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Emission Characteristics of Volatile Organic Compounds from Automobile Coating Industry in Chongqing City

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Abstract: To fully analyze emission characteristics of VOCs (Volatile Organic Compounds) from automobile coating industry in Chongqing city, related enterprises in the city were investigated in detail. The results show that the emission of VOCs from the automobile industry in Chongqing city was approximately 20,000 t in 2012, and the contribution rate of automobile coating was 83%. Most VOCs discharged from automobile coating industry were mainly from the use of coatings, diluents, curing agents and other materials containing VOCs. During the process of automobile coating, VOCs were mainly produced during the processes of spraying and drying. Besides, VOCs are partly produced during the processes of mixing paint, gluing, waxing and repairing. The major ingredients of VOCs discharged during the process of automobile coating were methylbenzene, dimethylbenzene, trimethylbenzene, cumene, ethyl acetate, butyl alcohol, n-butyl alcohol, isobutyl alcohol, formaldehyde, butanone, acetone, cyclohexanone and methyl ethyl ketone. Besides, the ingredients of naphtha and aromatic hydrocarbon in VOCs need to be determined. The maximum OFP (Ozone Formation Potential) of the above pollutants by the typical enterprises was 900-1,300 mg/m^3. The production of SOA (Secondary Organic Aerosol) was 3,636-11,073 t/a, which didn't include the contribution of aldehydes, ketones, ethers and esters to SOA. This research could provide scientific reference for the establishment of emission standard, control technology and management methods of VOCs from automobile coating industry that accord with economic and social development in Chongqing city.

Key words: Automobile coating, VOCs, environmental impact, ozone, SOA.

1. Introduce

The automobile industry is the pillar industry of national economy in Chongqing city, and its annual average growth rate has exceeded 20% in recent ten years. In 2012, Chongqing municipal government issued the Three-year Plan for Revitalizing Automobile Industry in Chongqing city which core was to increase investment in the automobile industry, to improve complete sets of equipment, and to speed up transformation and upgrading of the automobile industry. In 2014, auto production in Chongqing city ranked firstly, that was, 2.58 million autos were produced, increasing by about 28% compared with 2013.

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At the back of the splendid economic bill, the environmental impacts of VOCs (Volatile Organic Compounds) have become increasingly obvious with the rapid development of the automobile industry. In the automobile industry, workers' income is relatively high, so economic development and population growth are relatively rapid, and environmental complaints are more and more frequent in regions around accumulation areas of the automobile industry. In 2012, large-scale enterprises in the automobile industry, pharmaceutical manufacturing, petrochemical industry, organic chemical industry, package printing, food processing, furniture making, production of electronics and rubber manufacturing discharged around 6.5×10^4 t of VOCs, and the contribution rate of the automobile industry was up to

30%. The emission of VOCs from the automobile industry was mainly from automobile manufacturing industry, manufacturing industry of auto parts and automobile repair industry. In 2013, the emission of VOCs from automobile coating was 1.6×10^4 t, accounting for 83% of total emission of VOCs from the automobile industry.

As important precursors, VOCs take part in the formation of secondary aerosols [1-3], speed up the generation of ozone [4-7], and have damage to human health [8, 9], which is being widely known and deeply studied. Hence, it is urgent to determine the emission situation of VOCs from automobile coating and characteristic pollutants. In this paper, emission characteristics of VOCs from automobile coating industry in Chongqing city were surveyed in detail to provide important reference for the establishment of emission standard, control technology and management methods of VOCs from automobile coating industry that accord with economic and social development in Chongqing city.

