\mu\text{mol/mol}$ creatinine ($p < 0.001$) and the molar sum of phenantrenes and 1-OH-pyrene, as a surrogate for total PAH exposure, was 2.29 vs. 1.39 $\mu\text{mol/mol}$ creatinine ($p < 0.001$).

Defining tobacco smoking status as having cotinine urine levels of $>50 \mu\text{g/L}$, overall 20 individuals (17.9%) were smokers, 16 (22.2%) in the exposed group, and 4 (10%) in the non-exposed group ($p = 0.106$). Analyzing only non-smokers, the levels of all measured PAH metabolites were significantly higher in exposed compared to control participants, confirming the results of the overall analysis, as shown in Table 3.

A significant proportion of control participants reported driving as occupation. In most cases, no specification of driving activities was recorded. When we compared urinary PAH metabolite concentrations of control participants who worked as drivers to those with other occupations, we did not find any significant differences for any of the

Table 1
Participants' characteristics, socioeconomic, biometric and medical parameters.

Parameter (n = available datasets)	Total (n = 117)	Controls (n = 42)	Exposed (n = 75)	p
Age, years (n = 117)	25.8 (SD 8.8)	25.2 (SD 7.4)	26.1 (SD 9.6)	0.59
Female Gender (n = 117)	21 (18.0%)	8 (19.0%)	13 (17.3%)	0.82
<i>Socioeconomic parameters</i>				
Marital status (n = 116)				0.02
Married	41 (35.3%)	8 (19.5%)	33 (44%)	
Single	74 (63.8%)	33 (80.5%)	41 (56.7%)	
Divorced	1 (0.9%)	0	1 (1.3%)	
Education level (n = 115)				0.001
Primary education	68 (59.1%)	29 (70.7%)	39 (52.7%)	
Secondary education	20 (17.4%)	11 (26.8%)	9 (12.2%)	
Tertiary education	1 (0.9%)	0	1 (1.4%)	
No formal education	26 (22.6%)	1 (2.4%)	25 (33.8%)	
Able to speak and understand English (n = 108)	75 (69.4%)	34 (89.5%)	41 (58.6%)	0.001
Able to read and write English (n = 108)	62 (57.4%)	28 (73.7%)	34 (48.6%)	0.01
Religion (n = 117)				<0.001
Christian	45 (38.5%)	36 (85.7%)	9 (12%)	
Islam	69 (59%)	3 (7.1%)	66 (88%)	
Other	3 (2.6%)	3 (7.1%)	0	
Electricity in household (n = 117)	95 (81.2%)	28 (66.7%)	67 (89.3%)	0.003
Car in household (n = 117)	37 (31.6%)	17 (40.5%)	20 (26.7%)	0.12
Television in household (n = 117)	72 (61.5%)	26 (61.9%)	46 (61.3%)	0.95
Refrigerator in household (n = 117)	28 (23.9%)	14 (33.3%)	14 (18.7%)	0.07
Piped water in household (n = 117)	40 (34.2%)	7 (16.7%)	33 (44%)	0.003
Weekly income, USD (n = 114)	46.7 (SD 65.6)	57.0 (SD 93.4)	39.0 (SD 41.3)	0.24
Weekly alcohol consumption, units (n = 116)	0.6 (SD 2.4)	1.5 (SD 3.7)	0.1 (SD 0.6)	0.003
Meals usually taken at chop bars (n = 117)	100 (85.5%)	29 (69.0%)	71 (94.7%)	<0.001
Number of meals with fish weekly (n = 113)	5.8 (5.2)	6.7 (4.6)	5.4 (5.4)	0.18
Number of meals with meat weekly (n = 110)	6.5 (5.1)	6.2 (5.4)	6.6 (5.0)	0.66
Tobacco smoking ^a (n = 112)	20 (17.9%)	4 (10%)	16 (22.2%)	0.11
Worry about occupational risks (n = 113)	99 (87.6%)	27 (69.2%)	72 (97.3%)	<0.001
Usage of protection measures (n = 114)	17 (14.5%)	8/40 (20%)	9/74 (12.2%)	0.26
<i>Characterization of e-waste work (n = 75)</i>				
Residence at Agbogbloshie site			68 (90.7%)	–
Time working at Agbogbloshie, years			3.9 (SD 4)	–
Activities within recycling process ^b				
Burning of materials			27 (36%)	–
Collection			46 (61.3%)	–
Dismantling			36 (48%)	–
Ash/wire collection after burning			34 (45.3%)	–
Lead smelting			11 (14.7%)	–
<i>Biometric data and vital parameters</i>				
Weight, kg (n = 117)	67.3 (SD 10.1)	66.5 (SD 9.1)	67.8 (SD 10.6)	0.51
Height, cm (n = 116)	167.3 (SD 8.1)	166.9 (SD 7.7)	167.5 (SD 8.4)	0.68
Body mass index (n = 116)	24.1 (SD 3.3)	23.9 (SD 3.4)	24.1 (SD 3.3)	0.75
Systolic blood pressure, mm Hg (n = 113)	121.6 (SD 13.2)	125.1 (SD 13.7)	119.5 (SD 12.5)	0.03
Diastolic blood pressure, mm Hg (n = 112)	76.2 (SD 7.4)	78.6 (SD 7.4)	74.7 (SD 7.0)	0.007
Pulse, beats per minute (n = 111)	76.7 (SD 3.8)	76.5 (SD 4.4)	76.8 (SD 3.4)	0.66
<i>Symptoms (past 4 weeks)</i>				
Cough (n = 117)	52 (44.4%)	4 (9.5%)	48 (64%)	<0.001
Shortness of breath (n = 117)	1 (0.9%)	0	1 (1.3%)	0.45
Chest pain (n = 117)	19 (16.2%)	0	19 (25.3%)	<0.001
Fever (n = 117)	3 (2.6%)	2 (4.8%)	1 (1.3%)	0.26
Abdominal pain (n = 117)	18 (15.4%)	5 (11.9%)	13 (17.3%)	0.43
Nausea/vomiting (n = 117)	2 (1.7%)	1 (2.4%)	1 (1.3%)	0.68
Diarrhea (n = 116)	0	0	0	–
Skin eruptions/lesions (n = 117)	7 (6%)	2 (4.8%)	5 (6.7%)	0.68
Headaches (n = 117)	47 (40.2%)	12 (28.6%)	35 (46.7%)	0.06
Dizziness/vertigo (n = 117)	12 (10.3%)	0	12 (16%)	0.006
Loss of sensation/numbness in arms/legs (n = 117)	0	0	0	–
Concentration difficulties (n = 117)	1 (0.9%)	1 (2.4%)	0	0.18
Sleeping difficulties (n = 116)	12 (10.3%)	2 (4.8%)	10 (13.5%)	0.14
Other health problem (n = 117)	11 (9.4%)	3 (7.1%)	8 (10.7%)	0.53
Taking regular medication (n = 114)	45 (39.5%)	12 (28.6%)	33 (45.8%)	0.07
Taking herbal preparations (n = 110)	44 (40.0%)	8 (20%)	36 (51.4%)	0.001

