



Research Paper

Human exposure to BTEX emitted from a typical e-waste recycling industrial park: External and internal exposure levels, sources, and probabilistic risk implications

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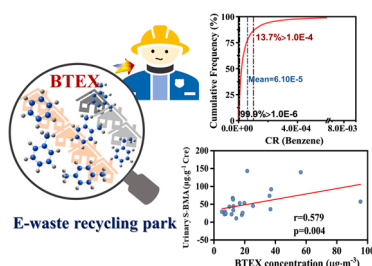
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HIGHLIGHTS

- Ambient and biomonitorial of BTEX were studied in a typical e-waste recycling park.
- Urinary metabolites levels were much higher than unmetabolized BTEX in workers.
- HA was found to be the absolute major component in urinary samples.
- Urinary S-BMA might act as a tracer for BTEX derived from e-waste disposal.
- Biomonitoring exposure to benzene showed 99.9 % cumulative carcinogenic risk.

GRAPHICAL ABSTRACT



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ABSTRACT

Benzene, toluene, ethylbenzene, and xylene (BTEX) can be released during extensive activities associated with the disposal of electronic waste (e-waste), which might pose deleterious health effects on workers. In this study, pollution profiles of BTEX in air and their urinary excretive profiles in occupational workers were investigated in a typical e-waste recycling industrial park. The results showed that the workers in the park were generally exposed to high levels of BTEX. The median levels of urinary metabolites were approximately 6-orders of magnitude higher than those of unmetabolized BTEX, indicating that pollutants efficiently metabolize at those occupational levels. The analytes presented differential profiles in external and internal exposure. Among the metabolites, significant correlation ($p < 0.05$) was observed between N-acetyl-S-benzyl-L-cysteine (S-BMA) concentration and atmospheric individual BTEX derived from the e-waste recycling area, suggesting that S-BMA is a potential marker for BTEX exposure to e-waste occupational workers. Notably, 95.2 % of all the workers showed a cumulative carcinogenic risk induced by BTEX exposure via inhalation, with 99.9 % of the carcinogenic risk distribution based on concentration of benzene metabolite (N-acetyl-S-(phenyl)-L-cysteine) exceeding 1.0E-

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6. This study holds potential in providing valuable inferences for the development of remediation strategies focusing on BTEX exposure reduction to protect workers' health at e-waste recycling industries.

1. Introduction

The remarkable global increase in electronic waste (e-waste) and its indiscriminate disposal will inevitably cause problems to human health and the ecosystem. As per a monitor report in 2020, the amount of global e-waste will increase to 74 million tons in 2030 (Forti et al., 2020). Strict regulations and recycling of e-waste offer a sustainable approach for both environmental and economic interests, while it embodies a wide variety of challenges. Many studies have reported the emission of different types of pollutants into the environment during the handling and processing of e-waste (An et al., 2014a; Guo et al., 2015; Liu et al., 2020; Ren et al., 2014). Various scientific investigations have revealed high levels of pollutants, such as halogenated flame retardants, polychlorinated dibenzo-*p*-dioxin and dibenzofurans, polybrominated dibenzo-*p*-dioxin and dibenzofurans, polycyclic aromatic hydrocarbons (PAHs) as well as heavy metals in and around e-waste recycling industries (Liu et al., 2020; Ren et al., 2014; Luo et al., 2014; Gangwar et al., 2019). Although the adverse effects of exposure to these pollutants are not fully understood, available scientific findings have confirmed that e-waste recycling considerably increases the incidence of disease, threatening both occupational workers and local residents (An et al., 2014a; Lu et al., 2016).

In recent decades, the problem of volatile organic compounds (VOCs) released from e-waste disposal processes has garnered attention due to their increasing concentration in the environment and toxic effects on human health and natural ecosystems. Many simulation studies on laboratory-scale have confirmed severe pollution of benzene, toluene, ethylbenzene, and xylene (BTEX) during the combustion of e-waste materials (Moltó et al., 2009; Alston et al., 2011; Chiang and Lin, 2014). The concentrations of benzene and toluene during pyrolysis and combustion of waste printed circuit boards (WPCBs) were up to 13.8 and 3.35 g·kg⁻¹, respectively, which were much higher than that for PAHs (2.77 ± 1.41 mg·kg⁻¹) (Cai et al., 2018; Ortuno et al., 2014). Moreover, field observations further illustrated that high levels of BTEX can accumulate in the air during WPCB dismantling and plastic waste recycling processes (An et al., 2014a; He et al., 2015; Chen et al., 2016a). Although e-waste recycling is an emerging source for recovering various materials, it might serve as a major contributor to atmospheric BTEX levels in addition to fuel combustion, solvent usage, and vehicular exhaust (Bolden et al., 2015; Mendoza-Cantu et al., 2006; Dominutti et al., 2019). However, limited data are available for the formal e-waste recycling industrial park, which embodies a variety of disposal instruments (Chen et al., 2021).

The environmental impacts associated with e-waste dismantling have translated directly into serious health threats to occupational workers, and the inhalation-related risks of pollutants at concentrations exceeding the permissible levels have been reported (An et al., 2014a; He et al., 2015; Chen et al., 2016a). However, previous external exposure assessment reports primarily focused on deterministic risk quantification, leading to either under- or overestimation of risk due to the specific variabilities among individuals (Tong et al., 2019). As the concentration of contaminants is highly variable in occupational settings, it is difficult to accurately predict health risks using deterministic methods (Tong et al., 2019). In comparison, probabilistic risk analysis may circumvent the shortcomings of the deterministic method and help in identifying the major factors for risk control. This approach has been successfully applied by various researchers in the cases of heavy metals (Yang et al., 2019), PAHs from agricultural soils (Tong et al., 2018), and VOCs during wooden furniture manufacturing (Tong et al., 2019).

