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A review of whole-process control of industrial volatile organic compounds in China

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ABSTRACT

Volatile organic compounds (VOCs) play an important role in the formation of ground-level ozone and secondary organic aerosol (SOA), and they have been key issues in current air pollution prevention and control in China. Considerable attention has been paid to industrial activities due to their large and relatively complex VOCs emissions. The present research aims to provide a comprehensive review on whole-process control of industrial VOCs, which mainly includes source reduction, collection enhancement and end-pipe treatments. Lower VOCs materials including water-borne ones are the keys to source substitution in industries related to coating and solvent usage, leak detection and repair (LDAR) should be regarded as an efficient means of source reduction in refining, petrochemical and other chemical industries. Several types of VOCs collection methods such as gas-collecting hoods, airtight partitions and others are discussed, and airtight collection at negative pressure yields the best collection efficiency. Current end-pipe treatments like UV oxidation, low-temperature plasma, activated carbon adsorption, combustion, biodegradation, and adsorption-combustion are discussed in detail. Finally, several recommendations are made for future advanced treatment and policy development in industrial VOCs emission control.

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Introduction

VOCs (volatile organic compounds) are currently regarded as important pollutants from the viewpoint of air pollution control. However, there is no uniform definition worldwide. The World Health Organization (WHO) defines them as organic compounds with melting points lower than room tem-

perature and boiling points between 50 and 260°C. The US Environmental Protection Agency (US EPA) defines VOCs as any carbon-containing compounds that participate in atmospheric photochemical reactions except for CO, CO₂, H₂CO₃ and metal carbides or carbonates. In China, however, according to the newly formulated standard named Unorganized Emission Control Standard for Volatile Organic Compounds (GB37822-2019), VOCs are defined as organic compounds that

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participate in atmospheric photochemical reactions or that could be identified according to relevant regulations. It is concluded that China now has a clearer understanding of VOCs, which can be expressed by the following: on the one hand, all those that participate in photochemical reactions are included, which focuses the emphasis on air pollution control; on the other hand, from the perspective of actual supervision, all that can be measured by instruments or identified by regulations are covered. With the enhancement of regulations and increasing determination capabilities in the future, more and more VOCs will be under supervision.

VOCs mainly include alkanes, alkenes, aromatic hydrocarbons, esters, ethers, ketones, aldehydes, etc. About 30% of VOCs are toxic and odorous compounds according to Chen et al. (2011). VOCs can irritate the eyes and upper respiratory system, and short-time exposure can cause headaches, nausea, vomiting, and weakness of the limbs; convulsions, coma, and memory loss may occur in some cases (Habebullah, 2015; Matysik et al., 2010). Studies also show that long-term exposure to certain concentrations of VOCs can cause damage to the human liver, kidneys, brain and nervous system (Bari and Kindziarski, 2017; Heibati et al., 2018; Masih et al., 2018; Sofuoglu et al., 2011; Tan et al., 2013). As for air pollution control, VOCs are key factors in that VOCs play an important role in the formation of ground-level $PM_{2.5}$ and O_3 (He et al., 2019a; Lu et al., 2012; Sun et al., 2013; Yuan et al., 2013; Zhao et al., 2013). Under solar illumination, VOCs react with other chemical components in the atmosphere such as NO_x to form ozone (Jiang et al., 2015; Zeng et al., 2020; Wang et al., 2022). Meanwhile, VOCs can also participate in a series of chemical reactions, and secondary organic aerosol can be generated (Li et al., 2020a; Wang et al., 2017a; Zhan et al., 2021). As mentioned above, VOCs are regarded as important precursors of $PM_{2.5}$ and O_3 from the viewpoint of air pollution prevention and control (Liu et al., 2021; Shao and Dong, 2013).

VOCs have a wide range of sources, including natural and anthropogenic sources. As for the former, biological emissions (such as those from vegetation, oceans, soil microorganisms, etc.) and non-biological processes (such as earth movement, forest burning, etc.) are the two main sources (Ye and Chen, 2017). As for the latter, industrial emissions, transportation emissions, agricultural releases and living source dispersion are the major contributors. At the global scale, the emissions from natural sources are greater than those from anthropogenic sources, but at the regional scale, especially in China, the emissions from anthropogenic sources are slightly higher than those from natural sources (Wei et al., 2008). Ye calculated the emissions of anthropogenic VOCs in China from 2011 to 2019 based on the emission coefficient method and the results showed that the emissions from anthropogenic sources increased from 22,976,500 to 27,974,500 tons with an increase of 21.75% (Ye et al., 2020), of which industrial activities accounted for more than 50% of total anthropogenic emissions, and the primary species of industrial VOCs were alkanes, aromatic hydrocarbons, chlorinated hydrocarbons, oxygenated hydrocarbons and so on. As for specific industries, VOCs vary depending on the raw materials, processes and treatments. For example, in the petrochemical industry, VOCs are characterized by alkanes and aromatic hydrocarbons with more than 70% contribution from pres-

sure reduction, sulfur recovery, delayed coking and oil storage units (Qi, 2018). By contrast, VOCs are characterized by halogenated hydrocarbons, aromatic hydrocarbons, and oxygenated species for the dye, coating, and pesticide industries, respectively (Shao, 2019). More details about VOCs emissions characteristics and the impact of treatments on emissions can be found elsewhere (Chen, 2011; Wang et al., 2013; He, 2016; Ke, 2020).

