Research article

Cutting down on the ozone and SOA formation as well as health risks of VOCs emitted from e-waste dismantlement by integration technique

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ABSTRACT

Elimination of volatile organic compounds (VOCs) in the e-waste dismantling industry by an integration technique of spray tower-electrostatic precipitation-photocatalysis was conducted to investigate its application possibility for reducing formation of O_3 and secondary organic aerosols (SOAs) as well as exposure risk. Results revealed the average $5.4 \times 10^2 \, \mu g \, m^{-3}$ of VOCs with the top two groups being aromatic hydrocarbons (AHs, 55.93%) and halogenated hydrocarbons (HHs, 33.33%), contributing to 1.3×10^3 and $3.0 \times 10^4 \, \mu g \, m^{-3}$ of the O_3 and SOA (OFP and SOAFP) formation potential, respectively. Furthermore, 86.47% of OFP and 99.87% of SOAFP were ascribed to AHs, in which toluene ranked first (35.30% and 48.07%). The highest removal efficiency (76.92%) for VOCs by the integrated technique resulted in excellent prevention efficiencies of OFP (71.54%) and SOAFP (80.62%). Occupational cancer risk assessment found that HHs (62.63%) and AHs (36.93%) were the top two contributors. After the treatment by the integrated technique, 55.44% of the total risk index was reduced with the accumulation of few low-concentrated and more toxic AHs (e.g. $6.6 \, \mu g \, m^{-3}$ benzene on average). All results suggest that controlling AH and HH emissions from the e-waste dismantling source could efficiently prevent atmospheric secondary pollution and human exposure risk to industrial emission.

1. Introduction

Volatile organic compounds (VOCs) consist of a diversity of organic species which are critical for increasing the O_3 atmospheric concentration and the formation of secondary organic aerosols (SOAs) (Kumar et al., 2018; Suarez-Bertoa et al., 2015; Wu and Xie, 2017; Wu et al., 2017; Ziemann and Atkinson, 2012). The formed O_3 and SOAs may affect the climate and have adverse effects on the ecosystem (Trivitayanurak and Adams, 2013; Wu et al., 2017). Apart from the important role in atmospheric chemistry, VOCs can also have direct and indirect negative effects on human health (Kumar et al., 2018). Given the significantly adverse effects on both the atmospheric environment and public health, ambient VOC measurements and their source apportionment are currently the subjects of grave concern among environmental scientific research (Huang et al., 2014; Kumar et al., 2018; Wu and Xie, 2017; Zhang et al., 2017; Zheng et al., 2009). Industrial processes (Kwon et al., 2015; Tsai et al., 2008; Wu and Xie, 2017; Zheng et al., 2013), biomass burning (Wang et al., 2014a), fossil fuel combustion (Wang et al., 2018), solvent usage (Kumar et al., 2018; Wang et al., 2014b) and vehicular exhaust (Kumar et al., 2018; Wu and Xie, 2017; Zheng et al., 2009) are known as the top five anthropogenic sources of VOCs. However, to date, the understanding of VOC emission from those representative sources, including the e-waste dismantling industry, is still incomplete.

In 2017, the global generation of e-waste waste was approximately 65.4 million tons (Breivik et al., 2014). Furthermore, greater than 70% of this e-waste is delivered to China, where its recycling is recognized as an important resource management strategy due to its huge economic contribution. However, safety concerns arise because these materials contain heavy metals and chemical products that are hazardous to human health and the environment (Guo et al., 2015; Lu et al., 2016; Xiao et al., 2016; Zhou et al., 2014). In our previous studies, VOC emission from e-waste dismantling processes was found to be a predominant contributor of anthropogenic VOCs in some locations (An et al., 2014; Chen et al., 2016a; Liu et al., 2017). Moreover, these emitted VOCs are important precursors for O_3 and SOAs (Shao et al.,

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2009b). Nevertheless, there is still lack of valuable data relating to O_3 and SOA formation from VOCs discharged from e-waste dismantling industry sources.

Since the O_3 and SOA formation process involves a series of photochemical reactions between VOCs and other atmospheric pollutants (Ding et al., 2017; Wu and Xie, 2017; Ziemann and Atkinson, 2012), they can be theoretically avoided by eliminating VOCs. Unfortunately, limited work has focused on VOC elimination in the e-waste recycling industry to avoid O_3 and SOA formation. In our early work, a middle scale spray tower-electrostatic precipitation-photocatalysis integrated technique demonstrated an excellent VOC elimination ability and subsequent risk reduction from the e-waste dismantling process (Liu et al., 2017). Moreover, the characterization of this decontamination technology only focuses on VOC concentration control and risk reduction. If these VOCs were directly emitted to the atmosphere, they would quickly participate in thousands of photochemical reactions that ultimately form O_3 and SOAs (Huang et al., 2017; Timonen et al., 2017; Ziemann and Atkinson, 2012). Therefore, to alleviate the formation potential of O_3 and SOAs associated with industrial source VOC emissions, the reactivity-based control approach might be more efficient than traditional concentration-based emission control approaches (Avery, 2006; Luecken and Mebust, 2008).

