



Preferential removal of aromatics-dominated electronic industrial emissions using the integration of spray tower and photocatalysis technologies

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ABSTRACT

Industrial emissions from the manufacturing of printed circuit boards (PCBs) are characterized by low concentration and high gas flow rates, and are one of the important sources of toxic and odorous volatile organic compounds (VOCs). They are deleterious to human health and considered as important precursors for the formation of ozone. Herein, we conducted a case study in which an integrated decontamination technique consisting of a spray tower (ST) and photocatalysis (PC) was applied to purify the waste gas emitted from a PCB manufacturing facility. Total of 66 VOCs including halogenated hydrocarbons (HHs), aromatic hydrocarbons (AHs), oxygen-containing hydrocarbons (OVOCs), and aliphatic hydrocarbons (AIHs) were quantified using GC-MS during four sampling events. The results showed that AHs were the largest group (contributing >85.0% of the total VOCs). The average removal efficiency (RE) of VOCs during the nine months' continuous treatment was 72.39%. High RE was attributed to the following three aspects: the capture efficiencies of HHs with slight water solubility (50.16% of RE) and AIHs with low saturation pressure (54.75% of RE) were improved by the process consisting of ST upstream of PC, whereas AHs was preferentially degraded by PC (61.34% of RE). In addition, the ozone formation potential and the health risk (cancer and non-cancer risks) greatly decreased after the ST-PC treatment. This study suggests that ST-PC technique is a promising approach for removing continuously emitted organic waste gas with low concentration and high gas flow rates, as well as reducing the ozone formation and risk of health hazards.

1. Introduction

Electronic manufacturing is an important industrial pillar of Chinese economy, and has contributed more than one fifth of the total gross domestic product (GDP) growth in 2019. However, abundant toxic and odorous organic compounds are discharged during the manufacturing process. According to the National Bureau of Statistics, the gas emitted from electronic industry is a dominant contributor to anthropogenic volatile organic compounds (VOCs) that adds around 1.3% of the total VOC emissions (<http://www.stats.gov.cn/>) and strongly impacts both the human health and the air quality (Chen et al., 2019; Kumar et al., 2018; Liang et al., 2020a; Shao et al., 2009; Tan et al., 2018; Wan et al.,

2011). Numerous studies have demonstrated that the inhalation and long-term exposure of VOCs can cause nasal, throat and lung irritations, and even promote the occurrence of serious diseases such as cancer and liver damage (Adamova et al., 2020; Colman Lerner et al., 2012; Huang et al., 2014; Maciua et al., 2012; Song et al., 2019; Soni et al., 2018; Zheng et al., 2020). More importantly, the released atmospheric VOCs have also been demonstrated as important precursors in the photochemical formation of ozone (Carter, 2010; Chen et al., 2016; Li et al., 2020; Ou et al., 2015; Zhang et al., 2020b). Among them, benzene, toluene, styrene and other substances need to be paid more attentions as substances with high ozone photochemical activity (Cheng et al., 2021; Li et al., 2021). Given the significant environmental and health impacts

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of VOC emissions, the evolution of environmental protection policies to minimize their environmental impact and health risks has recently attracted increasing attention. Therefore, effective VOC remediation technologies are highly desirable for efficiently control the VOC emissions as well as reducing the potential formation of ozone, thus reducing the health risks to residents exposed to these VOC emissions.

Electronic manufacturing involves many chemical processes and uses different types of organic compounds, which generally result in the emission of VOCs with low concentration and high gas flow rates (Babar and Shareefdeen, 2013; Lv et al., 2021; Wang et al., 2013). Moreover, various AHs, (such as xylene, toluene, ethyl benzene, and benzene), OVOCs (such as ketone, acetone, alcohol, and acetate), and AIHs (such as hexane, 2-methylpentane, 3-methylpentane, and n-pentane) are frequently emitted during the production processes of printed circuit boards (PCBs). The composition and concentration of these organic pollutants varies with the type of the final product and the manufacturing techniques employed in the process (Lv et al., 2021; Park et al., 2011; Shen et al., 2018). The control of such VOC emissions is a big challenge, not only due to low concentration and high gas flow rates, but also due to the complexity, diversity, odorous and disorganized characteristics of VOC emission sources (Liang et al., 2020a; Lv et al., 2021). Therefore, the selection of the most appropriate technology to treat the emitted VOCs is critically important. Many techniques are currently used to remove VOCs, including absorption, adsorption, biological treatment, alkali washing spray, incineration and catalytic oxidation (An et al., 2014; Azlah et al., 2017; Babar and Shareefdeen, 2013; Liang et al., 2020b; Ribeiro et al., 2019; Sansotera et al., 2019; Zhuang et al., 2021). Among these technologies, adsorption and biological treatment are widely used for the remediation of emitted VOCs with low concentrations and high gas flow rates (Detchanamurthy and Gostomski, 2012; Yang et al., 2019; Zhang et al., 2017). However, it should be noted that the adsorption is hindered by the unavailability of novel and advanced adsorbents having excellent adsorption performance, the complex recovery process of adsorbents and the inefficient recyclability. Alternatively, biological methods have gained much attention for the treatment of industrial emissions, particularly for the treatment of odorous emissions (An et al., 2010; Wan et al., 2011). However, biological treatment has always required much larger reactor volumes to increase the contact between VOCs and microorganisms. Furthermore, a previous study has reported that aromatic hydrocarbons (AHs) and aliphatic hydrocarbons (AIHs) were the major groups in the waste gas emitted from the electronic-related industry, which is hard to dissolve in water due to their highly hydrophobic properties (Palau et al., 2011). Therefore, the efficiency of mass transfer of these hydrophobic VOCs into the liquid phase needs to be further enhanced for achieving high removal efficiencies of VOCs using biological treatment (Vikrant et al., 2017). In this regard, alternative technologies should be explored for the remediation of emitted VOCs that are dominated by AHs.