Due to the public health threat, important impact on $PM_{2.5}$ and O_3 and notable contribution to anthropogenic sources, industrial emission is considered to be one of the most important sources of VOCs in China. Currently, China has successively issued a series of policies and regulations to strengthen the governance of industrial VOCs. In 2018, the State Council released the Three-year Action Plan to Fight Air Pollution, in which petrochemical, chemical, industrial coating, packaging and printing, gasoline storage, transportation and marketing were listed as the key industries that need to be comprehensively controlled. Leak detection and repair standards (LDAR) and technical guidelines for VOCs treatment are required. Production and usage of solvent-based coatings, inks, or adhesives with high VOCs content are prohibited in some key regions. By 2020, total emissions were mandated to decrease more than 10% compared with 2015. In 2019, the Ministry of Ecology and Environment released the Comprehensive Treatment Plan for Volatile Organic Compounds in Key Industries. This plan proposed to establish and improve the VOCs pollution prevention and control management system by 2020 and implement continuous actions for the improvement of ambient air quality (MEE, 2019). In 2020, the Ministry of Ecology and Environment announced Critical Treatment Battles against Volatile Organic Compounds Pollution, which pointed out that petrochemicals, chemicals, industrial coatings and others were the key objects. Substitution of low (no) VOCs content raw and auxiliary materials, collection enhancement for unorganized process emissions and in-depth treatment of end-pipe emission were required simultaneously (MEE, 2020). It is noted that local governments also issued relevant policy documents during the same period, which have put forward specific treatment plans for local key enterprises and key industries. The treatment of VOCs emissions from industrial sources has drawn unprecedented attention and great changes have taken place in VOCs treatment in recent years. On the one hand, end-pipe treatment has evolved from whether treatment facilities exist to whether such facilities work or not. On the other hand, the single end-pipe treatments of past years have been replaced by whole-process treatment, including source reduction, collection enhancement and in-depth treatment of end-pipe emissions. Besides, the number of detailed industrial emission standards has also increased remarkably. It can be concluded that the treatment of VOCs has stepped into a new age.

In this article, with the aim of introducing the current VOCs treatments for industrial sources, all works in the field of source reduction, collection enhancement, and in-depth treatment of end-pipe emissions are summarized, as well as the emission standards and technical specifications. Finally, the whole-control technology route and other suggestions are recommended with the purpose to provide references and sup-

port for the better whole process control of industrial VOCs in the future.

1. Source reduction

Compared with end-pipe treatment, source reduction has only developed in recent years and has drawn more and more attention since 2017. Source reduction is related to two different activities at present. One is LDAR and the other is material substitution. As for the former, LDAR is commonly implemented in the petrochemical and chemical industries. As for the latter, industrial practices such as the usage of coatings and organic solvents have vigorously reduced the usage of solvent-based paints and inks, and materials with lower VOCs content such as water-based inks and water-based coatings have been encouraged and developed.

LDAR refers to a system that detects and repairs leakage from units in the entire process of petrochemical or chemical production (Zhang and Zhang, 2018; Zhao and Chen, 2018). According to the Guidelines for Leak Detection and Repair of Petrochemical Enterprises (MEE, 2015), LDAR workflow mainly includes three steps: project establishment, on-site detection and leak repair. Generally speaking, units of devices such as valves, flanges, vacuum pumps and open pipelines are positioned and put into the system firstly, and then portable VOCs instruments with flame ionization or photo ionization detectors are employed to quantitatively detect or inspect those units to determine whether the units leak or not. If leaks are found, effective measures will be taken to repair the leak at a certain time with the aim of controlling the loss of material by leakage and reducing environmental pollution from the target unit. This technology is widely used in VOCs reduction from leakage in petrochemical and chemical industries (Pacsi et al., 2019; Ravikumar et al., 2020; Zhao and Chen, 2018).

Developed countries such as the United States of America (USA) and the European Union (EU) began to control VOCs emissions from the petrochemical industry through the implementation of LDAR in the 1980s, and significant VOCs reduction has been achieved (Lu et al., 2011). The US EPA has evaluated companies that implemented LDAR and concluded that about 63% of leakage was avoided by repairs and that the total VOCs emissions from petroleum refineries could be reduced by 56% (Pacsi et al., 2019). In China, however, LDAR was introduced at the beginning of the 21st century to effectively control the VOCs emissions of petroleum and petrochemical devices (Zhang, 2016). Ke (2020) analyzed and compared the application of LDAR in four oil refineries and six typical devices, and the results showed that about 0.2%–0.4% of total units were defined as leak points, which accounted for about 91.8% of total unit linkage. By repairing the devices, about 42%–57% reduction of total VOCs emissions could be achieved (Ke et al., 2020a). Similar results could also be seen in other studies (Chen et al., 2018; Ravikumar et al., 2020). Due to the significant contribution to VOCs emission reduction, LDAR is also employed in other industries such as coking, pesticide manufacture, pharmaceutical manufacture and other industries involving VOCs emissions, the Thirteenth Five-Year Plan for Volatile Organic Compounds Emission Reduction clearly requires that the coking industry must fully implement LDAR,

and that pharmaceuticals, pesticide, coatings, ink, and adhesives manufacture and other industries should start with LDAR gradually (MEE, 2019).

As for material substitution, its implementation was rather late compared with LDAR. In early years, VOCs control and supervision in China mainly focused on end-pipe emission treatment and rarely involved source control. Only in recent years was it realized that end-pipe emission reduction was not as satisfactory as expected, and that more attention should be given to material substitution (Ke, 2020). Besides, the potential for reducing VOCs emissions from industries related to usage of paints, inks and organic solvents is huge. In 2018, the VOCs emissions from industrial sources amounted to 12,698,000 tons, of which that related to the usage of paints, inks and organic solvents accounted for 59% (Liang et al., 2020). Therefore, it is possible and necessary to reduce the emission of VOCs by material substitution. At present, for the container manufacturing industry, the replacement rate of water-based coatings has reached about 90%, and the replacement rate of water-based coatings in the auto manufacturing industry for middle and base paint coating is about 50%, which is basically equivalent to that of the United States and lower than that of Europe, with a 60% replacement rate. In the engineering machinery manufacturing industry, the replacement ratio of powder- and water-based coatings falls in the range of 20%–25%, which is at the same level as the automotive metal parts manufacturing industry, with a 23% water-based coating replacement ratio. It is noted that although 100% replacement by water-based coatings is technically feasible in the furniture manufacturing industry, the actual replacement ratio is only 12%, which is lower than the average replacement ratio of 50% in Europe. It is thought that the low replacement ratio is mainly influenced by the 30% increase in cost compared with solvent-based coatings. For industries like steel structures, automotive plastic parts manufacturing and ship building, the coating processes are still dominated by solvent-based coatings and the replacement ratios of water-based coatings are all less than 3%. More details on material substitution in various industries can be found elsewhere (CNCIA, 2021).

