JOURNAL OF ENVIRONMENTAL SCIENCES 135 (2024) 546–559

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JOURNAL OF ENVIRONMENTAL SCIENCES

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Research Article

Species profile and reactivity of volatile organic compounds emission in solvent uses, industry activities and from vehicular tunnels

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ARTICLE INFO

Article history:
Received 23 March 2022
Revised 28 August 2022
Accepted 30 August 2022
Available online 11 September 2022

Keywords:
Volatile organic compounds (VOCs)
Species profile
BTEX ratio
Source reactivity

ABSTRACT

A survey was conducted of the volatile organic compounds (VOCs) released from sources of solvent use, industry activities and vehicle emissions in Guiyang, a capital city of China. Samples were collected by canisters and analyzed by GC-MS-FID. The species profiles of VOCs emitted from sources were obtained. Results showed that xylenes, ethylbenzene, acetone and dichloromethane were the characteristics species for painting, 2-propanol and ethyl acetate for printing, α-pinene for solid wood furniture manufacturing, and 2-butanone for biscuit baking. These characteristics species could be as tracers for the sources respectively. In most of samples from the solvent use, the benzene/toluene (B/T) ratio was less than 0.3, indicating that the ratio could be as the indicator for tracing the solvent use related sources. The results also suggested that the toluene/xylene (T/X) ratio be as the indicator to distinguish the VOCs sources of painting (<2) from the printing (>2). Aromatics contributed the most to ozone formation potential (OFP) of most painting and non-paper printing sources, and oxygen-containing VOCs (OVOCs) were major species contributing to OFP of the sources from food production and paper printing. The OFP of the VOCs emissions from vehicle in tunnels and from other manufactures were dominated by both aromatics and alkenes. The α-pinene could explain 56.94% and 32.54% of total OFP of the VOCs sources from filing cabinet and solid wood furniture manufacturing, which was rarely been involved in previous studies of VOCs source profiles, indicating that the species of concern for VOCs sources are still insufficient at present.

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https://doi.org/10.1016/j.jes.2022.08.035
1001-0742/

Introduction

As one of the precursors of ozone (O_3) formation, volatile organic compounds (VOCs) play a crucial role in ground-level O_3 concentration rising (Atkinson, 2000), especially in urban areas (Shao et al., 2009; Wang et al., 2009; Li et al., 2019), which now is the major air pollution problem in China (Li et al., 2020; Liu and Wang, 2020). Reduction of VOCs emissions has been proposed as an effective path to alleviate ozone pollution (Shao et al., 2009; Xue et al., 2014). However, the sources of VOCs are complex and strongly affected by human activities in urban areas, such as vehicle emission, solvent use, industry activities and combustion related processes. Species profiles of VOCs released from sources can visually reveal the emission characteristics and is a theoretical basis for developing VOCs emission control strategies.

Industry activities and solvent use are two key roles driving the increase of total anthropogenic VOCs emissions in China (Zhao et al., 2017; Li et al., 2019). Previous studies have reported the species profiles of VOCs emission from industry of petrochemical facilities, petroleum refineries, chemical synthesis pharmaceutical, synthetic resins and rubber footwear industries (Wei et al., 2014; Mo et al., 2015; Li et al., 2019; Cheng et al., 2021) and from solvent use of surface coating on automobiles, furniture, building and metal parts, and printing of gravure and offset processes (Liu et al., 2008; Yuan et al., 2010; Zheng et al., 2013; Wang et al., 2014; Zhong et al., 2017). However, industry activities and solvent use sectors are complex with diverse raw materials, processes and product types, and the specie profile of VOCs in many industries have not been revealed yet, such as painting and printing on different substrates, production of different kinds of foods and furniture etc. In addition, vehicle emission is also an important contributor to VOCs pollution in urban areas (Liu et al., 2016; Hui et al., 2018; Song et al., 2018). Species profile of VOCs emitted from vehicles have been widely reported, which obtained by using chassis dynamometers or by direct measurements in tunnels or roadsides (Liu et al., 2008; Mo et al., 2016; Tang et al., 2021; Liu et al., 2022). However, there are few reports focused on the species profile of VOCs from different vehicles mixed together on the actual road, especially the on-road vehicles on different speed-limited sections. It is no doubt that previous literatures have contributed to the local species profiles of VOCs in China, while VOCs emission sources are difference in spatial depending on industrial structure, energy consumption and urban function (Baudic et al., 2016; Song et al., 2018; Li et al., 2020; Xu et al., 2021). The surveying of VOCs sources emission in a few cities are not sufficient and the knowledge of species profile of VOCs emission is far from enough in covering all the VOCs sources. Thus, more studies are needed to fill the knowledge gap of species profile of VOCs in emission sources, such as solvent use, industry activities and vehicle emission sectors.

To enrich the database of VOCs species profile in China, field measurements of 114 VOCs species were conducted to obtain the source profile of VOCs in typical sectors of solvent use, industry activities and vehicle emission in Guiyang, a capital city in southwest China in 2019. Varieties of sources including solvent use of painting and printing on different substrates, industry activities of manufacturing of rubber products, asphalt, plastic woven fabrics, Chinese medicine, furniture, several foods, landfilling as well as vehicle emission from urban road tunnel and highway tunnel were investigated, covering main sources of VOCs in the city. The characteristics, potential tracers, special species (benzene, toluene, ethylbenzene, xylenes, BTEX) ratio and source reactivity of each emission source were discussed. These results will enrich the knowledge of VOCs species profile from emission sources and provide more information for VOCs pollution control.

1. Materials and methods

1.1. *Sampling*

Evacuated stainless steel canisters (Entech Instruments, California, USA, 1.4 L) were employed to collect the source air, which were cleaned three times with high purity nitrogen (> 99.999%) and finally evacuated to 10 mTorr prior to sampling. A filter was installed on the canister when sampling to remove particulate matters. Samples were collected in the workshop for painting, printing, the manufacturing of rubber products, asphalt, plastic woven fabrics, Chinese medicine, furniture, the foods as well as for landfilling. The sampling periods were approximately 1 min, as reported in the literature of Yuan et al. (2010). In our investigation, air in most of workshops are directly discharged into the environment through windows and ventilation fans. Although some workshops have gas collection devices, some gas still remains inside the workshop. And previous study found the removal efficiency of ten VOCs treatment methods commonly used was not satisfactory based on 130 industry enterprises, with average efficiency of 66.20% (Su et al., 2016). Therefore, we focused on the fugitive emission in each workshop in this study. Samples were also collected in an urban road tunnel and a highway tunnel. After sampling, the canisters were promptly shipped to laboratory for analysis.

For painting. Painting is widely used in most sectors of industries. Painting on various substrates has different purposes, including wear resistance, corrosion resistance, heat insulation, electrical conductivity, insulation, etc. The painting-related processes investigated in this study included the painting on different surfaces (wooden furniture, metal parts, color aluminum tile and anticorrosive steel), the electrostatic powder coating and paint manufacturing. Air samples were collected in the workshop during the normal working condition of painting.

For printing. Printing is a process that transfer ink to the surface of paper, textiles, plastic products and other materials. In this study, sampling was conducted in typical printing factories, with products of books, packaging cartons, cigarette boxes, teaching materials, plastic woven fabrics, nonwoven bag, etc. Samples were collected in the workshops during the normal working condition of printing machine, and covered the printing methods of offset, gravure and letterpress.

For rubber products manufacturing. The rubber products manufacturing involves three basic steps of mixing, shaping and vulcanization processes. In this study, samples were collected from four rubber product manufacturers, including two manufacturers for truck radial tire and two for automobile rubber parts and rubber belts and hoses. Samples were collected from each process during the routine manufacturing condition.

For asphalt processing. Asphalt is the by-product of petroleum refining. High temperature condition (usually above 110°C) is required for the production of asphalt mixture and VOCs released to the atmosphere are from this process. The samples were from the workshops of asphalt storage, mixing/melting furnace and discharge port of asphalt dump truck.