Furthermore, internal exposure is increasingly regarded as an ideal alternative to assessing the health risk of various pollutants because it

might accurately reflect actual environmental exposure without omission. Several studies have reported a series of internal exposure surveys using urine under different occupational settings (Caro and Gallego, 2008; Chen et al., 2018; Gao et al., 2015; Huang et al., 2021; Lu et al., 2020). Within e-waste recycling premises, high concentrations of urinary metabolites of PAHs and phthalate esters as well as heavy metals have been reported (Zhang et al., 2019; Li et al., 2019; Lin et al., 2020; Wang et al., 2011). Although BTEX have a short half-life, profiles of BTEX excreta were reported from individuals of several occupations, including oil refineries, beauty salons, healthcare waste autoclaves, and printing industries (Mendoza-Cantu et al., 2006; Erb et al., 2019; Moradi et al., 2019; Rafiee et al., 2018). Importantly, the results revealed that urinary excretion concentrations are markedly correlated with external exposure levels. Although studies have shown that BTEX are present during the e-waste dismantling processes (An et al., 2014a; He et al., 2015; Chen et al., 2016a), it is unknown whether BTEX affect the health of workers. Furthermore, the measured levels of urinary metabolites can finally be transformed into an individual's daily intake of the corresponding parent compounds, and this might allow quantitative identification of unknown exposure pathways (Itoh et al., 2007).

Therefore, it is necessary to study atmospheric BTEX and the associated occupational health risks for occupational workers. This study focuses on the emission and health effects of representative aromatic hydrocarbon pollutants from a typical e-waste recycling industrial park. First, the speciated profiles of atmospheric BTEX in the park and the resulting external exposure factors of the uncertainty and variability by inhalation were investigated. Second, the internal exposure of BTEX was evaluated by analyzing the urinary concentration of unmetabolized BTEX and metabolites. Finally, the relationship between internal and external exposure levels was assessed, intending to validate the comprehensive occupational exposure assessment in the e-waste dismantling area.

2. Materials and methods

2.1. Reagents and materials

The chemicals and materials used in this study are described in [Supplementary Information \(SI\)-Section 1](#).

2.2. Sample collection

Atmospheric and urinary samples were collected from a formal e-waste dismantling industrial park in South China. Detailed information is provided in [SI-Section 2](#). Two to five sampling sites were selected within different functional areas to capture the pollution of VOCs adjacent to the dismantling activity. Considering the spatial heterogeneity of VOC concentration around the e-waste dismantling area, the 2 × 2 km grid box was further divided into 25 smaller grid squares, and one sampling site was selected in every square based on the field survey. Air samples were sampled at a height of 1.5 m during a working time. A total of 198 atmospheric samples were collected seasonally every three months from October 2018 to October 2019 in pre-cleaned and pre-evacuated Summa canisters (ENTECH Instruments Inc., Silonite TM) within and around the recycling area. Occupational workers in this industrial park were recruited and formally interviewed to obtain general demographics ([Table S1](#)). Morning urine samples of volunteers (n = 57) were collected and stored in screw-cap-sealed polypropylene vials. After immediately analyzing creatinine (Cre), the sampled urine was transported to the lab and stored at -80 °C until further analysis. This study was approved by the ethics committee of the Guangdong University of

Technology. All the volunteers were clearly informed of the purpose of this study, and written consent was obtained from all of them.

2.3. Sample preparation and chemical analysis

The methods of measuring analytes in atmospheric samples followed the reference (Chen et al., 2021). The urinary unmetabolized BTEX were analyzed using an automatic Purge and Trap sampler (Atomx, Teledyne Tekmar, USA) coupled with Agilent 7890B-5977 C GCMS with a DB-VRX column (20 m × 0.18 mm × 1.0 μm, Agilent Technology, CA, USA) (Brčić and Skender, 2003). For urinary metabolites, the extraction and cleanup procedures were modified from our previous study (Kuang et al., 2019), and analysis was conducted using an ultra-high performance liquid chromatography system (Thermo Ultimate 3000) interfaced to triple quadrupole mass spectrometry (Thermo TSQ Endura). Cre was determined using a spectrophotometric method immediately to adjust the internal exposure of each individual. Detailed analysis parameters are provided in SI-Section 3.

2.4. Quality assurance and quality control

The collection and analysis procedure of atmospheric samples followed the references (An et al., 2014a; Chen et al., 2021). The method detection limits were determined by running 100 mL 1 ppbv standards seven times and multiplying by 3.14 with the range of 0.09–0.25 μg·m⁻³. Instrumental quality control (solvent blanks and standard solutions) and method quality control (procedural blanks and spiked matrices) were conducted with each batch of 10 field samples to ensure the analysis accuracy. The target analytes were confirmed at below 5 % in the blanks. Quantification was based on a multipoint calibration curve of standard solutions ($R^2 > 0.99$). The values of limits of quantification (LOQ) were 0.04 ng·mL⁻¹ for BTEX and 0.101–52.6 ng·mL⁻¹ for metabolites. The recovery of the target compounds in the standard spiked samples was 88.8–108% for BTEX and 83.3–139 % for metabolites (Table S2). The concentrations of target compounds in the field samples were not corrected by the recovery, but the corresponding procedural blank within the same batch.