Research has shown that for industrial coatings such as those used in typical furniture manufacturing and automobile manufacturing processes, the main components of VOCs are benzene series and esters for solvent-based coating usage, while alcohol ethers are the typical components of water-based coatings (Ke et al., 2020b). The substitution of solvent-based coatings by water-based ones can lead to 20%–70% reduction in total VOCs emission based on data from different industries (Ye et al., 2020). Besides, Gao's study also showed a similar change trend for VOCs emission components before and after substitution with water-based architectural coatings, but the result was more remarkable, with 90% VOCs emission reduction (Gao et al., 2021).

Besides, China has also shown its firm determination for control of VOCs emissions from source materials. In 2020, seven mandatory standards on the content limits of VOCs were released, which are Limit of Harmful Substances of Woodenware Coatings (GB18581-2020), Limit of Harmful Substances of Industrial Protective Coatings (GB30981-2020), Limit of Harmful Substances of Vehicle Coatings (GB24409-2020), Limit of Harmful Substances of Architectural Wall Coat-

Table 1 – Summary of standards for VOCs-containing materials.

| VOCs-containing Standards | Targets | Limit values (g/L)* |
|---|-----------------------------|--|
| Limit of Harmful Substances of Woodenware Coatings GB18581-2020 | Water-based paint | 250, 300 |
| | Solvent paint | 420, 450, 700 |
| | Radiation curing paint | 250, 420 |
| | powder paint | 0 |
| Limit of Harmful Substances of Architectural Wall Coatings GB18582-2020 | Water-based wall paint | 80, 100, 120 |
| | Decorative board paint | 120, 250, 580, 760 |
| Limit of Harmful Substances of Vehicle Coatings GB24409-2020 | Water-based paint | 250, 300, 350, 420, 480, 530 |
| | Solvent paint | 480, 500, 530, 540, 550, 560, 580, 600, 630, 680, 750, 770, 840 |
| Limit of Harmful Substances of Industrial Protective Coatings GB30981-2020 | Radiation curing paint | 150, 200, 400, 550 |
| | Solvent-free paint | 100 |
| | Water-based paint | 250, 300, 350, 420, 480 |
| | Solvent paint | 420, 480, 500, 520, 540, 550, 600, 630, 650, 680, 700, 720, 750, 780 |
| Limit of Volatile Organic Compounds Content in Adhesive GB33372-2020 | Radiation curing paint | 150, 200, 400, 550 |
| | Water-based paint | 50, 100, 150, |
| | Solvent-based | 250, 400, 450, 500, 600, 700, 850 |
| | Native adhesive | 20, 50, 100, 150 |
| Limit of Volatile Organic Compounds Content in Printing Ink GB38507-2020 | Water-based ink | 5, 15, 25, 30 |
| | Solvent ink | 75, 95 |
| | Offset printing ink | 3, 10 |
| | Energy curing ink | 2, 5, 10 |
| | Engraving gravure ink | 20 |
| Limit of Volatile Organic Compounds Content in Cleaning Agents GB38508-2020 | Aqueous Cleaning Agent | 50 |
| | Semi-aqueous Cleaning Agent | 300 |
| | Solvent Cleaning Agent | 900 |

* The unit of limit values in GB38507-2020 is %, others are all presented by g/L.

ings (GB18582-2020), Limit of Volatile Organic Compounds Content in Adhesive (GB33372-2020), Limit of Volatile Organic Compounds Content in Cleaning Agents (GB38508-2020), Limit of Volatile Organic Compounds Content in Printing Ink (GB38507-2020), respectively. These standards established limits on the VOCs contents of solvent-based, water-based and other materials, with more details shown in Table 1.

2. Enhanced collection

In terms of process control following the principle of collecting all that is possible, it is necessary to convert all the unorganized emissions into organized ones for better control. According to rule 45 in the Law of the People's Republic of China on the Prevention and Control of Atmospheric Pollution, VOCs from all industrial activities must be collected and treated before emission. With the implementation of the national Air Pollution Prevention and Control Action Plan (2013–2017), VOCs emissions have mainly been collected, and the emission of VOCs from such industrial activities without collection is rarely seen under the requirements for more and more stringent supervision. Therefore, effective collection of VOCs is a prerequisite for subsequent treatment. In actual VOCs emission control, with the aim of reducing the impact of VOCs on the environment, gas-collecting hoods and airtight compartments are often adapted for collection. A gas-collecting hood refers to a device used to collect the VOCs emissions from processes. Due differences in process operations, a gas-collecting hood can be designed in various forms. According to

the waste gas flow mode of the hood, hoods are divided into two categories: suction collecting hoods and blowing-suction collecting hoods. According to the relative position and location of pollution sources, suction collecting hoods are further divided into airtight hoods, exhaust cabinets, external collecting hoods and so on. Compared with a gas-collecting hood, an airtight compartment under positive or negative pressure is more effective, because cutting off the diffusion of VOCs yields less air flow interference and lower ventilation volume.

At present, industries have different methods for collecting unorganized VOCs. In the packaging and printing industry, most plants mainly adopt sealed and centralized ventilation collection, such as airtight cabinets during the drying process, but most of them do not set up small compartments for each process, and sealing areas still remain sources of fugitive emissions (Xu, 2019). For the coating industry, the collection method most commonly used is the top gas hood (Gao, 2015), but this collection method is not efficient, and fugitive VOCs are accumulated with high concentrations in the workshop and cause threats to the health of workers. As for the furniture manufacturing industry, problems such as improper storage and usage of VOCs containing raw and auxiliary materials and unreasonable layouts for VOCs hoods and pipelines are often seen (Dang et al., 2021). Generally speaking, only VOCs emissions from vehicle manufacture, electronic manufacture and several other industries are well collected by airtight compartments under positive or negative pressure during the whole coating processes. Most industries with VOCs emissions are not characterized by effective collection.