Data presented as mean (standard deviation) or number (proportion of the respective population). p-Values were calculated using unpaired, 2-sided t-test or chi-square test for categorical variables.

^a Status "Tobacco smoking" defined as urinary cotinine concentration of >50 µg/mL.

^b More than one activity possible.

Table 2

Descriptive statistical parameters of PAH-metabolite, creatinine and cotinine urine concentrations and renal function parameters in exposed individuals and controls.

PAH metabolite, urine concentration	Controls (n = 40)		Exposed (n = 72)		p
	Median	IQR [p25–p75]	Median	IQR [p25–p75]	
1-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.55	0.30–0.73	0.85	0.54–1.24	<0.001
2-/9-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.37	0.26–0.59	0.55	0.37–0.89	0.005
3-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.63	0.40–0.80	0.99	0.69–1.30	<0.001
4-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.11	0.07–0.16	0.22	0.17–0.34	<0.001
1-OH-pyrene [$\mu\text{g/g}$ creatinine]	0.54	0.29–0.80	1.33	0.78–2.52	<0.001
Molar sum of phenantrenes [$\mu\text{mol/mol}$ creatinine]	1.08	0.60–1.34	1.57	1.10–2.20	<0.001
Molar sum of phenantrenes and 1-OH-pyrene [$\mu\text{mol/mol}$ creatinine]	1.39	0.74–1.74	2.29	1.69–3.55	<0.001
<i>Other parameters</i>					
Cotinine [$\mu\text{g/l}$]	1.60	1.37–2.13	3.25	2.13–14.10	<0.001
Creatinine, urine [g/l]	1.71	1.24–2.19	1.25	0.78–1.85	0.02
Creatinine, serum [mg/dl] ^a	0.85	0.62–0.96	0.83	0.73–0.91	0.88
eGFR [ml/min] ^{a,b}	127.73	110.35–140.56	125.00	113.91–149.78	0.76

p-Values were calculated using Mann–Whitney U test.

