



Preferential purification of oxygenated volatile organic compounds than monoaromatics emitted from paint spray booth and risk attenuation by the integrated decontamination technique



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ABSTRACT

Purification and risk attenuation of mixed oxygenated volatile organic compounds (OVOCs) and monoaromatics (total concentration: 5.8×10^5 – $1.2 \times 10^6 \mu\text{g m}^{-3}$) from a paint spray booth of auto 4S shop were investigated. The applied integrated technique of spray tower (ST) with photocatalysis (PC) displayed 2.5 times higher elimination capacity (EC) to OVOCs ($309.9 \text{ g m}^{-3} \text{ h}^{-1}$) than monoaromatics ($124.5 \text{ g m}^{-3} \text{ h}^{-1}$), due to higher synergetic elimination of these two techniques. Specifically, approximately 4.3 times higher amount of OVOCs than monoaromatics was removed by ST, contributing to their two orders of magnitude higher Henry's law constants. Similar EC trend was also observed on PC ($774.5 \text{ g m}^{-3} \text{ h}^{-1} > 518.5 \text{ g m}^{-3} \text{ h}^{-1}$). The corresponding quantum chemical calculation revealed more negative adsorption energy of OVOCs ($-40.29 \text{ kcal mol}^{-1}$) than monoaromatics ($-33.32 \text{ kcal mol}^{-1}$) onto the catalyst, resulting in more spontaneous and easy adsorption, enrichment and later degradation. Furthermore, the integrated technique displayed preferential and efficient reduction ability to both acute inhalation and chronic occupational exposure risks of OVOCs than monoaromatics. This study provided an efficient and pertinent approach to atmospheric purification and human health protection in motorvehicle repair business.

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1. Introduction

Since the beginning of the automotive industry from 1890s, the global production of motorvehicles significantly increased from 54 million in 1997 to 91 million in 2015 (from The International Organization of Motor Vehicle Manufacturers). Consequently, motorvehicle repair business accordingly expands rapidly during last few decades. Generally, most of repair items have to receive surface coatings for damaged vehicle shell redecoration, while a mixture of binder, organic solvent, pigment and additives has been inevitably applied (Viguri and Irabien, 2005). More than 70% of the used solvent end up as gaseous pollutant emissions (mainly of volatile organic compounds (VOCs)), resulting in severe atmospheric pollution and human health threat (Pierucci et al., 2005).

Although the constituent of emitted VOCs varies with the type of paint utilized, dominant composition was clarified from a mixture of monoaromatics and oxygenated VOCs (OVOCs) (Mathur and Majumder, 2008). Notably, these two groups of VOCs showed very different physical and chemical properties, including solubility and polarity. As such, to design a general control technology that can efficiently and specially eliminate these two groups of VOCs emitted from motorvehicle repair process based on their physico-chemical property disparities should be highly meaningful for our environment protection.

Given its generation of powerful oxidant active species, photocatalysis (PC) technology has attracted considerable attention recently for gaseous pollutants elimination (An et al., 2012; Ananpattarachai and Kajitvichyanukul, 2016; Chen et al., 2011b, 2013; Chen and Chu, 2011). PC has been proved to possess excellent degradation ability toward monoaromatics, OVOCs (Aghighi and Haghghat, 2015; Blanco et al., 1996; Lim et al., 2009; Moulis

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and Krysa, 2013), and the mixture of them (Lichtin et al., 1996; Palau et al., 2012) in laboratory. Notably, high degradation activity order of ethyl acetate > toluene (Chen et al., 2011a) or butyl acetate > m-xylene (Palau et al., 2011) onto TiO₂ based photocatalyst was obtained within their mixture under the different initial concentration, consistent with the order of their dielectric constants. Obviously, PC prefers degrading OVOCs to monoaromatics on TiO₂ based photocatalyst, due to the polar nature of the catalyst and much higher polar properties of OVOCs. However, the direct and overwhelming evidence to prove the more positive effect of polarity on the adsorption and degradation activities of OVOCs than monoaromatics onto the catalyst is still rare. Furthermore, all available data were obtained under ideal lab environment, and only two kinds of VOCs have been separately investigated. Whether and how does the property of VOCs influence the adsorption and degradation efficiencies in real? The questions are still unanswered yet. In our recent work, higher PC removal activity to ethyl acetate than toluene from the real electronic waste recycling process was obtained, which, however, was mainly due to significantly lower inlet concentration of ethyl acetate ($45 \mu\text{g m}^{-3}$) than toluene ($513 \mu\text{g m}^{-3}$) (Liu et al., 2016). Different from the dominant amount of monoaromatics from electronic waste recycling process, surface coating operation of car inside paint spray booth (PSB) using organic solvents would generate equal and even higher level of OVOCs than monoaromatics (Bratveit et al., 2004; Pierucci et al., 2005). Thus, the investigation of whether and why the PC preferentially eliminates OVOCs than monoaromatics in PSB is meaningfully and credibly, which will then give a solid answer to the above questions. However, up to date, such kind of works was not attempted yet.