Table 2 – Summary of major methods for VOCs collection from industrial sources.

| Collecting method | Efficiency | Description and application |
|----------------------|------------|---|
| Gas-collecting hood | 60%–70% | Easy to implement with lowest cost and used in most industries like surface coating, chemical production, printing and others. Due to the limited collection efficiency, excessive emission of unorganized VOCs occurs in some cases. |
| Collecting cabinet | 80% | More expensive and more efficient than gas-collecting hood due to the design of three -sided fence, not so popular as gas-collecting hood. Some cases in laboratory waste gases collection are seen. |
| Airtight compartment | ≥ 90% | The most expensive and efficient among the existing collection methods with the least fugitive VOCs. Airtight compartments under negative pressure are often applied in furniture manufacturing, automobile manufacturing and others, and airtight compartments under positive pressure are applied in pharmaceuticals, food manufacturing and so on. |

According to the empirical determination method, the collection efficiency of the gas-collecting hood is about 60%-70% or even less, the collection efficiency of the collecting cabinet is about 80%, and the collection efficiency of the airtight compartment under positive or negative pressure is around 90%. Considering the application of widely used gas-collecting hoods and their limited collection efficiency, the Standard for Unorganized Emissions of Volatile Organic Compounds (GB37822-2019) clearly requires that the wind speed of the farthest collecting points should not be less than 0.3 m/sec under the gas-collecting hood mode. In addition, conveying pipelines should be sealed, and VOCs collection should be operated under negative pressure. If it is operated under positive pressure, leak tests should be done. It has been proved that the most effective way to collect VOCs is airtight compartments under negative pressure. However, due to the cost of operation and investment, the current most popular collection method still remains the gas-collecting hood (Ke, 2020). A summary of different collection methods can be seen in Table 2.

3. End-pipe treatment

Generally, VOCs end-pipe treatments can be divided into recovery and destruction technologies (Cui et al., 2016; Kamal et al., 2016). Based on differences in the physical and chemical properties of VOCs, enrichment and separation of VOCs are achieved by changing temperature, pressure or using selective absorbents. This mainly includes adsorption, absorption, condensation, membrane separation and so on (Bouchra et al., 2016; Hariz et al., 2017; Zhang et al., 2017). Destruction usually converts VOCs into harmless substances such as CO₂ and H₂O through combustion, including thermal combustion and catalytic combustion. In addition, low-temperature plasma, photo-oxidation and biodegradation are also employed in VOCs treatment (Chen et al., 2017; Khalilzadeh and Fatemi, 2016; Qi et al., 2013). Meanwhile, in recent years various combined technologies have developed rapidly, such as adsorptive concentration combined with catalytic combustion and zeolite concentration combined with thermal combustion (Xi et al., 2012; Luan et al., 2011, 2014; Mohamed et al., 2014). The characteristics and application

of major VOCs treatment technologies are summarized in Table 3. The following are the major technologies used frequently in industrial VOCs treatment.

3.1. Adsorption

Adsorption mainly depends on an abundant microporous structure on the surface of the adsorbent to trap VOCs molecules. At present, the most commonly used adsorbent substance is activated carbon. It is suitable for the removal of low and medium concentrations of VOCs. Activated carbon adsorption is simple, economical and can be used in most industries (Wang et al., 2015, 2020a; Yang et al., 2018a; Zhang et al., 2017). Quite a few studies focus on promoting the performance of activated carbon with larger surface areas and adjustable porous channels to obtain more adsorption capability, and others put emphasis on the adsorption mechanism for typical VOCs like benzenes and halohydrocarbons (Wang et al., 2012, 2019, 2020b; Li et al., 2020b; Zhu et al., 2020). Polymeric resins and zeolite are also used as adsorbents to a rather small extent (Wang et al., 2014a, 2021a). It has been noted that a disadvantage of adsorption application in coating industries is the presence of smog. If smog is not thoroughly pretreated, the existing smog could adsorb on the surface of the activated carbon and result in rapidly decreasing adsorption performance. Another factor that effects adsorption performance is the adsorption temperature. As is known, VOCs adsorption is classified as physical adsorption, and good performance is usually achieved at low temperature below 40°C (HJ 2026-2013). Therefore, in summertime, VOCs adsorption is not as satisfactory as expected, especially for treatments exposed to outdoor temperatures.

3.2. Recovery

As mentioned above, the choice of recovery depends on multiple factors, including the properties, concentration, volume, recovery efficiency, field conditions, recovery value, etc. For lower concentrations of VOCs below 5000 mg/m³, activated carbon adsorption combined with hot air or steam desorption is suitable. When the concentration exceeds 5000 mg/m³, dilution is required before treatment. For VOCs with huge vol-

Table 3 – Summary of current industrial VOCs treatment technologies.