^a Creatinine serum concentrations and eGFR were available for 34 controls and 66 exposed individuals.^b Estimated glomerular filtration rate (eGFR) was calculated with the Cockcroft–Gault equation.

PAH metabolites. No differences of PAH urine levels were found between participants performing specific activities within the recycling process, e. g. burning of e-waste materials (data not shown).

Results of the multivariate analysis are shown in Table 4. Exposure to the e-waste recycling process was significantly associated with higher concentrations of all measured PAH metabolites, after correction for sex and tobacco smoking. Although the associations were less significant, female sex was associated with a higher urinary molar sum of phenanthrenes and a higher molar sum of phenantrenes and 1-OH-pyrene only. Tobacco smoking was associated with higher urinary concentrations of 1-OH-phenanthrene, 3-OH-phenanthrene, 1-OH-pyrene, the molar sum of phenantrenes and the molar sum of phenantrenes and 1-OH-pyrene. Again, the association was less significant than observed for exposure to e-waste recycling.

4. Discussion

Environmental pollution from informal recycling of e-waste is an emerging problem in Africa. Among other toxins, PAHs are a major health concern for exposed individuals. We investigated the levels of PAH metabolites in the urine of individuals working on one of the largest e-waste recycling sites in Africa, and in controls from a suburb of Accra without direct exposure to e-waste recycling activities.

4.1. Biometric, socioeconomic and medical parameters

Most of the exposed participants in our study were young males who had moved to Accra from northern parts of the country in order to find work. Consequently, the level of education was lower in the exposed group compared to controls, which were long-time residents generally. The income was not significantly different between both groups, and a higher proportion of the exposed participants had electricity and piped water in the household. Although almost all of the exposed participants reported that they feared health risks associated to their work, only very few of them used any protective measures, most likely due to the lack of protective gear. According to the clinical examination, most participants appeared to be of good health and pathological findings were rare. However, a high proportion of exposed participants complained about respiratory symptoms (cough and chest pain) and about dizziness or vertigo. Most individuals were involved in e-waste recycling only since a relatively short period of time, and discussions with e-waste workers revealed that the work in e-waste recycling often has to be given up due to the occurrence of health problems.

4.2. Urine concentrations of PAH metabolites

This is the first study to report PAH concentrations in the urine of e-waste workers at an African recycling site, and of a control group

Table 3

Descriptive statistical parameters of PAH-metabolite, creatinine and cotinine urine concentrations and renal function parameters in non-smoking exposed individuals and controls.

PAH metabolite, urine concentrations	Controls (n = 36)		Exposed (n = 56)		p
	Median	IQR [p25–p75]	Median	IQR [p25–p75]	
1-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.55	0.29–0.73	0.80	0.54–1.26	<0.001
2-/9-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.36	0.26–0.60	0.53	0.37–0.91	0.008
3-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.61	0.34–0.84	0.99	0.68–1.24	<0.001
4-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.11	0.06–0.17	0.21	0.15–0.33	<0.001
1-OH-pyrene [$\mu\text{g/g}$ creatinine]	0.47	0.26–0.78	1.27	0.69–2.28	<0.001
Molar sum of phenantrenes [$\mu\text{mol/mol}$ creatinine]	1.02	0.54–1.33	1.54	1.08–2.14	<0.001
Molar sum of phenantrenes and 1-OH-pyrene [$\mu\text{mol/mol}$ creatinine]	1.28	0.70–1.71	2.24	1.62–3.23	<0.001
<i>Other parameters</i>					
Creatinine, urine [g/l]	1.68	0.99–2.19	1.21	0.66–1.74	0.02
Creatinine, serum [mg/dl] ^a	0.82	0.62–0.95	0.83	0.71–0.93	0.63
eGFR [ml/min] ^{a,b}	130.28	110.38–141.78	123.96	112.81–149.19	0.89

p-Values were calculated using Mann–Whitney U test. Non-smoking status was defined as urinary cotinine concentration <50 $\mu\text{g/mL}$.^a Creatinine serum concentrations and eGFR were available for 31 controls and 53 exposed individuals.^b Estimated glomerular filtration rate (eGFR) was calculated with the Cockcroft–Gault equation.

Table 4
Multivariable linear regression analysis of PAH metabolite urine concentrations and exposure to e-waste recycling activities, sex and smoking status.