2.5. Risk assessments and statistical analysis

Two occupational exposure scenarios for the occupational workers were considered for the health risk assessment. Indirect health impacts of BTEX in contaminated air via inhalation were obtained from USEPA (USEPA, 2009) as well as a previous study (Huang et al., 2021). Direct exposure assessment based on the urinary metabolites was calculated as per Fan et al (Fan et al., 2014). Hazard quotient (HQ) was applied to assess the non-carcinogenic health risk. Carcinogenic risk (CR) was applied to evaluate the incremental probability of an individual developing cancer over a lifetime. Probabilistic health risk assessment was conducted using Monte-Carlo simulation for the trials with a 95 % confidence interval (CI). Further information can be found in SI-Section 4 and Tables S3 and S4.

The statistical differences in atmospheric BTEX levels in different areas were evaluated using the Mann-Whitney *U* test. Spearman's test (two-tailed) was applied to assess the correlations among the analyte concentrations. Concentrations that were not detected were assigned a value of zero, and below LOQ were replaced by values equal to half of the LOQ for statistical analysis, respectively. A *p*-value < 0.05 was considered significant.

3. Results and discussion

3.1. Occurrence of atmospheric BTEX and metabolites

BTEX were detected in all atmospheric samples with total concentrations ranging from 3.51 to 148 μg·m⁻³ (median: 23.6 μg·m⁻³) within

the e-waste dismantling area. These values were significantly higher than those from the surrounding area (ranging from 0.536 to 854 with a median of 8.17 μg·m⁻³) (*p* < 0.001) (Table S5). As reported in our previous studies, various aromatics were released during the WPCB dismantling process (An et al., 2014a; Chen et al., 2016a). The range of BTEX concentrations in the surrounding areas was much higher than that within the dismantling area, which may be attributed to the high fluctuation of xylene. This demonstrates a high dispersion degree for BTEX in the surrounding areas, thereby suggesting more complex sources other than transmissions from the industrial park. A previous study demonstrated that e-waste dismantling and gasoline evaporation contribute to 50 % of the total VOCs, and these two emission sources steadily release VOCs, thus explaining lower standard deviations for BTEX concentrations within the dismantling area (Chen et al., 2021).

The mean BTEX level (8.95 ± 7.23 ppbv) in the present study was comparable with those from the industrial area (7.80 ppbv) (An et al., 2014b), oil/gas station (3.41 ppbv) (Zheng et al., 2018), petrochemical industry (3.07 ppbv) (Zheng et al., 2020), and the ship channel (2.03 ppbv) (Leuchner and Rappenglück, 2010) (Table S6). This indicates that the e-waste recycling industrial park has been largely affected by BTEX. Furthermore, the mean BTEX value (35.2 μg·m⁻³) (Table S5) within the e-waste recycling area was higher than that observed in an e-waste dismantling region (27.8 μg·m⁻³) previously (Chen et al., 2021). Thus, subsequent exposure risk occurs within the industrial park where the occupational workers were present during their worktime.

The highest concentration of toluene was found (median: 10.2 μg·m⁻³) to be almost 3–6 folds higher than of the other constituents (1.71–3.68 μg·m⁻³) within the e-waste dismantling area. A similar trend was observed in the surrounding areas, but the concentrations were significantly lower than those within the e-waste dismantling area (*p* < 0.001). The emission of toluene made a great contribution to the pollutant levels detected in dismantling various WPCBs (Chen et al., 2016b) and at plastic waste recycling granulation plants (Huang et al., 2013). The studies from other occupations, i.e., petrol station and beauty salons, also showed that toluene had the highest concentration (Moradi et al., 2019; Campo et al., 2016; Mukherjee et al., 2015). The toluene concentration in the present work was comparable to that in an enclosed multistorey car park (28.8 μg·m⁻³) (Taneepanichsku et al., 2017) but lower than that in beauty salons (98.5 μg·m⁻³ in indoor air) (Moradi et al., 2019).

Urinary unmetabolized benzene had the highest detection frequency (96.5 %), followed by ethylbenzene and *m,p*-xylene with the detection frequencies of 64.9 % and 59.6 %, respectively (Table 1). In comparison, toluene and *o*-xylene were detected with frequencies lower than 40 %. The concentration of urinary unmetabolized ∑₆BTEX ranged from a below detection limit to 6.72 μg·L⁻¹ (5.17 μg·g⁻¹ Cre) with a median value of 0.194 μg·L⁻¹ (0.210 μg·g⁻¹ Cre). The median urinary BTEX concentration in this study was nearly 40-fold lower than that for autoclave operators (7.69 μg·L⁻¹) (Rafiee et al., 2018). The highest concentration among studied compounds was found for benzene (median: 5.63 × 10⁻² μg·L⁻¹), followed by ethylbenzene and *m,p*-xylene with median values of 2.00 × 10⁻² and 2.00 × 10⁻² μg·L⁻¹, respectively. This is, however, contrary to the findings from other occupational settings, in which toluene and *o*-xylene accounted for the highest levels in urinary samples (Moradi et al., 2019; Rafiee et al., 2018). The inconsistencies might be explained by different exposure levels and endogenous metabolic rates under those conditions. Unmetabolized median benzene level in e-waste recycling industrial park workers was comparable to those in beauty salons (0.06 μg·L⁻¹) (Moradi et al., 2019) and oil refinery (0.07 μg·L⁻¹) (Erb et al., 2019) but significantly lower than those in healthcare waste autoclaves (2.04 μg·L⁻¹) (Rafiee et al., 2018) and petrol stations (0.34 μg·L⁻¹) (Campo et al., 2016).

Nine urinary metabolites of BTEX were detected with a frequency of 100 %, while N-acetyl-S-phenyl-L-cysteine (S-PMA) was detected in 94.7 % of samples (Table 1), indicating the ubiquitous occurrence of BTEX metabolites in occupational workers in e-waste recycling industrial

Table 1

Urinary concentrations of unmetabolized BTEX and metabolites within the occupational workers in the e-waste park.