Thus, in this work, the industrial scale spray tower-electrostatic precipitation-photocatalysis integrated equipment with a large treatment capacity of $6.0 \times 10^4 \, \mathrm{m^3 \, h^{-1}}$ was applied for e-waste dismantling emitted waste gas treatment. Firstly, the emission profile of VOCs from the e-waste dismantling process, as well as their contributions to O_3 and SOA formation, were evaluated to understand their quantitative relationship. Then, the ability to prevent these secondary pollutants forming from VOCs by the integrated large-scale reactor was measured. The elimination of VOCs to protect the formation of O_3 and SOAs were discussed in detail. Overall, the main objective of this study was to determine whether, and how, the elimination of VOC emissions affects the formation of O_3 and SOAs, as well as their quantitative contribution percentage variations in the e-waste recycling industry. In addition, organics associated with VOCs have already been proven toxic, posing various threats to humans (Rohr, 2013; Su et al., 2014). Moreover, our previous work indicated that the exposure of e-waste recycling emitted VOCs in the work environment might lead to chronic health effects, such as cancer (Chen et al., 2016b). Thus, occupational cancer risk of the workers exposed to VOCs was also investigated during the 180-day operation period before and after the treatment by the integrated technique.

2. Experimental

2.1. Equipment description

The experiment was performed on the roof of an e-waste dismantling building with twenty e-waste dismantling workshops from July 2015 to January 2016 in a small town in southern China, where printed circuit boards from television waste were manually dismantled by electric heating furnaces. The furnace usually offers an environmentally friendly alternative for processing due to the low incubation period (2 min) and heating temperature (260 °C). More information on e-waste and dismantling techniques can be found in our previous research (An et al., 2011, 2014; Zhang et al., 2011). To efficiently prevent the formation of both O_3 and SOAs, as well as reduce the risk from VOCs, an integrated reactor using spray tower-electrostatic precipitation-photocatalysis was designed (Fig. 1). The spray tower unit (6000 mm in height and 3200 mm in diameter) was artificially divided into four parts with each part filled to 500 mm height with commercial raschig rings. For the electrostatic precipitation unit, a total of 28 electrostatic dust collectors were equipped in a hermetically sealed stainless-steel box (13000 mm × 2500 mm × 2500 mm). Subsequently, the photocatalysis reactor consisted of 20 pieces of

microporous nickel foam (1000 mm × 1250 mm), coating with commercial TiO_2. In the photocatalysis (PC) reactor, 128 UV-lamps with a maximum wavelength at 254 nm and a minor emission at 185 nm were installed parallel with the vertical distance between two lamps being 200 mm. The UV-lamp was placed 10 cm away from the photocatalytic medium. Detailed parameters of the equipment are supplied in Table S1. The flow rate of the waste gas through the integrated equipment was $60000 \, \mathrm{m^3 \, h^{-1}}$, leading to residence times of 0.72 and 4.9 s in the spray tower and electrostatic precipitation-photocatalysis units, respectively.

2.2. Sampling and analytical procedure

VOCs were collected at the inlet and outlet of the spray tower unit, as well as at the outlet of the integrated reactor, at regular time intervals of 60 days with 2.7-L SUMMA canisters (ENTECH Instruments Inc., Silonite™), which were pre-cleaned five times using high-purity nitrogen and pre-evacuated using a canister cleaner before sampling. After sampling, identification and quantification of VOCs were performed with a pre-concentrator (Entech 7100A, Entech Instruments Inc., CA, USA) working in line with a gas chromatography-5975C mass spectrometer device (7890A GC-5975C MS, Agilent Technology, USA) based on the methods recommended by the US Environmental Protection Agency (USEPA) TO-15. Detailed sampling and analytical procedures and information are provided in supporting information (SI) and Table S2, as well as in our previous works (An et al., 2014; Chen et al., 2016a, 2017).

2.3. O_3 and SOA formation calculation

The maximum incremental reactivity (MIR) value expresses the O_3 formation potential (OFP) from different VOC species. OFP is used to describe the maximum contribution of VOC species to O_3 formation under optimum reaction conditions and to determine key species and sources for O_3 formation based on both emissions and reactivity (Ou et al., 2015; Wang et al., 2018; Wu et al., 2017). OFP can be calculated by multiplying the emission by the corresponding MIR value using Eq. (1).

$$OFP_i = C_i \times MIR_i \tag{1}$$

where OFP_i is the O_3 formation potential of VOC species i; C_i is the concentration of the compound ($\mu g \, m^{-3}$); and MIR_i is the maximum incremental reactivity of species i (Table S3).

