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Biomonitoring of Organohalogen Compounds and Metals in Vietnamese Female Electronic Waste Recyclers

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Abstract

Introduction—E-waste has increased exponentially in recent years. Unsafe e-waste recycling techniques can lead to unsafe chemical exposures.

Objectives—To measure metals and organohalogens in blood and urine from Vietnamese e-waste workers and a non-e-waste recycler group. We compare these with female participants of the U.S. National Health and Nutrition Examination Survey (NHANES).

Methods—This study enrolled 40 female recyclers from Vietnam and a group of 20 non-recyclers from neighboring villages. CDC laboratories performed the chemical analyses.

Results—Recyclers showed higher serum concentrations of certain PBDE congeners than non - recyclers. Surprisingly, PCB 138/158 and PCB 153 were elevated in 18–<38 year old non-recyclers. Arsenic levels did not differ between recyclers and non-recyclers, but were higher than in the matched US NHANES population. Blood and urinary lead concentration levels were higher in the recyclers. Compared to U.S. NHANES women, our results showed both Vietnamese cohorts had higher levels of certain PBDE congeners, DDT, arsenic, lead and mercury concentrations, but lower overall PCB concentrations. p,p'-DDE levels were higher in the non-recyclers than in the recyclers, but were lower overall when compared to US NHANES.

Conclusion—These findings are consistent with occupational exposure to PBDEs and lead among the recyclers. Median concentrations of certain metals, such as lead and arsenic were

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higher in the Vietnamese compared to U.S. NHANES women of similar age, consistent with environmental exposures. Improved recycling protocols may decrease e-waste exposure and environmental measures may decrease contamination of the general population.

Introduction

Global demand for electrical and electronic equipment has risen rapidly over recent decades. This demand has increased as a result of technological advances, decreasing manufacturing costs, demand, and reduced service life of many electronic products. Forty-two million metric tons of e-waste was generated globally in 2014, with an expected annual increase of 4% to 5%, making e-waste one of the fastest growing waste streams (Balde et al., 2015). Major components of this waste stream include cellular phones, televisions, printers, computer monitors and hardware, stereo systems, and other household appliances. An emerging concern with this increasing demand for updated electronic products is the subsequent increased generation of electronic waste (e-waste). Thus, there is a need to properly recycle e -waste for recovery of metals and plastics, which then may be reused in new electronic products.

Industrialized nations export 50% to 80% of their e-waste to developing countries, including some in Asia (Wong, 2007). E-waste recycling provides vital additional income to recycling workers and their families, but many of these facilities employ unsafe techniques to separate parts for recycling and salvaging. These unsafe techniques include open burning of electrical cables and acid leaching of the waste (Leung et al., 2006; Leung et al., 2007; Li et al., 2009; Tue et al., 2013; Wong et al., 2007). The presence of metals (eg. arsenic, cadmium, lead, and mercury), persistent organic pollutants or POPs (eg. polychlorinated biphenyls (PCBs)), and flame retardants such as polybrominated diphenyl ethers (PBDEs) in e-waste, constitutes an increased risk to workers engaged in recycling, especially where weak or loosely enforced worker protection rules and their enforcement exist. A number of studies have documented elevated concentrations of e-waste contaminants in both human and environmental matrices, including human blood, urine and breast milk (Schecter et al., 2013; Song and Li, 2014; Tue et al., 2013), and occupational matrices such as air, dust, soil, and sediment (Chen et al., 2013; Leung et al., 2006; Leung et al., 2007; Luo et al., 2011). In contrast, recycling done with attention to occupational guidelines can improve sustainability, recover valuable materials for reuse and preserve worker health (Thuresson et al., 2006b; Fela, 2010).

In Vietnam, e-waste recycling is frequently home-based and located near agricultural areas. Current Vietnamese rules and regulations concerning e-waste, some in effect since 2006, do not adequately prevent human and/or environmental contamination (Nguyen et al., 2009). Hence, a high probability of chemical exposure exists at hazardous levels since the e-waste recyclers work at home in the absence of enforced regulations (Matsukami et al., 2015). Potential long-term health impacts of these e-waste contaminants, such as neurodevelopmental toxicity in children (Chen et al., 2011), and reproductive or endocrine-disrupting effects in children and adults (Needhidasan et al., 2014)are concerns which require further study , but are outside the scope of the current work. Evidence suggests improved recycling safety precautions can be effective in reducing exposure. Improvements in worker safety measures have been shown to reduce the body burden of certain e-waste

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compounds, as was demonstrated with the reduction of serum PBDEs at a Swedish work site (Thuresson et al., 2006a).

PBDE congeners are frequently found in e-waste (Bi et al., 2009; Sjödin et al., 2004a). Three major commercial mixtures of the brominated flame retardant PBDEs have been used: PentaBDE, OctaBDE, and DecaBDE. PentaBDE contains mainly BDE-47, -99, -100, -153, and -154. OctaBDE usually contains the following congeners: BDE-153, 154, 175/183, -196, -197, -203, -206, -207, and -209. DecaBDE typically contains 97% BDE-209 (La Guardia et al., 2006). Although exposure to pesticides including DDE and DDT is not related to e-waste recycling, these persistent organic pollutants can also affect health, and were measured to study exposures of the Vietnamese women in our study, as at least one recent study has reported finding DDT in Vietnamese waterways (Pham et al., 2010). Additionally, certain metals, such as lead, cadmium, and mercury are characteristically found in electronic waste (Wang et al., 2015).