Recently, the degradation of VOCs using photocatalysis (PC), as a typical gas-solid heterogeneous reaction, has been validated as a promising technology for treating low concentration VOCs coming from the industrial emissions. The method requires relatively low investment, mild reaction conditions without high-temperature heating treatment, and results in efficient photocatalytic degradation of gaseous contaminants to harmless products (Jansson et al., 2016; Kamal et al., 2016; Mama-gani et al., 2018). More specifically, significant studies have demonstrated that PC is a promising technology for degrading AHs in laboratory, which is due to the high affinity of TiO_2 towards AHs. It is well known that TiO_2 has abundant Lewis acid sites, which are derived from the unsaturated Ti surface atoms, benefiting the adsorption of AHs through delocalized π bonds, and leading to the formation of cation- π bonds between the AHs and TiO_2 (Bui et al., 2011; He et al., 2019). However, the pilot-scale photocatalytic purification of exhaust gas that has abundant AHs is rarely reported in the literature, as aromatic compounds are widely used during the electronic manufacturing

process. Therefore, the application of PC in the treatment of real waste gas, discharged from electronic factory, is meaningful and credible, and will result in an efficient approach for the treatment of waste gas with abundance of AHs discharged from electronic industries.

Herein, the pollution profile of the VOCs emitted during the production of printed circuit boards (PCBs) was investigated with the aim to explore the application of PC for controlling the VOC emissions. Based on the analysis of VOC emissions, a pilot-scale integrated technique of spray tower and photocatalysis (ST-PC) (Scheme 1) with a high treatment capacity ($6.0 \times 10^4 \text{ m}^3 \text{ h}^{-1}$) was designed and employed to efficiently control the waste gas emitted from a PCB production facility. The ST unit was applied to remove the particle matters as well as the corrosive compounds from the exhaust gas, with the aim to prevent the deactivation of photocatalyst during the long-term operation of PC technology. The efficiency of integrated technique for removing VOCs was further evaluated. More specifically, the degradation selectivity of integrated technique towards AHs, AIHs, halogenated hydrocarbons (HHs) and oxygen-containing hydrocarbons (OVOCs) was studied. Furthermore, the potential health risks from various VOCs as well as the ozone formation potential (OFP) were evaluated before and after the 270-day operation of the process.

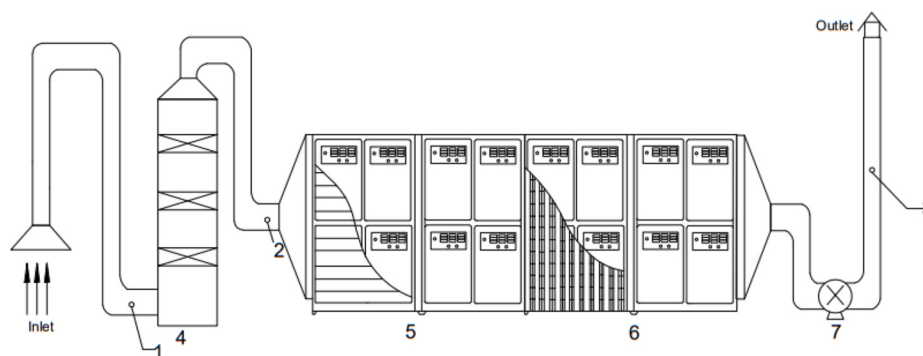
2. Experimental

2.1. Equipment description

The integrated ST-PC reactor was fixed on the roof of a PCB manufacturing building located in Guangzhou, China with the population of 15 million (children: 13.87%, adults: 86.13%). The workshop was mainly engaged in the production of high-density precision interconnected circuit boards as well as flexible and hard interconnected circuit board structure composite boards. During the production process, exhaust gas containing VOCs was produced. The produced waste gas was collected and introduced into the integrated equipment with the flow rate of $60000 \text{ m}^3 \text{ h}^{-1}$ using an exhaust fan mounted at the end of the system. The ST was made of stainless steel with the height and diameter of 7000 and 3800 mm, respectively. To enhance the absorption performance of ST for gaseous VOCs, polyhedral hollow sphere (diameter: 76 mm; specific surface area: $\geq 200 \text{ m}^2 \text{ m}^{-3}$; material: polypropylene) was introduced in the ST. The solution used in the spray tower was the tap water with the pH of 6.75. The photocatalytic reactor had the stainless-steel body (reactor dimensions: $10300 \text{ mm} \times 3000 \text{ mm} \times 3500 \text{ mm}$), which housed 192 pieces of $1000 \text{ mm} \times 420 \text{ mm}$ foam nickel mesh and 192 pieces of $500 \text{ mm} \times 420 \text{ mm}$ foam nickel mesh. A total of 120 ultraviolet lamps with the power of 150 W/branch and a wavelength range 185–254 nm were installed in the PC reactor.

