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Improved emission factors and speciation to characterize VOC emissions in the printing industry in China

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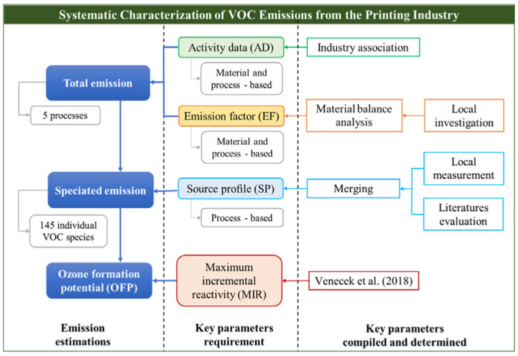
HIGHLIGHTS

- Raw material and process–based EFs were established through material balance analysis methods.
- Composite source profiles based on a variety of raw materials of the printing industry were compiled.
- Mass VOC emission for the period of 2010–2019, along with the speciated emission and the OFP in 2019 were estimated.
- Gravure printing and compound processes were key contributors for both VOC emissions and OFP.
- Toluene, ethyl acetate, 1,3-butadiene, isopentane, and 1-butene were the key species in terms of OFP.

GRAPHICAL ABSTRACT



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ABSTRACT

Printing industry is one of the most important sources of industrial volatile organic compound (VOC) emissions in China, and is thus a key industrial sector in terms of VOC control. However, process-based VOC emission and speciation from the printing industry have not been well identified, mainly owing to the poor emission factors (EFs) and diversity of source profiles. In this study, we systematically characterized process-based VOC emissions from the printing industry for the period of 2010–2019, through the establishment of improved emission factors and composite source profiles. VOC emissions from the printing industry were found to continuously increase from 2010 to 2018, reaching their maximum in 2018 at 939.8 Gg, but started to decrease afterwards. The substantial growth is driven by the large demand for ink and adhesive and the absence of effective control measures in the printing industry. The total VOC emissions and ozone formation potential (OFP) in China in 2019 were 916.1 Gg and 1834.5 Gg, respectively. Gravure printing and the compound process were the processes that contributed the most to both emissions and OFP. Rapidly developing provinces such as Guangdong, Jiangsu, and Zhejiang were the largest contributors to emissions. Oxygenated VOCs (OVOCs) accounted for most of the VOC emissions, followed by alkanes and aromatics, while aromatics were the dominant groups for total OFP, followed by alkenes/alkynes and OVOCs. Ethyl acetate, toluene, isopropanol, isopentane, and n-pentane were the top five VOC species in terms of emissions, while toluene, ethyl acetate, 1,3-butadiene, isopentane, and 1-butene were the top five species in terms of OFP. Scientific and precise control policy were proposed, involving four aspects: environmental access, emission standards, classification and management, and research on source substitution. We believe our study will provide an important reference for the systematic characterization and control policy of VOC emissions from other industries.

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1. Introduction

As a service industry supporting the national economy, the printing industry is important for regional economic development and people's livelihoods. Printed matter is used in the clothing, food, housing, and transportation sectors. According to the statistics provided by the Printing and Equipment Industry Association in China, in the period of 2010–2019, the average annual growth rate of printing industry value was 7.5 % in China. In 2019, the value of the printing industry was 1.3 trillion RMB, representing a year-on-year increase of 2.4 %. Organic raw materials such as inks, adhesives, and thinners generate large amounts of volatile organic compounds (VOCs) during the printing and compound processes (Song et al., 2022; Zhao et al., 2021; Wu et al., 2020; Zhang et al., 2020). VOCs are crucial precursors in the formation of ozone (O_3) and secondary organic aerosol (SOA) (Chen et al., 2020; Lewis et al., 2020; Yuan et al., 2013; Zhao et al., 2013). Effectively controlling VOC emissions is essential to alleviating O_3 and SOA pollution in China.