2. Emission Characteristics of VOCs during the Process of Automobile Coating

In this paper, 57 enterprises related to automobile coating (including 20 enterprises engaging in the whole automobile coating and 37 enterprises engaging in surface coating of auto parts) were investigated to obtain their production, consumption of materials containing VOCs, production processes, disposal facilities and efficiency in the form of spreadsheets. Meanwhile, 20 typical enterprises engaging in automobile coating in Chongqing city were surveyed, and the major survey objects included four enterprises engaging in car production (4×10^5, 2×10^5, 2.9×10^5 and 5×10^4 cars were produced respectively every year), two enterprises engaging in van production (the average number of box trucks and vans produced every year was 1.2×10^5 and 2.5×10^4 respectively), and one enterprise engaging in producing special vehicles (2×10^3 special vehicles were produced in each year). The investigation content include written material (such as environment impact assessment reports, monitoring reports, transhipment shipping bills of hazardous wastes, the discharge, treatment and utilization situation of pollutants by industrial enterprises, input and output lists of raw materials, MSDSs (Material Safety Data Sheets) of paint and cleaning agents, etc.), technicians' on-site explanations and answering questions about the production process of an enterprise, the thickness of each coating, disposal techniques of pollutants, production conditions, development expectation, etc., and image data on painting lines, drying rooms, assembly lines, water curtain adsorption, RTO (Regenerative Thermal Oxidizer), etc..

Automobile manufacturing often includes stamping, welding, coating, final assembly, inspection and other processes, during which VOCs are primary pollutants, and they are mainly discharged during the process of coating [10]. During the process of coating, raw materials discharging VOCs include paint (containing diluents, curing agents, etc.), cleaning agents and sealants (internal board rubber, external board rubber and PVC), etc.. Large amounts of VOCs are produced during the coating process of paint. Automotive coatings from the inside to the outside of a vehicle are primer (electrophoretic paint), intermediate paint, finish paint, varnish and refinishing paint, and they have different components. Typical layers of paint coated on a vehicle from the bottom to the top and their thickness are shown in Fig. 1.

2.1 Processes of Automobile Coating in Chongqing

Processes of automobile coating in Chongqing city can be divided into two parts: one process is metal surface treatment before automobile coating, such as phosphating, oxidation or passivation treatment for removing stains; the other process is coating, such as spraying and drying primer, gelatinizing, spraying intermediate paint and finish paint, levelling and drying. Cars are typical representatives in the

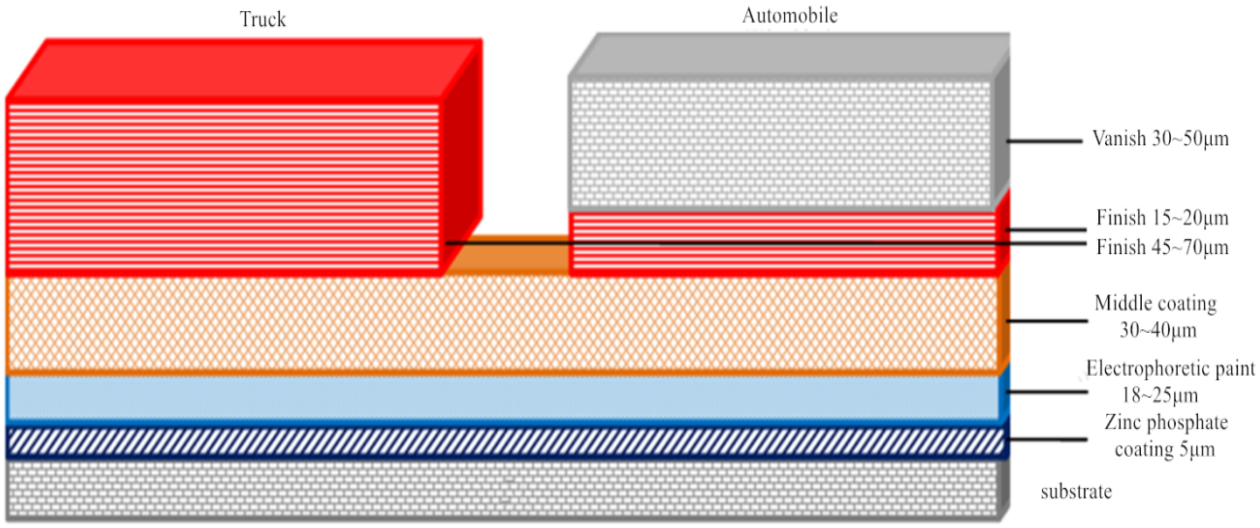

Fig. 1 Lacquer layers of automobile coatings and their thickness.