2.2. Sampling methodology and analysis

VOC samples were collected from the inlet and outlet of ST, and the outlet of the PC equipment using 2.7 L SUMMA canisters (Entech Instruments Inc., Silonite™, USA). Before collecting the sample, the SUMMA canisters were pre-cleaned for at least five times using continuous pre-evacuation and high-purity nitrogen washing. Sampling was performed in the morning of June 1, 2020 (moderate rain, 30°C), June 9, 2020 (moderate rain, 29°C), September 17, 2020 (moderate rain, 30°C), and March 23, 2021 (cloudy, 18°C) during the continuous operation of the equipment, and denoted as 1st, 2nd, 3rd and 4th, respectively. After sampling, the composition and concentration of VOCs were identified and qualified using gas chromatography-mass spectrometry (7890A GC-5975C MS, Agilent Technology), which was equipped with a pre-concentrator (ENTECH 7100A, ENTECH Instruments Inc., CA, USA). To this end, the methods of TO-15 (the US Environmental Protection Agency) and PAMs (Linde Spectra Environment Gases, USA) were employed. The detailed procedure for the analysis is described in the Supporting Information (SI).



Scheme 1. Schematic diagram of integrated spray tower -photocatalysis reactor. (1–3: sampling points; 4- spray tower; 5- dehumidification system; 6-photocatalysis; 7- exhaust fan).

2.3. Odorous VOC analysis using electronic nose

A portable electronic nose (e-nose) Pen3.5 (win muster airsense (WMA) analytics Inc., Germany) was applied for on-line analysis of odorous VOCs before and after the waste gas treatment. The system consisted of a detector unit with 10 gas sensor arrays (Table S1), a sampling device and a model recognition software (Baietto et al., 2010). The odorous VOC analysis program consisted of the following steps. First, before the operation, preheating was conducted for 10 min, followed by cleaning for 200 s. The sampling was done for 60 s. Air was used as the carrier gas and the flow rate was set at 200 ml min^{-1} . The data obtained were analyzed using PCA principal component analysis, after which the VOCs in the whole process section were distinguished (Benedetti et al., 2008; Zhang et al., 2020c).

2.4. Calculation of the ozone formation potential (OFP)

Ozone is the major by-product formed by the reaction of VOCs with free radicals in atmosphere. Therefore, the contribution of VOCs to the generation of ozone is estimated by calculating the OFP. The OFP of VOCs was calculated based on maximum incremental maximum incremental reactivity (MIR) of each VOC, as given by Eq. (1).

$$\text{OFP}_i = C_i \times \text{MIR}_i \quad (1)$$

where OFP_i is the ozone formation potential of i th VOC species, C_i is the concentration of the i th VOCs ($\mu\text{g m}^{-3}$), and MIR_i is the maximum incremental reactivity of i th VOC species (Table S2).

2.5. Health risk assessment

The lifetime cancer risk (LCR_i) and non-cancer risk (hazard ratio (HR_i)) were calculated to estimate the reduction of health risk after treatment of the emitted VOCs. The values of LCR_i and HR_i are obtained using Eqs. (2) and (3), respectively.

$$\text{LCR}_i = C_i \times \text{UR}_i \quad (2)$$

$$\text{HR}_i = C_i / \text{RfC}_i \quad (3)$$

where UR_i is the unit risk of the i th VOC, C_i is the daily concentration of the i th VOC ($\mu\text{g m}^{-3}$), and RfC_i is the reference concentration ($\mu\text{g m}^{-3}$) of the i th VOC. The reference concentration of each VOC and the unit risk data were both obtained from the USEPA Integrated Risk Information System (Ramirez et al., 2012), and listed in Tables S3 and S4, respectively.