The use of volatile chemical products (VCPs)—including coatings, inks, adhesives, pesticides, cleaners, and personal care products, was found to be emerging as a major contributor of VOC emissions in the United States and Europe as transportation emissions have declined rapidly (McDonald et al., 2018; von Schneidemesser et al., 2016). In China, the latest results of anthropogenic VOC emission inventory from MEIC show that the use of VCPs has also become the largest source with the change of energy structure and the control of VOC emission from vehicles (Li et al., 2019). The use of VCPs was therefore highly focused by researchers for the emission inventory, chemical composition and environmental impact of VOCs, due to its increasing contribution to VOC emissions (Tanzer-Gruener et al., 2022; Coggon et al., 2021; Gkatzelis et al., 2021a, 2021b; Mo et al., 2021; Stockwell et al., 2021; Pearson, 2019). The printing industry, an important branch of the use of VCPs, also lead to substantial emissions of VOCs to the atmosphere. The printing industry was found to be one of the most important contributor (nearly 10 %) of the industrial VOC emissions in China, of which emissions was second only to that of industrial coating (27 %) (Liang et al., 2017, 2020). Therefore, it has thus always been a key area for national VOC emission control efforts in China. In recent years, the Chinese government issued a series of control policies and standard guidelines enabling the printing industry to carry out comprehensive rectification work (Ministry of Ecology and Environment (MEE), 2020a, 2020b, 2021, 2022), including the *Guidelines on feasible technologies for pollution prevention and control in the printing industry* (HJ 1089–2020) and *Emission standard of air pollutants for printing industry* (GB 41616–2022).

Great efforts have also been made to characterize VOC emissions from the printing industry for China, including emission factors (EFs), source profiles, and emission inventories. However, these studies had several limitations. First, existing emission factors lack applicability: the consumption of total organic raw materials was used to indicate the activity data when calculating most EFs (i.e., $\mathrm{VOC/C}_{\text{Total organic raw materials}}$) (Liang et al., 2019; Wang et al., 2018). However, these EFs were not suitable for macro-estimation of VOC emissions from the printing industry, as activity data for organic raw materials other than inks and adhesives are currently unavailable (e.g., thinners, and wetting fluids). Due to the inapplicability of the current EFs, researchers often use the VOC standard limit of ink and adhesive in China as the substitute (Mo et al., 2021; Li et al., 2019; Wu et al., 2016; Wu and Xie, 2017), which will make the estimation far from the actual one. Second, source profiles should be systematically improved: most source profiles have focused on the processes such as lithography and gravure printing (Liang et al., 2020; Wu et al., 2020; Yang et al., 2020; Zhang et al., 2020; Liu et al., 2019; Li et al., 2018; Shen et al., 2018; Xie et al., 2018; Zheng et al., 2013; Yuan et al., 2010). Important factors affecting the source profiles were rarely considered, including raw material type (e.g., solvent- or water-based), specific processes (e.g., compounding) and control techniques. Moreover, since the raw materials used in the printing industry are diverse, a large and varied source profile exists in the printing industry, indicating that the use of any single VOC source profile would probably lead to the omission of some key VOC species. But comprehensive source profiles for the entire printing industry have not been determined. Finally, present VOC emission inventories cannot represent the situation regarding VOC emissions from the printing industry: most previous studies estimated VOC emissions from the two printing processes of gravure and lithography, without considering compound process (Mo et al., 2021; Li et al., 2019; Sun et al., 2018; Wu and Xie, 2017; Wu et al., 2016). Additionally, almost all above inventories ignore the effects of diluents with VOC content approaching 100 % on VOC emissions, using only the VOC content of the ink or adhesive itself as the EFs. This is an important factor in terms of the total amount of VOCs emitted from the printing industry that should not be ignored. Furthermore, to the best of our knowledge, no studies that have systematically and comprehensively characterized VOC emissions from the printing industry; existing studies only considered source profile data or an emission inventory, for example. Therefore, the overall emissions of the printing industry remain poorly understood and it is difficult to provide technical support for pollution control.