In a prior study, elevated levels of certain metals and halogenated organics were reported from a cohort of 10 Vietnamese female home based e-waste recyclers and 10 non-recyclers with no known occupational exposure for the previous five years (Schecter et al., 2013). This current Vietnam-based study includes different 40 e-waste recyclers and 20 non-recyclers who volunteered blood and urine samples.

The objectives of the current study are (1) to present data on exposure of both select metals and organics among groups of e-recyclers and non-recyclers; (2) to suggest improvements in occupational safety among e-waste recyclers if requested; (3) comparing observed biomonitoring data to the National Health and Nutrition Examination Survey (NHANES), to identify instances where biomonitoring data indicate exposures of the investigated population that is higher than the US general population and where intervention may be beneficial; (4) to use these findings to highlight the usefulness of future exposure and health studies to better understand health risks associated e-waste work.

Methods

This study took place in rural northern Vietnam, approximately 130 kilometers northeast of Hanoi. Forty e-waste recyclers with at least 5 consecutive years of work were selected from villages in the Bảo Đài commune, while twenty non-recyclers were recruited from nearby hamlets in the Cấm village. Inclusion criteria for both non-recyclers and workers required that they be self-reported “healthy” women. In addition, the non-recyclers could not have worked as e-waste recyclers for at least the previous five years. Basic demographics including age, weight, height and occupation were collected by research staff administered questionnaires. We collected whole blood, serum, and urine using supplies from the Centers for Disease Control and Prevention (CDC) lots screened for chemical contamination. Specimens were frozen immediately, maintained at −20° to −70°C, and shipped on dry ice to the CDC in Atlanta, GA, USA.

Exposures to halogenated organics (eg. PBDEs and PCBs), persistent organochlorine pesticides such as DDT, and 16 common metals and metal species including lead, cadmium,

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mercury, and arsenic were measured to document exposure to e-waste, in order to determine if there were elevations in recycling workers compared to non-recyclers. Analyses included measurements at the same CDC laboratories used for the generation of NHANES data. Non-recyclers to age-matched US women were performed using age and gender matched values from a recent NHANES report, representative of the US general population (CDC, 2003–2004 & CDC, 2011–2012).

The Institutional Review Board (IRBs) at the University of Texas Health Science Center at Houston and the Ethics Board of the Hanoi School of Public Health reviewed and approved the protocol for this study. It was determined at the CDC that the agency was not engaged in human subjects' research.

Laboratory analysis

PBDEs, PCBs, and 2,2',4,4',5,5'-hexabromobiphenyl (PBB-153) measurements were conducted at CDC using standard methods including gas chromatography, isotope dilution, and high resolution-mass spectrometry (Sjödin et al., 2004b, Jones et al., 2012). PBB-153, an older brominated flame retardant has been measured in parallel with PBDEs for many years. There was no expectation that it would be elevated in this study but it was included in these analyses. Eleven PBDE congeners (17, 28, 47, 66, 85, 99, 100, 153, 154, 183, and 209) were measured in this study. PCB congeners measured were: 28, 44, 49, 52, 66, 74, 87, 99, 101, 105, 110, 114, 118, 123, 128, 138–158, 146, 149, 151, 153, 156, 159, 167, 170, 172, 177, 178, 180, 183, 187, 189, 194, 195, 196–203, 199, 206, 209. Pesticides measured were hexachlorobenzene (HCB), β-hexachlorocyclohexane (β-HCH), γ-hexachlorocyclohexane (γ-HCH or Lindane), oxychlordane, trans-nonachlor, p,p'-DDE, p,p'-DDT, and Mirex. Concentrations of organohalogen compounds were expressed as ng/g serum lipid. Serum lipid concentrations were determined using commercially available test kits from Roche Diagnostics Corp. (Indianapolis, IN)for the quantitative determination of total triglycerides (Roche test name: Triglycerides GPO-PAP, reagent product no. 11730711216) and total cholesterol (Roche test name Cholesterol CHOD-PAP, reagent product no. 11491458216). Final determinations were made on a Hitachi Modular P Chemistry Analyzer (Hitachi; Tokyo, Japan). The total lipids concentration was calculated from the total triglyceride and cholesterol concentrations (Phillips et al. 1989).

Metals were measured using inductively coupled plasma dynamic reaction cell mass spectrometry (ICP-DRC-MS). All elemental analysis methods used can be found in NHANES (CDC 2013–2014). Sixteen elements were analyzed in one panel (antimony, arsenic, barium, beryllium, cadmium, cesium, cobalt, lead, manganese, molybdenum, platinum, strontium, thallium, tin, tungsten, and uranium) in urine. Iodine and mercury were measured in urine. We measured five elements in whole blood (lead, cadmium, mercury, selenium and manganese) also with ICP-DRC-MS. Three elements were measured in serum (copper, zinc and selenium). Seven urinary species of arsenic were analyzed using high performance liquid chromatography (HPLC) coupled to ICP-DRC-MS. Blood mercury species (inorganic mercury, ethyl mercury, and methyl mercury) were measured using Triple Spike Isotope Dilution Gas Chromatography-Inductively Coupled Plasma Dynamic Reaction Cell Mass Spectrometry (TSID-GC-ICP-DRC-MS). Urinary creatinine

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concentrations were determined by using a Roche/Hitachi Modular P Chemistry Analyzer (Caldwell et al., 2013). Urine metal concentrations were adjusted by using creatinine concentrations to correct for variable water excretion rates at the time of spot urine specimen collection. All data passed expanded Westgard quality control rules, which have been described previously (Caudill et al., 2008).