Compounds	Concentration percentile			Median ^b	Mean ^b	Range ^b	DF ^b
	Median ^a	Mean ^a	Range ^a				
Benzene	5.63×10^{-2}	9.17×10^{-2}	n.d. – 0.535	5.62×10^{-2}	8.43×10^{-2}	n.d. – 0.644	96.5
Toluene	n.d.	0.467	n.d. – 6.19	n.d.	0.352	n.d. – 4.76	38.6
Ethylbenzene	2.00×10^{-2}	5.71×10^{-2}	n.d. – 0.388	2.44×10^{-2}	5.27×10^{-2}	n.d. – 0.254	64.9
<i>m,p</i> -Xylene ^c	2.00×10^{-2}	0.288	n.d. – 1.75	2.80×10^{-2}	0.198	n.d. – 1.25	59.6
<i>o</i> -Xylene	n.d.	9.67×10^{-2}	n.d. – 0.734	n.d.	7.17×10^{-2}	n.d. – 0.741	21.1
Σ U-BTEX	0.194	1.00	n.d. – 6.72	0.210	0.758	n.d. – 5.17	98.2
<i>t,t</i> -MA	236	372	29.3 – 2.37×10^3	224	289	19.5 – 1.24×10^3	100
CA	2.85×10^3	2.82×10^3	559 – 4.53×10^3	2.36×10^3	3.05×10^3	192 – 1.12×10^4	100
S-PMA	1.38	3.32	n.d. – 19.2	1.11	2.39	n.d. – 10.1	94.7
S-BMA	48.5	72.2	5.95 – 521	41.2	55.4	10.6 – 319	100
HA	2.95×10^5	3.96×10^5	1.58×10^4 – 2.50×10^6	2.03×10^5	3.84×10^5	2.41×10^4 – 3.58×10^6	100
PGA	1.81×10^3	2.42×10^3	38.4 – 1.46×10^4	1.57×10^3	1.73×10^3	25.1 – 7.61×10^3	100
MA	3.59×10^3	3.90×10^3	484 – 1.55×10^4	2.76×10^3	3.20×10^3	345 – 1.58×10^4	100
2-MHA	328	658	38.1 – 5.56×10^3	304	511	15.1 – 2.24×10^3	100
3/4-MHA ^d	1.07×10^3	3.21×10^3	167 – 3.24×10^4	902	2.37×10^3	113 – 1.31×10^4	100
Σ Metabolites	3.01×10^5	4.09×10^5	2.04×10^4 – 2.50×10^6	2.20×10^5	3.95×10^5	2.78×10^4 – 3.59×10^6	100

DF: detection frequency; n.d.: not detected; *t,t*-MA: trans, trans-muconic acid; CA: catechol; S-PMA: N-acetyl-S-phenyl-L-cysteine; S-BMA: N-acetyl-S-benzyl-L-cysteine; HA: Hippuric acid; PGA: Phenylglyoxylic acid; MA: Mandelic acid; MHA: Methylhippuric acid; a Concentrations of excreta in urine (ng·mL⁻¹), respectively; b Concentrations of excreta in urine (μg·g⁻¹ Cre), respectively; c *m,p*-Xylene: both *m*-Xylene and *p*-Xylene could not be separated chromatographically and were quantified together; d 3/4-MHA: both 3-MHA and 4-MHA could not be separated chromatographically and were quantified together.

park. Compared with those of unmetabolized BTEX, these metabolites had much higher detection frequencies. Additionally, the concentrations of total metabolites ranged from 2.78×10^4 to 3.59×10^6 μg·g⁻¹ Cre with a median value of 2.20×10^5 μg·g⁻¹ Cre, which was almost 6-orders of magnitude higher than those of unmetabolized BTEX (0.210 μg·g⁻¹ Cre). This demonstrates that BTEX absorbed by the workers exist in the form of metabolites at that occupational level, leading to accumulated health risks during the process of eliminating BTEX. Unmetabolized BTEX in urine may represent specific indicators for the evaluation of their bioavailability. Many studies have reported the urinary metabolites of BTEX, and all show that hippuric acid (HA) is the dominant excretion in occupational workers exposed to BTEX (Mukherjee et al., 2015; Taneepanichsku et al., 2017; Kim et al., 2011). The median concentration of HA in workers from the e-waste recycling industrial park was 2.03×10^5 μg·g⁻¹ Cre, which was 1-order of magnitude higher than that of occupationally exposed workers in the car park (2.74×10^4 μg·g⁻¹ Cre) (Taneepanichsku et al., 2017). Furthermore, the results were comparable to those from shipbuilding (Kim et al., 2011) and higher than those reported in traffic areas and petrol pump (Mukherjee et al., 2015). Our results further illustrated that working in this e-waste recycling industrial park induced a significant exposure risk caused by BTEX exposure.

3.2. Composition profiles of atmospheric and urinary excretive BTEX

Differences existed in the isomer profiles of BTEX between

atmospheric and urinary samples (Fig. 1). Toluene was the most predominant atmospheric congener, accounting for 42.9 % and 44.3 % in the e-waste dismantling area and surrounding area, respectively. However, these results were inconsistent with the observations made during the e-waste dismantling processes (An et al., 2014a; Chen et al., 2016a; Liu et al., 2017). This might be attributed to the fact that BTEX from the fresh emission sources were associated with different disposal conditions. The redistribution may greatly influence the atmospheric compositional profiles of freshly emitted BTEX during their transportation. Additionally, the implementation of purification technology might alter the profile of atmospheric BTEX while effectively reducing VOCs emitted from thermal processes (Chen et al., 2016a; Liu et al., 2017).