The specific objectives of this work were to (1) develop raw material and process–based EFs through material balance analysis method, while considering the VOC emission from diluents and wetting fluids, (2) establish local source profiles based on process, raw material and terminal control technology, then evaluate available VOC source profiles and merge them to establish composite profiles, (3) compile mass VOC emissions inventories for the printing industry in China during 2010–2019, (4) and estimate the speciated VOC emissions and total OFP for the printing industry in 2019. Scientific and precise VOC control strategies for the printing industry in China were also discussed based on the above systematic characterization. On the road of deep emission reduction of VOCs in China, this study will not only provide a theoretical basis for the scientific and accurate emission reduction of VOCs from printing industry, but also provides a reference for the estimation and control of VOCs from other industries.

2. Methods and data

Fig. 1 shows the overall methodology framework designed for the systematic characterization of VOC emissions from the printing industry in this study. The total emission, speciated emission and OFP of VOCs from printing industry were estimated successively. To characterize the above emissions, activity data (AD), EFs and source profile were compiled and determined respectively, based on investigation and measurement. The specific methods of each characterization process are as follows:

2.1. Emission estimations

2.1.1. Total emission inventory

The VOC emissions from the printing industry in China for the period of 2010–2019 were estimated based on emission factor method. The estimation covered the emissions caused by the use of main organic raw materials in the printing industry such as inks, adhesives, thinners, and wetting fluids. The VOC emissions were calculated as follows:

$$E = A \times EF \times (1 - \zeta \times \eta \times \theta) \tag{1}$$

where E is the VOC emissions (unit: g); A is the activity data (unit: g), i.e., the consumption of ink or adhesive (Section 2.2); EF is the emission factor before treatment facility (unit: g VOCs g^{-1} ink or adhesive) (Section 2.3); ζ is the effective coverage ratio of treatment facilities (unit: %); The exhaust from printing industry need to be collected before entering treatment facilities, and the collection efficiency varies with different collection devices. η is the average collection efficiency of the exhaust gas (unit: %); and θ is the average removal efficiency of treatment facility for the collected exhaust gas (unit: %).

In the period of 2010–2017, ζ and θ were directly obtained from Mo et al. (2021). Mo et al. (2021) estimated the effective coverage ratio of control facilities and average removal efficiency of control facility for the industrial solvent sources during 2000–2017. The estimation were based

