Characteristics of VOCs Emission Components in Typical Solvents Source Industries in Tianjin

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Abstract. Based on the demand for volatile organic compounds (VOCs) emissions controlled in Tianjin, VOC emissions compositions from four typical solvent use industries (automobile painting, furniture painting, wood-based panel manufacturing, Packaging & Printing) were measured by field sampling and analysis, the influence of different treatment facilities on VOCs composition and the ozone formation potentials (OFP) of species were analysed. The results showed that the major VOCs emissions species were aromatics and oxygenated VOCs. Among all waste gas treatment facilities, the catalytic combustion system could reshape the VOCs profile from different industries, and which made the proportion of ethylene increased from 5.81% to 28.30%. Aromatics had a great influence on OFP. The VOCs species with the largest OFP contribution was m/p-xylene, which contributed 16.36% of the total OFP.

Keywords: VOCs; solvent use industries; source profiles; treatment facility; OFP.

Volatile organic compounds (VOCs) which is one of the main contributors to regional atmospheric compound pollution in China at present, are the main participants in the transformation of atmospheric chemical composition and important precursors of ozone and secondary aerosol (SOA). VOCs are emitted from many sources, and the related research on their composition characteristic component spectrum has become a research hotspot at home and abroad. Since the 1980s, a large number of spectrum studies on the source component of VOCs have been carried out in developed countries [1]. Europe has compiled a source component spectrum database [2], while the US Environmental Protection Agency has collected the VOCs source emission component spectrum from the US and Canada and established a SPECIATE database, which has now been updated to Version4.5 [3]. In China, there are many VOCs emission sources and complex emission components. At present, domestic scholars[4] have constructed the anthropogenic VOCs emission inventory with species allocation information in China, and the Yangtze River Delta, Pearl River Delta, Beijing and other regions have also carried out VOCs species composition analysis of main solvent use pollution sources[5]. However, due to the complexity of solvent sources, VOCs components vary greatly among different regions and industries. Solvent components and waste gas treatment facilities are also important factors affecting VOCs emission composition [6].
As the economic center of circum-bohai sea region and the core area of northern international shipping, Tianjin is facing greater development opportunities. However, it is also one of the main haze areas in China, and the air pollution prevention and control pressure is great. In recent years, Tianjin has issued a number of policy documents on the prevention and control of volatile organic compounds, making it the focus of air pollution prevention and control in Tianjin. Some scholars have carried out studies on the VOCs component characteristics of some key industries in Tianjin [7-9], but the study on the VOCs emission component characteristics of solvent sources in Tianjin is still blank.

VOCs emissions from solvent sources in Tianjin are mainly from four industries [10]: automobile spraying (36%), furniture spraying (28%), packaging printing (14%), and wood-based panel production (12%). Therefore, this paper selects two representative enterprises from each of the four typical solvent using industries to study the VOCs source composition spectrum and identify the differences in VOCs components between different industries. Explore the influence of different waste gas treatment facilities on VOCs components; To study the contribution of different components to ozone generation potential, in order to provide scientific basis for the formulation of effective pollution control and emission reduction measures, and better serve the improvement of regional air quality.

1. Materials and Methods

1.1. Sample Collection

In this study, there are 8 typical solvent-use-industry were selected as industry representatives to carry out VOCs source composition spectrum research. The types of enterprises, production processes, waste gas treatment facilities and related parameters are shown in Table 1.