3. Results and discussion

3.1. VOC emission profile

Analyzing the compositions and concentrations of VOC emissions is helpful for accurately selecting the purification technology. Therefore, samples were collected using pre-evacuated steel canisters at the inlet of integrated equipment, and subsequently analyzed within 24 h using pre-concentrated GC-MS. In total, 66 kinds of VOCs with the average total volatile organic compound (TVOC) concentration of 23.35 mg m^{-3} were detected and quantified during four sampling intervals, including 16 kinds of AHs, 12 kinds of HHs, 21 kinds of AIHs and 6 kinds of OVOCs, as listed in Table S5. Fig. 1 displays the variation in VOCs' concentrations during the four sampling events. It can be observed that the TVOC concentrations of each sampling event varied from 13.61 to 26.81 mg m^{-3} , which can be ascribed to the different unit operations. Among the emitted VOCs, AHs displayed the highest concentration (11.48 – 21.93 mg m^{-3} , with an average value of 17.69 mg m^{-3}), followed by AIHs (0.031 – 2.61 mg m^{-3} , with the average concentration of 0.91 mg m^{-3}), OVOCs (0.35 – 1.21 mg m^{-3} , with the average concentration of 0.85 mg m^{-3}), and HHs (0.11 – 2.17 mg m^{-3} , with the average concentration of 0.57 mg m^{-3}), as displayed in Table S5. On the basis of above analysis, it can be found that AHs were the dominant group, which contributed 78.47%–97.77% to the total VOCs (Fig. S1), suggesting that AHs were the dominant VOCs emitted during the PCB production. Among the detected AHs, 1,2-dimethyl-4-ethylbenzene, 1,2,3,4-tetramethylbenzene, 1,2,3,5-tetramethylbenzene, 5-ethylm-dimethylbenzene, 1,3-dimethyl-4-ethylbenzene, 1,3-dimethyl-2-ethylbenzene, 1,3,5-trimethylethylbenzene, 1-methyl-4-(2-propene) benzene, methyl (isopropyl) benzene, p-diethylbenzene, and 2,4-dimethylstyrene were the top 10 AHs and the total concentration of these compounds exceeded 1 mg m^{-3} , indicating high health risk posed by them to the residents. The high concentration of AHs could be attributed to the widespread application of phenol resin and other raw materials used in the PCB manufacturing

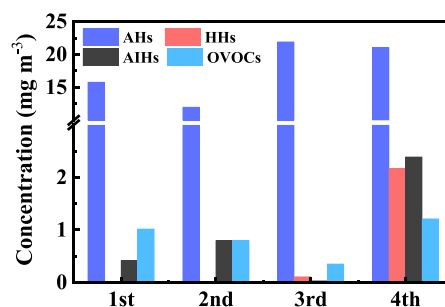


Fig. 1. Concentrations of VOC species sampling at the inlet of integrated equipment.

process (Guo et al., 2012). Meanwhile, AIHs (accounting for 0.15%–8.92% of the total VOCs) exhibited a higher proportion than the OVOCs (accounting for 1.58%–5.92% of the total VOCs) and HHs (accounting for 0.02%–8.09% of the total VOCs). For HHs, chloride organic compounds were the major components, which were probably generated by the decomposition of flame retardants during the production process (Zhou et al., 2017).

3.2. Performance of the pilot-scale ST-PC in treating VOC emissions

Furthermore, the removal of emitted VOCs was carried out using the integrated ST-PC technique. As observed from Fig. S2, the removal efficiencies of TVOCs at the four sampling events were 77.06%, 72.80%, 71.51%, and 73.08%, respectively, demonstrating that the employed integrated ST-PC technique exhibited outstanding capability and high stability to purify VOCs emitted from the PCB factory during the 270-day treatment period. The average concentrations of TVOCs decreased from 16.32 to 4.35 mg m⁻³, with the average removal efficiency of 73.45%. It should be noted that there was significant fluctuation of VOC concentrations (13.61–26.81 mg m⁻³) during the four sampling events as displayed in Fig. S3, which might be due to the different unit operations of factory. The TVOC concentration of the 4th sampling event was approximately two times of the 1st sampling event. However, the removal efficiency of the 4th sampling event (73.08%) was similar to that of the 1st sampling event (77.06%) even after the 270-day continuous operation of the integrated process, demonstrating that the integrated ST-PC technique displayed outstanding stability and resistance for the purification of VOCs with significant fluctuations in the VOC concentrations. Furthermore, the removal efficiencies of TVOCs using ST, PC and the integrated ST-PC technology were also analyzed. It can be observed that the average removal efficiency using ST unit was 19.12% (Fig. S2). This is mainly due to higher removal efficiency of relatively hydrophilic VOCs or VOCs with low saturation pressure VOCs, which is due to the reason that ST uses H₂O as a solvent (Liang et al., 2020b) and is mainly used to remove particles (Liu et al., 2017). In contrast, the average removal efficiency was 58.28% using the PC unit, which is much higher than that of the ST unit, demonstrating that PC is an effective technology for the degradation of gaseous VOCs emitted from the PCB facility, even though there were significant fluctuations in the concentrations of VOCs.