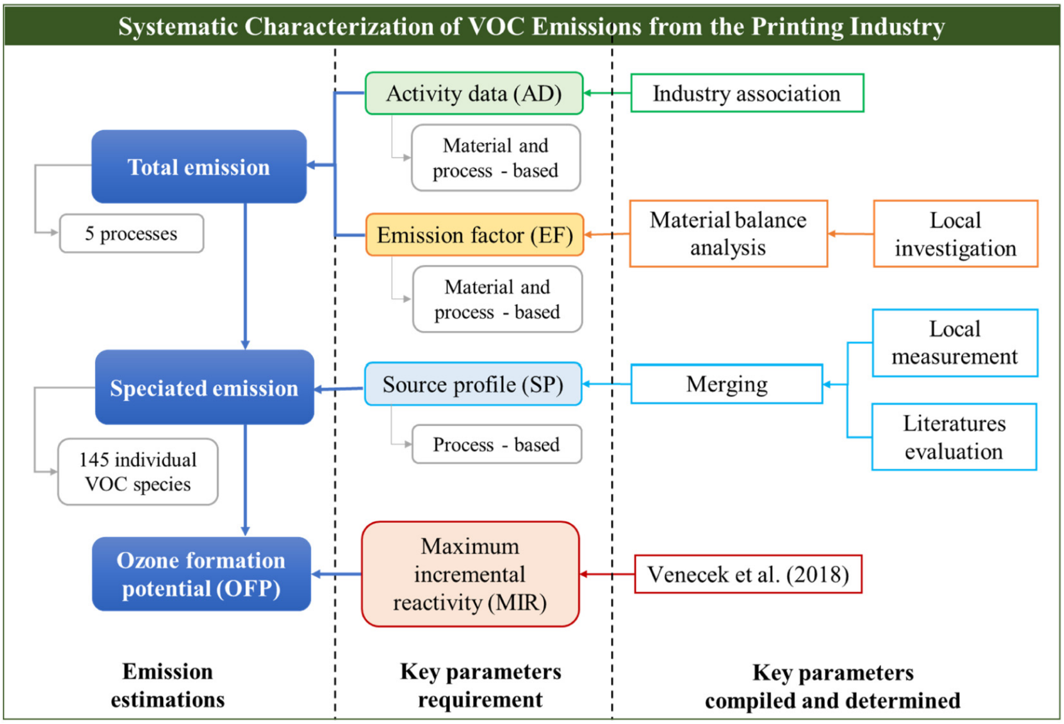

Fig. 1. The methodology framework for systematic characterization of VOC emissions from the printing industry.

on implemented policy, the production values for organic exhaust gas treatment devices, and market shares of VOC control techniques and their control efficiency. From 2017, China has released a series of VOC control measures in key industries such as the printing industry, emphasizing the installation and efficient operation and maintenance of treatment facilities. The effective coverage ratio of treatment facilities has already experienced a rapid growth before 2017 (Mo et al., 2021), therefore, we assumed that its growth rate in 2018–2019 was reduced from 10 % in 2015–2017 to 5 %. And due to the policy to implement more efficient operation and maintenance of treatment facilities, we also assumed that the annual growth of comprehensive governance efficiency will increase from 4.3 % in 2015–2017 to 6 % in 2018–2019 (Fig. 2). For η, the average collection efficiency (i.e., 20 %) of the exhaust from printing industry for the year of 2018 was obtained from our recent study (Liu et al., 2021). η for other year were estimated based on the value in 2018 and assumed growth rate. We divided the period of 2010–2019 into three stages with that reported by Mo et al. (2021), i.e., before 2013, 2014–2017, and 2018–2019, assuming slow (0.5 %), moderate (2 %), and fast (10 %) increase rates for the three stages, respectively (Fig. 2). Specific annual value of ζ, η and θ for printing industry during 2010–2019 are presented in Table S1.

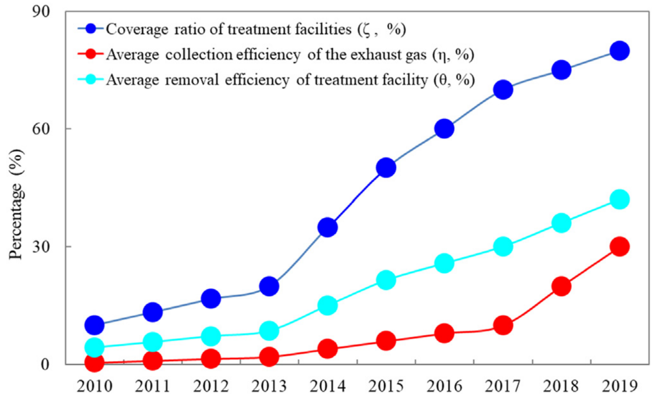

Fig. 2. Annual value of coverage ratio of treatment facilities, collection efficiency of the exhaust gas and removal efficiency of treatment facility for printing industry during 2010–2019.

2.1.2. Speciated emission inventory

The total VOC emissions from the printing industry in 2019 were then broken down for a given process into individual species to create a speciated VOC emissions inventory, as shown in Eq. (2):

$$E_{i,k} = \Sigma_k E_i \times R_{i,k} \tag{2}$$

where $E_{i,k}$ is the total VOC emissions of species k in process i; E_i is the total VOC emissions of process i; and $R_{i,k}$ is the weight percentage of species k to process i (Section 2.4.2).

2.1.3. Ozone formation potential

(1) The OFP emissions intensity (OFPEI)

The OFPEI can quantify the OFP emissions intensity and reactivity per unit emissions of each source (Liang et al., 2020). We calculated the OFPEI of each process as follows:

$$OFPEI_i = \Sigma_k R_{i,k} \times MIR_k \tag{3}$$

where $OFPEI_i$ is the OFP emissions intensity of process i and MIR_k is the maximum incremental reactivity of species k. The updated MIR values reported by Venecek et al. (2018) were used in this study.