Table 1. VOCs Sampling in Different Industries

| Serial number | Industry categories | Coating type | Production process | Treatment method | Design removal rate | Design treatment air volume/(m³·h⁻¹) | Numbe of samples | Sampling location |
|---------------|---------------------|--------------|--------------------|------------------|-------------------|--------------------------------------|-----------------|------------------|
| 1#            | Automobile Paint    | Mixed paint  | Paint spraying     | Catalytic combustion | 97%              | 7 000                                 | 6               | Equipment exit, entrance |
| 2#            | Furniture paint     | Water paint  | Paint spraying     | Activated carbon adsorption | 95%              | 5 000                                 | 3               | Equipment exit |
| 3#            | Oil paint           | Oil paint    | Topcoat spraying   | Spray scrubber activated carbon adsorption | 60%              | 2 500                                 | 3               | Equipment exit |
| 4#            | Wood-based          | Formaldehyde-free resin adhesive | Heat setting section | Spray scrubber activated carbon adsorption | 95%              | 10 000                                | 6               | Equipment exit, entrance |
| 5#            | Formaldehyde        | Heat setting section | Catalytic combustion | 60%              | 2 500                                 | 3               | Equipment exit |
| 6#            | Formaldehyde        | Heat setting section | Heat setting section | 97%              | 2 500                                 | 3               | Equipment exit, entrance |
| 7#            | Packaging print     | Water-based ink | Gravure printing | Spray scrubber activated carbon adsorption | 60%              | 7 000                                 | 6               | Equipment exit, entrance |
| 8#            | Water-based ink     | Water-based ink | Gravure printing | Activated carbon adsorption | 95%              | 5 000                                 | 3               | Equipment exit |

The samples were collected in a stainless steel sample tank (Entech SUMMA tank) with a polished inner surface and salinized inner surface. The volume of the sample tank is 3.2L and the maximum pressure is about 0.3MPa. The sampling time was from July to August 2018, and the sampling frequency was 20 min/ time. Samples are collected under the normal operation of the production equipment and
production process. During sample collection, a stainless steel tube is connected to the SUMMA tank, and a glass filter tube filled with glass wool and anhydrous sodium sulfate is connected in the pipe to remove water vapor and particulate matter in the exhaust gas. The glass filter tube is used after high temperature baking in muffle oven before filling, and the stainless steel tube is deep into the sampling port of the exhaust tube for sample collection. Connect the pre-cleaned and vacuumed sampling tank to the dynamic diluent meter (American Entec 4600), open the high purity nitrogen and high purity air valves, and close the sampling valve and cylinder gas valve when the pressure of the sampling tank reaches 101 kPa, and take them as blank control samples. Considering that sampling time, working conditions and other influences may vary to some extent, this paper collects 3 samples from each sampling point and takes the average data of 3 times for follow-up research.

1.2. VOCs Analysis Method

1.2.1. Analysis Conditions of Chromatography-mass Spectrometry. The starting temperature of GC column was 10 °C and kept for 3 min. the temperature was raised to 40 °C at 10 °C / min and then to 250 °C at 6 °C / min for 5 min. The scanning range of MS is 40 ~ 550 amu, the voltage is 0.7 kV, and the EI temperature of ion source is 250 °C. Split injection was used.

1.2.2. Quality Control and Assurance. In the process of analysis, strict quality control and quality assurance procedures were carried out. The standard gases with different concentration gradients (0.5 nmol/mol, 2.5 nmol/mol, 5.0 nmol/mol, 10.0 nmol/mol, 20.0 nmol/mol) were prepared by dynamic dilution gas distributor, and the working curves were established. The R2 of the working curves of each component are greater than 0.99. The experiment was repeated for 3 times for each concentration gradient and the average value was calculated to obtain the corresponding relationship of each component in the standard gas, so as to quantify the sample. The standard gases used for quantitative analysis were 65 TO-15 and 57 PAMs mixed standard gases. The measurement accuracy of components is guaranteed to be less than 10%. System blank test and daily calibration are carried out every day in the analysis process. The ratio between calculated concentration and theoretical concentration of daily calibration is maintained within the range of 0.8 ~ 1.2, which indicates that the instrument is stable in operation.