For Chinese medicine processing. Two Chinese medicine manufacturers were investigated in the workshops involving process of extracting Chinese herbal and filling medicines into capsules. Samples were collected close to the work line in capsules and extraction workshop.

For plastic woven fabrics manufacturing. The plastic woven fabrics manufacturing use polypropylene as the raw material, with process of tape cutting, weaving, drawing, textiles, printing and so on. In this study, a plastic woven fabrics factory was selected to collected air samples. The main products of the factory are plastic filaments, ropes and woven fabrics, with the production capacity of 4000 ton/year. Samples were collected for the entire production processes from raw materials to products. The sampling was closed to the devices with working condition.

For landfilling. Landfill gas generated by wastes decomposition. In this study, two landfills were investigated with the average daily garbage disposal capacity of 600 tons and 1800 tons respectively. Sampling conducted on site in the landfill and leachate treatment workplace.

For furniture manufacturing. The samples of furniture manufacturing were collected from workshops of mattress, ceramic tiles, filing cabinet and solid wood furniture manufactures.

For food production. The samples collection of food production covered bacon production, bread making, biscuit making, beer, white wine and fruit wine production in this study.

In tunnels. An urban road tunnel with 970 m length, 60 km/hr speed limit and a highway tunnel with 630 m length, 80 km/hr speed limit were selected for this study. For the urban road tunnel, the samples were collected in the morning (8:00–9:00), noon (13:00-14:00), afternoon (17:00-18:00) and evening (23:00–24:00) for three days (December 24, 25 and 28, 2019). For the highway tunnel, hourly samples were collected during 8:30-18:30, December 30, 2019, with 7 times in the day. Samples were collected at the tunnel entrance, exit and inside in both two tunnels and the sampling periods were one hour controlled by a restricted sampler. Totally, 36 samples were obtained from the urban road tunnel and 21 from the highway tunnel.

The source profiles name and corresponding number of samples in each source were given in Appendix A Table S1.

1.2. Samples analysis

A total of 114 VOCs, including species of PAMS (56 compounds), TO-15 (65 compounds) and seven terpenes, covering 29 alkanes, 18 alkenes, 18 aromatics, 12 OVOCs, 35 halogenated, one alkyne and one sulfide, were measured using GC-MS-FID system (Trace 1310/ISQ7000, Thermo Fisher Scientific Inc, USA) coupled with pre-concentrator instrument (7200, Entech Instruments, California, USA). Samples were pre-concentrated by the 7200 using CTD model and then injected in GC-MS-FID system. After injection, the sample was first separated by a TG-1MS capillary column (60 m × 0.25 mm × 1.0 μm, Thermo Fisher Scientific Inc, USA) with helium and then split in two ways by dean-switch (Deans, 1981; Tranchida et al., 2012), one was a TG-BOND Alumina column (Na_2SO_4, 30 m × 0.32 mm × 5.0 μm, Thermo Fisher Scientific Inc, USA) followed by a FID detector, and the other was a 4.31 m × 0.18 mm × 0 μm capillary column followed by MS detector. All column were kept isothermal at 5 °C for 7 min, then heated to 190 °C at the rate of 5 °C/min and finally maintained at 190 °C for 10 min. Light carbon components (ethane, ethylene, acetylene, propane and propylene) were quantified by FID, and the rest were quantified by MS. Both full scan and TSIM mode were used for MS quantitative determination in mass range of 26 to 270 *m/z*. The ion source temperature was set at 320 °C and transfer line temperature was set at 300 °C.

Method detection limit (MDL) was calculated for a signal-to-noise ratio of 3:1 or 3 times the standard deviation of blank samples, which ranged from 0.2 ppt to 45 ppt. Precision was determined by seven replicated measurements of calibration gas. The relative standard deviation (RSD) values were within 6% for all target species. The detail category information, MDL and RSD of each species were listed in Appendix A Table S2.

1.3. Data analysis

1.3.1. Species profile

The species profiles of VOCs were present by mass proportion of each specie to total VOCs. Species profile of VOCs emission from sources were obtained by the proportion-average and calculated as Eq. (1) in follows, which proposed by USEPA (2004).

$$Xi = \frac{\sum_{j=1}^{n} x_{i,j}}{n} \tag{1}$$

where, X_i is the average mass proportion of specie *i*. $x_{i,j}$ is the mass proportion of specie *i* in the individual profile *j*, and *n* is the number of individual profile of same type source, as shown in Appendix A Table S1.

1.3.2. Ozone formation potential

The ozone formation potential (OFP) of unit mass VOCs from sources were evaluated to estimate the source reactivity. The total OFP were the sum of species OFP calculated by multiplying the species proportion by the maximum incremental reactivity (MIR) and VOCs emissions (Na and Kim, 2007; Carter, 2010; Yuan et al., 2010). The species OFP was calculated as Eq. (2) as follows:

$$OFP_i = X_i \times MIR_i \times E \tag{2}$$

where, OFP_i was the ozone formation potential of specie i, which unit was g O_3/g VOCs. X_i was the average mass proportion of specie i, obtained from Eq. (1). MIR_i was the maximum increment reactions value of specie i, as proposed by Zhang et al. (2021), Venecek et al. (2018) and Carter (2010), which summarized in Appendix A Table S2. E was the total VOCs emissions from sources with unit of g, here defined as unit mass.

2. Results

A total of 29 species profiles were obtained in this study and each source profile included 114 species. The proportion of species in profiles and total VOCs concentration of each source were listed in Appendix A Table S3. We divided the 114 species into seven groups including alkanes, alkenes, alkynes, aromatics, halogenated, OVOCs and others, and the source profile of seven groups of VOCs in each emission source were showed in Appendix A Fig. S1. Detail information of VOCs species profile in each source were discussed below. Due to the inconsistent VOCs species between source profiles observed in previous studies and in this study, we reclassified the "Others" group in the sub-section below into other alkanes, other alkenes, other alkyne, other aromatic, other halocarbon, other OVOCs and carbon disulfide (CS_2), for comparing with reported studies. The five species of VOCs (myrcene, 2-carene, α-terpinene, limonene and γ-terpinene) in this study were classified as other alkenes. Detailed classification was shown in Appendix A Table S3.

2.1. *Species profile in solvent use*

2.1.1. *In painting*

In this study, species profiles of VOCs from painting on different surfaces (wooden furniture, metal parts, color aluminum tile and anticorrosive steel), the electrostatic powder coating and paint manufacturing were obtained and results are showed in Fig. 1. It is found that species profiles of VOCs from painting on the surface of wooden furniture and metal parts were similar. Aromatics and OVOCs were main VOCs groups and xylenes, ethylbenzene, ethyl acetate and toluene were the dominant species in VOCs emission from wooden furniture and metal parts painting processes. The results were comparable with previous studies of painting (Yuan et al., 2010; Zheng et al., 2013; Wang et al., 2014; Zhong et al., 2017; Zhang et al., 2020). While the proportion of m/p-xylene (25.21%), o-xylene (17.34%), ethylbenzene (16.15%) and ethyl acetate (36.25%) in wooden furniture painting were higher than those in metal parts painting, with proportion of 5.47%, 3.99%, 3.10% and 14.31%, respectively. However, dichloromethane was the most species in metal parts