(2) The OFP emissions inventory

An OFP-based VOC emissions inventory for the printing industry was compiled based on the speciated VOCs emissions inventory and MIR values. It was calculated as follows:

$$OFP_i = \Sigma_k E_{i,k} \times MIR_k \tag{4}$$

where OFP_i is the total OFP emissions of process i; and $E_{i,k}$ is the total VOC emissions of species k in process i.

2.2. Activity data (AD)

The apparent consumption (output + import - export) of raw material-based inks and adhesives were used to represent the activity data due to the lack of authoritative statistics, and were obtained from the National Ink Standardization Technical Committee, Printing and Equipment Industry Association and Adhesive and Adhesive Tape Industry Association in China. Fig. S1 shows the consumption of inks and adhesives in China from 2010 to 2019. China is the world's second largest ink producer and consumer. In 2019, China's ink consumption was 780 Gg, an increase of 32.4 % from the 589 Gg in 2010. With the improvement of living standards, the requirements for product packaging have gradually increased, and the printing market structure and demand have changed accordingly. The proportion of gravure printing inks has gradually increased, accounting for 42.5 % of all ink use in 2019 and exceeding the proportion for flat ink (Fig. S2 a). China also produces large amounts of adhesives; its annual output thereof accounts for about one-third of the global total. In 2019, the consumption of adhesives reached 8720 Gg, an increase of 92.5 % from the 4530 Gg in 2010. Downstream applications of adhesives in China are mainly in the fields of construction, packaging (label), and wood, which account for 28.7 %, 21.1 %, and 13.8 % of the total use in 2019, respectively (Fig. S2 b). Fig. S3 presents the market consumption structure of the above two types of solvent products used in printing industry. Lithographic inks are mainly environmentally friendly inks such as vegetable oil-based offset inks and radiation-curable inks, accounting for 90 %. Gravure inks are still mainly solvent-based, accounting for 80 %. Most of the flexographic printing inks have been water-based, with a water-based ratio of 60 %. Stencil printing inks and other types of inks account for a small proportion in the market, and their market consumption structure refers to flexographic inks. In the printing adhesive market, water-based adhesives used in wet compound process account for 60 % of the total consumption of compound adhesives, solvent-based adhesives used in dry compound process account for only 10 %, and the rest are other types of adhesives such as hot-melt type and solvent-free type.

2.3. Emission factors (EFs)

Unlike the EFs reported in previous studies, which were based on the consumption of the total organic raw materials (i.e., VOC/C $_{\text{Total organic raw materials}}$), we used inks or adhesives as the EF benchmark (i.e., VOC/C $_{\text{Ink or adhesive}}$), which are more suited to the establishment of regional and national emission inventory due to the lack of consumption data of thinners and wetting fluids. The EFs in this study were the total uncontrolled VOC emissions from inks, thinners, cleaning agents, adhesives, wetting fluids, and other raw materials, covering printing, drying, cleaning, dampening, compounding, and other discharge-producing processes. We determined the EFs based on field investigations and the widely used material balance analysis technique (Liang et al., 2019). A total of 14 typical printing enterprises including 20 process objects were selected, covering six specific processes and 10 types of raw material (Table 1). Two to four process objects were considered for the main processes of the printing market in China, including gravure printing, lithography, and dry and wet compound processes. For letterpress and stencil printing, which have a small market share, only one object was selected for each process. In addition, solvent-free and co-extrusion compound process were not considered to be limited by the field investigation conditions. The EFs were calculated based on a material balance analysis using Eq. (5):

$$EF_{i,j} = \frac{\Sigma_j E_{i,j}}{\sum C_{i,Ink\ or\ Adhesive}} = \frac{\Sigma_j C_{i,j} \times WT_{i,j}}{\sum C_{i,Ink\ or\ Adhesive}} \tag{5}$$

where $EF_{i,j}$ represents the emission factor of process i based on raw material j; $E_{i,j}$ is the VOC emission from process i based on raw material j; $C_{i,\ Ink\ or\ Adhesive}$ represents the annual consumption of ink or adhesive in process i, after taking into account the solvent recovery; $C_{i,j}$ is the annual consumption of raw materials j in process i; and $WT_{i,j}$ is the VOC content of raw material j in process i, obtained from the average VOC content in the material safety data sheet (MSDS) provided by the enterprise. Detailed example of the material balance analysis for solvent-based gravure printing was shown in the Table S2.