Statistical Analysis

Data analysis was performed using SAS, v9.3 SAS Institute Inc. (Cary, NC, USA). All concentrations less than the limit of detection (LOD) were substituted with LOD/√2. Statistical significance between the e-waste recyclers and comparison groups was assessed with the median test using the SAS procedure "PROC NPAR1WAY" with the exact *p*-option enabled because of the number of subjects included in the study (20 non-recyclers and 40 e-waste recyclers). Group differences were assessed for all subjects and stratified by the median age of the control group, (eg., 18–<38 and ≥38–52 years). Median concentration for any analyte with less than 60% detection was not calculated. This cutoff was chosen to assure that the reported median would be based on detectable results.

Results

All participants in this study were self-reported non-smoking Vietnamese women who reported being in good health. There were no statistically significant differences in median age (39 [range: 19–50] and 37 [range: 18–52] years), weight (50 [range: 40–72] and 50 [range: 44–70] kg), and body mass index (BMI) (20.8 [16.6–30.4] and 20.3 [17.9–27.3] kg/m^2), between the e-waste workers and non-recyclers, respectively. All e-waste recyclers reported "farmer" as their primary occupation while in the non-recycler group only 50% of the women reported "farmer" as their primary occupation. Other occupations the non-recycler group reported were tailor (n=6), nurse (n=2), student (n=1), and accountant (n=1).

PBDE congeners and select organochlorine contaminants (e.g., PCB138/158, PCB153, p,p'-DDE, p,p'-DDT) that had greater than 60% detection frequency in the serum of either the e-waste recyclers or the non-recyclers are shown in Table 1. In general, certain PBDE congeners were measured at significantly lower concentrations in the non-recycler group than in the e-waste recyclers (Table 1). The PBDE congener profiles among the e-waste recyclers were dominated by PBDE congeners with 6 to 10 bromines typically present in commercial OctaPBDE and DecaPBDE commercial mixtures (BDE-153, BDE-183, and BDE-209), as previously reported in similar Vietnamese e-waste recycling workers (Tue et al., 2013). In non-recyclers, PBDE-209 was less than the LOD of 3.2 ng/g lipid, while in recyclers PBDE -209 was measured at a median concentration of 73.3 ng/g lipid. One worker had a serum concentration of 1,420 ng/g lipid, almost 20 times higher than the median concentration of the e -waste recycling group. Among the non-recycler population the only PBDE congeners with a detection frequency over 60% required to calculate a median concentration were PBDE-153 (1.2 ng/g lipid) and PBDE-183 (0.75 ng/g lipid). The median concentrations of these congeners were 10-fold higher in the recyclers compared with the non-recyclers. Among the recycling workers we measured higher concentrations for PBDE-153 (13.0 ng/g lipid) and PBDE-183 (7.3 ng/g lipid) than for other congeners with 4

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to 6 bromines present in the commercial PentaBDE mixture (range: 0.5–1.5 ng/g lipid). All PBDE congeners were found at a significantly higher concentration in the e-waste workers than in the non-recyclers (Wilcoxon rank-sum Test, Table 1).

Of the 37 PCB congeners measured, only 16 of the congeners (43%) had greater than or equal to 60% detection in both non-recyclers and e-waste recyclers. The serum concentrations of PCB-138/158 and PCB-153 are shown in Table 1, stratified by age (18-<38, ≥38–52, and all ages combined). The concentration of PCB138/158 and PCB153 in the comparison group were 2.2 (PCB-138/158) and 2.5-fold (PCB-153) higher than in the e-waste recyclers, for the age group 18–<38 years ($p<0.05$, Table 1). As shown on Table 1, concentrations of PCBs were in general higher among U.S. NHANES women than in both Vietnamese non-recyclers and workers and for the ≥38–52 year age group the 95% confidence intervals for the median did not overlap.

p,p'-DDE levels were observed at significantly higher concentrations in the comparison population (182.5 ng/g lipids) relative to the e-waste workers (132.0 ng/g lipids) (Table 1) while no statistical difference was observed for p,p'-DDT. Of note, DDT levels were higher in both recyclers and non-recyclers in contrast to NHANES (see Table 1).

Lead was significantly higher in both whole blood and urine of e-waste workers than in the non-recycler population (Table 2). The non-recycler median lead urine was (2.31 μg/g Cre) and the whole blood concentration was (2.93 μg/dL). In contrast, recyclers had significantly elevated median lead urine and whole lead blood concentrations of (3.22 μg/g Cre) and (4.82 μg/dL whole blood) respectively. Although lead levels were lower in the non-recyclers in contrast to the recyclers, lead levels were higher in both Vietnamese cohorts than they were in the median lead NHANES matched estimates (Table 2). This observation is consistent with what we found in our earlier group of 10 e-waste recycling workers and 10 matched controls where lead was elevated in both groups as compared to NHANES (Schecter et al., 2013). Although there was no statistical difference between recyclers and non-recyclers for cadmium, mercury, or arsenic, all metals tested were higher in the Vietnamese than in the age matched American women.