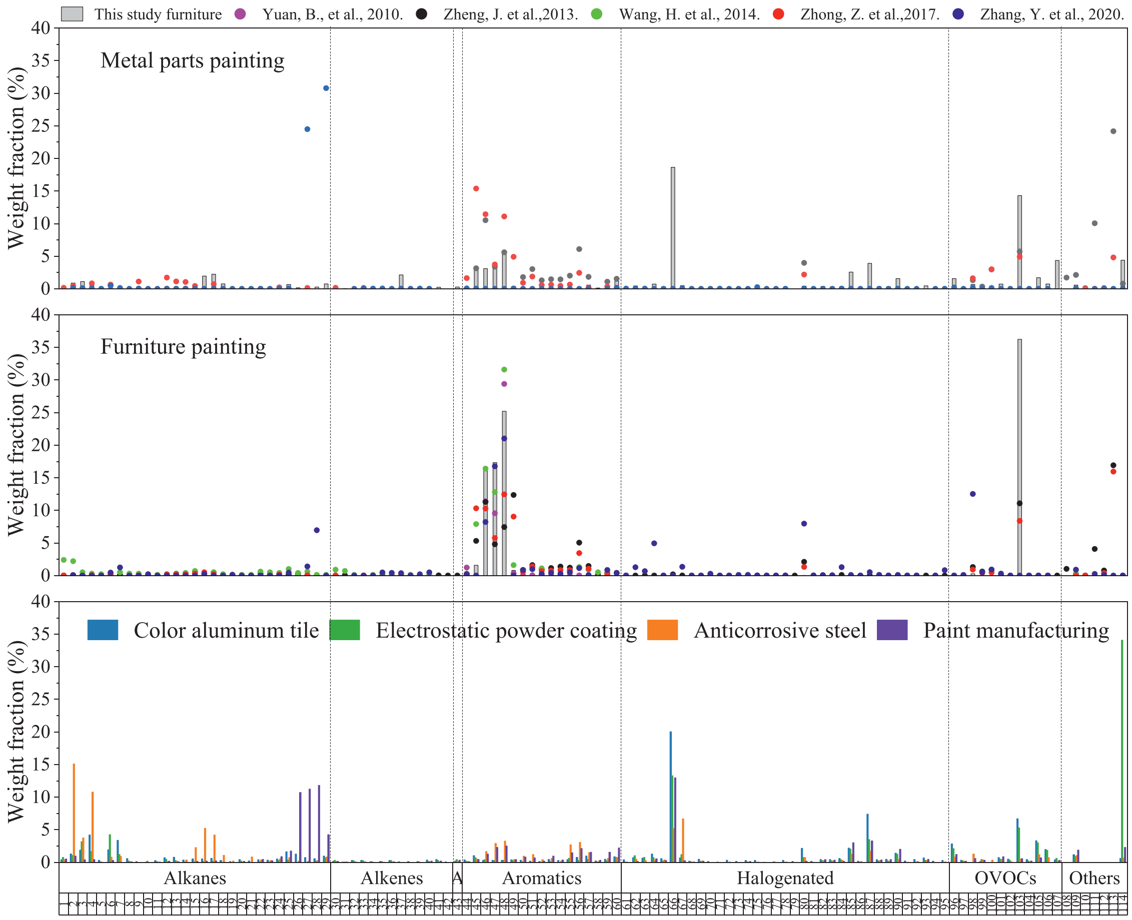

Fig. 1 – Species profiles of VOCs released from different painting processes ("A" represents "Acetylene" in the X-axis. The species name corresponding to serial number in the figure are the same as those in Appendix A Table S3. Detail species profiles was given in Appendix A Table S3. Hereinafter the same.)

painting and accounted for 18.64% to total VOCs, which was significantly higher than that from wooden furniture painting with proportion of 0.13%. So, dichloromethane could be used as a marker to distinguish VOCs emission from wood furniture painting and metal parts painting. Moreover, species profiles of VOCs from painting on surface of color aluminum tile and anticorrosive steel were quite different from that of wooden furniture and metal parts. As shown in Fig. 1, halogenated (44.62%) were dominant group of VOCs in color aluminum tile painting and dichloromethane (20.04%), 1,3-dichlorobenzene (7.40%), ethyl acetate (6.69%) and butane (4.20%) were the top four VOCs. While alkanes (49.96%) were the most VOCs group in anticorrosive steel painting and propane, butane, methyl chloride, 2-methylhexane, dichloromethane were the top species in anticorrosive steel painting with proportion of 15.10%, 10.78%, 6.68%, 5.22% and 5.18% respectively.

In fact, ethyl acetate and xylenes are the typical components of paints and diluent for painting on surface of wooden furniture and metal parts (Zheng et al., 2013; Zhong et al., 2017), and dichloromethane is used as a solvent in paint strippers, metal cleaning and finishing (USEPA, 2016). Water-borne zero VOCs paints mainly emit oxygenate (for example acetone) and almost no aromatics exhausting (Stockwell et al., 2021), which is used as paints in the surveyed color aluminum tile painting factory. While solvent-borne paints are mainly composed of alkanes (<C10), cycloalkanes and aromatics (especially xylenes and ethylbenzene) (Stockwell et al., 2021), which are used in painting on anticorrosive steel we investigated. The difference of species profiles in painting on surface of four substrates (wooden furniture, metal parts, color aluminum tile and anticorrosive steel) indicated that the species profiles highly correlated with the raw materials (Zheng et al., 2013; Zhong et al., 2017).

Electrostatic powder coating is one of the ways for surface treatment, which uses the principle of corona discharge to directly adsorb the atomized paint on the surface of the substrate. Species profile of VOCs from electrostatic powder coating had rarely been involved in previous researches. Unlike painting processes, sulfides (34.09%) was the most abundant group in VOCs emission from electrostatic powder coating, followed by halogenated (26.97%), alkanes (15.99%) and OVOCs (14.65%), while aromatics only accounted for 5.06% to total VOCs (Fig. 1). CS_2, dichloromethane and ethyl acetate were the dominants species with proportion of 34.09%, 13.29% and 5.29% respectively. In addition, air samples from paint manufacturing were also analyzed. In general, alkanes (49.96%), halogenated (25.27%) and aromatics (19.35%) were the major groups in paint manufacturing workshop and dichloromethane, nonane, decane and undecane were dominant species with the proportion of 12.97%, 10.72%, 11.25% and 11.81% respectively (Fig. 1).

2.1.2. *In printing*

Generally, VOCs emission from printing processes derive from the use of raw and auxiliary materials, such as ink, cleaning agent, diluent, fountain solution and adhesive. Fig. 2a shows the species profiles of VOCs from different printing processes of offset, gravure and letter printing. Results showed that 2-propanol, ethyl acetate, methyl methacrylate and dichloromethane were the dominant species in all VOCs source profiles of offset, gravure and letter printing. Ethyl acetate and methyl methacrylate are mainly released from ink and isopropanol is mainly used as a thinner for adhesives (Zheng et al., 2013). The results were consistent with studies in VOCs source profiles of printing from literatures (Zheng et al., 2013; Shen et al., 2018; Alabdulhadi et al., 2019), which also reported 2-propanol, ethyl acetate and methyl methacrylate were the main VOCs components in offset and gravure printing processes (Fig. 2a). But different composition characteristics were observed from letter printing by Zheng et al. (2013), which reported that aromatics were the most abundant group with proportion of 41.89%, and among which benzene (14.76%), toluene (12.77%) and ethylbenzene (4.85%) contributed the most. Aromatics mainly come from solvent-based ink. With the development of ink types, water-based ink which do not contain strong toxic aromatics, are more and more widely used. The workshops of letter printing we investigated also used water-based inks. It was the main reason why the concentration of aromatics observed by us was lower than that observed by Zheng et al. (2013).

Species profile of VOCs from printing on different substrates were also investigated. The substrates for printing of offset, gravure and letter we discussed above were all paper products. We also compared the differences in species profile of VOCs from printing on substrates of plastic and non-woven fabrics. As shown in Fig. 2b, toluene, benzene, 2-propanol and ethyl acetate were found to be the dominant VOCs species in plastic printing, contributing 55.00%, 11.94% and 7.94% of the total VOCs respectively. Toluene (19.11%), ethyl acetate (15.04%) and propane (9.34%) were main VOCs species in non-woven fabrics printing, which were similar with the results of cloth printing reported by Zhong et al. (2017) and Zhang et al. (2020) (Fig. 2b). The difference of species profile of VOCs among printing on paper, plastic and non-woven fabrics could be explained by the type of ink and the adsorption capacity of ink for different substrates. Besides, air samples from adhesive binding workshop were also analyzed. As shown in Fig. 2b, OVOCs was dominant VOCs group in binding workshop, with proportion of 67.89%, followed by aromatics (17.45%) and alkanes (8.89%), and 2-propanol contributed the most to the total VOCs, accounting for up to 65.13%.