A decreasing order of airborne exposure to toluene, *m,p*-xylene, benzene, ethylbenzene, and *o*-xylene was observed in occupational settings. In contrast, the urinary excretion order was benzene, ethylbenzene, and *m,p*-xylene for unmetabolized BTEX in urine, accounting for 51.7 %, 25.8 %, and 22.5 %, respectively. In comparison, the excretion pattern of urinary metabolites followed the order toluene > ethylbenzene > benzene > *m,p*-xylene > *o*-xylene. HA was the most extremely abundant isomer, accounting for 96.1 % among all metabolites. The other metabolites accounted for < 4 % of the total metabolites. This was inconsistent with previous findings in beauty salons (Moradi et al., 2019), healthcare waste autoclaves (Rafiee et al., 2018), petrol pumps, and traffic zones (Mukherjee et al., 2015). Additionally, catechol concentration was higher than that of the other two metabolites, which

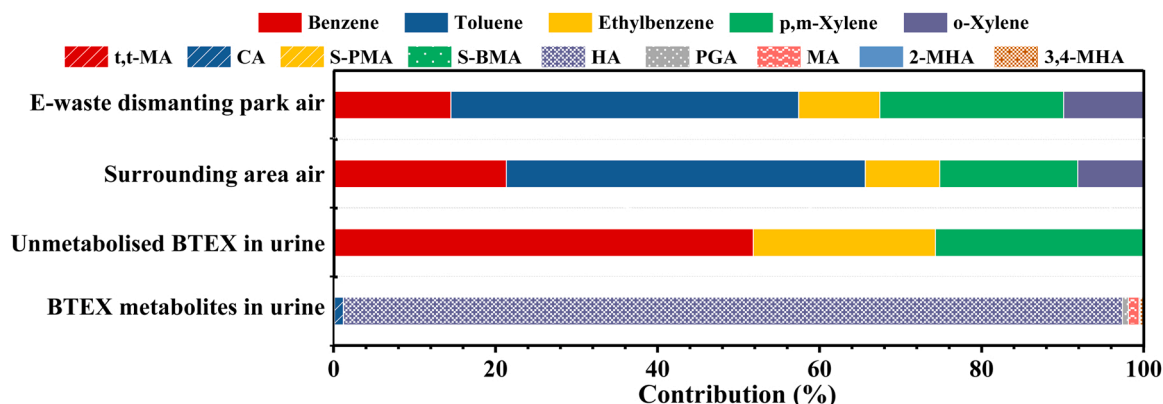


Fig. 1. Composition profiles of atmospheric BTEX and corresponding urinary excretion of the exposed workers.

is inconsistent with a previous study (Mukherjee et al., 2015). The different excretion profiles of urinary metabolites may rely on the different VOC exposure levels. For benzene exposure, median values of dose-specific levels of total metabolites were reduced by approximately 9-folds between 0.03 and 88.9 ppm, except for S-PMA (Kim et al., 2006).

3.3. Correlation between external and internal exposure

The correlations among individual atmospheric BTEX, urinary BTEX, and metabolites collected from the industrial park were examined using Spearman correlation analysis. Significant positive correlations were found between atmospheric individual BTEX ($r = 0.808\text{--}0.976$, $p < 0.01$), indicating the existence of the same source of these compounds, i.e., the emission from the e-waste disposal processes (Table S7). Large amounts of BTEX were discharged into the surrounding environment because of primitive e-waste recycling activities (An et al., 2014a; He et al., 2015; Chen et al., 2016b). In addition, significant positive relationships were observed in 5 out of 10 urinary BTEX ($r = 0.324\text{--}0.702$, $p < 0.05$) and 26 out of 36 metabolites ($r = 0.268\text{--}0.945$, $p < 0.05$), implying that the sources of workers' exposure to BTEX in this industrial park were common or related (Table S8).

The association between the concentrations of atmospheric BTEX and urinary excretion was further analyzed in the participants who were directly engaged in the e-waste recycling area ($n = 30$). Significant positive relationships ($p < 0.05$) between N-acetyl-S-benzyl-L-cysteine (S-BMA) and individual atmospheric BTEX were observed ($r = 0.479\text{--}0.626$) (Fig. 2). Among the biomarkers of the other urinary excreta, nothing was observed following the occupational exposure, indicating these compounds were poor markers for assessing exposure to BTEX in the e-waste dismantling site. These results demonstrate that S-

BMA may act as a qualified indicator of occupational BTEX exposure in the e-waste recycling area.

The findings indicating a significant correlation between urinary S-BMA and individual atmospheric BTEX found in the e-waste recycling area were inconsistent with those found in petrol pump workers and petrol station attendants, in which atmospheric benzene is correlated significantly with trans, trans-muconic acid (*t,t*-MA) and post-shift urinary benzene of exposed workers, respectively (Campo et al., 2016; Mukherjee et al., 2015). These results demonstrate that occupational workers exposed to different levels of BTEX might present specific metabolism. BTEX can be metabolically activated by cytochrome P450 enzymes, which might produce metabolic interactions in BTEX mixtures (Cheng and Bois, 2011). Furthermore, the biotransformation potential of occupational exposure to benzene is suppressed by co-exposure to toluene and vice versa (Inoue et al., 1988). However, no modification in the metabolism is induced by the combined exposure when the exposure level is low (Kim et al., 2006; Kawai et al., 1992). A previous study reported that the intake of high levels of sorbic acid originating from food may also lead to significant correlations between benzene exposure and *t,t*-MA in urine (Hoet et al., 2009). Moreover, the inhalation pathway of BTEX is not solely dependent on atmospheric levels. When the exposure levels of aromatic hydrocarbons decreased, the respiratory absorption factors also decreased in a logarithmic manner (Huang et al., 2017).