A truck covered with monochromatic paint coating; A vehicle covered with monochromatic paint and metallic paint; Varnish; Finish paint; Intermediate paint; Electrophoretic paint; Zinc phosphate; Substrate.

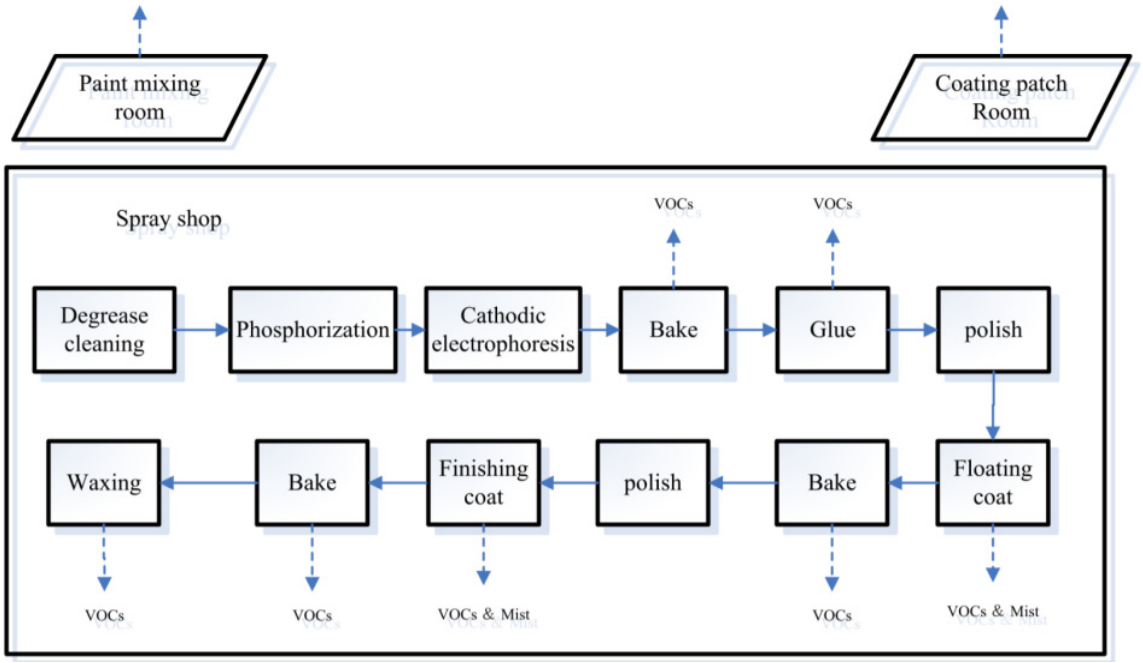

Fig. 2 Typical processes of car coating in Chongqing city.

Paint mixing room; Repair room; Degreasing; Cleaning; Phosphating and washing; Cathode electrophoresis; Drying electrophoresis; Gluing; Polishing electrophoresis; Waxing; Drying finish paint; Spraying finish paint; Polishing; Spraying intermediate paint; Drying intermediate paint; Polishing; Spraying finish paint; Drying finish paint; Waxing; Paint mist.

automobile industry in Chongqing city, and the typical processes of car coating are shown in Fig. 2.

2.2 Generation of VOCs during the Process of Automobile Coating in Chongqing City

Seen from Fig. 2, during the process of automobile coating, VOCs are mainly produced during the processes of spraying and drying. In addition, VOCs are partly produced during the processes of mixing paint, gluing, waxing and repairing [11-16]. The production of VOCs in such processes was shown in Table 1.

Table 1 Production of VOCs during the process of automobile coating in Chongqing city.