2.2. *Species profile in industry activities*

2.2.1. *Rubber products manufacturing*

Rubber industry is an important resource-based industry sector. In China, the rubber industry has maintained stable development. The rubber consumption, total output value of rubber industry and tire output are all ranked first in the world (CRIA, 2020). The species profile of VOCs from rubber products manufacturing obtained in this study are shown in Fig. 3a. C6-C8 alkanes, especially heptane and isomers of heptane (4.01%-5.14%), dichloromethane (5.54%), naphthalene (3.74%), 4-methyl-2-pentanone (3.60%), acetone (3.64%) and CS_2 (11.61%) were dominant species in VOCs emission from rubber products manufacturing. It had been reported that water-based adhesives used in rubber products manufacturing emitted complex mixture of normal and branched alkanes (Girman et al., 1986), and naphthalene, ethylbenzene, xylenes and CS_2 were commonly used as rubber solvents

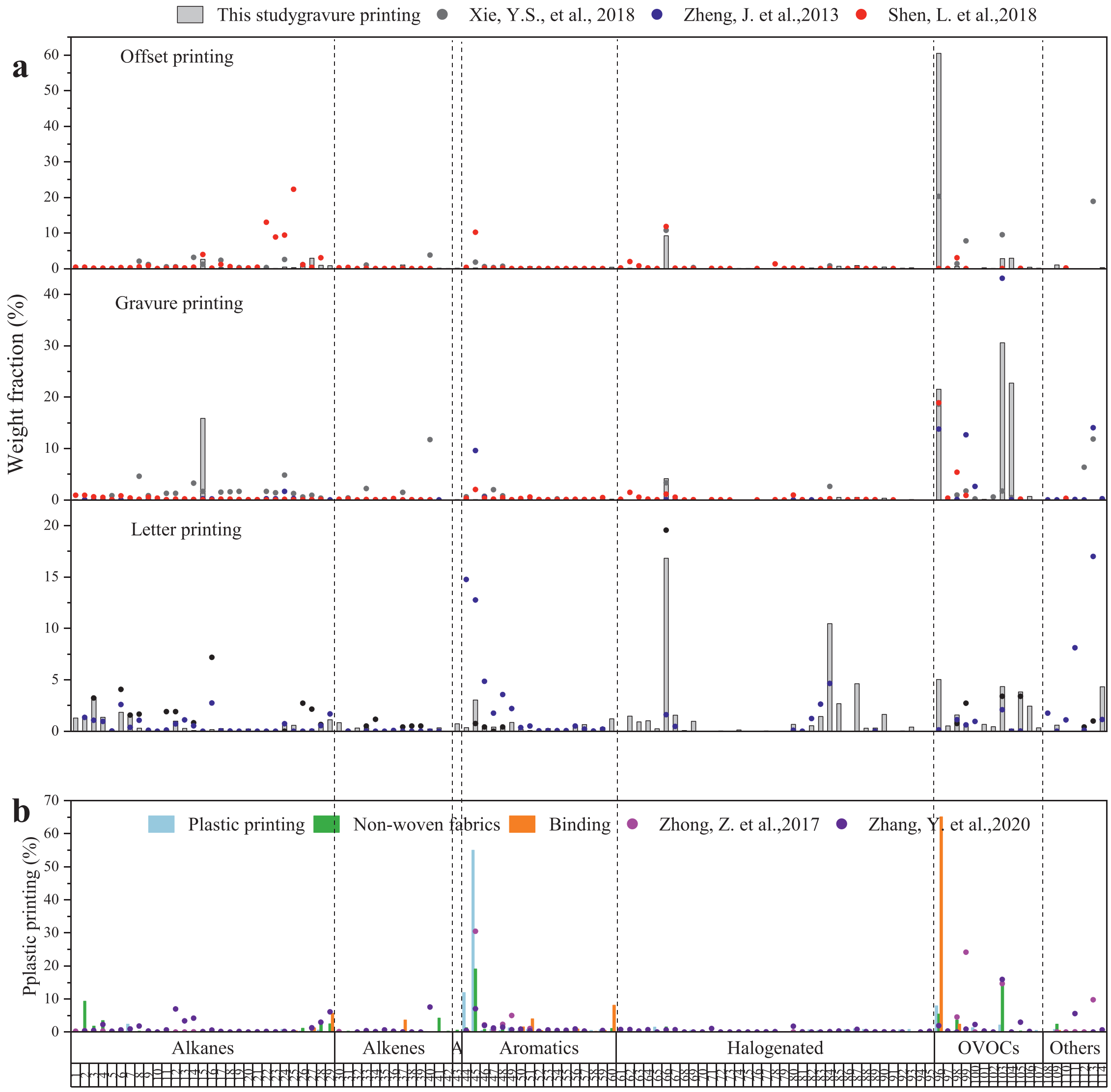

Fig. 2 – Species profiles of VOCs released from different printing processes.

(Adams et al., 1990). The results we obtained were similar with the VOCs source profile of rubber manufacturing in USEPA SPECIATES 5.1 database (USEPA, 2020), which suggested that the uncontrolled VOCs emission from rubber manufacture were heptane and isomers of heptane. Wu et al also reported a VOCs source profile of rubber manufacture, which found m/p-xylene (14.81%), ethylbenzene (12.39%), styrene (9.70%), heptane and isomers of heptane (3.16%-4.42%) were major species (Wu and Xie, 2017) of VOCs emitted from rubber manufacture.

2.2.2. *Asphalt processing*

Asphalt is a complex black–brown mixture and consist of hydrocarbons and their nonmetallic derivatives. The mixtures are generated by mixing asphalt and mineral aggregate at 100–130°C in asphalt mixing plants (Qu et al., 2021). Species profile of VOCs from asphalt manufacturing we observed is shown in Fig. 3b. In fugitive emissions of VOCs from asphalt manufacturing, alkanes and OVOCs were the major groups, and ethyl acetate was the most abundant species, accounting for 23.04% to total VOCs, followed by naphthalene, toluene, dodecane, propane, iso-pentane and cyclohexane. The results were similar with other reported in asphalt mixture plant, which was found that alkanes was the major group with proportion of 46.98% to total VOCs (Li et al., 2020).

2.2.3. *Chinese medicine processing*

The extraction purification of Chinese herbal and the sub-packaging of capsules are two main processes in the Chinese medicine processing. Species profile of VOCs from Chinese medicine processing were obtained and results are shown in Fig. 3c. OVOCs was the most group with the proportion of 38.14% to total VOCs, followed by halogenated (21.16%), alkanes (14.28%), aromatics (13.37%) and the rest. 2-Propanol (25.68%) was the most abundant species in VOCs emission, followed by dichloromethane (6.91%) and dodecane (4.08%). The species profile of VOCs was quite different from that of previous studies. Acetone (68.37%) and pentane (16.04%) were found to be the major VOCs in air above the exhaust ducts