2.4. Source profiles

2.4.1. Measurement of source profiles

There is a lack of chemical profiles of VOC emissions for some key processes and raw materials in the printing industry in China due to the large variety of production processes. After a preliminary investigation, five representative enterprises were selected to measure the chemical composition of VOC emissions from the printing industry. According to the emission status of the printing industry, the production process, and the characteristics of production and discharge, the following basic principles were followed when selecting the enterprises for measurement. In terms of the production process, sub-sources that made the largest contributions to VOC emissions from the printing industry were selected, including compound processes, gravure printing, and lithography; in terms of enterprise scale, three different scales were covered: large, medium, and small; and in terms of raw materials and the end treatment, both solvent-based and water-based raw material types were considered, and the end treatment typically used, i.e., absorption, has a relatively large market share among industrial treatment technologies. Fugitive and stack emission samples were collected from 13 raw material-based processes. Specific sampling information for

Table 1

Emission factors of VOC for the printing industry.

Processes		Raw materials		Number of process objects	Emission factors (g VOCs g^{-1} ink or adhesive)		
					This study		Guideline range (Ministry of Ecology and Environment (MEE), 2020b)
					Average value	Grade	
Printing	Gravure printing	Solvent-based ink		4	1.57	II	1.50–2.00
		Water-based ink		2	0.22	III	0.10–0.30
	Lithography	Radiation curable ink/vegetable oil-based lithography ink	Traditional wetting fluid	4	0.52	II	0.50–0.80
			No/low alcohol wetting fluid	2	0.12	III	0.05–0.30
	Letterpress printing	Solvent-based ink		1	1.55	III	1.00–1.20
		Water-based ink		1	0.17	III	0.05–0.30
	Stencil printing	Solvent based ink		1	0.75	III	0.60–1.00
		UV ink		1	0.11	III	0.05–0.10
Compounding	Dry compounding	Solvent-based adhesive		2	1.65	III	1.00–1.20
	Wet compounding	Water-based adhesive		2	0.10	III	0.03–0.05
	Solvent-free compounding	Solvent free adhesive		0	0.01	IV	≤0.01
	Co-extrusion compounding	Hot melt adhesive		0	0.01	IV	≤0.01

the measured enterprises is presented in Table 2, including the product type, scale, raw materials used, end treatment technique, and sampling location. To the best of our knowledge, almost no domestic VOC speciation measurements have been conducted for most of these raw materials and end treatment-based key processes; thus, our study was expected to improve existing domestic source profiles for the printing industry. Stack and fugitive emissions samples from each source were collected in 3.2 L Summa canisters, which were obtained under normal operating conditions. For stack emission samples, we connected a Teflon tube to the canister; the other end of the tube was equipped with a salinized filter head to remove particulate matter and moisture. The filter head was extended into the center of the flue, away from the vortex area, during sampling. Under a controlled flow, the sampling time was roughly 10 min, i.e., the time taken for the pressure in the canister to become ambient. For fugitive emission samples, the canister was placed near the production equipment during printing and compounding, and each sample was collected over approximately 8 min with a flow-limiting valve. Repeated sampling was conducted for each process to reduce error. All samples were delivered to the laboratory within 1 week for analysis. The VOC samples were analyzed based on the United States Environmental Protection Agency (USEPA) TO-15 method using gas chromatography–mass spectrometry (GC–MS; 7890A/5975C; Agilent, USA). The VOC species were identified using the Photochemical Assessment Monitoring Stations (PAMS) network and TO-15 standard mixtures. Detailed descriptions for the VOCs analysis system and quality control are available in our previous study (Liang et al., 2020).