SOA formation potential (SOAFP) was calculated using the secondary organic aerosol potential (SOAP, toluene weighted mass contributions) method (Derwent et al., 2010; Zhang et al., 2017). $SOAP_i$ is the SOA formation potential parameter for species i, in which toluene is set to equal 100. The $SOAP_i$ value from Derwent et al. is preferred to serve as a data source for detected VOC species (Derwent et al., 2010), as shown in Table S3. SOAFP (ug m^{-3}) can be calculated by Eq. (2).

$$SOAFP_i = \frac{\sum (VOC_i \times SOAP_i)}{100} \times FAC_{toluene} \tag{2}$$

where VOC_i is the mass concentration of a VOC source for species i ($\mu g \, cm^{-3}$); and $FAC_{toluene}$ is the fractional aerosol coefficient of toluene (%), which was chosen as 5.4% in this study.

2.4. Occupational exposure cancer risk evaluation

The occupational exposure risk (E_i) for each VOC was calculated as an individual concentration divided by its time-weighted average (TWA_i) (Eq. (3)), as listed in Table S4. The reference data were obtained from previous research (An et al., 2014; Chen et al., 2016a; Liu et al., 2017).
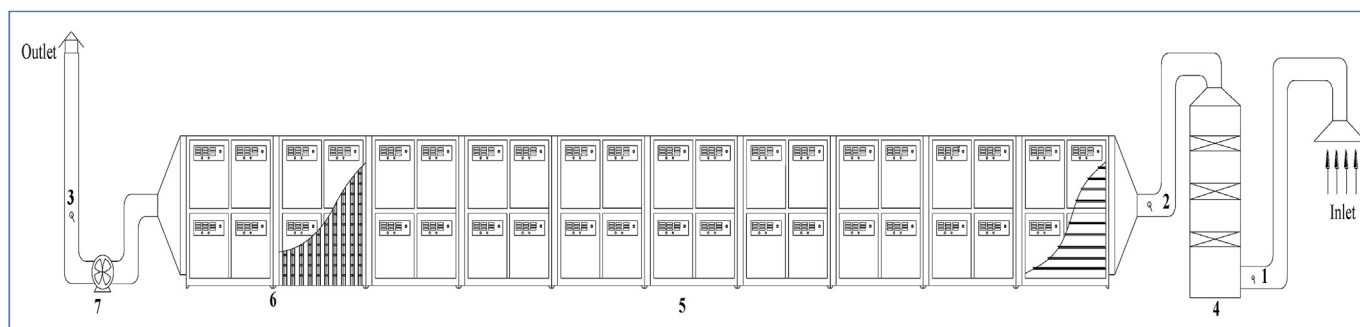
Fig. 1. Schematic diagram of integrated spray tower-electrostatic precipitation-photocatalysis reactor. (1–3: Sampling points; 4: Spray tower; 5: Electrostatic precipitation, 6: Photocatalysis, 7: Centrifugal pump).

$$E_i = \frac{C_i}{TWA_i'} \tag{3}$$

3. Results and discussion

3.1. VOC emission profiles and contributions to O₃ and SOA formation

In the present study, a total of 39 VOCs was identified and quantified during three sampling intervals, including fifteen aromatic hydrocarbons (AHs), ten halogenated hydrocarbons (HHs), ten aliphatic hydrocarbons (AlHs) and four oxygenated VOCs (OVOCs) (Table S5). Fig. 2a shows the variation of their concentrations during the three sampling intervals of six months. As expected, AHs were still the most dominant group from the e-waste dismantling process with concentrations ranging from 1.3×10^2 to $6.4 \times 10^2\,\mu g\,m^{-3}$ ($3.0 \times 10^2\,\mu g\,m^{-3}$, on average), followed by HHs (53–$3.8 \times 10^2\,\mu g\,m^{-3}$, $1.8 \times 10^2\,\mu g\,m^{-3}$, on average), AlHs ($12$–$72\,\mu g\,m^{-3}$, $35\,\mu g\,m^{-3}$, on average) and OVOCs (6.3–$55\,\mu g\,m^{-3}$, $24\,\mu g\,m^{-3}$, on average). The corresponding percentages accounted for 55.93% (AHs), 33.33% (HHs), 6.51% (AlHs) and 4.23% (OVOCs) (Fig. 2d). This conclusion is consistent with our previous work on the pollution profiles of VOCs, which indicated that AHs and HHs were the two predominant groups from the e-waste dismantling processes using the electric heating furnace

technique due to the decomposition of phenol resin (An et al., 2014; Liu et al., 2017) and chlorinated fire retardants, such as Dechlorane Plus (Qiu et al., 2007; Xiao et al., 2013).