In this study, no significant correlation was found between urinary BTEX concentrations and body mass index or age, and similar result was also found for metabolite (Table S9). However, unmetabolized urinary toluene, *m,p*-xylene, and *o*-xylene were weakly positively correlated with working time ($p = 0.005$, 0.031 , and 0.008 , respectively). Additionally, *t,t*-MA was negatively correlated with working time ($p = 0.028$). Thus, occupational exposure might result in age-related accumulation in urinary excreta of BTEX exposure of such

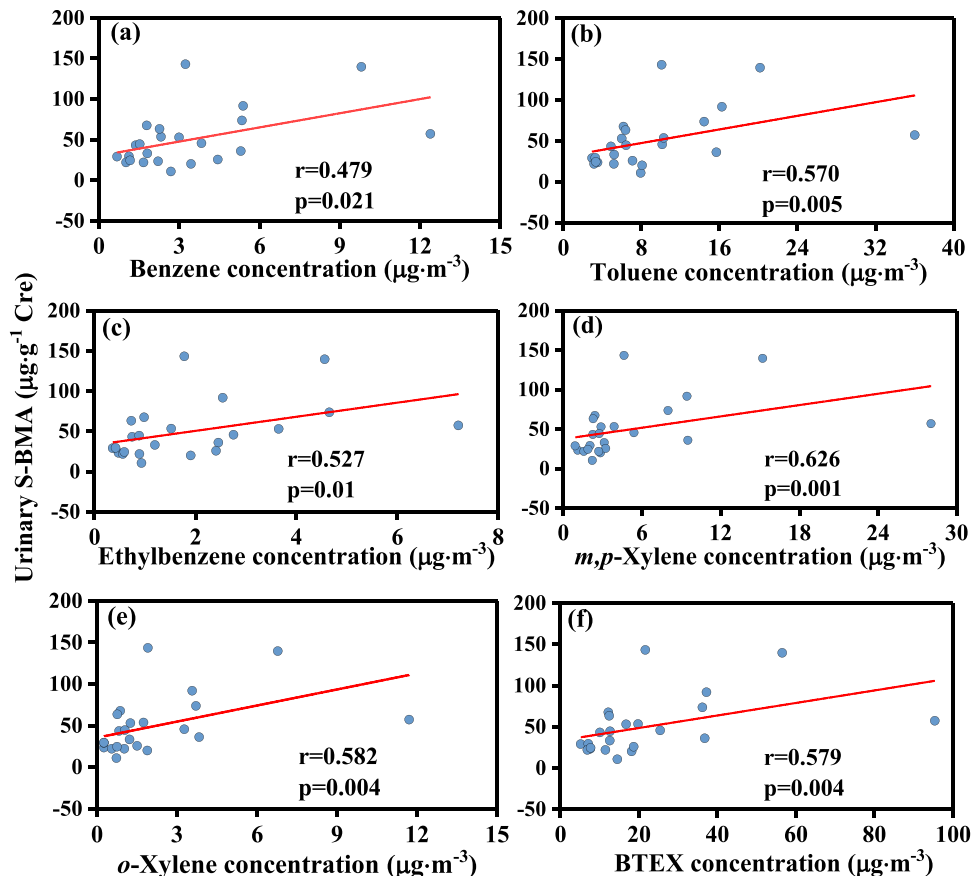


Fig. 2. Exposure to BTEX and relative excretion of S-BMA evaluated by simple linear regression analysis.

occupational workers. Furthermore, smoking is a non-ignorable factor when evaluating BTEX exposure (Rafiee et al., 2018; Taneepanichsku et al., 2017). To determine the influence of smoking on BTEX exposure in the industrial park, the differences in urinary BTEX excreta levels between non-smokers and smokers were also evaluated (Fig. S1). Urinary toluene and *o*-xylene were not examined due to their very low concentrations. Significant differences were observed in urinary *m*, *p*-xylene concentrations between non-smoker and smoker occupational workers but not in other urinary BTEX excreta levels. This demonstrates that smoking is an unimportant contributor to BTEX exposure in the e-waste recycling industrial park. The results of this study were consistent with those of previous studies (Rafiee et al., 2018; Fustinoni et al., 2011), further confirming that smoking is an unimportant contributor among occupational exposure populations. This result suggests that strong BTEX emissions from occupational conditions reduce the effect of smoking habits.

3.4. Human exposure and health risk assessment

Both non-CR and CRs were computed to assess the health risks posed to the workers by external exposure to atmospheric BTEX (Table 2). For non-CR, the mean HQ value of BTEX for occupational workers in the e-waste industrial park was < 1.0. The basic trend of the mean HQ value was benzene > *p*-xylene > *o*-xylene > *m*-xylene > ethylbenzene > toluene. Additionally, the probability of all the BTEX compounds was below the acceptable level (i.e., 1.0), while 7.09 %, 0.18 %, 0.03 %, and 0.05 % of HQ for benzene, *p*-xylene, *m*-xylene, and *o*-xylene, respectively, exceeded a possible non-CR of 0.1 (Fig. S2). Overall, the mean total hazard index (HI) for the occupational workers was found 6.51E-2 (95 % CI: 2.38E-2 – 1.41E-1), which was significantly lower than the USEPA guideline value, 1.0. Further, the probability value of HI in this study surpassing 0.1 was 14.4% (Fig. 3a). Thus, the results indicate that inhalational exposure to BTEX in this e-waste recycling industrial park would result in possible non-carcinogenic hazards for occupational workers.