1.2.3. Ozone Formation Potential (OFP) Assessment Method. The activity of VOCs in atmospheric chemical reactions can be measured by the ozone formation potential (OFP) and the incremental reactivity of VOCs. In this paper, the maximum incremental reactivity (MIR) was used to measure the ozone formation potential (OFP) of VOCs. MIR reflected the contribution capacity of VOCs to ozone generation of emitters [12]593. The calculation formula is as follows.

\[
\text{OFP} = \sum_{i=0}^{n} c_i \times \text{MIR}_i
\]

In this formula, OFP is the total ozone formation potential, mg•m-3; Ci is the mass concentration of component “i”, mg•m-3.

\[
\text{MIR} = \sum_{i=0}^{n} \text{MIR}_i \times x_i
\]

In this formula, Mir represents the maximum reaction increment of a source component spectrum, g•g-1; MiRi represents the maximum reaction increment of component I, g•g-1VOCs; Xi is the mass fraction of component i.
2. Result and Discussion

2.1. VOCs Source Composition Spectrum of Different Industries

In this study, 73 VOCs components were analyzed, including 28 alkanes, 12 alkenes, 16 aromatic hydrocarbons and 17 oxygen-containing VOCs. The emission components of the top 8 VOCs in each enterprise are shown in Table 2.

| Industry Categories | Enterprise Numbers | Characteristic VOCs Components |
|---------------------|--------------------|--------------------------------|
| Car painting        | 1#                 | Ethylene (28.30%), Propylene (8.71%), M/p xylene (8.01%), 1 - butylene (7.20%), n - undecane (6.30%), Isopropyl alcohol (4.32%), Styrene (3.11%) |
|                     | 2#                 | 1 - butanol (26.20%), Ethyl acetate (18.50%), 2 - butanone (13.3%), O-xylene (9.5%), 2-methyl hexane (5.2%), 1 - butylene (5.11%), Propylene (5.02%), 1,2,4 - three methyl benzene (4.85%) |
| Furniture spray     | 3#                 | N-butyl acetate (27.90%), Ethyl benzene (22.00%), Cyclohexanone (9.31%), O-xylene (9.20%), Cyclohexane (7.20%), M/p xylene (6.81%), M-ethyl toluene (6.42%), Toluene (1.82%) |
|                     | 4#                 | Ethyl benzene (27.40%), Ethyl acetate (21.20%), cyclohexanone (11.20%), N-butyl acetate (7.40%), cyclohexane (5.80%), M/p xylene (5.72%), o-xylene (5.37%), styrene (4.87%) |
| Artificial plate production | 5#         | Ethanol (23.50%), Acetone (17.41%), Toluene (12.22%), Styrene (4.10%), Cyclohexane (3.80%), Ethyl acetate (3.67%), Isopropyl alcohol (3.41%), Are aoi alkanes (2.21%) |
| Packaging and printing | 6#         | 2,4 - dimethyl pentane (27.50%), Sec-butyl acetate (17.31%), Acetone (16.81%), O-ethyl toluene (9.82%), 1 - butylene (8.00%), n - undecane (7.42%), Dodecanoic (6.31%), Styrene (2.52%) |
|                     | 7#                 | Ethyl acetate (62.61%), Butyl acetate (9.31%), Acetone (6.22%), Cyclohexane (4.32%), n - undecane (2.61%), O - xylene (2.50%), Isopropyl alcohol (2.47%), N-heptane (2.12%) |
|                     | 8#                 | n - undecane (43.20%), N - decane (16.80%), cyclohexane (10.21%), Methyl cyclohexanone (6.51%), Ethyl acetate (4.80%), M/p xylene (3.7%), Sec-butyl acetate (3.51%), Isopropyl alcohol (3.02%) |

Note: The values in the table are the mass fractions of VOCs characteristic components.

Figure 1. Components of VOCs for automobile spraying industry

Figure 2. Components of VOCs for furniture spraying industry
2.1.1. VOCs Composition of Automobile Spraying Industry. VOCs emissions from the automotive spraying industry mainly come from the volatilization of raw and auxiliary materials containing VOCs such as coatings used in the spraying process, diluents and curing agents [11].