To have a deep understanding of the synergistic effect between the ST and PC techniques for the treatment of waste gas emitted from the PCB facility, the authors further evaluated the removal efficiencies of AHs, AIHs, HHs, and OVOCs after treatment using individual techniques. As shown in Fig. 2, the average concentrations of AHs (decreased from 17.69 to 14.96 mg m⁻³), HHs (decreased from 0.57 to 0.32 mg m⁻³), AIHs (decreased from 0.91 to 0.43 mg m⁻³), and OVOCs (decreased from 0.84 to 0.61 mg m⁻³) decreased after treatment using ST, thus corresponding to the average removal efficiencies of 15.50%, 50.16%, 54.75% and 22.51% for AHs, HHs, AIHs, and OVOCs, respectively, during the four sampling events. Clearly, ST unit is an effective technology for the removal of HHs. This is probably due to higher hydrophilicity of HHs as compared to other VOC groups. Particularly, 1,2-dibromoethane and trans 1,3-dichloro-1-propene are only slightly soluble in water (Tolgyessy and Hrivnak, 2006). In contrast, AIHs, such as n-dodecane, n-octane and n-nonane with lower saturated vapor pressure as compared to water are easily transported from gaseous phase to liquid phase during the ST treatment process, leading to the reduced concentration of AIHs in the gaseous phase (Bravo et al., 2017; Liang et al., 2020b; Liao et al., 2021).

The waste gas treated by ST was further introduced into the PC unit. As displayed in Fig. 2b, the PC unit's contribution to average removal efficiency was 61.34%, 24.79%, 33.70% and 43.16% for AHs, HHs, AIHs, and OVOCs, respectively, during the 270-day continuous treatment. Obviously, PC unit displayed higher affinity and stability for the removal of AHs as compared to other VOC groups during the four

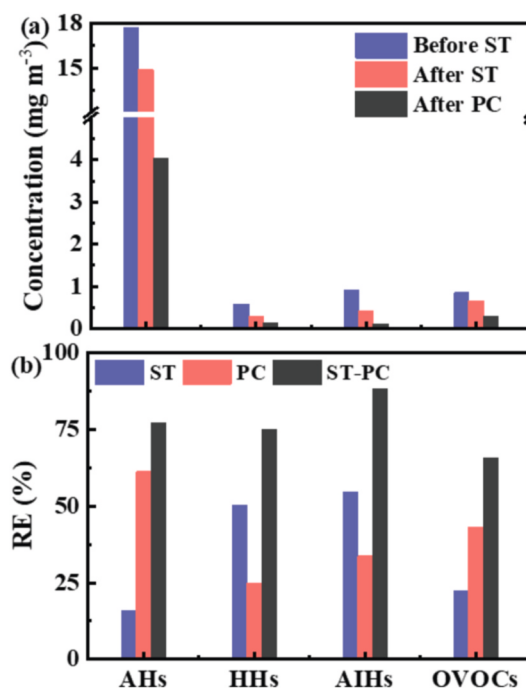


Fig. 2. The average concentration of AHs, HHs, AIHs, and OVOCs at different sampling events before ST, after ST and after PC treatment (a); The average removal efficiency of AHs, HHs, AIHs, and OVOCs at different sampling events after ST, PC and ST-PC integrated treatment (b).

sampling events. In the case of degradation of individual AHs, high removal efficiencies were obtained using the PC treatment for benzene (95.34%), ethylbenzene (88.74%), p-xylene (96.35%), 1,2,3-trimethylbenzene (96.73%) and 1,2,4-trimethylbenzene (74.14%), as demonstrated in Fig. S4. The great variation in removal efficiencies across these VOC groups can be attributed to their different degrees of affinities towards TiO₂. It is well known that TiO₂ has abundant Lewis acid sites, which are derived from the unsaturated surface of Ti atoms that are beneficial for the adsorption of AHs with delocalized π bond and therefore, lead to high photocatalytic degradation of AHs (Bui et al., 2011; Chen et al., 2019; Godlewski et al., 2012; Zhang et al., 2020a). On the basis of above analysis, it can be concluded that ST-PC is a promising technology for the purification of waste gas emitted from the PCB facility with a very high flow rate of 6.0×10^4 m³ h⁻¹.

Beside the common VOCs, considerable unpleasant VOCs, such as styrene (with the concentration of 0.08–0.34 mg m⁻³ during the four sampling events) and other AHs are also emitted during the PCB production process. Therefore, the capability of integrated ST-PC process in removing odorous VOCs was also assessed. PEN3 e-nose was applied to monitor the odorous VOCs at different sampling points using PCA (principal component analysis) (Arasradnam et al., 2011; Delgado-Rodríguez et al., 2012; Ravi et al., 2013). As shown in Fig. 3, the odorous VOCs at the inlet of ST unit were distinguished from the environment, suggesting that there was a significant variation between the VOCs in the waste gas and the environment. Higher signals were observed from the W2W, W5C and W3C sensors, which were used to discriminate AHs at the inlet of PC site compared to environment-1, environment-2 and the outlet of ST-PC equipment, thus suggesting that the AHs were the dominant groups contributing the most to the odorous VOCs. This result is also consistent with the pollution profiles of VOCs analyzed using GC-MS. The intensity of these sensors in the exhaust gas after ST-PC treatment obviously weakened. The odor response intensity at the outlet of ST-PC equipment was similar to that of the environment, suggesting that the integrated ST-PC technology is also very promising for removing odorous VOCs emitted during the production process of PCBs.