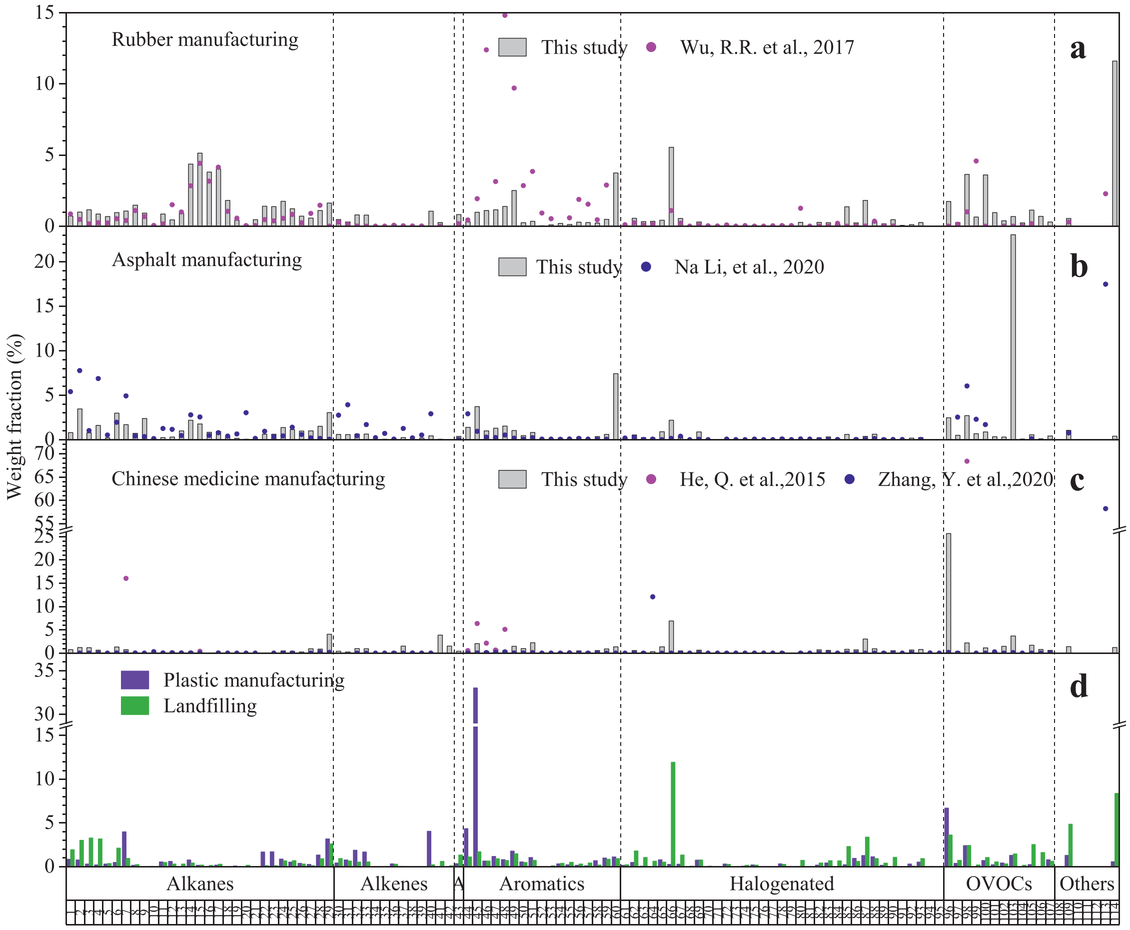

Fig. 3 – Species profiles of VOCs released from rubber products manufacturing (a), asphalt processing (b), Chinese medicine processing (c), plastic manufacturing and landfilling (d).

of production lines in a medicine producing industry reported by He et al. (2015). While acetone (15.66%), chloroform (10.56%) and ethanol (58.96%) were the dominant VOCs in the fugitive emission air from workshops of chemical reaction pharmaceutics and extraction purification in pharmaceutical manufacturing found by Zhang et al. (2020). Extraction purification process commonly used only one kind of extraction agent, such as ethanol, acetone or 2-propanol, which had a significant effect on the source profile of VOCs. However, the effects of different chemical pharmaceutical reaction processes on the composition of VOCs were minimal (Zhang et al., 2020). So, the difference of species profile of VOCs from pharmaceutical manufacturing among studies were related to the different extraction agent used in different factories.

2.2.4. *Plastic woven fabrics manufacturing*

Species profile of VOCs from plastic woven fabrics manufacturing are shown in Fig. 3d. In fugitive emissions, toluene was found to be the most abundant VOCs, accounting for 33.01% to total VOCs, followed by 2-propanol, benzene, 1-hexene, pentane and dodecane, with proportion of 6.71%, 4.35%, 4.05%, 3.98% and 3.18% respectively. It had been found that toluene, ethyl-benzene and xylenes were major species emitted from melting and powdering processes in the polypropylene (PE) and polyethylene (PP) plastic waste recycling, with proportion of 40% to total VOCs (Tsai et al., 2009). While toluene only contributed 5.66% and 2.11% to the total VOCs emissions from PP and PE producing reported by He et al. (2015).

2.2.5. *Landfilling*

Based on two landfills the species profile of VOCs from landfilling were obtained and the result is shown in Fig. 3d. Halogenated, alkanes, OVOCs and aromatics were major VOCs groups, and dichloromethane was found to be the most abundant VOCs, accounting for 11.95% to total VOCs, followed by CS_2 (8.93%), 2-propanol, propane, butane and isobutane, with proportion of about 3%. In addition, other alkenes, such as terpenes of myrcene, limonene, carene and terpinene contributed 4.85% of total VOCs. Aromatics and terpenes are usually emitted from food waste, wood, yard waste and personal care products (e.g., air freshener and body wash), while OVOCs and alkanes are the representatives of the comprehensively degrading waste and halogenated compounds symbolize xenobiotic compounds (Scaglia et al., 2011; Tan et al., 2017; Lim et al., 2018; Nair et al., 2019). In fact, the composition of domestic waste transferred to landfill vary extremely at different regions, resulting in regional variations in VOCs emissions due to the differences in living habits, geographical

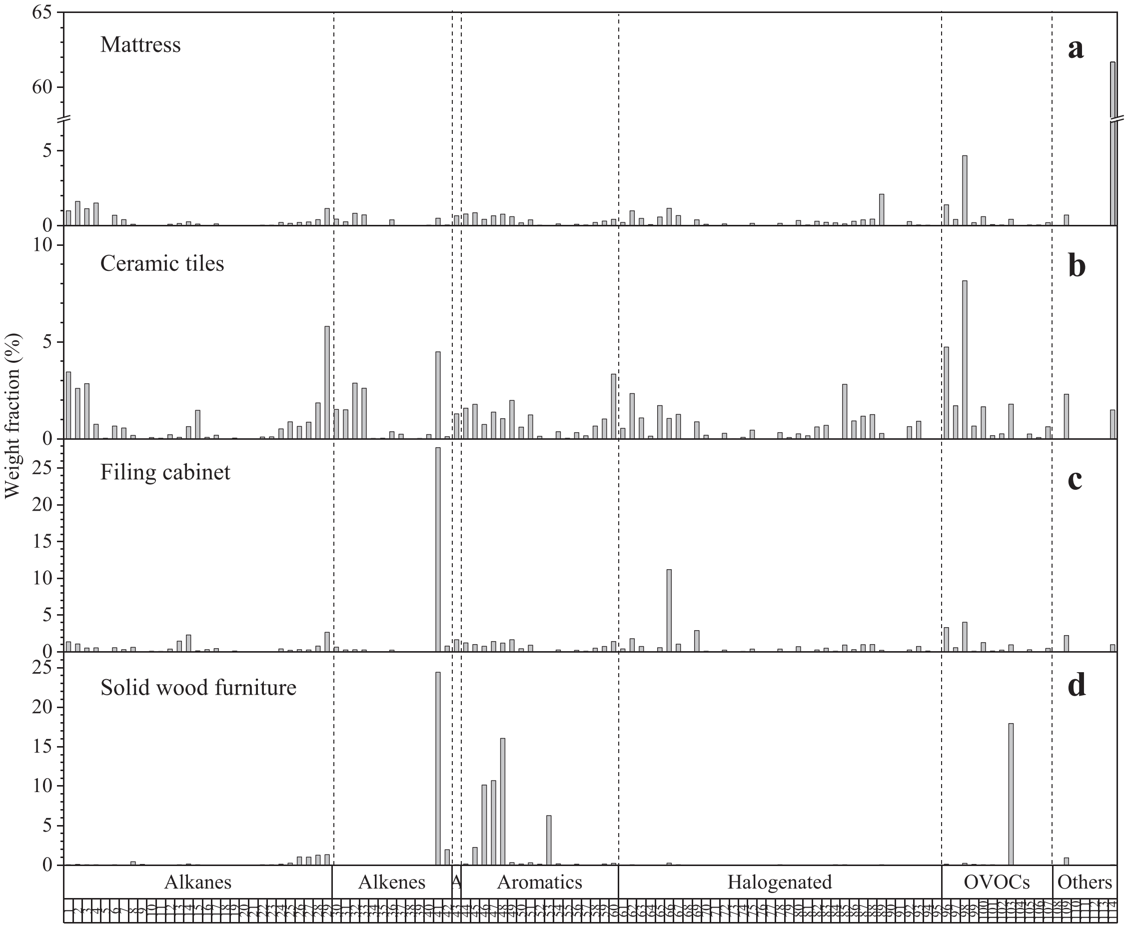

Fig. 4 – Species profiles of VOCs released from manufacturing of Mattress (a), Ceramic (b), Filing cabinet (c) and Solid wood furniture (d).

location and climatic conditions. Ethyl acetate (53.86%) was the highest VOCs in a compression transfer station in Guangzhou (Li et al., 2013) and ethanol (67.71%) contributed the most of VOCs emission in a waste transfer station located in Suzhou (Liu and Zheng, 2020).