As can be seen from Figure 1, in the VOCs component of 1# enterprise, the proportion of alkanes/alkynes is the highest, reaching 51.26%, followed by oxygen-containing VOCs and aromatic hydrocarbons, accounting for 20.11% and 16.77% respectively, and the proportion of alkanes is only 11.86%. In 2# enterprise, the proportion of oxygen-VOCs was 59.00%, followed by aromatic hydrocarbons and ene/acetylene, which accounted for 20.00% and 13.60%, respectively. Compared with other domestic automotive spraying enterprises, 1# enterprise has the highest content of alkene/alkyne, which is the same as the conclusion in literature [6] that alkenes is the main component of VOCs in automotive spraying industry. The VOCs component of 2# enterprise is mainly composed of oxygen-containing VOCs, which is similar to the VOCs source component spectrum results of Beijing automotive spraying enterprises in literature [12]. The oxygen-containing VOCs are the component with the highest contribution rate in generating VOCs, and the proportion of aromatic hydrocarbons decreases.

In terms of the main VOCs components (Table 2), ethylene and propylene are the main components in 1# enterprise, which is similar to the result of ethylene and propylene as the main VOCs components in the automotive spraying industry in Changjiang Delta, and may be related to the generation of olefin components through catalytic combustion of VOCs [6]. 2# enterprise took 1-butanol, ethyl acetate and 2-butane as the main VOCs components, which was similar to the research results of Beijing and Zhengzhou that took alcohols and esters as the main VOCs components, probably because this enterprise used water-based coatings with higher content of oxygen compounds to replace solute coatings with higher content of benzene series [14].

2.1.2. Composition of VOCs in Furniture Spraying Industry. The VOCs emitted by spraying enterprises are similar to those of automobile spraying enterprises, which mainly come from the volatilization of raw and auxiliary materials containing VOCs such as coatings used in the coating process, diluents and curing agents; In addition, the industrial characteristics of intermittent painting operation will also lead to certain differences in composition spectra of furniture spraying enterprises established under different working conditions.

As can be seen from Figure 2, the main VOCs components of furniture spraying 3# and 4# enterprises are all aromatic hydrocarbons, accounting for 47.48% and 46.37% respectively, followed by oxygen-containing VOCs, accounting for 37.72% and 39.86% respectively. Compared with other domestic studies, the results of 3# and 4# enterprises are similar to those of literatures [14], all of which take aromatic hydrocarbons as the main VOCs component, but different from the results of literatures [11] of furniture spraying enterprises that take oxygen-containing VOCs as the main component. It shows
that the proportion of aromatic hydrocarbons in the furniture spraying industry in Tianjin is still higher than that in the areas with stronger control.

In terms of main VOCs components, n-butyl acetate was the primary VOCs pollutants in 3# enterprise, accounting for 27.90%, followed by ethylbenzene (22.00%) and cyclohexanone (9.31%) o-xylene (9.20%). Ethylbenzene was the main VOCs emission component in 4# enterprise, accounting for 27.40%, followed by ethyl acetate and cyclohexanone, accounting for 21.20% and 11.00%, respectively. The research and literature [14], [15] results similar to that in benzene series and ketone ester as the main components of VOCs, but with the Beijing city furniture coating components of VOCs in ethanol, ethyl acetate and acetone as the main component of research differences [11] 4399, which related to the Beijing furniture coating using water-based coating process, so the spray of tianjin furniture industry should take advantage of water-based coating process to replace solvent-based coating process.

2.1.3. Composition of VOCs in Wood-based Panel Industry. In the wood-based panel production industry, VOCs emissions mainly come from the use of adhesives, surface coatings and dyes in the processing process, and the use of raw materials and wood-based panels will also release different degrees of VOCs.

As can be seen from Figure 3, in the VOCs generated by the hot pressing section of No.5 Company, oxygen-containing VOCs accounted for the highest proportion, up to 58.70%, followed by aromatic hydrocarbons and alkanes, accounting for 20.67% and 15.13% respectively. In 6# enterprise, the proportion of alkanes was 42.50%, followed by oxygen-containing VOCs and aromatic hydrocarbons, which accounted for 36.39% and 12.91%, respectively. Compared with other domestic studies, the results of 5# enterprise are similar to those of literature [14], which all take oxygen-containing VOCs as the main VOCs component. Alkanes are used as the main VOCs component in # 6 enterprise, which may be related to the generation of alkane components after catalytic combustion of VOCs [13]6.