| Technology categories | Treatment concentration (mg/m ³) | Treatment volume (m ³ /hr) | Treatment temperature (°C) | Target industries | Investment (CNY, 10,000 m ³ /hr) | Operating cost (CNY, 1000 m ³ /hr) | VOC removal efficiency |
|--------------------------------|--|---------------------------------------|----------------------------|--|---|---|------------------------|
| Physical adsorption | 0–100,000 | 1000–50,000 | < 40 | Industries with low and intermittent emissions like auto repair, plastic fabrication and so on | 10,000 | 0.4–0.5 | < 70% |
| Chemical absorption | 0–100,000 | 1000–10,000 | < 40 | Alcohol, Sewage treatment, Chemicals, Pharmaceuticals, etc. | 2000–10,000 | 0.1–0.5 | 5%–90% |
| Condensation | 10,000–100,000 | 1000–10,000 | < 150 | Oil and gas recovery | 300,000 | 1.0–2.0 | 70% |
| Membrane separation | 10,000–100,000 | 100–4000 | < 40 | Oil and gas recovery | 10,000 | 0.4–0.6 | 90% |
| Thermal combustion | 2000–80,000 | 1000–100,000 | ≥ 0 | Chemical industry, Shoemaking, Tanning, etc. | 100,000 | 0.4–1.2 | 97% |
| Regenerative thermal oxidation | 2000–12,000 | 1000–100,000 | ≥ 0 | Petroleum refining, Petrochemical, coating, Petrochemicals, etc. | 100,000 | 0.4–0.8 | 95%–98% |
| Catalytic oxidation | 2000–8000 | 1000–100,000 | < 400 | Coating, Various chemical processes. | 80,000–100,000 | 0.4–1.0 | 90%–95% |
| Biodegradation | 120–2000 | 1000–120,000 | 10–45 | Sewage treatment, Composting, Chemicals, Pharmaceuticals, etc. | 10,000–40,000 | 0.6–1.2 | 50%–80% |
| UV oxidation | 100–300 | 1000–50,000 | < 80 | Sewage treatment, Rubber manufacturing, etc. | 2000–5000 | 0.1–0.2 | 10%–30% |
| Low-temperature plasma | 120–500 | 1000–60,000 | < 80 | Sewage treatment, Rubber manufacturing, etc. | 6000–10,000 | 0.1–0.3 | 50%–70% |
| Adsorption-recovery | 300–120,000 | 5000–150,000 | < 40 | Oil and gas recovery, Pharmaceuticals, Petroleum refining, Petrochemicals, etc. | 100,000–420,000 | 4–8 | 80%–90% |
| Adsorption-combustion | 120–2000 | 1000–180,000 | < 40 | Coating, Printing, Chemical industry, Shoemaking, Tanning, Electronics manufacture, etc. | 30,000–60,000 | 0.4–0.6 | 80%–90% |

CNY: Chinese Yuan.

ume, it is necessary to install desorption equipment on site. For small-volume applications, the saturated activated carbon can be transported and treated in a centralized facility. The problem is that the waste liquid produced is not easy to handle and is generally regarded as hazardous waste. Condensation has a better treatment effect on VOCs with high boiling points. For those with medium and high volatility, it does not work well. Condensation is suitable for VOCs of 50,000 mg/m³ or even higher, under low-temperature and high-pressure operation conditions, and non-condensable gas is generated during the treatment process. In fact, condensation is rarely used alone due to the high cost of investment and operation and low recovery efficiency. Membrane separation is more suitable for processing VOCs below 1000 mg/m³. In most cases, fluctuation of the temperature, pressure, flow rate and VOCs concentrations within a certain range requires recovery devices with a certain level of adaptability, and membrane separation can meet this requirement and work well. The selection of cycling technology depends on the specific application. Activated carbon adsorption mainly deals with emission to the atmosphere and recycling is just an auxiliary process to save in-

vestment. Membrane separation mainly focuses on recovery. Due to the high recovery rate, economically valuable VOCs are basically fully recovered and the amount of unrecovered material is very small. Membrane separation is usually used for oil and gasoline recovery in the petrochemical industry, and unreacted monomers and organic solvents in the pharmaceutical industry (Kang, 2015; Maryam et al., 2007).

3.3. Combustion

Combustion is a traditional treatment for VOCs control. VOCs are converted into CO₂ and H₂O by combustion or oxidation at rather high temperatures. Combustion is commonly considered to be a highly efficient method for VOCs removal, and there are three types of combustion treatments presently used, which are thermal oxidation (TO), regenerative thermal oxidation (RTO) and catalytic oxidation (CO), respectively (He et al., 2019b; Kamal et al., 2016). Thermal combustion carried out at 700–900°C or even higher temperature is often required for VOCs with high concentration or high calorific value. In most cases, other fuels like diesel oil or natural gas

need to be added for complete combustion. Compared with TO, RTO is more popular in that the heat is reusable and the operation cost is relatively economical. As for catalytic oxidation, the operation cost is the least due to the conservation of the catalyst during the oxidation. In most cases, the temperature of catalytic oxidation is about 300–350°C, in which the catalyst plays an important role (Wang et al., 2017b; Huang et al., 2013; He et al., 2012, 2018; Sun et al., 2019). Numerous researchers focus on preparing catalysts with good performance, especially for the chlorinated and sulfur-containing VOCs, which could easily make the catalyst inactive (Abdullah et al., 2006; Huang et al., 2011; Darif et al., 2017; He et al., 2016; Wang et al., 2014b; Xing et al., 2020; Li et al., 2020c). Others mainly study the mechanism during the catalytic oxidation process (Liu et al., 2012, 2017; Pan et al., 2017; Li et al., 2016, 2021; Yue et al., 2013; Yang et al., 2018b; Jian et al., 2018). Although the daily operating cost of CO is lower, the operation conditions are more demanding and efficiency is lower than RTO, which results in less satisfactory application compared with RTO. Meanwhile, it should be pointed out that combustion is characterized by high VOCs removal efficiency, but this method may be contrary to the present hot topic and policy of carbon reduction due to the production of carbon dioxide during combustion and high cost during operation.