In addition, complex real-situations would deactivate the catalyst during the long-term operation of photocatalytic technology, mainly due to the coexisted particulate matters within waste gas, such as the mists from paint spray process. Therefore, to maintain the high efficiency and stability of PC technique, and reduce the adverse effects of them on human health, the particle should be pretreated ahead of this technology. Spray tower (ST) with water is one of the most widely used control devices to remove the oil mist or particles from industrial effluent gases, mainly due to its easy operation, simple construction and high efficiency (Almuhanna et al., 2009; Mohan et al., 2008). For instance, nearly all the micron-size particles could be removed by ST technology under optimal operating conditions (Darcovich et al., 1997). Besides, this technology also showed its preference to remove hydrosoluble gaseous organics (Kim et al., 2000).

Therefore, in this study, these two efficient and selective techniques were contrapuntally united to develop an integrated ST-PC technique for OVOCs and monoaromatics mixture elimination as well as for their risks reduction in PSB. Based on the available physico-chemical data of VOCs and quantum chemical calculations, this study also will try to verify the hypothesis that whether and why ST, PC and their integrated techniques preferred to reduce the OVOCs as well as their acute and chronic risks than monoaromatics. The obtained data would provide very useful guidance for pertinently VOCs pollution and risk control in motorvehicle repair business.

2. Experimental

2.1. Study site and set-up

The experiments were conducted in a PSB of an auto 4S shop in Guangdong, China, which was mainly engaged in repainting damaged car shells. During the working period, high-pressure airbrush was used to spray the designed paint mixture onto the

car shell surfaces, and gaseous VOCs were discharged into atmospheric environment.

An integrated ST-PC reactor (Fig. S1) was fabricated to purify the emitted waste gas. The gas was firstly fed into the ST reactor (Height × Diameter, approximately 4500 mm × 2000 mm) and then to PC unit (1900 mm × 1900 mm × 1500 mm) by a centrifugal pump mounted at the outlet of PC reactor to obtain a stable flow-rate of $16,000 \text{ m}^3 \text{ h}^{-1}$. Detail information of set-up is provided in Supporting Information (SI).

2.2. Sample collection and analysis

VOCs samples were collected from inlet, outlet of ST and outlet of PC, respectively, using 2.7-L stainless Summa canisters (ENTECH Instruments Inc, Silonite™) during the paint spray processes for three different times (denoted as 1st, 2nd and 3rd). Then the samples were qualitatively and quantitatively analyzed on a Entech 7100 pre-concentrator (Entech Instruments Inc., CA, USA) followed by gas chromatography-mass spectrometry (7890-5975 GC-MS, Agilent technologies, USA) according to the US Environmental Protection Agency (USEPA) TO-15 method (USEPA, 1999). A control point was selected at about 10-m outside of the PSB. Detail sampling and analysis information as well as quality assurance and quality control were provided in the SI and in our previous work (He et al., 2015).

2.3. The calculation of adsorption configuration and adsorption energy

The adsorption configuration and energy of different VOCs molecules onto TiO₂ were also calculated using the Gaussian 09 software package and the detail description of the computation method is provided in the SI.

2.4. Risk assessment

The acute and occupational exposure risks of VOCs emitted during the paint spray processes were evaluated to understand their potential acute and chronic effects on the workers after short- and long-time inhalation, and the corresponding risk evaluation methods were provided in the SI.

3. Results and discussion

3.1. VOC pollution profile and risk evaluation

The components and concentrations of VOCs emitted during the paint spray processes in the PSB were firstly investigated. As Table 1 shows, a total of 23 VOCs were detected and categorized into two groups of monoaromatics and OVOCs for convenience. Fig. 1a and b displays the concentration and percentage of these VOCs, respectively. As can be seen, the sum of VOC concentrations was in the range of 5.8×10^5 to $1.2 \times 10^6 \mu\text{g m}^{-3}$, which was at least six times higher than Guangdong provincial emission pipe emission standard of VOCs for surface coating of automobile manufacturing industry ($9.0 \times 10^4 \mu\text{g m}^{-3}$, DB44/816-2010). This indicated that the atmosphere environment in the PSB was highly contaminated by VOCs, and must be purified before emission. Meanwhile, higher percentage of OVOCs was found (64.8% in average) than that of monoaromatics (35.2% in average) among these VOCs. Similar results were also obtained in other PSBs (Martinez-Soria et al., 2009; Qi et al., 2005). Notably, approximately $5.9 \times 10^3 \mu\text{g m}^{-3}$ of VOCs were detected at the control point, which was almost three times higher than Guangdong fugitive emission standard of VOCs for surface coating of automobile manufacturing industry

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