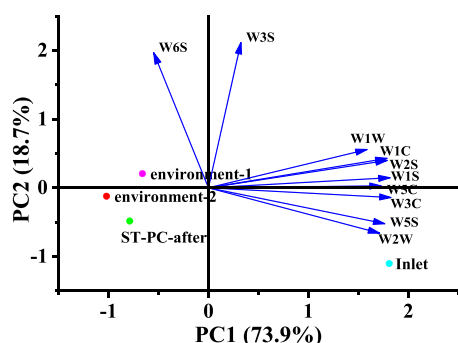


Fig. 3. Principal component analysis (PCA) based on the average change of VOC mixture composition at different sampling points was used to identify VOCs before and after ST-PC treatment. PC1 and PC2 accounted for 73.9% and 18.7% of the variation, respectively.

3.3. Prevention of ozone formation by VOC abatement using the integrated technique

Recently, ozone pollution has become very serious all over the world (Fiore et al., 2002). VOCs are one of the most important precursors for the formation of ozone in the tropospheric atmospheric chemistry (Luo et al., 2020). Especially, AHs and alkanes serve as important precursors for the formation of ozone due to their high MIR values (Carter, 2012). The capability of integrated ST-PC process in preventing the formation of ozone was also evaluated. The average OFP concentrations for the four categories of VOCs before and after the ST-PC treatment are shown in Fig. 4. The average OFP_i values of the four sampling events before the treatment using ST-PC process were calculated to be 132086, 1051, 4.31, and 2048 $\mu\text{g m}^{-3}$ for AHs, AIHs, HHs, and OVOCs, respectively. Obviously, the largest contributor for the OFPs was the AHs, followed by OVOCs, AIHs and HHs. The corresponding values for AHs, AIHs, HHs, and OVOCs were 97.70%, 0.78%, 0.0031%, and 1.52%, respectively. Although AHs contributed 88.36% to the total VOC concentration, their OFP contribution was approximately 97.70% of the total ozone formation, suggesting that AHs played an important role in the formation of ozone due to their high MIR values. For instance, 1,2,4-trimethylbenzene, 1,2,3,5-tetramethylbenzene, 1,3,5-trimethylbenzene, 1,2,3,4-tetramethylbenzene, and 5-ethylm-dimethylbenzene were the top five species among all detected AHs, and accounted for 67.44% to the formation of ozone, as shown in Fig. S5. These results are also consistent with our early work (An et al., 2014), demonstrating that the highest contributor to the formation of ozone is the AHs during the electronic waste dismantling process. Fortunately, the ST-PC process is effective for the treatment of organic waste gas with low concentration and high flow rates. The average OFP_i values during the four sampling events reduced from 135191 to 28014 $\mu\text{g m}^{-3}$, corresponding to the prevention efficiency of 79.28%. The average prevention efficiencies for AHs, AIHs,

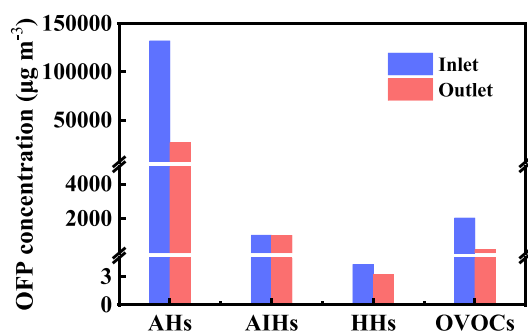


Fig. 4. The average OFP concentration from four sampling events of AHs, HHs, AIHs and OVOCs before and after ST-PC integrated treatment.

HHs, and OVOCs were calculated to be 79.73%, 0.62%, 25.04% and 90.18%, respectively, as displayed in Fig. S6. This trend is highly consistent with the concentration removal efficiency (Fig. 1). In addition, the prevention efficiencies for 1,2,4-trimethylbenzene (87.65%), 1,3,5-trimethylbenzene (84.12%), 1,2,3,5-tetramethylbenzene (83.48%), 1,2,3,4-tetramethylbenzene (82.21%), and 5-ethylm-dimethylbenzene (89.68%) were relatively high, accounting for 72.67% of the total prevention efficiency (Fig. S7). On the basis of the above analysis, the integrated ST-PC technology was demonstrated as an effective strategy for preventing the formation of ozone due to AHs-dominated exhaust gas discharged from the PCB facility.