We observed no significant difference in urine mercury levels between non-recyclers (0.34 μg/g Cre) and e-waste recyclers (0.52 μg/g Cre). However, total mercury was higher in whole blood concentrations of non-recyclers (3.46 μg/L) versus e-waste (2.49 μg/L). The non-recycler population also had significantly higher methyl mercury concentrations. The non-recyclers' methyl mercury median concentration was 4.16 μg/L while the e-waste recyclers had a methyl mercury concentration of 2.76 μg/L respectively. Of note, whole blood and urine cadmium and mercury results were also higher in the Vietnamese women than in matched NHANES women. These findings will be examined further in the discussions section of this paper.

In both the non-recyclers and recyclers, each arsenic species shown on our table has higher concentrations than those found in NHANES. Total urinary arsenic was higher in both non-recyclers (46.94 μg/g Cre) and e-waste recyclers (42.35 μg/g Cre) when compared to their age-matched U.S. NHANES values (6.84 μg /g Cre) (Table 2).

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Discussion

We hypothesized that we would find significant elevations of certain metals and halogenated organics, such as PBDEs and lead, in women working in e-waste recycling compared to non-recycler women with similar backgrounds. Pictures of typical work situations for the recycling workers studied are given in Figure 1 and 2. With respect to PBDEs, BDE-153, BDE-183, and BDE-209, congeners present in the commercial Octa-and DecaBDE mixtures, tended to be the largest contributors to the serum concentration in these e-waste recyclers, which is consistent with the use of these mixtures in electronics (Zhao et al., 2010). Median serum lipid BDE-153, BDE-183, and BDE-209 concentrations in the comparison group were significantly lower than their corresponding median concentrations in the e-waste recyclers, consistent with their use in electronics. BDE-209 was below the limit of detection in all but four of the women in the comparison group, whereas it was detected in 100% of the e-waste recyclers. These findings are consistent with e-waste studies performed in China at four different recycling sites, which reported finding higher serum concentration of Hepta-to Deca -BDE congeners in male e-waste recyclers vs. the non-recyclers. (Bi etal., 2007; Zhao et al., 2010). Despite BDE -209 having a relatively short biological half-life of 2 weeks in humans (Geyer et al., 2004, Thuresson et al., 2006b), it was detected in higher concentrations throughout the e-waste women's blood specimens, therefore suggesting that these e-waste recyclers were recently exposed to BDE-209 through their occupation.

Commercial PentaBDE was mainly used in polyurethane foam containing the congeners, BDE- 47, 99, 100, 153, and 154 (Zota et al., 2008). Consistent with an earlier study of Vietnamese males (Tue et al., 2013), PentaBDE congeners were measured at significantly lower serum lipid levels in our non-recyclers than in the e-waste recycler women. Further, these lower brominated PBDE congeners were detected in less than 20% of the non-recycler group, and the median concentrations for these congeners in the non-recyclers were lower than the level of detection. This is consistent with increased exposure of recyclers. It has been reported that PentaBDE can be found in older circuit boards, protective electronic coating, and accumulation of indoor dust, known to contain PentaBDE congeners in electronics and subsequent mobilization of this dust during recycling (Hazrati & Harrad, 2006; UNEP, 2015). We hypothesized these findings may be attributable to the accumulation of indoor dust, and the possibility that the breakdown of higher brominated congeners within the electronics potentially created PentaBDE congeners. This is in agreement with previous studies that found elevation of PentaBDE congeners in the air in Vietnamese e-waste recycling areas and in indoor air at an electronics recycling facility in Sweden (Tue et al., 2013; Thuresson et al., 2006a).

Pesticides, PCBs, and metals not characteristically found in e-waste, such as arsenic and mercury, were also analyzed to provide further exposure and health related data on mixtures of metals and organics in Vietnam. Serum PBDE levels did not correlate with serum PCB levels, supporting the conclusion that the exposure comes from different sources. With the exception noted below, none of the seventeen PCB congeners with detection rates over 60% were significantly different between the two groups, which is consistent with studies that examined serum levels in male Chinese e-waste workers (Bi et al., 2007; Zhao et al., 2010).

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An interesting finding however is that for PCB 138/158 and PCB 153 in 18-<38 year old non-recycling women, these levels were higher compared to recyclers. Furthermore, the median serum PCB levels in both Vietnamese populations were lower compared to the corresponding NHANES median levels. PCBs were once commonly used in transformers and capacitors in many countries until they were banned and/or phased out during the last quarter of the twentieth century. Most e- waste currently being recycled and presumably manufactured after the 1970s does not contain PCBs, although PCBs have been known to persist and bioaccumulate in human and environmental matrices for years (Sjödin et al., 2004a).

With regards to persistent pesticides, p,p'-DDE levels were statistically significantly higher in the non-recyclers compared to the recyclers. Specifically, p,p'-DDE was measured at a concentration 95% higher in the 18 to < 38 year old non-recyclers compared to similarly aged e-waste recyclers. This is an interesting finding since proportionally more of the e-waste workers stated farming as their primary occupation when contrasted with the non-recyclers. Overall, p,p'-DDE levels of both the non-recyclers and recyclers were similar to that found in NHANES. It should be noted, the recyclers and non-recyclers, although not statistically different from one another, showed higher levels of p,p'-DDT than NHANES(Table 1) . This could possibly be explained by the use of DDT after 1995 when it was banned in Vietnam (Pham et al., 2010).