2.4.2. Evaluation and merging of source profiles

Due to the diversity of raw materials in the printing industry, the VOC source profiles measured in this and previous studies were collected, systematically evaluated and merged to establish a composite source profile for the printing industry in China. We assessed the quality of the source profiles, including in terms of their authoritativeness, representativeness, completeness, and accessibility. Source authoritativeness refers to the quality of journals where the source profiles were published. Source profiles in low-quality publications were not accepted. Representativeness refers to whether the source profiles represent the current emission characteristics of VOCs in the printing industry, including the timeliness, sampling objects, and quantity. Source profiles from 10 years ago, and those derived from an unknown printing process with a small number of samples, were not considered. Completeness refers to whether the main contributors to the source profiles were characterized [e.g., whether OVOCs, which are currently important VOCs in the printing industry, were omitted]. If they were omitted, the data were not considered (Yuan et al., 2010).

After the evaluation, the selected source profiles were fused into one composite profile to represent a certain process. The source profiles in this study based on both processes and raw materials were first combined into a source profile for processes, based on the market share of raw materials and EFs, using Eq. (6). The source profiles of the same processes were then combined to yield a composite profile representing the sector (using Eq. (7), which was reported in Sha et al., 2021). Source profiles of orifice and other printing methods was not considered due to its small market share and lack of local research, and composite chemical components combining gravure, lithography, and letterpress printing were selected for the subsequent establishment of a speciated emissions inventory.

$$SP_{p_i} = \Sigma_j SP_{p\&m_{i,j}} \times \frac{W_{p\&m_{i,j}} \times EF_{i,j}}{\Sigma_j W_{p\&m_{i,j}} \times EF_{i,j}} \tag{6}$$

where i represents the processes of the printing industry, j represents the raw material type, $SP_{p\ i,}$ is the source profile of process i, $SP_{p\&mi,j}$ is the source profile of process i based on raw material type j, $W_{p\&mi,j}$ is the market share of process i based on raw material type j (obtained from the National Ink Standardization Technical Committee and Adhesive and Adhesive Tape Industry Association in China), and $EF_{i,j}$ is the emissions factor of process i based on raw material type j (estimated in Section 2.3).

$$R_{i,k} = \frac{\Sigma_n R_{i,k,n}}{N} \tag{7}$$

where k represents the species; n is the number of source profiles to be merged; $R_{\ i,k}$ is the ratio of species k in process i; $R_{i,k,n}$ is the ratio of species k in process i of the source profile n; and N is the total the number of profiles adopted to develop the composite profile.

3. Results

3.1. Emission factors

The EFs of the six processes and 10 types of raw material were calculated for the VOCs emitted from printing, and are given in Table 1. Generally, the EFs of processes based on solvent-based raw materials were larger than those for processes based on water-based and UV-based raw materials, indicating that source substitution of no- or low-VOC-based raw materials can substantially reduce the emission of VOCs for each process. This phenomenon was also consistent with results reported in recent studies conducted in China (Wang et al., 2018; Liang et al., 2019). This was due to the large VOC content of solvent-based raw materials, as well as the large amounts of diluent used in these processes. Therefore, processes and enterprises using solvent-based raw materials tend to be the focus of VOC emission management and control. Substitution with low-VOC raw materials can effectively reduce VOC emissions from printing. The EFs for both solvent-based and water-based raw materials from gravure and letterpress

Table 2
Details of the enterprises studied and sampling locations.