2.2.6. *Furniture manufacturing*

The furniture manufacturing process involves different products made of different materials such as wood, metal, ceramic, plastic, etc. Most of previous studies in VOCs source profile of furniture manufacturing focused on the VOCs emission from furniture painting (Zheng et al., 2013; Zhong et al., 2017). In this study, species profiles of VOCs from manufacture of mattress, ceramic, filing cabinet and solid wood furniture were obtained and the results are shown in Fig. 4. CS_2 was found to be the most abundant VOCs in the species profile of mattress manufacturing, with proportion of 61.68% to total VOCs (Fig. 4a), followed by acetone (4.68%), chlorobenzene (2.10%), C2-C4 alkanes (0.99%-1.52%) and some aromatics (such as toluene, ethylbenzene and xylenes, 0.43%-0.87%). Similar species profile of VOCs was found from ceramic tiles manufacturing (Fig. 4b), in which acetone (8.16%), C2-C4 alkanes (2.60%-3.44%) and some aromatics (such as toluene, ethylbenzene and xylenes, 0.74%-1.78%) were also major VOCs species. However, the proportion of dodecane (5.80%) and 2-propanol (4.73%) in profile of ceramic tiles manufacturing were higher than that in mattress manufacturing, and proportion of CS_2 in the profile of ceramic tiles manufacturing was only 1.48%. Interestingly, α-pinene was found to be the most abundant species in the profile of filing cabinet manufacturing (Fig. 4c) and solid wood furniture manufacturing (Fig. 4d), with proportion of 27.79% and 24.42% respectively. It had been reported that terpenes was the most abundant group of total VOCs emission from solid wood furnishings (Czajka et al., 2020) and air-dried wood itself could be release α-pinene (Risholm-Sundman et al., 1998; Manninen et al., 2002). Therefore, α-pinene could be used as a tracer for manufacturing of wood-based furniture. In addition, dichloromethane (11.20%), acetone (4.04%) and 2-propanol (3.32%) were also major species in filing cabinet manufacturing, which had been proved that these species were often used as solvents for painting on filing cabinets or metal cleaning (Zheng et al., 2013; USEPA, 2016). Moreover, xylenes, ethylbenzene, n-propylbenzene and ethyl acetate were major species in solid wood furniture manufacturing, which were mainly released from the process of surface coating (Zheng et al., 2013; Zhong et al., 2017) mention at section 2.1.1.

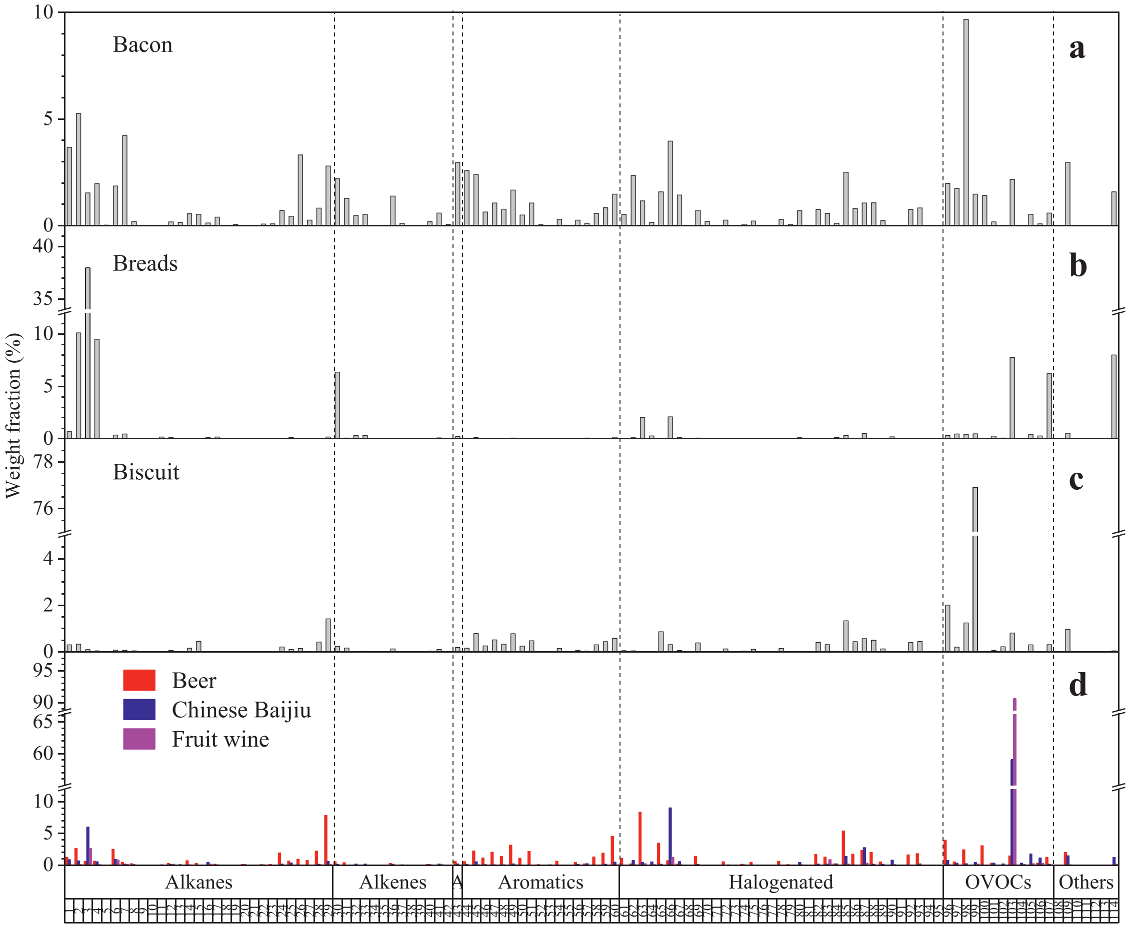

Fig. 5 – Species profiles of VOCs released from the production of Bacon (a), Breads (b), Biscuit (c) and Wine (Beer, Chinese Baijiu and Fruit wine) (d).

2.2.7. Food production

The previous studies on species profiles of VOCs from food production of vinegar, brewery, pickles and instant noodle found that OVOCs was the most abundant VOCs group in these sources, with proportion of 69.6%-98.9% (Gao et al., 2019). In this study, the species profiles of VOCs from food production of bacon, bread, biscuit and different types of wine were presented. As shown in Fig. 5a, acetone (9.68%) was the most abundant species in bacon production, followed by ethane, propane, pentane, nonane, dodecane, ethene, acetylene, benzene, toluene and dichloromethane with proportion of 2%-5%. While propane (10.11%), butane (37.95%), iso-butane (9.51%), ethene (6.36%), ethyl acetate (7.76%), tetrahydrofuran (6.21%) and CS_2 (7.99%) were major species in VOCs emission from breads baking (Fig. 5b). And 2-butanone was the most abundant species in the profile of biscuits making with proportion of 76.90% of total VOCs (Fig. 5c). As shown in Fig. 5d, ethyl acetate was the dominant species in Chinese Baijiu and fruit wine production, accounting for 59.06% and 90.61%, respectively. It had been reported that esters were formed during fermentation (Gao et al., 2019), which was the main process in winemaking. While dodecane, naphthalene, Freon 11 and 2-propanol were major species in VOCs emission from a workshop of beer production. The filling, fermentation and cooling equipment of the beer production was being cleaned when sampling, resulting in the significantly different among species profiles of VOCs from beer production, Chinese Baijiu and fruit wine production.