3.4. Biodegradation

Biodegradation is a relatively new technology, which was first applied to waste gas deodorization. In recent years, biodegradation has gradually been applied to the treatment of VOCs (Lu et al., 2021; Malakar et al., 2017; Yoshikawa et al., 2017). Compared with other technologies, it has the advantages of simple equipment, low investment and operating costs and no secondary pollution. However, due to the low degradation rate, it is economical only for low-concentration VOCs treatment. In addition, biodegrading bacteria are highly selective and only water-soluble VOCs are suitable for removal by biodegradation. According to the operation modes, biodegradation mainly consists of three modes, which are filtering, washing/scrubbing and trickling. The washing mode is suitable for the removal of VOCs with small volume, high concentration, easy dissolution and slow biological metabolism rate. For high-volume, low-concentration VOCs, a filtering bed is more suitable; for VOCs yielding acidic substances after degradation, a trickling filter bed is suitable. Generally, due to the advantages of green environmental protection and lower treatment cost, domestic research on the treatment of VOCs by biodegradation has made rapid progress in recent years (Miao et al., 2019; Qin et al., 2020; Khoramfar et al., 2020; León et al., 2020; Du et al., 2021), which make it possible that biodegradation will be a hot topic in VOCs treatment in the future.

3.5. Low-temperature plasma

Plasma is called the fourth form of matter and is composed of electrons, ions, free radicals and neutral particles. Low-temperature plasma technology uses the plasma generated by a dielectric discharge to repeatedly bombard the VOCs

molecules at a very high rate to deactivate, ionize, and crack the molecules through a series of complex oxidation processes (Feng et al., 2018). Since C–S and S–H bonds are relatively easily broken, low-temperature plasma has good performance in the removal of odors. For the removal of VOCs, the results are not as good as for odors. The key to this technology is whether the design of the plasma generator is reasonable. As a rather new technology, current research on the mechanism of action is not sufficient (Ma, 2019), and there is no general understanding of how to design plasma generators for different VOCs in different industries. In general, the VOCs removal efficiency is rather low, which limits practical application.

3.6. UV photo-oxidation

UV photo-oxidation was the one of the most popular treatments in past years, and has been recommended by some local authorities due to its low investment, low cost and eco-friendly operation. However, it has now been proven to be invalid. According to theory, the chemical bonds of VOCs could be broken under irradiation by light with wavelength less than 185nm, and the breaking of double and triple bonds of VOCs like alkenes and alkynes needs more energetic wave lengths less than 170 nm. However, UV lamps generating 170 nm light have not been developed up to now, and 185 nm sources only account for a small proportion of the total in the current UV light market, at less than 3%. So, the irradiated energy is not powerful enough to break the bonds of VOCs, especially with the presence of detected alkenes and alkynes in some cases. In fact, UV photo-oxidation technology was banned by the Ministry of Ecology and Environment in 2019 and 2020 due to its low removal efficiency and secondary ozone pollution (Wang et al., 2021b).

3.7. Adsorption-combustion

Adsorption is suitable for the treatment of low-concentration VOCs, while combustion technology is suitable for high-concentration VOCs. At present, VOCs emissions from most industries are characterized by low concentration and large volume, which makes direct combustion expensive. For this reason, adsorptive concentration combined with catalytic oxidation or high-temperature oxidation technology has been developed. When VOCs emissions do not contain poisoning substances, catalytic combustion is usually used for post-treatment; otherwise, high-temperature combustion is used. Generally, the adsorbent containing VOCs is purged and regenerated with a small amount of hot gas flow, and the purged-out VOCs enter the combustion chamber for oxidation without extra energy for heating due to the highly concentrated VOCs. The heat from the combustion process not only can be used for regeneration of the adsorbent after temperature adjustment, but also can be used for heating fresh air. In this way, the calorific value of VOCs can be fully utilized, and the operating cost can be significantly reduced.

Honeycomb activated carbon is usually used as the adsorbent. It has the advantages of low pressure resistance and good kinetic performance, but it also has some serious flaws.

Firstly, safe operation is not guaranteed. When the temperature of the regenerated hot air stream exceeds 100°C, the adsorbent is likely to burn due to its low ignition temperature. Secondly, VOCs with high boiling point will remain on the surface of activated carbon due to the regeneration temperature beyond 120°C, which makes the adsorption capability decrease gradually. Finally, activated carbon has a strong water absorption capacity. When the relative humidity of VOCs exhaust is high (over 70%), the adsorption capacity decreases rapidly. In fact, Japan began to study the use of modified silica-alumina molecular sieve instead of activated carbon in the 1990s. Molecular sieve adsorbents are characterized by good safety, can be desorbed and regenerated at high temperatures (up to 220°C) and can be used for most VOCs. These adsorbents have been introduced in China in recent years. At present, the problems of preparation and molding of modified molecular sieves have been basically resolved, and they are expected to be widely applied in the treatment of VOCs in industries.

Although all the technologies mentioned are represented in the treatment cases, there is a clear difference in the application ratio. Technologies with high VOCs removal efficiency such as combustion or adsorption-combustion are not as popular as single activated carbon adsorption or low-temperature plasma. Gao et al. (2015) conducted field investigations on 285 typical enterprises in the Pearl River Delta region and revealed that activated carbon adsorption, solution absorption, low-temperature plasma, and water spray combined with activated carbon adsorption accounted for 45%, 21%, 15%, and 10%, respectively (Gao et al., 2015b). Su and others selected 130 companies in six key industries to investigate the application of VOCs treatments, and the results showed that the most popular treatment was activated carbon adsorption, accounting for 43.08%, followed by low-temperature plasma, water spray combined with activated carbon adsorption and UV oxidation, which accounted for 23.85%, 19.23%, and 6.15%, respectively (Su et al., 2016). On the basis of investigating a large number of industrial VOCs treatment cases, the study showed that catalytic combustion, activated carbon adsorption and biodegradation are the most popular treatments, followed by thermal combustion and low-temperature plasma (Xi et al., 2012). Although typical industries have great potential for reducing VOCs, most companies generally reduce the emissions of VOCs by simple treatments such as activated carbon adsorption, low-temperature plasma, UV oxidation and so on, which have been reconsidered as being too simple and inefficient by the central government. The treatment of industrial VOCs requires consideration of multiple factors including policies, standards, emission characterization, technical support and so on. Only in this way can VOCs from end-pipe emissions be controlled efficiently.