Meanwhile, a high contribution to OFP and SOAFP from AHs was observed. About $1.3.11 \times 10^3\,\mu g\,m^{-3}$ on average of OFP (Fig. 2b) were ascribed to AHs emitted during the e-waste dismantling process, accounting for 86.47% of the total OFP (Fig. 2d), which was significantly higher than OVOCs ($2.1 \times 10^2\,\mu g\,m^{-3}$ or 9.10%), AlHs ($39\,\mu g\,m^{-3}$ or 2.43%) and HHs ($28\,\mu g\,m^{-3}$ or 2.00%). Fanizza et al. also reported that the highest contributors to O₃ production in Rome were alkenes and AHs (Fanizza et al., 2014). Although the emission concentration of OVOCs was about 7.41 and 1.42 times lower than that of HHs and AlHs, respectively, its OFP contribution was approximately 7.45 and 5.35 times higher than the latter two species, suggesting the important role of OVOCs in O₃ formation in present work. The potentially high contribution of OVOCs to OFP may be due to its high OH radical loss rate and the subsequent chemical reactivity (Liu et al., 2015b; Shao et al., 2009a). Thus, besides AHs, low-concentrated OVOCs should also be paid attention to for their contribution to O₃ pollution in the e-waste recycling region.

Similar to OFP, AHs also contributed to 99.87% of the total SOAFP, with a concentration of $3.0 \times 10^4\,\mu g\,m^{-3}$ (Fig. 2d), revealing its absolutely dominant function for SOA formation during the e-waste
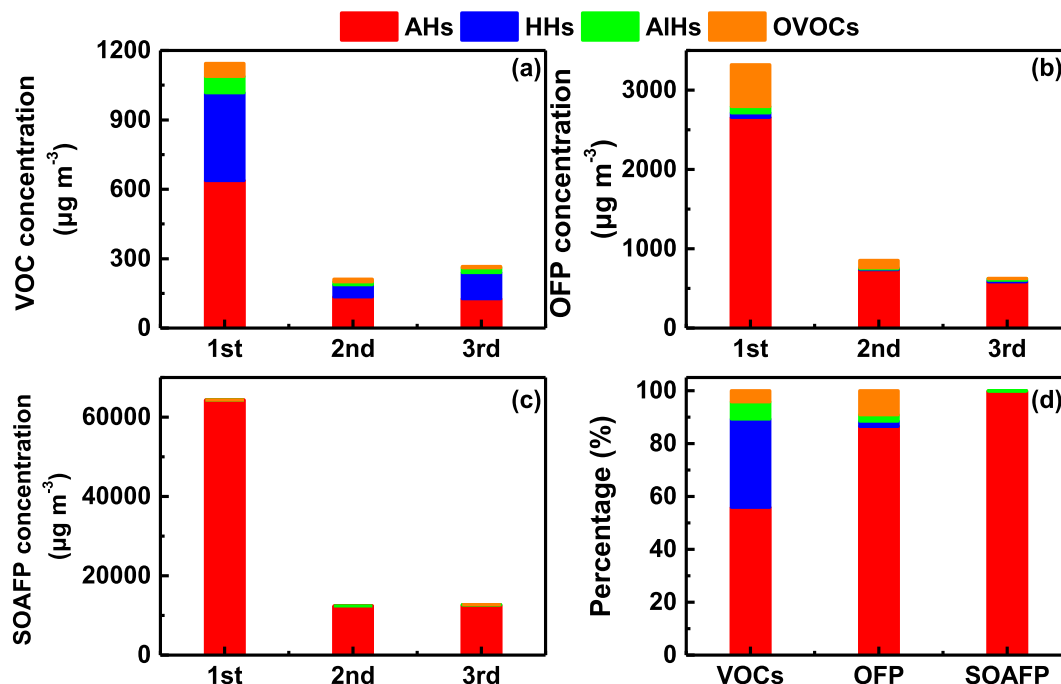