Lead levels in both whole blood and urine were significantly higher for the recyclers in compared to the non-recyclers. This finding is consistent with other studies that reported elevated lead concentrations in both blood and urine samples from male e-waste recyclers (Wang et al., 2011; Yang et al., 2013b), although those studies reported higher blood lead concentrations than found in this study. However, both groups had elevated levels compared to matched U.S. NHANES levels, which might pose a health concern due to current warnings suggesting that there is essentially no safe level of lead (AAP, 2016; HHS, 2012). Other studies have described significantly elevated levels of lead in children who work at or reside near e -waste facilities (Song & Li, 2014). Children living or working near the vicinity of e-waste facilities are therefore at higher risk for health effects such as neurodevelopmental pathology from lead toxicity (Chen et al., 2011). The elevated lead documented in these recyclers and also to a lesser degree in the Vietnamese non-recyclers, is consistent with occupational exposure, although the source of the environmental lead is not clear. This finding is worthy of further clarification to characterize and address elevated lead levels in both groups in Vietnam.

Cadmium levels in both blood and urine were not significantly different between recyclers and non-recyclers, but the median concentrations found in both Vietnamese populations were higher when compared to NHANES. Cadmium, a known human carcinogen, has been linked to workplace exposure, environmental airborne dust, and smoking. Cigarette smoking is very rare among Vietnamese women, and each of our study participants denied cigarette smoking. However, there was no data collected on exposure to passive cigarette smoke.

As reported in the results, we observed no significantly higher urine mercury levels in the e-waste recyclers, but total mercury was higher in the whole blood concentrations of non-

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recyclers when compared to the recyclers, likely attributed to increased methyl mercury levels found in the non-recyclers. Both whole blood total mercury and methyl mercury levels were higher in the Vietnamese populations when compared to the age-matched American women reported in NHANES. Urine mercury is primarily inorganic mercury, whereas whole blood total mercury includes both inorganic *and* organic forms of mercury, such as ethyl and methyl mercury. Methyl mercury, which biomagnifies up the aquatic food chain, was higher in the non-recyclers than in the e-waste recyclers. Urinary mercury concentrations tend to suggest chronic exposure to inorganic mercury in e-waste recyclers, whereas, the higher level of methyl mercury in non-recyclers is possibly due to greater fish consumption.

We had previously reported in our earlier study that urinary total arsenic concentrations were higher in the e-waste recyclers than the non-recyclers but this was not statistically significant (Schecter et al., 2013). Our findings here are consistent with our earlier study: urinary total arsenic levels were not significantly different between e-waste recyclers and non-recyclers. However median urinary arsenic levels were higher in Vietnamese females in relation to age - matched U.S. NHANES values. This arsenic elevation is partially explained by two studies in the Red River Delta of Northern Vietnam where elevated concentrations of arsenic in groundwater occurs (Agusa et al., 2013; Nguyen et al., 2009). Many rural Vietnamese derive their water from wells. Other studies have reported significantly greater arsenic contamination in well-water than surface water in Vietnam (Hoang et al., 2010; Hug et al., 2008; Nguyen et al., 2009). The findings in recyclers and non-recyclers are of concern regarding the toxic arsenic species, arsenous acid (As III), monomethyl-arsonic acid (MMA) and dimethylarsenic acid (DMA), which have concentrations in these Vietnamese women greater than those found in NHANES.

The elevated arsenic levels in our volunteers are consistent with arsenic contamination in well water in Vietnam and are presumably un related to e-waste recycling(Agusa et al., 2013). This suggests that environmental remediation could also lower body burden.

In some developing countries, there are new environmental measures for arsenic treatment of well-water such as barrel filters designed to remove arsenic developed by researchers at MIT, in cooperation with the Environment and Public Health Organization of Nepal, and Rural Water Supply and Sanitation Support Programme. These filters are designed to remove arsenic, iron, odor, and pathogens found in potable water, based on slow sand filtration and iron hydroxide adsorption principles (using rusty nails changed once each year) (Ngai et al., 2005).

This study has strengths as well as limitations. A strength of this study is the comparison of Vietnamese women to age matched women in the U.S. general population (CDC, 2009–2010). However, direct comparisons between the two counties should be interpreted with caution due to the many differences between Vietnam and the U.S. general population including but not limited to genetic, socio-economic, geographic, climate, cultural occupational and life style differences. The samples from both an earlier study and the current extension of that study were analyzed in the same CDC laboratories which performed the NHANES analyses, enhancing the validity of the results. In terms of limitations, this study includes a relatively small number of subjects in part due to the

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considerable effort involved in finding and obtaining permission from workers, employers, and government officials. Further, e-waste recycling was not necessarily the main occupation for the female recyclers performing home based recycling ;many women report ed being farmers in both the e -waste workers and non-recycler groups, whereas some in the non-recycling group reported different occupational titles. Although these apparent inconsistencies in reported occupation are a reflection of the demographics of subjects performing home based recycling of electronics in Vietnam, these inconsistencies may have confounded some of the results for measured contaminants not present in e-waste such as DDE/DDT and arsenic. This study includes a convenience sampling of rural northern Vietnamese women e-waste recyclers and comparisons, and may not necessarily be found in other populations. Another limitation was our inability to better assess dietary factors, such as consumption levels of fish and well-water, which may have confounded concentrations of various pollutants measured in both the non-recyclers and recyclers. Future exposure assessments in Vietnamese women should also take into account the dietary contribution to these exposures.