4. Authority management

Compared with Europe and the United States, the promulgation of VOCs regulations in China lags behind. The Air Pollution Prevention and Control Law of the People's Republic of China is the fundamental basis for overall air pollution management, but there are no clear VOCs control requirements. The original Integrated Emission Standard

of Air Pollutants (GB16297-1996) only restricted the emission of benzene, toluene, xylene, phenols and formaldehyde. The Emission Standards for Odor Pollutant (GB14554-1993) mainly focuses on odorous VOCs including styrene, methyl mercaptan, methyl sulfide, dimethyl disulfide and trimethylamine. As for emission standards for specific industries, in 2007 and 2008, Emission Standard of Air Pollutant for Bulk Gasoline Terminals (GB20950-2007), Emission Standard of Air Pollutant for Gasoline Transport (GB20951-2007), Emission Standard of Air Pollutant for Gasoline Filling Stations (GB20952-2007) and Emission Standard of Pollutants for Synthetic Leather and Artificial Leather Industry (GB21902-2008) were released. Since then, emission standards related to VOCs from key industries have been rapidly put in place. The statistics show that about eight new standards have been established and two revised since 2015. The new standards mainly deal with petroleum refining (GB31570-2015), petroleum chemistry (GB31571-2015), synthetic resins (GB31572-2015), pharmaceutical (GB37823-2019), paint, ink and adhesive (GB37824-2019), foundry (GB39726-2020), pesticide manufacture (GB39727-2020) and onshore oil and gas exploitation and production (GB39728-2020) industries; more details on the mentioned standards are summarized in Table 4. In addition, in terms of VOCs treatment technology, the ministry of Ecology and Environmental has issued Technical Specifications of Adsorption Method for Industrial Organic Emissions Treatment Project (HJ2026-2013), Technical Specifications of Catalytic Combustion Method for Industrial Organic Emissions Treatment Project (HJ2027-2013), and Technical Specifications for Industrial Organic Waste Gas Treatment by Regenerative Thermal Oxidation (HJ1093-2020) with the aim of guiding the design, construction, acceptance, operation and management of VOCs treatment projects.

Since the issue of VOCs control has only received attention in recent years, there exist some deficiencies in VOCs management. Attention should be addressed to the following questions.

Firstly, the control and management of VOCs in China is overdue. On the one hand, management of VOCs should be carried out right now for better air quality; on the other hand, the environmental treatment costs are relatively high, and neither the authority nor the enterprise can obtain enough funding to support the treatment. In some cases, specific funding is used for other purposes instead of environmental protection, which makes it more difficult to control VOCs emissions. In addition, there is significant imbalance between the costs of law compliance and law breaking, which lessens the importance of controlling VOCs emissions from the industries' point of view.

Secondly, there is a weak linkage between VOCs management and policy-making, and few existing regulations/standards have taken industry-oriented VOCs into account. General management of VOCs is not satisfactory due to the lack of consistency, which is somewhat simple and partial. When it comes to specific industrial sectors, more emission standards still need to be made due to the large numbers of industries, and more emphasis should be put on the control of unorganized emissions. Accord to the reference (Liang et al., 2020), there are at least 20 key industries with relatively huge VOCs emissions, but only ten

Table 4 – Summary of VOCs emission standards for industries.

| Emission standards | Specific species | Integrated VOCs | Organized emission | Unorganized emission |
|--|---|----------------------|--------------------|----------------------|
| Integrated emission standard of air pollutants (GB16297-1996) | Vinyl chloride, Benzene, Toluene, Xylene, Chlorobenzene, Nitrobenzene, Aniline, Phenol, Formaldehyde, Acetaldehyde, Acrolein, Acrylonitrile, Methanol | NMHC | EC, ER | BC |
| Emission standards for odor pollutants (GB14554-93) | Styrene, Methyl mercaptan, Methyl sulfide, Dimethyl disulfide, Trimethylamine | Odor | ER | BC |
| Emission standard of pollutants for synthetic leather and artificial leather industry (GB21902-2008) | Benzene, Toluene, Xylene, Dimethylformamide | TVOC | EC | BC |
| Emission standard of pollutants for petroleum refining industry (GB31570-2015) | Benzene, Toluene, Xylene | NMHC | EC, RE | LD, BC |
| Emission standard of pollutants for petroleum chemistry industry (GB31571-2015) | N-hexane, Cyclohexane, Halogenated hydrocarbons, 1,3-butadiene, Vinyl chloride, Benzene, Toluene, Methanol, etc. | NMHC | EC, RE | LD, BC |
| Emission standard of pollutants for synthetic resin industry (GB31572-2015) | Styrene, Acrylonitrile, 1,3-butadiene, Epichlorohydrin, Phenols, Formaldehyde, Acetaldehyde, TDI, MDI, IPDI, PAPI, Acrylic acid, Methyl acrylate, Butyl acrylate, MMA, Benzene, Toluene, Ethylbenzene, Dichloromethane, Tetrahydrofuran, PA | NMHC | EC | LD, BC |
| Emission standard of air pollutants for pharmaceutical industry (GB37823-2019) | Benzene, Formaldehyde | Benzenes, NMHC, TVOC | NMHC, ER, RE | LD, BC |
| Emission standard of air pollutants for paint, ink and adhesive industry (GB37824-2019) | Benzene, Isocyanates, 1,2-dichloroethane, Formaldehyde | Benzenes, NHMC, TVOC | ER, RE | LD, BC |
| Emission standard of air pollutant for bulk petroleum terminals (GB20950-2020) | | NMHC | EC, RE | LD, BC |
| Emission standard of air pollutant for gasoline filling stations (GB20952-2020) | | NMHC | EC | |
| Emission standard of air pollutants for foundry industry (GB39726-2020) | Acrylonitrile, Benzene, Formaldehyde, Phenols, Chlorobenzenes | Benzenes | NHMC, ER, RE | LD, BC |
| Emission standard of air pollutants for pesticide industry (GB39727-2020) | Phenols, Toluene, Chlorobenzenes, Acrylonitrile | NMHC | | BC |
| Emission standard of air pollutants for onshore oil and gas exploitation and production industry(GB39728-2020) | | NMHC | EC, ER | LD |

NMHC: non-methane hydrocarbon; TVOC: total VOCs; EC: emission concentration; ER: emission rate; RE: removal efficiency; BC: boundary concentration.
LD: leak detection.

industries are supervised by emission standards. Therefore, it is necessary to establish more standards for VOCs emissions from different industries and update current standards and regulations.