Process	General situation of processes
Spraying primer	Electrophoretic paint is usually sprayed on cars, most small and middle coaches and trucks by robots, and the coating efficiency is up to 85%-100% [17]; alkyd, phenolic or epoxy primer is often sprayed on special vehicles, trailers and big coaches by robots and people, and the coating efficiency is 30%-65% [17].
Drying primer	Electrophoretic paint is dried at about 180 °C.
Gelatinizing	A layer of PVC anti-stone-bumping sealing coating (1-3 mm in thickness) is sprayed on the baseboard of the body of an automobile, weld sealant and PVC paint are solidified and dried in an oven. Small quantities of VOCs are discharged during the process of solidification.
Spraying intermediate paint and finish paint	The processes of spraying intermediate paint and finish paint include automatic reciprocating spraying, paint cup-type robot spraying, electrostatic spraying and manual spraying. Intermediate paint and finish paint are solvent based coatings, and the coating efficiency is also 30%-65% [17].
Levelling and drying	Levelling and drying are conducted at 100-150 and about 200 °C respectively.
Mixing paint	Paint, diluents and curing agents are mixed according the demands of their proportions. Some enterprises have relatively dense paint mixing rooms but no waste gas processing unit.
Repairing	After manual spraying, paint is baked by a hot lamp. Some enterprises have relatively dense paint mixing rooms but no waste gas processing unit.

Table 2 Discharge of the exhaust gases in each section.

Section	Discharge of the exhaust gases VOCs
Painting room	Major harmful ingredients of the exhaust gases are aromatic hydrocarbon, alkane, ester, alcohol, aldehyde, ketone and ether organic solvents and a small quantity of paint mist, and about 60% of VOCs are produced in a painting room.
Levelling room	The composition of the exhaust gases in a levelling room is similar to that of a painting room, but there is no paint mist in the levelling room. During the process of spraying intermediate paint and finish paint, around 25% of the exhaust gases VOCs are produced in the levelling room. In the section, the total concentration of the organic exhaust gases is often about two times higher than that of a painting room. After being mixed with the exhaust air from a painting room, the exhaust gases are processed. When the blowing rate is 1×10^4 -1×10^7 m^3/h, the concentration of the discharged organic exhaust gases is often smaller than 100 mg/m^3. The exhaust gases are processed by mains of water curtain, venturi, water rotation, etc. to mainly remove paint mist, but the treatment efficiency of VOCs is low [18], and the running effect of facilities is not ideal.
Drying room	The dried exhaust gases have complex composition and contain thermal decomposition products and reaction products besides organic solvents, plasticizer and resin monomers. Exhaust gases are produced during the process of drying electrophoretic paint and solvent based coating, but there are big differences between them in terms of composition and concentration. electrophoretic paint. Electrophoretic paint is water-based paint, but the exhaust gases discharged during the process of drying the paint still contain certain quantities of organic ingredients. Besides a small amount of alcohol and ether organic matter, the exhaust gases contain thermal decomposition products (such as small molecules of aldehydes and ketones) produced during the process of drying the paint. In addition, the exhaust gases discharged during the process of drying the paint also contain closed isocyanate curing agent that can release sealants with small molecules, such as methyl ethyl ketone and many mixtures of alcohol and ether. Total concentration of organic matter in the exhaust gases discharged during the process of drying electrophoretic paint is 500-1,000 mg/m^3, lower than that of exhaust gases discharged during the process of drying solvent based coating. During the process of spraying intermediate paint and finish paint, 10%-20% of VOCs are discharged with car film in a drying room, and the blowing rate is relatively small (only 1,000-10,000 m^3/h), while the concentration is high. The exhaust gases are often processed by means of direct combustion and catalytic combustion, and the treatment efficiency of VOCs is 95%-99% [19-21].
Others	VOCs are discharged during the processes of gelatinizing, cleaning with cleaning agents and repairing. Sealant solid accounts for above 95%, while the content of VOCs is small, and corresponding exhaust gases are processed in a drying room. A large amount of paint is used, and organic exhaust gases will volatilize and diffuse if waste paint residue collected from a paint mixing room, repairing room and waste paint barrel as well as paint mist are processed in time.

2.3 Discharge of VOCs during the Process of Automobile Coating in Chongqing City

During the process of automobile coating in Chongqing city, the exhaust gases VOCs are mainly discharged from a painting room, levelling room, drying room and other processes (Table 2).

3. Variety of VOCs Discharged during the Process of Automobile Coating in Chongqing City

The production scale and process level of automobile manufacturers are different, and there is a variety of paint, so car production that develops rapidly was investigated in detail. According to various production scale, process level and management level, the most representative enterprises A, B and C were chosen. In 2013, they produced 0.63 million cars in total, accounting for 58% of total production of cars.