PAH metabolite, urine (log10)	Exposure ^a		Female sex ^b		Tobacco smoking ^c	
	Coefficient	p-value	Coefficient	p-value	Coefficient	p-value
1-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.47	<0.001	0.39	0.07	0.31	0.04
2-/9-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.36	0.005	0.30	0.06	0.29	0.08
3-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.41	0.001	0.23	0.12	0.36	0.02
4-OH-phenanthrene [$\mu\text{g/g}$ creatinine]	0.69	<0.001	0.20	0.18	0.24	0.11
1-OH-pyrene [$\mu\text{g/g}$ creatinine]	0.96	<0.001	0.41	0.05	0.50	0.02
Molar sum of phenantrenes [$\mu\text{mol/mol}$ creatinine]	0.44	<0.001	0.29	0.04	0.32	0.03
Molar sum of phenantrenes and 1-OH-pyrene [$\mu\text{mol/mol}$ creatinine]	0.61	0.001	0.33	0.03	0.40	0.01

^a vs. no exposure to e-waste.

^b vs. male sex.

^c vs. no recent tobacco smoking (cotinine urine concentration <50 $\mu\text{mol/L}$).

without direct exposure to e-waste recycling activities. We found high levels of PAH metabolites in both, the exposed and the non-exposed group, exceeding the concentrations reported from industrialized countries by far. In a survey conducted in the USA, the median concentration of 1-OH-pyrene, as biomarker for PAH exposure, was 0.074 $\mu\text{g/g}$ creatinine for adults, which is about 7 times lower than in our unexposed study population (CDC, 2009). The reference value for urinary 1-OH-pyrene concentrations in the non-smoking general population (aged 3–69 years) in Germany was defined as 0.5 $\mu\text{g/L}$ (corresponding to 0.3 $\mu\text{g/g}$ creatinine) (Wilhelm et al., 2008). Levels of PAH metabolites have been shown to be inversely associated with the distance to industrial sites. Nevertheless, 1-OH-pyrene levels in smoking women living in an industrial city in Germany were still distinctly higher compared to non-smoking women of the same region (medians 0.48 vs. 0.15 $\mu\text{g/g}$ creatinine), indicating a higher influence of smoking in contrast to environmental PAH immissions (Gündel et al., 1996). Another study from Germany found urinary 1-OH-pyrene levels of 0.1 $\mu\text{g/g}$ creatinine in non-smokers and 0.195 $\mu\text{g/g}$ creatinine in smokers (Heudorf and Angerer, 2001).

Higher PAH levels have been reported from developing countries, and were mostly attributed to indoor wood burning fireplaces and ingestion of smoked meat (Hemat et al., 2012; Johnson et al., 2009; Viau et al., 2000). In Ghana, the median 1-OH-pyrene concentration in the urine of non-smokers was reported to be 1.24 $\mu\text{g/g}$ creatinine (Johnson et al., 2009), which is distinctly higher than in our control group.

4.3. Urine concentrations of PAH metabolites according to exposure group

In our study, creatinine based urinary concentrations of all measured PAH metabolites, and the molar sum of phenantrenes and pyrene, as surrogate for total PAH exposure, were significantly higher in the exposed group compared to the control group. The creatinine urine concentration, as indicator of urine concentration and thus hydration status of participants, was significantly lower in the exposed compared to the non-exposed group. We have no sound explanation for this finding, indicating a higher dilution of the urine in exposed participants, but possible reasons might include the higher proportion of households with piped water as source for drinking water. We found no evidence of differences in renal function as possible explanation. Exposure to e-waste recycling activities was strongly associated to urinary PAH concentrations in multivariate linear regression analysis, after adjusting for sex and tobacco smoking.

A high burden of environmental toxins has been reported from the Agbogbloshie e-waste recycling site (Bridgen et al., 2008), but there exist only very limited data on human exposure, indicating high levels of multi-trace elements in e-waste recycling workers from this site (Asante et al., 2012). E-waste recycling might also contribute to the city-wide burden of environmental pollutants, as reported in some previous studies (Asante et al., 2011, 2012; Gioia et al., 2011). It is not clear, in how far the more distant areas around the e-waste recycling site are

affected by the contamination, but it is known that there is a long-range atmospheric movement of PAH from point sources of pollution (Aamot et al., 1996; Meharg et al., 1998). The prevailing wind in the study area is from the south or southwest, thus rather unpolluted air is brought from the sea to the e-waste recycling site, and the toxins released during the e-waste recycling processes are transported towards northern areas of Accra and the coastal region of Ghana, as shown for polychlorinated naphthalenes (Hogarh et al., 2012). However, although a contamination of the control site originating from the e-waste site cannot be completely ruled out, the high PAH levels observed in the unexposed group are more likely to be associated with indoor use of biomass fuel for cooking and air pollution from road traffic, considering the distance between both sampling sites. Cooking with biomass fuel has previously been reported as important source of PAH exposure (Li et al., 2011). The PAH exposure of Afghan adults was several times higher compared to the general population in developed countries, and mostly attributed to air pollution from car emissions and to burning of biomass fuels for cooking and household energy (Hemat et al., 2012). This explanation is in agreement with the association of female sex with higher concentrations of two of the measured PAH metabolites, because women and children, spending more time at home and in the kitchen, bear the main burden of indoor air pollution from burning biomass fuel as source of domestic energy (Fullerton et al., 2008).