A major strength of our study is that we analyzed a number of previously not studied or understudied metals and organic compounds in blood and urine samples at CDC laboratories, which also analyzed the NHANES samples. Most metal levels measured in our study, such as cadmium, mercury and arsenic, were not different between the recyclers and non-recyclers, but were elevated compared to age-matched US NHANES values. More importantly, our results indicate that halogenated organics, including certain PBDE flame-retardants (BDE-153, BDE-183, and BDE-209) and metals such as lead, were detected at higher concentrations in e-waste recyclers compared to non-recyclers. In this context, our study contributes to the body of information of occupational exposure risks for these e-waste recyclers and underscores the need for further study of risk factors for exposure in this occupation and in this environment.

Conclusions

Exposure to certain halogenated organics and metals were found to vary between recyclers and non-recyclers. For example, elevated lead and PBDE levels in workers and their families could be of concern , especially for exposed pregnant women and children. Other studies have documented the ability of improved worker safety measures to reduce occupational exposure to PBDEs (Thuresson et al., 2006a), emphasizing the importance of improved industrial hygiene standards and enforcement of these standards to reduce exposure to at least some of the compounds elevated in the recyclers we studied when compared to non-recyclers.

Furthermore, both worker and comparison populations had levels of blood mercury, urine cadmium, and urine arsenic species higher than the corresponding levels reported in NHANES. The elevated arsenic levels in our volunteers are consistent with arsenic contamination in well water in Vietnam and are presumably un related to e-waste recycling(Agusa et al., 2013).

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Although this study reported a larger number of metals and organics in Vietnamese recyclers and non-recyclers than previously reported, there are still more toxic compounds not examined in this study which may be found in Vietnamese recyclers or non-recyclers. For example, this study did not include some major flame-retardants other than PBDEs, such as some halogenated flame retardants and also organophosphate flame-retardants, which can be found in some electronics (Hoffman et al., 2015).

Finally, to the best of our knowledge, this is the first study measuring both organics, and metal exposure in recyclers and non-recyclers. Further study as well as dissemination of these findings may be beneficial for Vietnamese health authorities in evaluating where scarce resources might best be directed.

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Figure 1.
Two e-waste recyclers and a child without protective gear.

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Figure 2.
A female recycling worker and her husband exposed to fumes from burning e-waste without protection.

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Table 1

Median concentrations (ng/g lipid) of halogenated organics in e-waste workers, non-recyclers and NHANES.

Compound	Age Group	LOD[a]	Non-recyclers (n=20)		E-waste recyclers (n=40)		Median test[b]	NHANES[c]
			Median (95%CI)	PCT >LOD	Median (95%CI)	PCT >LOD	p-value	Median (95%CI)
Polybrominated Diphenyl Ethers (PBDEs)								
PBDE-47	***All***	***0.4***	***<LOD***	***15%***	***1.5 (1.1–2.4)***	***88%***	***<0.0001***	19.4 (15.0–23.8)
	18–<38		<LOD	10%	1.5 (0.7–3.6)	93%	<0.01	22.9 (18.8–26.6)
	≥38–52		<LOD	20%	1.4(0.9,2.5)	84%	<0.01	13.5 (11.0–21.3)
PBDE-99	***All***	***0.4***	***<LOD***	***0%***	***0.7 (0.4–0.9)***	***65%***	***<0.0001***	<LOD
	18–<38		<LOD	0%	0.8 (0.4–1.2)	67%	<0.05	<LOD
	≥38–52		<LOD	0%	0.6 (0.4–0.9)	64%	<0.01	<LOD
PBDE-100	***All***	***0.4***	***<LOD***	***0%***	***0.55 (0.4–0.7)***	***60%***	***<0.001***	3.5 (3.0–4.3)
	18–<38		<LOD	0%	<LOD	53%	0.056	4.3 (3.3–5.0)
	≥38–52		<LOD	0%	0.6 (0.4–0.8)	64%	<0.01	2.9 (2.1–3.8)
PBDE-153	***All***	***0.4***	***1.2 (1.0–1.7)***	***95%***	***13.0 (10.2–18.8)***	***100%***	***<0.0001***	4.2 (3.3–5.1)
	18–<38		1.0 (0.8–1.2)	100%	12.2 (5.6–18.8)	100%	<0.0001	5.1 (4.2–6.0)
	≥38–52		1.7(0.9,2.5)	90%	13.2 (9.5–20)	100%	<0.001	3.1 (2.5–4.2)
PBDE-154	***All***	***0.4***	***<LOD***	***0%***	***0.5 (0.4–0.7)***	***60%***	***<0.0001***	<LOD
	18–<38		<LOD	0%	<LOD	53%	0.078	<LOD
	≥38–52		<LOD	0%	0.5 (0.4–0.7)	64%	<0.001	<LOD
PBDE-183	***All***	***0.4***	***0.75 (0.4–1.2)***	***65%***	***7.3 (6.1–10.0)***	***100%***	***<0.0001***	<LOD
	18–<38		0.85 (0.4–1.3)	60%	8.8 (6.2–15.9)	100%	<0.001	<LOD
	≥38–52		0.65 (0.3–1.4)	70%	6.6 (4–10.7)	100%	<0.001	<LOD
PBDE-209	***All***	***3.2***	***<LOD***	***25%***	***73.3 (32.4–138.2)***	***100%***	***<0.0001***	n/a
	18–<38		<LOD	30%	82.4 (22.2–271.8)	100%	<0.01	n/a
	≥38–52		<LOD	20%	68.8 (19.8–138.2)	100%	<0.001	n/a
Polychlorinated Biphenyl (PCBs)								
PCB-138/158	***All***	***0.4***	***7.95 (5.4–9.9)***	***100%***	***7.7 (6.8–8.5)***	***100%***	***>0.5***	11.6 (9.9–14.2)
	18–<38		7.85 (5.0–9.9)	100%	3.5 (2.6–7.5)	100%	<0.05	7.4 (6.3–8.2)
	≥38–52		8.3(3.8–13.8)	100%	8.1 (7.7–11.7)	100%	>0.5	20.1 (16.6–23.8)
PCB-153	***All***	***0.4***	***8.1 (4.0–22.9)***	***100%***	***7.8 (2.4–18.9)***	***100%***	***>0.5***	15.5 (14.1–18.4)