3.1 Types of Raw and Auxiliary Materials Containing VOCs

Raw and auxiliary materials containing VOCs used by the three enterprises are primer, intermediate paint, finish paint, varnish, diluent and sealant. Among them, all primer is epoxy resin; intermediate paint and finish paint are acrylic paint, polyester paint and amino paint; diluent and cleaning agents are aromatic hydrocarbon solvent, alcohols solvent and ester solvent; all sealant is PVC. The proportion of raw and auxiliary materials containing VOCs is shown in Table 3.

3.2 Variety and Emission of Discharged VOCs

The treatment capacity of VOCs subtracted from the production of VOCs in various sections is the total emission of VOCs. The emission of VOCs from PVC is relatively small, so it was regarded as 0 during the process of calculation. The spraying efficiency is calculated according to the data of enterprises and EPA car coating standard [17], and its formula is shown as:

$$E_{\text{total}} = \sum_i E_i \times （1 - R_i）$$

Where E_{total} is the total emission of VOCs; E_i is the emission of VOCs in each section (t); R_i is the treatment efficiency in each section (%).

After the total emission of VOCs was calculated, it was shared according to the quantity of used paint that was calculated based on the input and output lists and MSDS of paint. The results show that the major ingredients of VOCs discharged during the process of automobile coating were methylbenzene,

Table 3 The proportion of all kinds of paint in typical automobile manufacturing enterprises in Chongqing city.

Paint	Plant A		Plant B		Plant C	
	Type of paint	Proportion (%)	Type of paint	Proportion (%)	Type of paint	Proportion (%)
Primer	Epoxy resin	36	Epoxy resin	27	Epoxy resin	34
Intermediate paint	Acrylic, polyester, and amino	5	Acrylic, polyester, and amino	10	Polyester and amino	13
Finish paint	Acrylic, polyester, and amino	15	Acrylic, polyester, and amino	13	Polyester and amino	13
Varnish	Acrylic, polyester, and amino	10	Acrylic, polyester, and amino	10	Polyester and amino	9
Diluent	Aromatic hydrocarbon	1	Aromatic hydrocarbon	6	Aromatic hydrocarbon and alcohol	21
Cleaning agent	Aromatic hydrocarbon	14	Aromatic hydrocarbon	3	Aromatic hydrocarbon and alcohol	9
Sealant	PVC	19	PVC	31	PVC	1

Table 4 The proportion of emission of identified VOCs from typical automobile manufacturing enterprises in Chongqing city.

Ingredient	Plant A	Plant B	Plant C
Methylbenzene	0.03	-	0.87
Dimethylbenzene	6.85	0.11	38.65
1, 3, 5-trimethylbenzene	-	4.68	2.61
Ethyl acetate	-	-	10.27
Butyl alcohol	13.82	5.54	4.69
n-butyl alcohol	8.33	10.45	0.58
2-methyl-1-propanol	1.73	-	19.60
Methyl isobutyl ketone	1.73	-	-
Methanol	-	-	0.94
Propylene glycol	-	3.64	-
Propylene glycol monoethyl ether	-	-	4.68
Dimethyl succinate	-	-	0.94
Dimethyl adipic acid	-	-	1.87
Dimethyl glutarate	-	-	2. 81
Methyl ethyl ketone	-	-	5.59
Diethylene glycol monobutyl ether	-	-	2.81
Formaldehyde	-	-	0.94
2-butoxy ethanol	-	-	0.87
2-propyl alcohol	-	5.54	-
n-heptane	-	1.66	-
Aromatic hydrocarbon	43.53	57.62	1.28
Naphtha	23.98	10.76	-

dimethylbenzene, trimethylbenzene, cumene, ethyl acetate, butyl alcohol, n-butyl alcohol, isobutyl alcohol, formaldehyde, butanone, acetone, cyclohexanone and methyl ethyl ketone. Besides, the ingredients of naphtha and aromatic hydrocarbon in VOCs need to be determined.