Fig. 2. Concentration of (a) VOC emission; (b) OFP; (c) SOAF and (d) corresponding percentage before treatment.
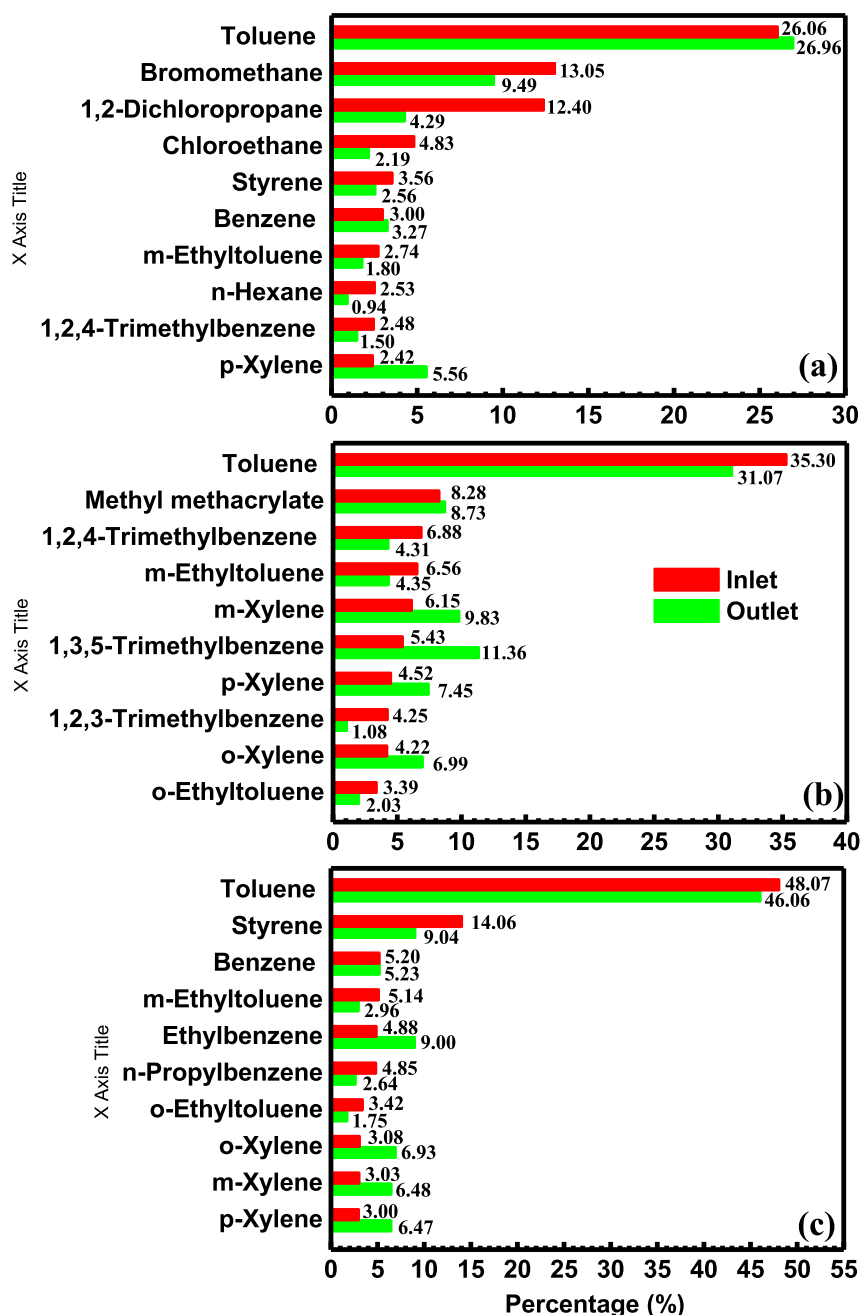
Fig. 3. Top ten VOC species in VOC emission (a), OFP (b) and SOAFP (c) before treatment.

dismantling process in this study. After considering the emission data of VOCs, it can be concluded that the higher emission of AHs leads to the higher formation potential of both O_3 and SOA during the e-waste dismantling process. Thus, our results are strongly consistent with previous work (Ou et al., 2015).

The dependence of the top ten VOCs with high emission concentrations for OFP and SOAFP are discussed and displayed in Fig. 3. As seen from the figure, these ten VOCs accounted for 73.07%, 84.98% and 94.73% of the total VOCs, OFP and SOAFP, respectively, half of which were ascribed to AHs, again confirming the significant contribution of AHs to secondary atmospheric pollutant formation in this study. Further observations reveal the highest contributor was toluene among these ten compounds, accounting for 26.06% of the emission concentration, 35.30% of OFP and 48.07% of SOAFP. Some studies also reported the high contribution of toluene to the formation of O_3 and SOA in China (Guo et al., 2012; Wu et al., 2017). In addition, xylenes