3.4. Health risk assessment of VOCs before and after the treatment

VOCs are not only detrimental to the environment, but also negatively affect the health of workers exposed to VOCs (Bari and Kindzierski, 2018). Therefore, the cancer and non-cancer risks of workers due to the inhalation of VOCs were simultaneously assessed using the previously reported health risk assessment methods (Moulis and Krýsa, 2013). The average non-cancer risks (Fig. 5) and cancer risks (Fig. 6) before and after the comprehensive treatment were analyzed. Total of 19 kinds of VOCs (1,2,4-trichlorobenzene, p-xylene, o-xylene, 1,2-dibromoethane, chlorotoluene, dibromochloromethane, 1,1,2-trichloroethane, 1,3-dichloroethane, 1,1,2-trichloroethane, methyl methacrylate, trichloroethylene, dichloromethane, butadiene, vinyl chloride, 1,2,3-, 1,2,4-, 1,3,5-trimethylbenzene, cumene, and ethylbenzene) were selected to evaluate the non-cancer risks. Moreover, a total of 10 VOCs, (1,2-dibromoethane, dibromochloromethane, 1,1,2-trichloroethane, 1,3-dichloropropene, trichloroethylene, dichloromethane, butadiene, vinyl chloride, ethylbenzene, and benzene) were selected to evaluate the cancer risks. The available reference concentrations and unit risk values are listed in Tables S3 and S4. In the case of non-cancer risk assessment, the average HRs of all 19 VOCs were higher than 0.1 during the four sampling events and the total HR was up to 369.05, suggesting potential non-cancer threats with these constituents according to the assessment standard (McCarthy et al., 2009). In particular, the average HRs of 16 kinds of VOCs (1,2,4-trichlorobenzene, p-xylene, o-xylene, 1,2-dibromoethane, chlorotoluene, dibromochloromethane, 1,1,2-trichloroethane, 1,3-dichloroethane, 1,1,2-trichloroethane, trichloroethylene, butadiene, 1,2,3-, 1,2,4-, and 1,3,5-trimethylbenzene) were higher than unity during the four sampling events, suggesting that these constituents may pose significant non-cancer threats to the workers (Li et al., 2013).

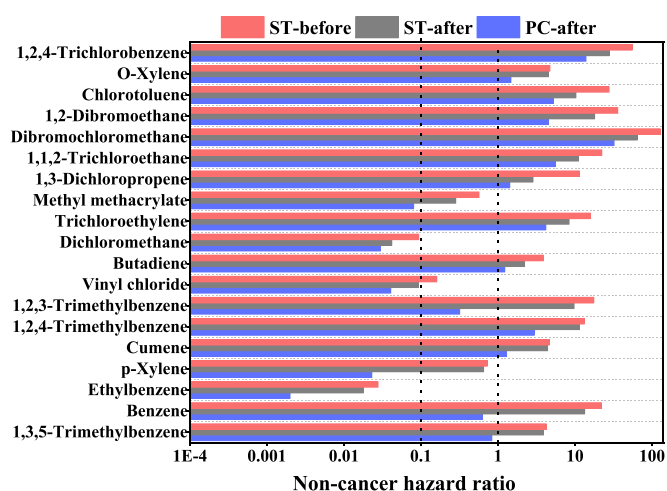


Fig. 5. Change of non-cancer risk rate before and after comprehensive treatment. Compounds with hazard ratios more than 1, between 0.1 and 1, and less than 0.1 were labeled as definite, probable or possible and negligible health risks, respectively.

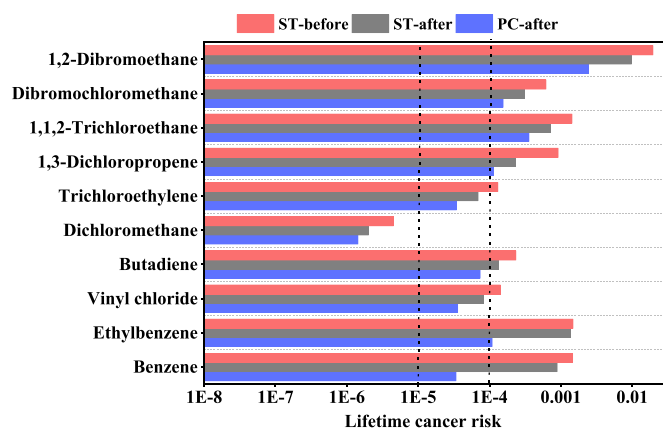


Fig. 6. The change of cancer risk rate before and after comprehensive treatment. Compounds with LCR more than 10^{-4} , between 10^{-4} and 10^{-5} , between 10^{-5} and 10^{-6} , and less than 10^{-6} were labeled as “definite, probable, possible and negligible risk”, respectively.

Fig. 6 displays the individual lifetime cancer risks (LCRs) of nine kinds of detected and quantified VOCs. The results show that the average LCRs of these VOCs were within the range of 8×10^{-6} – 1.92×10^{-2} during the four sampling events and the total LCR was around 2.4×10^{-2} , indicating that, if not treated properly, the VOCs produced during the production process would lead to obvious harmful effects on human health (Li et al., 2013). For the individual VOCs, the LCRs of 1,2-dibromoethane (1.94×10^{-2}), dibromochloromethane (6.155×10^{-4}), 1,1,2-trichloroethane (1.43×10^{-3}), 1,3-dichloropropene (9.12×10^{-4}), butadiene (2.33×10^{-4}), ethylbenzene (1.47×10^{-3}), trichloroethylene (1.31×10^{-4}) and benzene (1.46×10^{-3}) were all higher than 10^{-4} , demonstrating a definite cancer risk at current exposure levels. Overall, the VOCs discharged from the PCB facility displayed high levels of cancer and non-cancer risks, indicating that significant attention is needed to address this challenge.