Furthermore, for CR, it was observed that the mean total cancer risk (TCR) to the workers attributed to the inhalation pathway was 3.13E-6 (95 % CI: 1.01E-6 – 7.09E-6) (Table 2). Benzene and ethylbenzene almost had equal shares of TCR, with the mean CR values of 1.67E-6 (95 % CI: 3.96E-7 – 4.38E-6) and 1.48E-6 (95 % CI: 1.01E-6 – 7.09E-6), respectively. As shown in Fig. S3, for benzene and ethylbenzene, 60.6 % and 50.6 % of their probabilistic risk, respectively, exceeded the

Table 2
Summary statistics for health risk results via Monte Carlo simulation.

Risk	VOCs	Matric	Mean (median)	SD	95 % CI
HQ	Benzene		4.43E-2 (3.28E-2)	3.94E-2	(1.09E-2, 1.15E-1)
			8.63E-4 (6.12E-4)	8.41E-4	(1.78E-4, 2.41E-3)
	Ethylbenzene		1.02E-3 (7.09E-4)	1.06E-3	(1.62E-4, 2.90E-3)
			9.25E-3 (5.60E-3)	1.20E-2	(1.07E-3, 2.88E-2)
	<i>m</i> -Xylene		4.64E-3 (2.59E-3)	7.51E-3	(3.89E-4, 1.51E-2)
	<i>o</i> -Xylene	Air	5.67E-3 (3.39E-3)	7.34E-3	(6.05E-4, 1.81E-2)
HI	Total		6.51E-2 (5.34E-2)	4.30E-2	(2.38E-2, 1.43E-1)
CR	Benzene		1.67E-6 (1.22E-6)	1.50E-6	(3.96E-7, 4.38E-6)
			1.48E-6 (1.01E-6)	1.55E-6	(2.37E-7, 4.23E-6)
TCR	Total		3.13E-6 (2.55E-6)	2.13E-6	(1.01E-6, 7.09E-6)
CR	Benzene	Urine	6.10E-5 (2.04E-5)	1.82E-4	(2.61E-6, 2.23E-4)

acceptable level of 1.0E-6. Overall, 95.2 % of TCR was greater than 1.0E-6 (Fig. 3b), confirming that occupational workers in the e-waste dismantling industrial park are exposed to a high level of inhalation-related cancer risk during their working hours. Thus, monitoring benzene and ethylbenzene concentrations in the occupational exposure environment is important for assessing the CR in future studies. However, the authentic unit risk values for some BTEX are not available and may underestimate the aggregate risks in this study.

To determine the internal exposure health risk of tested e-waste workers to BTEX, daily intake level based on the urinary concentrations of their metabolites were tested. Only the risk of benzene exposure was evaluated in the present study. This was because no percentage data were available for other BTEX excreted through urine. Additionally, urinary S-PMA was chosen as a specific biomarker in urine to determine the risk of benzene exposure. As shown in Fig. 4 and Table 2, benzene had the mean CR value of 6.10E-5 (95 % CI: 2.61E-6 – 2.23E-4), which was approximately 60 times higher than the acceptable level of 1.0E-6. In addition, 99.9 % of the CR distribution based on S-PMA exceeded 1.0E-6. This further confirmed that workers in the e-waste dismantling industrial park were exposed to considerable BTEX levels, besides other hazardous pollutants (Zhang et al., 2019; Li et al., 2019; Lin et al., 2020; Wang et al., 2011). When compared to the risk assessment via external exposure of inhalation, a much higher level was for internal exposure. This suggests that the unassigned exposure pathway might be attributable to dermal uptake because of the lipophilic nature of these compounds, contributing to the internal dose in addition to inhalation. This has not been confirmed yet, but the following suggestive evidence has been reported for other VOCs. Previously, a finding pointed out that the contribution of dermal exposure was predictive of the urinary metabolites of styrene derived from thermoplastic panels (Creta et al., 2018). Furthermore, the urinary trihalomethane uptake of swimmers was higher than that of workers in their study (Caro and Gallego, 2007). Thus, the assessment of dermal exposure in workplaces is intended to have a high added value besides the atmospheric and biological monitoring. The risk assessment results in this work emphasize that the increased health risks caused by BTEX exposure in polluted air for the workers should not be ignored.

Fig. S4 presents the sensitive assessment results of critical parameters of non-CR and CR estimates caused by external exposure for occupational workers in the e-waste dismantling industrial park. The concentration of benzene (C_{benzen}) contributed the most to the HI sensitivity (0.85), followed by $C_{p\text{-xylene}}$, $C_{o\text{-xylene}}$, exposure time, and $C_{m\text{-xylene}}$ with sensitivities of 0.27, 0.19, 0.16, and 0.16, respectively. Exposure frequency, $C_{\text{ethylbenzene}}$, and C_{toluene} contributed less than 0.1 to the HI. For TCR, C_{benzen} and $C_{\text{ethylbenzene}}$ were the most influential parameters with the same value of 0.65, while much lower sensitivities were observed for other variables with the following trend: exposure time (0.14) > exposure frequency (0.060). Similarly, the derived sensitivity demonstrated that C_{toluene} has stronger influence on the CR of individual benzene (Fig. S5). On combining the external and internal exposure results, C_{benzen} was selected as the top prior parameter for both high non-carcinogenic and carcinogenic risks. This comprehensive research on BTEX levels can substantially aid future studies to refine occupational exposure criteria. Some countermeasures by controlling the concentration of benzene could greatly mitigate the health risk caused by BTEX exposure.