According to Table 4, plants A and B discharged a large amount of naphtha and aromatic hydrocarbon, that is, there were much benzene series and alkanes. Meanwhile, butyl alcohol and n-butyl alcohol were discharged from the two plants. Plant C mainly discharged dimethylbenzene, ethyl acetate, and isobutyl alcohol, and other esters, ethers and ketones were discharged. To control pollutants, the emission and treatment efficiency of aromatic hydrocarbons such as trimethylbenzene and n-propylbenzene, esters such as ethyl acetate and butyl acetate, alcohols such as n-butyl alcohol and 2-propyl alcohol, ketones and aldehydes should be studied deeply to determine control measures and management methods and establish more fine standard.

4. Environmental Impact

4.1 Production of Secondary Organic Aerosol

SOA (Secondary Organic Aerosol), the product of VOCs during the process of oxidation in the atmosphere, is the general name of organic pollutants in gas, liquid, solid, sol and dissolved states. The vapor pressure of VOCs such as high-carbon alkanes, alkene and aromatic hydrocarbons in an oxidation state in the atmosphere is lower than that in a reduction state, so they have the potential to be oxidized into SOA, that is, they can react with OH free radical, NO_2 in an excited state and O_3 in the atmosphere to generate SOA [22-24]. Based on aerosol production coefficient ($M_{SOA\_potential} = M_{VOC\_initial} \times FAC \times Fvocr$) produced by Grosjean, D. [25], the annual average emission of aromatic

hydrocarbons and alkanes from the investigated enterprises were estimated, and the production of secondary organic aerosol from the two substances was obtained.

Seen from Table 5, the emission of VOCs from each enterprise accounted for 74%, 75% and 43% of total emission of VOCs from the three enterprises. Esters, alcohols, aldehydes and ketones were not included here, so that the estimated value of SOA was small. In addition, the reaction between OH and initial VOCs was considered, SOA calculated by using FAC production rate method accounted for only 0.5%-50.0% of the actual value [26]. Therefore, the total production of SOA in Table 5 was lower than the actual production of SOA produced by VOCs. Aromatic hydrocarbons contributed greatly to the generation of SOA, while the contribution of alkanes to it was small. There are few studies on FAC values of alcohols, esters, aldehydes and ketones, so the production of SOA produced by them could not be calculated to determine their contribution. In respect of SOA control, automobile spraying and coating enterprises should control the use of dimethylbenzene, trimethylbenzene and other aromatic hydrocarbons and try to use heptane with small FAC and Fvocr.

4.2 Ozone Formation Potential

Under natural conditions without man-made interference, the photolytic cycle of NO, NO_2 and O_3 in the troposphere forms, and O_3 will not accumulate. Under the effect of VOCs and OH free radical, the balance will be damaged, and NO will be transformed into NO_2 continuously. As a result, O_3 accumulates and pollutes ambient air. MIR (Max Incremental Reactivities), PE (Propy-Equivconcentration), OH consumption rate (L^{OH}), etc. [27-30] have been used to analyze the numerical relation between VOCs and ozone.

Based on the latest parameter list of MIR of USEPA (70 FR 53930) [31], namely the revised edition of California in 2010 [32], the ozone formation potential of automobile coating enterprises OFP ($C_{i\_OFP} = C_{i\_VOC} \times MIR_{i\_index}$) was calculated. According to Table 4 and annual average emission of each enterprise, their annual average concentration was calculated, and OFP was calculated according to the recommended values of MIR. The MIR value of naphtha was the arithmetic mean of MIR values of 20 hydrocarbon mixed solvents, and the MIR value of aromatic hydrocarbons was the arithmetic mean of MIR values of 4 aromatic solvents (Table 6).

Major ingredients contributing to ozone production greatly are shown as: major ingredients were aromatic hydrocarbons (69%), dimethylbenzene (13%) and naphtha (7%) in plant A, aromatic hydrocarbon (76%), trimethylbenzene (11%) and n-butyl alcohol (6%) in plant B, and dimethylbenzene (69%), isobutyl alcohol (9%) and trimethylbenzene (7%) in plant C respectively. In respect of ozone control, automobile spraying and coating enterprises in Chonging city should control the use of trimethylbenzene and try to use ethyl acetate, butyl acetate and isobutyl alcohol to replace dimethylbenzene.