In terms of the main VO Cs components, the main VOCs group of No.5 enterprise is divided into ethanol and acetone, accounting for 23.50% and 17.41%, respectively. In 6# enterprise, 2, 4-dimethylpentane was the main VOCs component, accounting for 27.50%. The results of formaldehyde as the main component of VOCs in 5# and 6# enterprises are significantly different from those in literature [14], indicating that the formaldehyde free resin adhesive used in Tianjin replaces the urea formaldehyde resin adhesive that releases more formaldehyde in hot pressing section.

2.1.4. Composition of VOCs in Packaging and Printing Industry. VOCs emitted by the packaging and printing industry mainly come from the volatilization of raw and auxiliary materials containing VOCs such as inks used in the printing production process, diluents, moistening solution, cleaning agents, varnish and adhesives [28] 4401.

As can be seen from Figure 4, among VOCs produced by No.7 Enterprise, oxygen-containing VOCs accounted for the highest proportion, up to 81.10%, followed by alkanes and aromatic hydrocarbons. 8# Company takes alkanes as the main VOCs component, accounting for 71.00%, which is consistent with the results of Beijing and Chengdu printing industries that take alkanes as the main VOCs component [5] 376.

In terms of major VOCs components, ethyl acetate is the main VOCs component in 7# Enterprise, accounting for 62.61%. The main VOCs group of 8# enterprise is divided into n-undecane, accounting for 43.20%. This indicates that in recent years, the water-based ink printing process in Tianjin has replaced the solvent-based ink process, which makes the VOCs produced by it mainly consist of esters or alkanes, and the content of benzene series has been greatly reduced.

2.2. Influence of Waste Gas Treatment Facilities on VOCs Components

This section studies the effects of three types of waste gas disposal devices, activated carbon adsorption, water curtain spray and catalytic combustion device, on the treatment efficiency of VOCs waste gas. The components of VOCs before and after treatment of various waste gas treatment facilities in 1#, 4#
and 7# enterprises were selected as the research object, and the parameters of various treatment facilities obtained were shown in Table 3.

As can be seen from Table 3, the treatment efficiency of the catalytic combustion exhaust gas treatment facility of 1# enterprise for VOCs is 85%; 4# enterprise activated carbon waste gas treatment facilities for VOCs treatment efficiency is only 59%, while the design treatment efficiency of 97%, it can be seen that the treatment effect is not ideal, may be because the activated carbon used by the enterprise has not been replaced in time, so that the activated carbon adsorption capacity is low; The spray tower treatment facility used by 7# enterprise has a poor VOCs treatment effect of only 13%, mainly due to the high content of non-condensable gases in the VOCs spray, which leads to low treatment efficiency.

Figure 5 (a) shows the composition changes of VOCs produced by 1# Enterprise before and after the catalytic combustion treatment. Before the treatment of VOCs, benzene series such as m-xylene and styrene were the main components, while after the treatment of catalytic combustion facilities, VOCs were mainly composed of short-chain alkenes such as ethylene and propylene, with ethylene accounting for 28.30%. This is because VOCs are treated by catalytic combustion to produce CO2 and H2O, and at the same time generate short-chain olein [6]1947.

Fig. 5 (b) shows the change of components of VOCs produced by 4# enterprise before and after adsorption by activated carbon. The proportions of each component showed no significant change, but the proportions of ethyl acetate and cyclohexanone increased after adsorption by activated carbon, which may be due to the analytical effect of activated carbon [16] 37. The proportion of ethyl benzene, m-p-xylene and styrene decreased after adsorption by activated carbon, indicating that activated carbon has a good adsorption effect on benzene series, which is similar to previous studies on the adsorption effect of activated carbon on furniture spraying industry [16] 34.