have also been reported to largely contribute to the formation of O_3 (Wu and Xie, 2017; Zheng et al., 2009) and SOAs (Liu et al., 2015a; Wang et al., 2013). In this study, about 14.89% of OFP and 9.11% of SOAFP also originated from three xylenes (Fig. 3b–c), ranking the second and third species among all detected VOCs, thus confirming their importance in facilitating O_3 and SOA formation. Although the emission concentration percentage of several HHs were relatively high, such as bromomethane (13.05%), 1,2-dichloropropane (12.40%) and chloroethane (4.83%), its OFP and SOAFP were not in the top ten, indicating their small contribution to O_3 and SOA formation in this work. On the contrary, the OFP contribution of methyl methacrylate was found to be ranked as second (8.28%) among all detected VOCs (Fig. 3b), even though its concentration was not in the top ten, again proving the high O_3 formation devotion from low-concentrated OVOCs during the TV e-waste dismantling process by electric heating furnaces method.
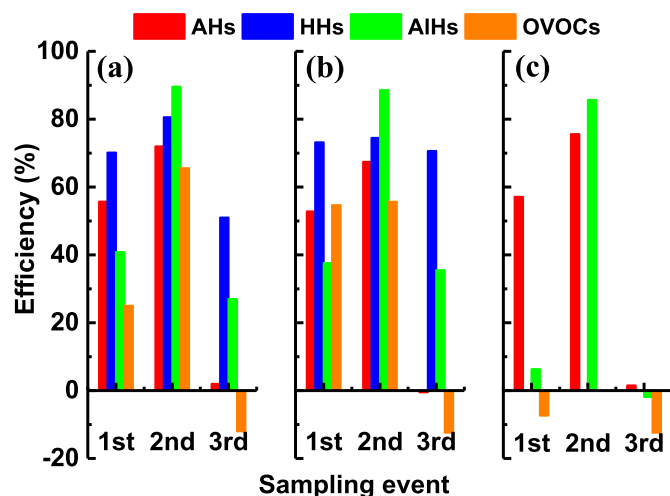
Fig. 4. Efficiency of VOCs removal (a), O_3 (b) and SOA (c) formation prevention by the integrated technique.

3.2. Prevention of O_3 and SOA formation from VOC abatement by integrated technique

Activated carbon adsorption has been established as the most popular technique for low-concentrated VOC removal in industry (Jeon and Jeon, 2017; Li et al., 2008). However, the high cost of direct VOC recovery after adsorption and secondary hazardous production from the inefficient desorption recovery process significantly limits its wider application. Alternatively, in our previous research (Chen et al., 2016a, 2016b, 2017; Liu et al., 2017), the photocatalysis-based integrated technology has shown excellent elimination ability for VOCs from the e-waste dismantling process. Thus, in this study, the integrated reactor for spray tower-electrostatic precipitation-photocatalysis is continuously applied for VOC elimination to prevent O_3 and SOA formation.

Fig. 4a shows the removal efficiency (RE) of four VOC groups after treatment by the integrated technique during a 180-day operation. After the first 60 days, about 55.70% of AHs, 70.14% of HHs, 40.83% of AlHs and 25.01% of OVOCs were removed, which then increased to 71.96%, 80.58%, 89.59% and 65.54%, respectively, after continuous treatment for an extra two months, possibly due to the significant decrease in the inlet VOC concentration (Fig. 2a). However, after another 60 days operation, REs for AHs, HHs, AlHs and OVOCs decreased to 2.00%, 51.00%, 27.00% and −12.00%, respectively, even with a similar VOC supply. In this study, due to a very high flow rate of $6.0 \times 10^4 \, m^3 \, h^{-3}$, the significantly decreased efficiency may be attributed to the faster generation and accumulation of recalcitrant, strongly bound reaction intermediates on the catalyst surface, resulting in catalyst deactivation (Einaga et al., 2002; Guo et al., 2008; Lewandowski, 2003; Lewandowski and Ollis, 2003; Sun et al., 2010), which suggests the necessary regeneration or replacement of the catalyst in the reactor after about six months of continous treatment.

Furthermore, the percentage of four VOC groups varies accordingly after the treatment (Fig. S1), leading to the different prevention efficiencies of the integrated technique toward O_3 and SOA formation (Fig. 4b–c and S2-S3). In the case of OFP, the highest prevention efficiency for O_3 formation was found to be for HHs, ranging from 70.63% to 74.48%, followed by AlHs (from 35.50% to 88.58%), AHs (from −0.49% to 67.43%) and OVOCs (from −12.42% to 55.65%). This trend is highly consistent with the concentration elimination efficiency, indicating that the removal of VOC emission can efficiently prohibit O_3 formation. However, after the treatnment both AHs (87.00%) and OVOCs (10.13%) were still the two dominant contributors to O_3 formation with their percentage increasing 0.53% and 1.03%,

respectively, indicating their residual potentially negative effect on the atmospheric environment, especially for AHs. For SOAFP, about 44.74% of SOA generation can be efficiently prohibited from AHs after the treatment, which is higher than AlHs (30.03% on average) and OVOCs (−6.60% on average), suggesting an excellent and efficient prevention ability of the applied integrated technique for SOA formation. However, there was still about 926.07 g of SOA formation per hour from the e-waste dismantling process, which may significantly influence regional climate change, suggesting further efficiency promotion of the integrated technique should be done.