4.4. Limitations of the study

One important limitation of our study is the cross-sectional design, which is not suitable to establish a causal relationship between elevated PAH exposure and e-waste related activities. Due to the unspecific character of PAHs, no information on potential sources can be provided. Furthermore, there are relevant differences between both study groups, such as in religious affiliation and ethnicity, which might be associated to PAH exposure and thus represent potential confounders. Our exposed group consisted mostly of Muslim migrants from northern parts of Ghana, whereas the control group consisted mainly of Christians. Given that diet and method of food preparation might differ by religion and region in Ghana, diet is a potential confounding variable in this study, which cannot be controlled for. However, Viau et al. (2002) demonstrated in one study that dietary pyrene intake does not affect the urinary 1-hydroxypyrene excretion. Furthermore, there was no difference in the frequency of meat or fish consumption between our study groups, but we did not document details on food preparation methods.

Exposure to indoor air pollution from burning biomass fuel is another possible confounding factor in our study, which cannot be controlled for. However, the vast majority (94.7%) of exposed participants reported that they usually take their meals in informal restaurants (chop bars) rather than preparing their meals at home, compared to 69% of controls. Thus, a significant additional PAH exposure of e-waste workers from indoor air pollution is unlikely, and controls may have a higher PAH exposure due to indoor air pollution from cooking on biomass fuel.

Tobacco smoking has been shown to be a major source of PAH exposure (Gündel et al., 1996; Heudorf and Angerer, 2001). A higher 1-OH-pyrene excretion in smokers has been mainly reported in populations with low PAH background levels, whereas smaller or no differences in 1-OH-pyrene were detected between smokers and non-smokers in populations with high PAH exposure (Buchet et al., 1992; Johnson et al., 2009; Levin, 1995). The proportion of smokers and urinary cotinine levels in our study were higher in the exposed group compared to the control group. In order to rule out that the observed differences in PAH exposure between the groups were caused by the effect of tobacco smoking, we conducted a separate analysis of urinary PAH concentrations, including only non-smokers. The analysis confirmed the results of the overall analysis, showing significantly higher urinary PAH concentrations for all measured metabolites in exposed individuals.

Road traffic pollution also has to be considered as possible source of PAH exposure. The extent of exposure to traffic exhaust fumes was not directly assessed in our study, and although working as driver was not associated with higher PAH urine concentrations in our control group, a confounding effect cannot be excluded.

It must be noted, that the age of our study cohort was not representative of e-waste workers at Agbogbloshie, since we included only adults, but many of the e-waste workers are children and adolescents. Children might be at particular risk for health effects due to higher exposure and susceptibility to the deleterious effects of PAH and other toxins during childhood development (Chen et al., 2011; Edwards et al., 2010; Perera et al., 2007).

Considering the deleterious effects of PAH on human health and especially on childhood development, a thorough assessment of the geographical extent of PAH exposure in the surroundings of the Agbogbloshie e-waste recycling site is warranted. Several schools, food processing industries and markets are in the direct vicinity of the recycling site.

Studies from China reported heavily PAH-contaminated agricultural soils in the surroundings of e-waste recycling sites, and showed that vegetables which were grown near e-waste recycling sites contain high concentrations of PAH (Tang et al., 2010; Y. Wang et al., 2012).

The investigation of other toxins in individuals exposed to e-waste recycling activities is also of importance. Concepts for effective pollution control measures are currently being developed by national and international stakeholders and need to be implemented.

5. Conclusions

This is the first study to report urinary levels of PAH metabolites in individuals working and living on an e-waste recycling site. In spite of an elevated PAH exposure of the control group compared to general populations of developed countries, we found distinctly higher urinary PAH levels in individuals exposed to e-waste recycling processes compared to unexposed controls. Moreover, individuals, who were exposed to the emissions of the e-waste recycling process complained more frequently about clinical symptoms as cough, chest pain and vertigo. Our findings suggest that involvement in the e-waste recycling process is associated with additional PAH exposure. The impact of e-waste recycling on the health of individuals living in the surroundings of the e-waste recycling site, including numerous schools and food processing industries, needs to be assessed.

Conflict of interest

The authors declare no conflict of interest.

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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.scitotenv.2013.06.097>.

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