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Compound	Age Group	LOD[a]	Non-recyclers (n=20)		E-waste recyclers (n=40)		Median test[b]	NHANES[c]
			Median (95%CI)	PCT >LOD	Median (95%CI)	PCT >LOD	p-value	Median (95%CI)
	18–<38		8.8 (4.8–10.6)	100%	3.55 (2.8–6.6)	100%	<0.05	8.8 (7.8–10.0)
	≥38–52		8.1 (4.4–15.2)	100%	8.7 (7.8–13.0)	100%	>0.5	25.8 (22.8–30.7)
Persistent Pesticides								
p,p'-DDE	***All***	***2.3***	***182.5 (139.3–258.7)***	***100%***	***132(108–187)***	***100%***	***<0.05***	163 (138–220)
	18–<38		158.3 (94.4–258.7)	100%	81.1 (57.9–141)	100%	<0.05	124 (96.3–155)
	≥38–52		209.0 (164.1–306.1)	100%	171 (131–275)	100%	0.15	260 (176–350)
p,p'-DDT	***All***	***1.6***	***26.1 (22.8–32.0)***	***100%***	***21.3 (19.0–25.7)***	***100%***	***0.3***	<LOD
	18–<38		25.0 (18.2–39.3)	100%	19.7(16.2–31.7)	100%	0.22	<LOD
	≥38–52		28.5 (20.0–48.5)	100%	24.8 (18.2–26.0)	100%	0.47	<LOD

Abbreviations: Limit of detection (LOD; ng/g lipid), detection frequency is given and p-values for the comparison of the two Vietnamese groups. LOD, Limit of Detection; 95% CI, 95% confidence interval; PCT >LOD, percentage of results above the limit of detection; PBDE-47, 2,2',4,4'-tetrabromodiphenyl ether; PBDE-99, 2,2',4,4',5-pentaBDE; PBDE-100, 2,2',4,4',6-pentaBDE; PBDE-153, 2,2',4,4',5,5'-hexaBDE; PBDE-154, 2,2',4,4',5,6'-hexaBDE; PBDE-183, 2,2',3,4,4',5',6-heptaBDE; PBDE-209, decaBDE; PCB138/158, 2,2',3,4,4',5'-hexachlorobiphenyl and 2,3,3',4,4',6-hexaCB; PCB153, 2,2',4,4',5,5'-hexaCB; p,p'-DDE, 2,2-Bis(4-chlorophenyl)-1,1-dichloroethene; p,p'-DDT, 2,2-Bis(4-chlorophenyl-1,1,1-trichloroethane.

[a]Limit of detection (LOD) is given as a median LOD since depended on available serum amount;

[b]Median test for the comparison of comparison group and E-waste recyclers;

[c]The US median population estimate for females from the National Health and Nutrition Examination Survey (NHANES) for the survey years 2003 and 04 (Sjödin et al 2008 and CDC 2003–2004).

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Table 2

Median concentration (μg/L or μg/dL whole blood or μg/g creatinine) of metals in e-waste recyclers.