2.3. The species profile of VOCs in tunnels

Species profiles of VOCs from urban road tunnel and highway tunnel obtained in this study are shown in Fig. 6. The two species profiles were basically the same. Alkanes was the most abundant VOCs group with proportion of 36% in the two tunnels, followed by halogenated, aromatics, OVOCs, alkenes and others. Propane, butane, pentane, ethane, isobutane, benzene, toluene, dichloromethane and acetone were the major species. The difference between the two source profiles was that the proportion of propane and ethyl acetate were slightly higher in highway tunnel (8.71% and 3.08%) than that in urban road tunnel (5.89% and 1.76%). Based on our observation during the sampling period, the proportion of trucks in the highway tunnel (23%) was higher than that in the urban road tunnel (3%), which should be responsible for the difference between the two source profiles. Previous studies reported that propane, pentane, most species of aromatics and acetone were major species in VOCs emission from gasoline

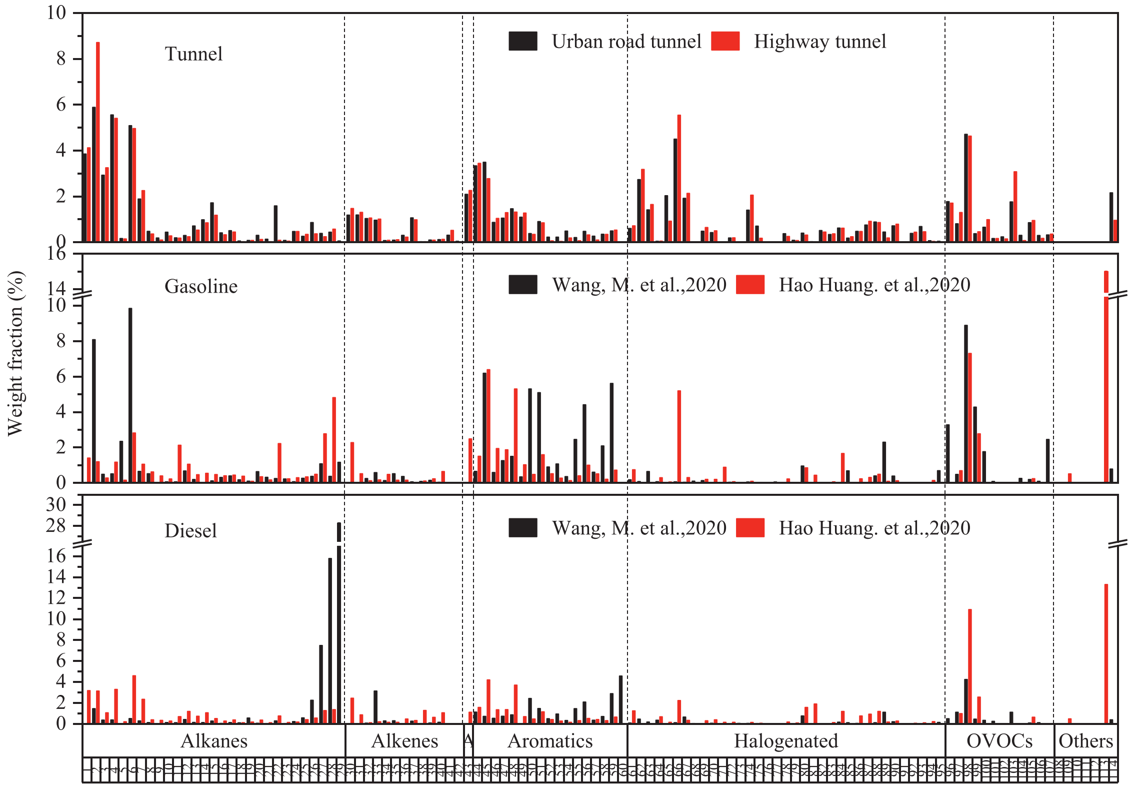

Fig. 6 – Species profiles of VOCs in tunnels and in gasoline and diesel vehicle emission.

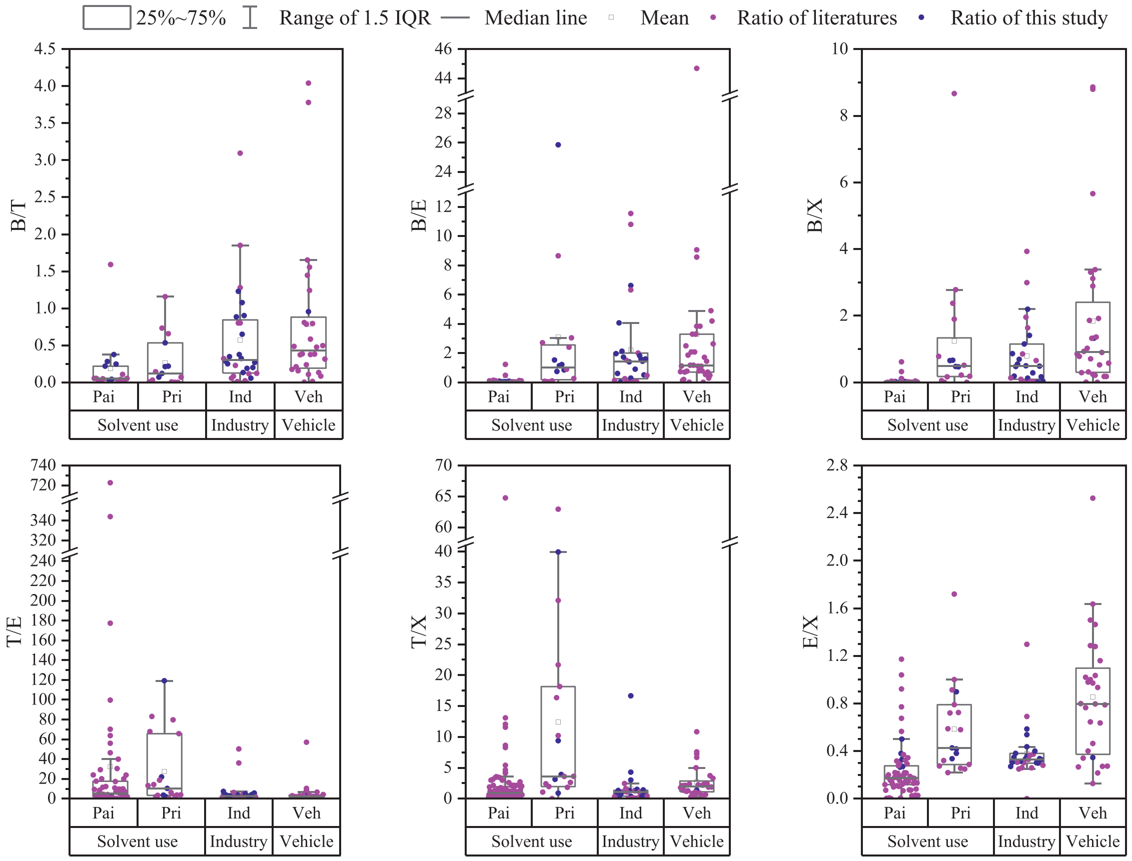

Fig. 7 – The ratio of B/T, B/E, B/X, T/E, T/X and E/X for different emission sources.