For the top ten VOCs after the treatment (Fig. 3), toluene was still found as the most predominant contributor to concentration (26.96%), OFP (31.07%) and SOAFP (46.06%). Further observations indicated that the increase in the toluene concentration percentage of 0.90% resulted in the decrease of its contribution to OFP and SOAFP (4.23% and 2.01%, respectively) after the treatment, indicating the enhanced contribution of OFP and SOAFP from other VOCs, especially for other AHs. For example, the concentration percentage of p-xylene rose 2.30 times (from 2.42% to 5.56%) after treatment by the integrated technique, leading to the promoted contribution of 1.65 times to OFP (from 4.52% to 7.45%) and 2.16 times to SOAFP (from 3.00% to 6.47%). The other two xylenes, as well as 1,3,5-trimethylbenzene, also showed a similar a phenomenon. Galval et al. have also reported the significant contribution of xylenes to OFP in industrialized urban areas of Brazil (Galvao et al., 2016).

Overall, the strategy of controlling industrial source emitted VOCs, especially for AHs, to efficiently prevent both O_3 and SOA formation was practicable in the e-waste dismantling industry using the integrated technique, which could be applied to other industries.

3.3. Occupational cancer risk reduction from VOC abatement by integrated technique

Besides the possibly negative effect on the atmospheric environment (O_3 and SOA formation), the potential cancer risk of exposure to VOCs through inhalation by the workers in the dismantling workshop is simultaneously measured based on the previously reported assessment method (An et al., 2014; Chen et al., 2016a), as well as available reference values (Table S4). The average E_i values of the four VOC groups from the inlet were all below 0.1 (Fig. 5a–b), suggesting that all VOCs at the current concentration level pose little cancer threaten to the dismantling workers during the e-waste dismantling process. However, about 2.0×10^{-2} of the E_i value was obtained for HHs (Fig. 5a), accounting for 62.63% of the total risk index (Fig. S4). Furthermore, its contribution to cancer risk was more than two times higher than AHs (9.4×10^{-3}), which was significantly different from the OFP and SOAFP trends. These results suggest that HHs should be simultaneously controlled to prevent human health threat, along with AHs, in the e-waste recycling industry. However, surprisingly, AlHs and OVOCs show negligible cancer risk (Fig. 5b). For individual species (Fig. 5c), bromomethane (62.02%) and benzene (32.86%) were the two highest percentage contributors to total E_i values, which is inconsistent with their concentration ranking (Fig. 3a), again implying the low emission concentration VOCs would lead to high occupational cancer risk in this study. As benzene and bromomethane are in the list of International Agency for Research on Cancer carcinogens, these two high risk VOCs should be paid much attention on in the e-waste dismantling process.