Thirdly, quite a few enterprises are at the bottom of the whole industrial manufacturing chain and most belong to small- and middle-scale enterprises with limited profits. When reducing the cost by technical promotion is impossi-

ble, the only remaining way is to reduce the payment or VOCs treatment investment to ensure their interest. Otherwise, extra investment in VOCs treatment would increase the cost, making them less competitive or even yield negative profits. In most cases, enterprises have not taken VOCs treatment into account, or the installed VOCs treatment devices are only operated in case of inspection.

5. Conclusions and prospects

As for VOCs control from industries, it is suggested that refined requirements be proposed for the whole process including source reduction, enhanced collection, and in-depth end-pipe treatment. Generally speaking, water-borne and less VOCs-containing materials are the keys to source substitution in industries related to coating and solvent usage, LDAR should be implemented firmly as an efficient way to achieve source reduction in refining, petrochemical and other chemical industries. As for VOCs collection, airtight collection under negative pressure yields the best collection efficiency. Current end-pipe treatments like adsorption-combustion technology should be encouraged for better VOCs removal efficiency. From the viewpoint of emission control on the national scale, emphasis should be given as follows.

Firstly, it is urgent to establish more emission standards and regulations, which are the foundation of industrial VOCs control. Some argue that industrial VOCs emission be controlled and managed at regional scale (Wang et al., 2011). However, it is difficult to identify responsibilities for VOCs pollution in that VOCs tend to transport from one place to another by diffusion. Secondly, economic development differences generally lead to different control levels for industrial VOCs emissions. Thus, others suggest that VOCs control and management should be carried out in industries one by one. Where there is an important industry, there should be related VOCs emission standards and regulations. VOCs emission standards and management regulations at the national level should be established in key industries like the coal chemical industry, electronics manufacturing, furniture manufacturing, automobile manufacturing, packaging and printing, rubber products, auto repairing, catering and so on, and then gradually expand to other industries.

Secondly, policy adjustments should be made. As discussed above, the major goal of enterprises is to pursue profits. Thus, it might be impossible for them to reduce VOCs emissions voluntarily and the cost is also high for them to reduce VOCs emissions. However, the cost of treatment would be much less if the enterprises participate and reduce VOCs emissions voluntarily. The reason is that enterprises have detailed information like the annual amounts of materials and products, the operation parameters, VOCs emission concentrations and amounts and so on, giving them more flexibility to decrease VOCs pollution by taking feasible and innovative measures. In the view of cost-benefit balance, if policy changes could bring the enterprises profits from the reduction of VOCs emissions, the work of reduction would be carried out smoothly. VOCs subsidy determined by tonnage of emission reductions is efficient to promote voluntary emission reduction by enterprises. More emission reduction would result in more subsidies, which could promote the enthusiasm of enterprises to control VOCs and effectively reduce the total amount of VOCs emissions.

Thirdly, more and novel reduction technologies should be developed. VOCs emission treatments include source reduction, enhanced collection and end-pipe treatment. In China, more emphasis is focused on the end-pipe treatment, which mainly includes adsorption, catalytic incineration, condensa-

tion, bio-degradation, plasma decomposition and so on. Each has its own advantages and disadvantages and is applied under different conditions. The most frequently used technology in industrial VOCs treatment in China is carbon adsorption. The common problems are low efficiency, high operation cost and tendency to cause secondary pollution, which generally limits its application. Therefore, it is necessary to develop technologies for reduction of individual industrial VOCs, which could be realized by two different paths. One is setting up centralized operation processing centers to realize an assembly line process and to make VOCs emission continuous and stable. A relatively efficient and mature technology like combustion or adsorptive concentration combined with combustion could be adopted to achieve high-efficiency VOCs emission reduction. Meanwhile, for companies characterized by small scale, low concentration and intermittent emission, third-party operation and maintenance could be introduced and innovated, in which standardized activated carbon adsorption equipped with mobile desorption treatment could be adopted, and enterprisers would only need to make the investment for standardized activated carbon devices in the early stage and the cost for each subsequent desorption. Only in this way can the investment and maintenance costs of the enterprise be optimized and VOCs emissions meet the standards in the long term.

Finally, as for administrative departments, a long-term VOCs supervision system should be established. VOCs emissions from important sources including petroleum refining, coal chemical industry, electronics manufacturing, furniture manufacturing, automobile manufacturing, packaging and printing, rubber products, auto repair, catering and others should be monitored, dangerous and volatile organic materials like coatings, inks, cleaning agents, adhesives should be supervised by a two-dimensional code marking system, and compilation of a menu of hazardous substances should be carried out. Special funding should be provided to form a VOCs reduction-emission association and support the research, development, and application of major popular treatments. The approval procedure for new projects should be stricter, those with predictably high VOCs emissions should be forbidden principally, and the same or similar industrials should be encouraged to locate at the same site to take advantage of concentric emission, which would significantly reduce the cost of VOCs control. Furthermore, it is suggested that the government strengthen the policies supporting enforcement of industrial VOCs emission control, and strengthen the linkage among administrative department, public, enterprise and VOCs pollution treating organizations. Establishing an effective trinity system composed of the leadership of government environmental regulations, assistance of public participation and cooperation of enterprise environmental management is necessary for better VOCs control in the near future.

Declaration of Competing Interest

We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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