Compound	Age Group[a]	LOD	Non-recyclers (n=20) Median (95%CI)	E-waste recyclers (n=40) Median (95%CI)	Median test[b] p-value	NHANES[c] Median (95%CI)
Whole blood measurements						
Cadmium (ug/L)	***All***	***0.1***	***0.59 (0.41 – 0.78)***	***0.59 (0.53 – 0.68)***	***>0.5***	0.30 (0.28 – 0.32)
	18–<38		0.54 (0.36 –0.83)	0.47 (0.40 –0.55)	>0.5	0.26 (0.24 – 0.29)
	≥38–52		0.64 (0.35 – 0.88)	0.72 (0.65 – 0.84)	>0.5	0.35 (0.32 – 0.39)
Lead (ug/dL)	***All***	***0.07***	***2.93 (2.62 – 3.88)***	***4.82 (4.52 – 5.96)***	***<0.0001***	0.69 (0.63 – 0.75)
	18–<38		2.70 (2.24 – 20.89)	4.64 (3.95 – 6.08)	0.23	0.54 (0.51 – 0.58)
	≥38–52		3.44 (2.45 – 3.97)	4.99 (4.52 – 7.00)	<0.001	0.91 (0.87 – 0.96)
Methyl mercury (ug/L)	***All***	***0.12***	***4.16 (2.93 – 5.08)***	***2.76 (2.30 – 3.22)***	***<0.05***	0.50 (0.41 – 0.64)
	18–<38		3.23 (2.58 – 5.35)	2.38 (2.01 – 4.10)	>0.5	0.44 (0.35 – 0.54)
	≥38–52		5.04 (4.13 – 10.88)	2.76 (2.47 – 3.81)	<0.001	0.61 (0.46 – 0.83)
Mercury, total (ug/L)	***All***	***0.28***	***3.46 (2.50 – 3.88)***	***2.49 (2.08 – 2.90)***	***0.055***	0.68 (0.59 – 0.81)
	18–<38		3.02 (2.37 – 3.88)	2.48 (2.00 – 3.08)	0.11	0.59 (0.50 – 0.68)
	≥38–52		3.84 (1.87 – 4.80)	2.60 (2.05 –3.07)	0.14	0.85 (0.64 – 1.05)
Urine measurements (μg /g Creatinine)						
Arsenic, total	***All***	***0.26***	***46.94 (38.56 – 65.70)***	***42.35 (37.54 – 49.55)***	***0.17***	6.84 (5.95 – 8.04)
	18–<38		44.59 (23.07 – 69.51)	43.38 (35.64 – 58.83)	0.43	6.49 (5.30 – 8.52)
	≥38–52		48.94 (38.56 – 116.22)	41.47 (36.22 – 49.55)	0.15	6.88 (6.39 – 9.28)
Arsenobetaine	***All***	***1.16***	***7.62 (2.32 – 24.90)***	***6.96 (5.37 – 10.29)***	***>0.5***	2.15 (1.75 – 2.90)
	18–<38		3.19 (1.91 – 49.24)	7.22 (2.65 – 30.65)	>0.5	1.95 (1.47 – 2.69)
	≥38–52		9.32 (2.13 – 24.90)	6.86 (3.91 – 10.85)	0.42	2.80 (1.81 – 3.36)
Arsenous Acid	***All***	***0.12***	***1.91 (1.63 – 3.26)***	***1.79 (1.52 – 2.64)***	***0.39***	<LOD
	18–<38		1.82 (1.61 – 4.86)	2.52 (1.71 – 3.74)	0.18	<LOD
	≥38–52		2.32 (1.42 – 3.99)	1.64 (1.43 – 2.48)	0.26	<LOD
Monomethylarsonic Acid	***All***	***0.2***	***3.63 (3.21 – 4.25)***	***3.78 (2.93 – 4.64)***	***>0.5***	0.91 (0.75 – 1.15)
	18–<38		3.63 (2.47 – 4.80)	3.78 (2.74 – 5.73)	>0.5	0.84 (0.71 – 1.07)
	≥38–52		3.78 (2.67 – 5.55)	3.71 (2.67 – 5.07)	>0.5	1.03 (0.68 – 1.31)
Dimethylarsenic Acid	***All***	***1.91***	***26.24 (19.51 – 30.06)***	***27.50 (22.69 – 33.05)***	***>0.5***	4.23 (3.71 – 4.59)

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Compound	Age Group[a]	LOD	Non-recyclers (n=20) Median (95%CI)	E-waste recyclers (n=40) Median (95%CI)	Median test[b] p-value	NHANES[c] Median (95%CI)
	18–<38		23.36 (18.62 – 39.32)	31.57 (21.94 – 37.82)	>0.5	3.65 (3.39 – 4.28)
	≥38–52		29.14 (16.89 – 41.71)	24.45 (22.52 – 36.35)	>0.5	4.52 (4.23 – 4.82)
Cadmium	***All***	***0.036***	***0.832 (0.484 – 1.070)***	***0.998 (0.816 – 1.249)***	***0.41***	0.192 (0.172 – 0.219)
	18–<38		0.574 (0.325 – 0.981)	0.812 (0.603 – 1.098)	>0.5	0.148 (0.126 – 0.171)
	≥38–52		1.058 (0.472 – 2.238)	1.217 (0.891 – 1.418)	>0.5	0.307 (0.275 – 0.342)
Lead	***All***	***0.03***	***2.31 (1.73 – 2.68)***	***3.22 (2.69 – 3.99)***	***<0.05***	0.34 (0.31 – 0.40)
	18–<38		2.43 (0.98 – 28.54)	2.89 (2.36 – 3.68)	<0.05	0.29 (0.25 – 0.33)
	≥38–52		2.17 (1.21 – 3.53)	3.98 (2.34 – 4.94)	0.26	0.49 (0.35 – 0.58)
Mercury, total	***All***	***0.13***	***0.34 (0.23 – 0.71)***	***0.52 (0.42 – 0.80)***	***0.21***	0.41 (0.33 – 0.46)
	18–<38		0.32 (0.22 – 0.76)	0.58 (0.37 – 1.00)	0.062	0.33 (0.25 – 0.46)
	≥38–52		0.46 (0.17 – 1.13)	0.49 (0.36 – 0.83)	>0.5	0.45 (0.41 – 0.58)

Abbreviations: Limit of detection (LOD) and the p-value for the comparison of the two exposure groups are given. NHANES median estimates are given as a comparison. LOD: 95% CI, 95% confidence interval; PCT >LOD, percentage of results above the limit of detection.

[a]Stratification for age; the all category contains all ages while the 18–<38 and ≥38–52 years were defined as stratification by the median of the control group.

[b]Median test for the analysis of comparison group and E-waste recyclers;

[c]The US median population estimate for females from the National Health and Nutrition Examination Survey (NHANES) for the survey years 2011 and 2012 using the same age stratification as for the Vietnamese women (Sjödin et al 2008 & CDC 2011–2012).

[d]84% detection rate.

[e]75% detection rate.

[f]90% detection rate.