Therefore, the ability of the ST-PC process to reduce the health risks was also examined. As displayed in Figs. 5 and 6, the HRs of all examined VOCs slightly decreased after treatment with the ST unit. However, the HRs of the most studied VOCs (1,2,4-trichlorobenzene, p-xylene, o-xylene, 1,2-dibromoethane, chlorotoluene, dibromochloromethane, 1,1,2-trichloroethane, 1,3-dichloroethane, 1,1,2-trichloroethane, trichloroethylene, butadiene, and 1,2,3-, 1,2,4-, 1,3,5-trimethylbenzene) still exceeded the value of unity. These results confirmed that ST had a poor ability for reducing the health risks posed by these HRs. Fortunately, the HRs of all VOCs decreased significantly after treatment using the PC unit, as displayed in Fig. 5. The highest non-cancer risk reduction was found for AHs. Specifically, the non-cancer risks of benzene (decreased from 3.20 to 0.0029), 1,2,3-trimethylbenzene (decreased from 5.93 to 0.023), 1,3,5-trimethylbenzene (decreased from 2.63 to 0.65) decreased, suggesting that there was no obvious non-cancer risk posed by them. These results were consistent with high removal efficiencies of benzene (95.34%), 1,2,3-trimethylbenzene (96.73%) and 1,3,5-trimethylbenzene (75.29%) (Fig. S4).

In the case of reducing cancer risk using ST, the LCRs of some VOCs including 1,2-dibromoethane, dibromochloromethane, 1,1,2-trichloroethane, butadiene, ethylbenzene, and benzene still exceeded the value of 10^{-4} (Fig. 5). These results indicate that ST had a poor efficiency in reducing the cancer risks from these VOCs. It should be mentioned that LCRs of some VOCs after treatment using the PC unit, including 1,2-dibromoethane (decreased on average from 9.7×10^{-3} to 2.43×10^{-3}), dibromochloromethane (decreased on average from 3.09×10^{-4} to 1.54×10^{-4}), 1,1,2-trichloroethane (decreased on average from 7.12×10^{-3} to 3.56×10^{-4}), 1,3-dichloropropene (decreased on average from 2.28×10^{-4} to 1.14×10^{-4}) exceeded the value of 10^{-4} . These results indicate the limited effect of PC technology on reducing the

cancer risks. Furthermore, it was found that this result was consistent with the poor photocatalytic degradation of HHs (Fig. 2), indicating the poor capability of PC technique for reducing the cancer risk derived from HHs. A previous work also reported that PC technique has a limited effect in reducing the cancer risk of HHs (Liu et al., 2017). In contrast, the definite LCRs of ethylbenzene and benzene after the PC unit treatment decreased obviously from 1.36×10^{-3} and 8.82×10^{-4} to 1.08×10^{-4} and 3.38×10^{-5} , respectively, leading to a very significant decrease in the cancer risk after the PC treatment.

It can be concluded that the integrated ST-PC process significantly reduces the negative effect on health induced by AHs. However, the risks still remained to some extent for HHs, especially, the average HRs and LCRs of dibromochloromethane, 1,2-dibromoethane, and 1,1,2-trichloroethane after ST-PC treatment were 32.17 and 1.54×10^{-4} , 4.50 and 2.43×10^{-3} , and 5.57 and 3.56×10^{-4} , respectively, which were still regarded as definite non-cancer and cancer risks. Therefore, strategies to improve the removal efficiency of HHs should be explored in future studies.

4. Conclusions

In this study, it was found that AHs-dominated exhaust gas with high OFP and high health risk was produced during the production of PCBs. The AHs contributed the highest OFP and non-cancer health risks as compared to other three VOC groups. Therefore, a pilot-scale integrated ST-PC process was developed and employed to treat AHs-dominated exhaust gas with low VOC concentration and high gas flow rates to prevent the OFP and reduce the health risk. The results indicate that ST-PC displayed outstanding removal efficiencies and durability for VOC elimination. The average removal efficiencies of 76.84%, 74.95%, 88.45% and 65.67% for AHs, HHs, AIHs, and OVOCs were obtained during the 270-day continuous treatment, respectively. In particular, the PC unit displayed high capability for degrading AH, resulting in the effective prevention of OFP and reducing the health risk. The successful application of integrated ST-PC technology for eliminating exhaust gas derived from the production of PCBs provides an efficient strategy to improve the air quality and relieve toxicological impacts on human health. Although the high removal efficiency was obtained, the average HRs and LCRs of dibromochloromethane, 1,2-dibromoethane, and 1,1,2-trichloroethane were still regarded as definite non-cancer and cancer risks, which could strongly impact human health and air quality after being discharged into air. Therefore, efforts should be devoted to study the diffusion and transformation of these toxic compounds and their effect on the health and air quality under different climates.

CRedit authorship contribution statement

Xiaolong Zhang: Methodology, Formal analysis, Writing – original draft. **Meicheng Wen:** Methodology, Data curation. **Kangwei Liao:** Visualization. **Guiying Li:** Writing – review & editing. **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. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2022.132706>.

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