Fig. 5 (c) shows the composition changes of VOCs produced by No.7 Enterprise before and after being treated by the spray tower. The proportion of main substances before and after treatment is similar, with the proportion of ethyl acetate and butyl acetate slightly increasing, indicating that the spray tower does have difficulties in removing volatile organic compounds that are difficult to dissolve in water [16] 44.

In summary, among the VOCs waste gas treatment facilities, the catalytic combustion device has the highest processing efficiency for VOCs and has a greater impact on the components, while the activated carbon adsorption and spray tower has a lower processing efficiency for VOCs and has a smaller impact on the components.

Therefore, when Tianjin controls the VOCs produced by the spraying industry, in addition to encouraging water-based coating process, it is also necessary to control the VOCs treatment facilities and regularly check whether the activated carbon is replaced in time. Besides, in order to reduce environment content of VOCs, Tianjin should be vigorously promoted combined application of waste gas treatment facilities, such as water spray tower + activated carbon adsorption, condensation - catalytic combustion device, adsorption and photocatalytic device.

### Table 3. Effect of Different Treatment Facilities

| Enterprise Numbers | Type of waste gas treatment facility | Monitoring of the concentration/(mg·m⁻³) | Actual handling of wind flow/(m³·h⁻¹) | Processing efficiency (%) |
|--------------------|------------------------------------|----------------------------------------|--------------------------------------|--------------------------|
|                    |                                    | Before                                 | after                                |                          |
| 1#                 | Catalytic combustion               | 124.01                                 | 18.60                                | 5 500                    | 85                       |
| 4#                 | Activated carbon adsorption        | 46.18                                  | 19.08                                | 8 000                    | 59                       |
| 7#                 | spray scrubber                     | 32.99                                  | 28.60                                | 5 500                    | 13                       |
2.3. Ozone Formation Potential Analysis of VOCs Components

OFP values calculated from 8 typical enterprises were used to calculate the OFP values of different components in 4 industries, as shown in Fig. 6. Among them, the OFP value of furniture spraying industry is the highest, and that of wood-based panel industry is the lowest. The contribution of different components to ozone was investigated by summing up OFPs of various components in the four industries. The contribution of aromatic hydrocarbons, olefin, oxygen-containing VOCs and alkanes to the total OFP was 50.69%, 20.55%, 20.20% and 8.56%, respectively. It can be seen that aromatic hydrocarbons are the component with the greatest potential for ozone formation.

Fig. 7 shows the 10 VOCs that contribute the most to OFP in the four types of typical solvent source industries in Tianjin, and the contribution of the top 10 species to the total OFP is 71.98%. M-p-xylene, o-xylene, ethylbenzene, m-ethyl toluene and styrene were all aromatic hydrocarbons, accounting for 47.24% of the total OFP, and m-p-xylene accounted for 16.36% of the total OFP, which was the component contributing the most to ozone formation potential. The results of this study are similar to the conclusion of literature [12] and [19] that m-xylene is the largest component contributing to the potential of ozone formation.

Fig. 5 Effect of different treatment facilities on VOCs component
3. Conclusion

(1) The VOCs emission compositions of four typical solvent using industries in Tianjin are significantly different. Olefins and oxygenated VOCs are the primary components of VOCs in the automotive spraying industry. Aromatic hydrocarbon is the main component of VOCs in furniture spraying industry; Oxygen-containing VOCs and alkanes in the wood-based panel industry are the primary components of VOCs; Oxygen-containing VOCs and alkanes in the packaging and printing industry are the primary components of VOCs.

(2) Different waste gas treatment facilities have different treatment efficiency on VOCs and influence on components. The catalytic combustion unit has a higher processing efficiency for VOCs, and has a greater influence on the composition of VOCs, especially the content of benzene series and olefin of VOCs components has obvious changes. Activated carbon adsorption and spray tower have low efficiency for VOCs treatment, and have little effect on VOCs components.

(3) The OFP values of furniture spraying and automobile spraying industries were higher, and aromatic hydrocarbons were the components with the largest contribution to the potential of ozone formation, among which m-xylene was the component with the largest contribution to the potential of ozone formation, accounting for 16.36% of the total OFP.

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