Table 5 The production of SOA caused by VOCs from typical automobile manufacturing enterprises.

Ingredient	FAC (%)	Fvocr	Production of SOA (kg/a)		
			Plant A	Plant B	Plant C
Methylbenzene	5.40	0.12	3	-	38
Dimethylbenzene	3.77	0.31	994	16	3,063
1, 3, 5-trimethylbenzene	2.90	0.74	-	1,250	380
n-heptane	0.06	0.14	-	2	-
Aromatic hydrocarbon	4.95	0.36	9,626	12,768	155
Naphtha	1.08	0.14	450	202	-
Total			11,073	14,238	3,636

Table 6 The ozone formation potential of typical automobile manufacturing enterprises by discharging VOCs.

Ingredient	MIR	OFP (mg/m^3)		
		Plant A	Plant B	Plant C
Methylbenzene	0.72	0.06	0.00	1.32
Dimethylbenzene	7.74	146.31	2.15	631.17
1, 3, 5-trimethylbenzene	11.76	-	137.86	64.83
Ethyl acetate	0.63	-	-	13.64
Butyl alcohol	0.83	31.65	11.52	8.21
n-butyl alcohol	2.88	66.20	75.35	3.54
2-methyl-1-propanol	1.36	6.48	-	78.74
Methyl isobutyl ketone	3.88	18.47	-	-
Methanol	0.67	-	-	1.32
Propylene glycol	2.58	-	23.48	-
Propylene glycol monoethyl ether	3.09	-	-	30.49
Dimethyl succinate	0.23	-	-	0.45
Dimethyl adipic acid	1.8	-	-	7.11
Dimethyl glutarate	0.42	-	-	2.49
Methyl ethyl ketone	1.48	-	-	17.46
Diethylene glycol monobutyl ether	2.39	-	-	14.30
Formaldehyde	9.46	-	-	18.67
2-butoxy ethanol	2.9	-	-	5.34
2-propyl alcohol	0.61	-	8.46	-
n-heptane	1.07	-	4.45	-
Aromatic hydrocarbon	6.48 (3.82-7.64)	777.97	934.69	17.58
Naphtha	1.12 (0.48-1.98)	74.10	30.16	-
Total		1121.25	1228.11	916.64

5. Conclusions

(1) In automobile coating industry of Chongqing city, VOCs are mainly discharged during the processes of spraying and levelling, while the emission of VOCs during the process of the drying stage is relatively small. Automobile spraying and coating enterprises in Chonging city should transform and upgrade the processes of spraying and levelling firstly and treat VOCs discharged during the processes of spraying and levelling deeply to reduce the emission of VOCs.

(2) The major ingredients of VOCs discharged during the process of automobile coating in Chongqing city were aromatic hydrocarbons such as dimethylbenzene and trimethylbenzene, esters such as ethyl acetate and butyl alcohol, alcohols like n-butyl alcohol, ethers, aldehydes and ketones. The emission and treatment efficiency of the above exhaust gases discharged from automobile coating should be studied deeply to determine control measures and management methods and establish finer standard.

(3) Among the major ingredients of VOCs discharged during the process of automobile coating in Chongqing city, aromatic hydrocarbons contributed to the production of SOA greatly, while the contribution of alkanes to it was small. There are few studies on FAC values of alcohols, esters, aldehydes and ketones, so the production of SOA produced by them could not be calculated. Automobile spraying and coating enterprises should control the use of dimethylbenzene, trimethylbenzene and other aromatic hydrocarbons and try to use heptane with small FAC and Fvocr.

(4) Among the major ingredients of VOCs discharged during the process of automobile coating in Chongqing city, aromatic hydrocarbons and alkanes

contributed to ozone production greatly. In addition, alcohols and ethers also had certain contribution. Automobile spraying and coating enterprises in Chonging city should control the use of trimethylbenzene and try to use ethyl acetate, butyl acetate and isobutyl alcohol to replace dimethylbenzene.

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