It is important to note that there were uncertainties and limitations for the exposure assessment in the present study. First, grab sampling of atmospheric BTEX might increase bias owing to the inevitable fluctuation of BTEX levels, processing techniques, raw materials, and other contextual factors. It would be better to establish a harmonious sampling protocol for future regional pollution and risk evaluation. In addition, temporal variability of urinary levels derived from the short elimination half-lives of BTEX metabolites would not rule out the possibility of chances in biomarker excretion within the exposure time. Because of practical issues, only urine spot samples were collected. In an ideal

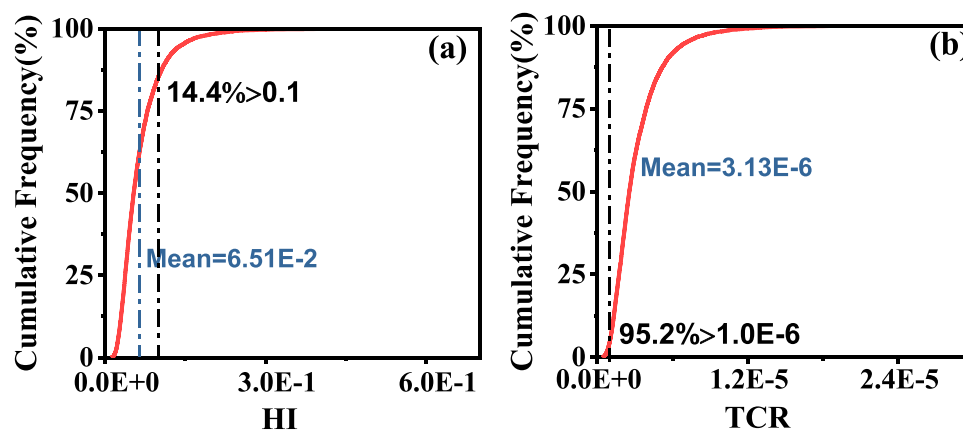


Fig. 3. Probability distribution of HI (a) and TCR (b) (the blue dashed vertical lines presented the mean values whereas the black presented the guideline values that of 0.1 and $1.0E-6$ for non-cancer and cancer risk, respectively).

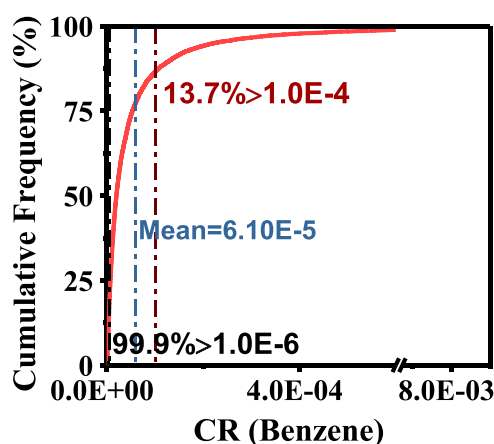


Fig. 4. Probability distribution of CR based on S-BMA (the blue dashed vertical lines presented the mean values whereas the black presented the guideline values that of $1.0E-6$).

manner, urine should be sampled at different time spots, allowing evaluation of kinetics and possible accumulation over the working time. Second, the internal exposure results are prone to uncertainty because of the potential interactions of multiple pollutants. Concentration-addition effects have been reported for A549 cells exposed to BTEX at different concentration ratios (Liu et al., 2014). However, an inhibiting effect between different chemicals could overestimate the total health risks to some extent if the estimation of each constituent was merely added together (Inoue et al., 1988). Finally, uncertainty might arise from the choice of exposure parameters. The inclusion of the respiratory coefficient into the exposure assessment may help obtain a reliable internal dose. However, this also has uncertainties with the assumption of constant transdermal coefficient, which could vary significantly among exposure levels (Huang et al., 2017). Thus, further risk studies should be conducted considering these points to better understand the unequivocal trend of health risks.

4. Conclusions

The pollution profiles of atmospheric and urinary excreta of BTEX were comprehensively investigated, and corresponding health risks were evaluated in an e-waste dismantling industrial park. High levels of urinary metabolites demonstrated the ubiquitous exposure of workers to these pollutants. Significant differences were found in the congener profiles of BTEX between external and internal exposure. Moreover, the orders of magnitude difference between urinary BTEX and their

metabolites showed that humans metabolize these pollutants efficiently at those occupational levels, leading to the accumulation of health risks during the process of BTEX elimination. Additionally, a specific correlation of urinary S-BMA concentration with corresponding environmental BTEX levels suggested that S-BMA might be a marker of BTEX exposure for e-waste recycling processes. Furthermore, a combination of probabilistic risk simulation based on inhalational exposure and urinary metabolites indicated that the workers face potential health risk from BTEX exposure, for which atmospheric benzene concentration should be particularly considered an indicator parameter.

Environmental implication

Atmospheric BTEX and urine samples were collected to explore external and internal loadings as well as the relationships between external and internal for e-waste recycling workers. Urinary metabolites levels were 6-orders of magnitude higher than unmetabolized BTEX. Atmospheric toluene, urinary unmetabolized benzene and HA were dominated. S-BMA positively correlated with atmospheric individual BTEX. 95.2 % of workers showed a cumulative carcinogenic risk induced by BTEX exposure via inhalation, while biomonitoring exposure to benzene showed 99.9 % cumulative carcinogenic risk. Our study indicates that BTEX exposure is non-negligible, which should be considered in future exposure and risk assessments.

CRedit authorship contribution statement

Ranran Liu: Methodology, Formal analysis, Writing – original draft. **Shengtao Ma:** Methodology. **Daijin Chen:** Methodology, Formal analysis. **Guiying Li:** Writing – reviewing & editing. **Yingxin Yu:** Visualization, Investigation. **Ruifang Fan:** Visualization, Investigation. **Taicheng An:** Conceptualization, Supervision.

Declaration of Competing Interest

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

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.129343.

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