Fortunately, after the treatment by the integrated technique, the risk indexes for all VOCs decreased to different degrees. As shown in Fig. 5a–b, the highest risk reduction was found for AlHs (70.75%), followed by HHs (64.34%), AHs (52.23%) and OVOCs (34.42%). Meanwhile, the top two contributors to the risk index were still HHs and AHs. However, their content gap significantly decreased from 25.70% to 5.48% (Fig. S4). The possible reason may be due to the obviously increased risk percentage of benzene (8.76%) and the decreased risk of bromomethane (11.91%) (Fig. 5c–d). Previous research
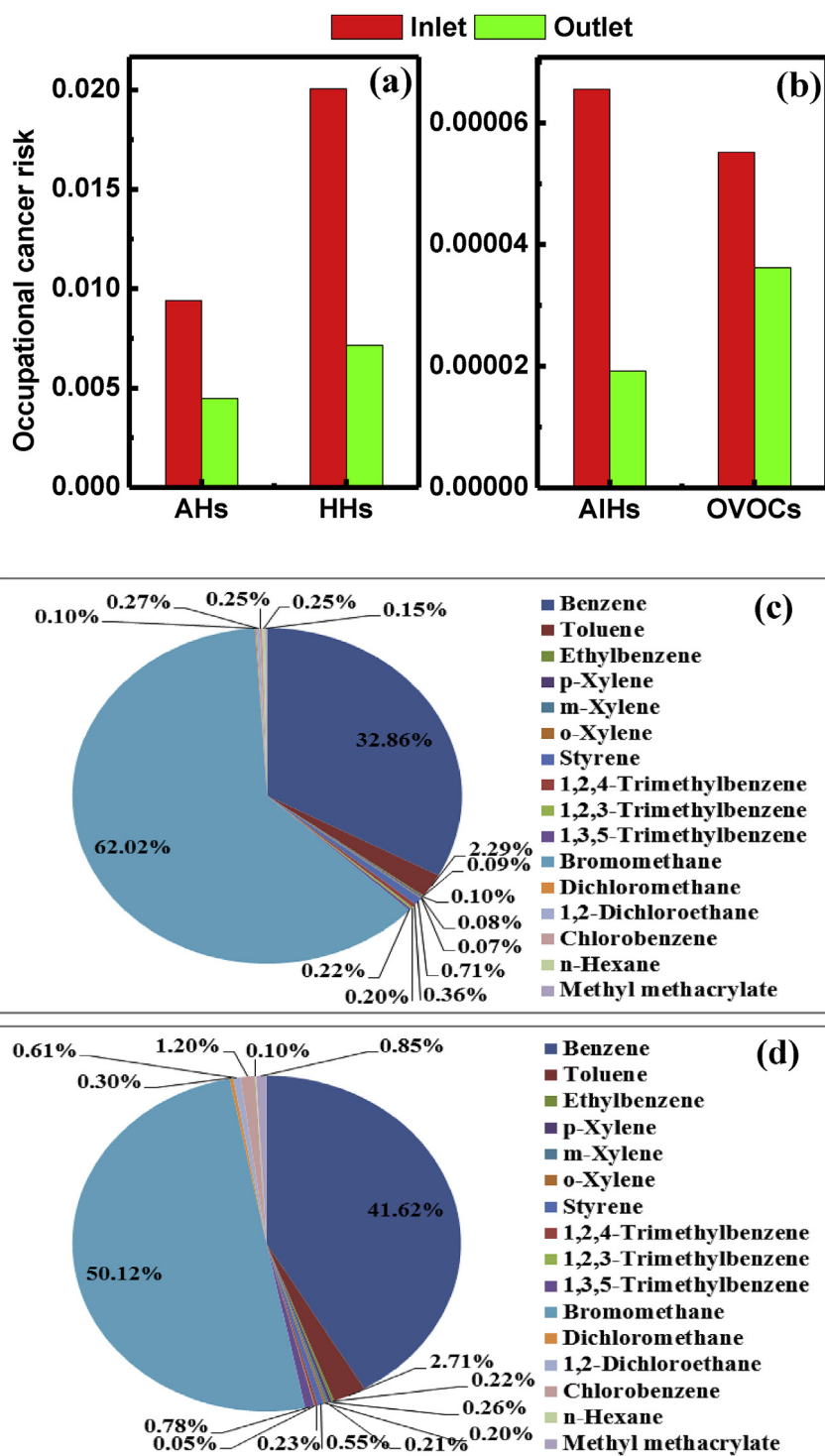
Fig. 5. Occupational cancer risk before and after the treatment by integrated technique.

has proven that photocatalytic decomposition of AHs with more branches can definitely generate benzene (Dhada et al., 2016; Pei and Leung, 2015; Sun et al., 2010), leading to its increased concentration content from 3.00% to 3.27% (Fig. 3a) and a corresponding risk contribution from 32.86% to 41.62%. The risk evaluation results indicate that after treatment by the integrated technique, even though the total risk index of generated VOCs during the e-waste dismantling process is significantly reduced, low-concentrated and more toxic compounds (e.g. 6.6 $\mu g\,m^{-3}$ of benzene) may still accumulate in the reactor and then discharge with the high-speed flow, further threatening workers'

health. Thus, a detailed investigation of the toxic effects of these intermediates to human beings should be conducted to comprehensively assess the safety prevention ability of the integrated technique in the near future.

4. Conclusions

During the e-waste dismantling process, highly concentrated VOCs were produced, and AHs and HHs were identified as two dominant groups. Meanwhile, much higher O_3 and SOA pollution was found due

to AHs, while HHs contributed to the highest occupational cancer risk. Fortunately, all these pollutants could be efficiently prevented and reduced by a self-designed integrated technique of spray tower-electrostatic precipitation-photocatalysis during a 180-day operation of a large-scale application, revealing that reduced VOC emissions from e-waste dismantling sources might facilitate the prevention of O_3 and SOA formation, as well as reduce potential risk. This study can provide an efficient approach to the targeted prevention of secondary atmospheric pollutant formation and improve the protection of human health from the industrial sources of VOCs.

Acknowledgments

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jenvman.2018.07.034.

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