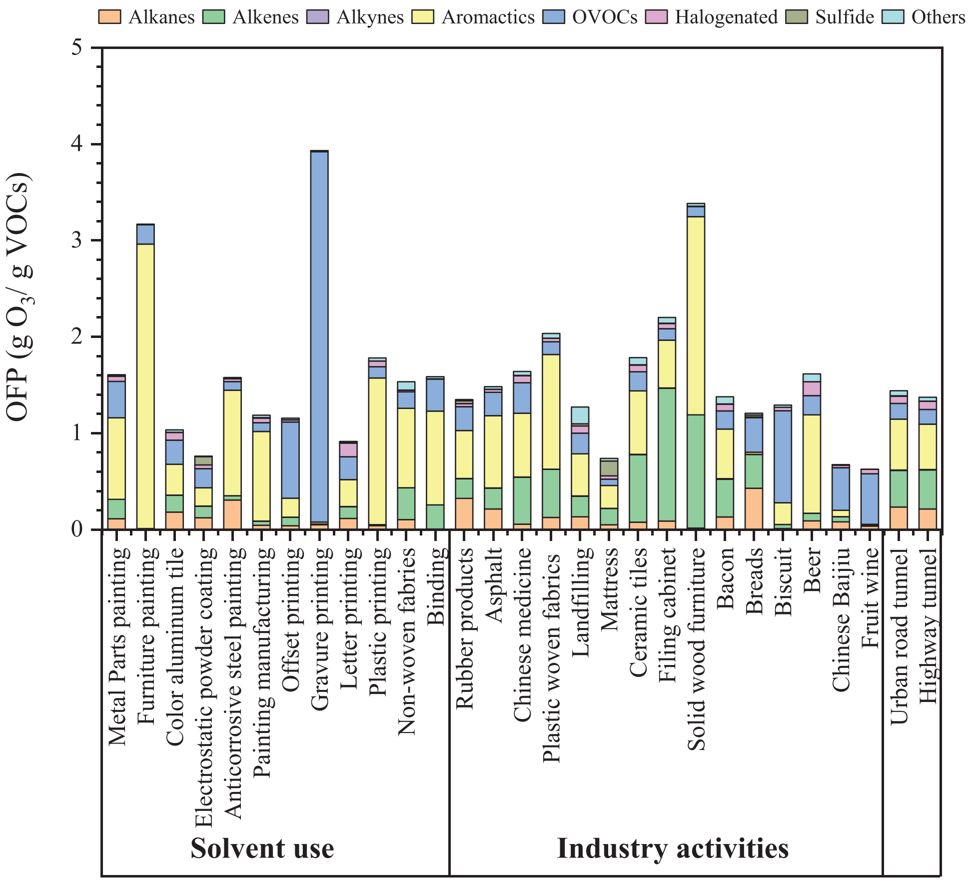

Fig. 8 – Comparison of the ozone formation potential (OFP) of per unit mass VOCs emission from the sources in this study.

vehicle, while high proportion of decane, undecane and dodecane were found in profile of VOCs emission from diesel vehicle (Fig. 6) (Huang et al., 2020; Wang et al., 2020).

3. Discussion

3.1. *The ratio of BTEX species*

The species which were unique or presented in high proportion in a certain emission source could be selected as characteristics species, and these characteristics species could be as the tracers for the source (Liu et al., 2008; He et al., 2015). According to the results of species profiles of VOCs from varieties sectors in this study, the potential tracers for each emission sources were listed in Appendix A Table S4. The potential tracers for each VOCs emission source will help to identify the attribution of ambient VOCs sources. However, some VOCs species are emitted from more than one source, and the overlapping emission sources of the species result in uncertainty in ambient VOCs sources attribution. The special ratio between VOCs species is a possible diagnose approach to distinguish different emission source (Liu et al., 2008). Previous studies reported that the ratio of benzene to toluene (B/T) was around 0.5 (wt/wt) in vehicular emissions and more than 1.0 in biomass combustion emission (Barletta et al., 2005; Tan et al., 2012), while it was 0.01-0.3 in solvent use emission (He et al., 2015). The paired ratios of BTEX (benzene, toluene, ethylene and xylenes) for each source were analyzed using the data from this work and previous reported and the results are shown in Fig. 7. Detail ratios of each source profile were given in Appendix A Table S5. It was found that B/T ratio in most painting and printing sources were ranged from 0.02 to 0.37, and in vehicle-related sources the ratio was ranged from 0.3 to 1.65 and in industry activities the ratio was 0.05-3.09. Previous studies showed that the B/T ratio of industrial processes involving solvent use was less than 0.2, because toluene was used as a solvent in many industries (Barletta et al., 2008). Thus, specific B/T ratio of less than 0.3 could be as the indicator for tracing the solvent use related sources. While industry activities and vehicle-related sources overlapped with each other and hardly distinguish them well in detail. In addition, in most printing processes, the ratio of T/X (>2) was higher than that of painting processes (<2). Although 2-propanol and ethyl acetate were the characteristic VOCs in printing processes, T/X ratio could be used as an indicator to distinguish painting and printing sources in absence of observed OVOCs. There were more overlaps for ratio of B/E, B/X, T/E and E/X for sectors of solvent use, industry activities and vehicle emissions as well as sub-sectors of each sector, so they were not suitable as indicators of different sources.

3.2. Source reactivity

The reactivity of the various emission sources investigated in this work were compared by calculating the OFP due to per unit mass emission of VOCs from each source. As shown in Fig. 8, the OFP of VOCs emissions from gravure printing was the highest among the various sources, with value of 3.91 g O_3/g VOCs. While the OFP of VOCs emissions from fruit wine production was the lowest (0.63 g O_3/g VOCs), because ethyl acetate with low reactivities ($MIR = 0.54$) contributed the most to the total VOCs emission. The OFP for other sources were in the range of 0.67–3.38 g O_3/g VOCs. The key species of VOCs that contributed the most to OFP varied from different emission sources. Aromatics was the VOCs group contributed the most to reactivity for most painting processes and non-paper printing processes, especially in furniture painting and plastic printing, with contribution of more than 85% to total OFP and consisting with the results reported by Wang et al. (2014). While OVOCs was major group contributed the most to total OFP in processes of foods production and paper printing. The OFP of VOCs emissions from ceramic tiles and filing cabinet manufacture were dominant by alkenes and from vehicle emissions in tunnels and other manufactures were dominant by both aromatics and alkenes. Toluene, xylenes, trimethylbenzene, 1,3-butadiene and 1-butene were the key reactive species commonly found in solvent use and industry activities and contributed significantly to total OFP in this study. However, terpenes were found to be the major contributors to total OFP in some sources. Limonene (11.51%) contributed the most to total OFP in landfilling and α-pinene could explain 56.94% and 32.54% of total OFP in filing cabinet and solid wood furniture manufacturing respectively. Terpenes were rarely been involved in previous studies of emission source profiles, indicating that the species of concern for emission sources are still not sufficient at present.

4. Summary

In this work we obtained the species profiles of VOCs released from varieties of sources, including solvent use of painting and printing, industry activities (such as rubber products, asphalt, plastic woven fabrics, Chinese medicine, furniture, several foods), landfilling and vehicle emission in urban road and highway tunnels by investigation. The data could represent a typical situation in capital cities in China. The characteristics species for each source were identified, such as xylenes, ethylbenzene, acetone and dichloromethane in painting, 2-propanol and ethyl acetate in printing, α-pinene in solid wood furniture manufacturing, as well as 2-butanone in biscuit baking etc. In the most of samples from the solvent use, the B/T ratio was less than 0.3, and the T/X ratio was less than 2 in most painting processes and more than 2 in most printing processes. Moreover, OFP due to per unit mass emission of VOCs from the various emission sources investigated in this work ranged from 0.63-3.91 g O_3/g VOCs, and the α-pinene could explain the 56.94% and 32.54% of total OFP of VOCs from the sources of filing cabinet and solid wood furniture manufacturing respectively, which was rarely been involved in previous studies of emission source profiles, indicating that the species of concern for emission sources are still not sufficient at present.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We gratefully acknowledge the Guiyang Research Academy of Eco-Environmental Science for partial support of this project.

Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2022.08.035.

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