Enterprise code	Product type	Enterprise scale	Production process	Raw material	Exhaust treatment techniques	Sample code	Sampling location	Sample type
A	Composite film packaging	Large	Gravure printing/ Compound process	Solvent-based ink/solvent-based adhesive	Gravure printing process: rotary concentration + oxidation incineration. Compound process: activated carbon adsorption + UV photolysis / adsorption concentration + condensation recovery	S1 S2 S3 S4	Gravure printing Gravure printing Compound process Compound process	Fugitive emissions Stack emissions Fugitive emissions Stack emissions
B	Cigarette packaging	Large	Gravure printing/ Compound process	Water-based ink/ alcoholic ink /water-based adhesive	Honeycomb activated carbon adsorption	S5 S6 S7	Gravure printing Gravure printing Compound process	Fugitive emissions Stack emissions Fugitive emissions
C	Cartons, cartons	Medium	Lithography	Plant-based inks/water-based inks	UV photolysis + granular activated carbon adsorption	S8 S9	Lithography Lithography	Fugitive emissions Stack emissions
D	Cartons, cartons	Medium	Lithography	Solvent-based ink	UV photolysis + granular activated carbon adsorption	S10 S11	Lithography Lithography	Fugitive emissions Stack emissions
E	Carton	Small	Lithography	Plant-based inks/solvent-based adhesives	Granular activated carbon adsorption	S12 S13	Lithography Compound process	Stack emissions Fugitive emissions

printing were similar, with values of 1.57 g VOCs g^{-1} ink and 1.55 g VOCs g^{-1} ink for their solvent-based materials, and 0.18 g VOCs g^{-1} ink and 0.17 g VOCs g^{-1} ink for the water-based materials, respectively. Although the VOC content of lithographic ink (i.e., radiation curable ink/vegetable oil-based lithography ink) is extremely low, the use of wetting fluid in the printing process still results in large VOC emissions. The EFs obtained when using traditional wetting fluid in lithography were about four times larger than those obtained when using no/low alcohol wetting fluid. The EFs of solvent-based and low-VOC-based raw materials for stencil printing were smaller than those for the other printing processes, except lithography. This was consistent with the conclusions of Liang et al. (2019). For compound processes, the EF of dry compounding based on solvent-based adhesives was about 15 times that of wet compounding based on water-based adhesives, indicating large VOC emissions from the dry compound process.

Due to the inapplicability of the current EFs to available activity data, the VOC limits of inks and adhesives in the standards in China were usually used by researchers to estimate the VOC emissions (Mo et al., 2021; Li et al., 2019; Wu et al., 2016; Wu and Xie, 2017). We compared the EFs with the VOC limits in the standards in China, i.e., GB 38507–2020 (National Public Service Platform for Standards Information (NPSPSI), 2020a) for inks and GB 33372–2020 (National Public Service Platform for Standards Information (NPSPSI), 2020b) for adhesives. As shown in Fig. 3 and Table S3, The VOC EFs of each process in this study, covering other organic raw and auxiliary materials such as diluents, were somewhat different from the VOC limit in the standards. The EFs for lithography using traditional wetting fluid, gravure printing using solvent-based ink, letterpress printing using solvent-based ink and dry compounding using solvent-based adhesives, were about 2–5 times larger than the corresponding VOC limits of the standards. The main reason was that the VOC content of ink or adhesive in the standards is in factory state (ink or adhesive itself), not in the ready-to-use state (adding diluent or wetting solution according to actual use requirements). In other words, the VOC limit in the standards does not take into account VOC emissions from the use of diluents and wetting fluids in the printing industry. This indicates that the VOC emissions in these processes would be seriously underestimated if the VOC limits in the standard were used to estimate the emissions. The EFs of other processes were close to the standard limit, and would not bring a large gap to the emission estimation. The EFs were also compared with those from the "*Guideline on available techniques of pollution prevention and control for printing industry*" (HJ 1089–2020) (Ministry of Ecology and Environment (MEE), 2020b). As shown in Table 1, the EFs for almost all processes and raw materials were within or near the thresholds of the guidelines. The difference between the EFs in this study and the guidelines was mainly due to differences in the survey targets and raw materials used. The EFs for solvent-free and co-extrusion compound processes were not considered in this study due to the limitations of the field investigation conditions, and the maximum value of the corresponding EFs of the guidelines was selected for subsequent emission inventory estimates.

According to the accuracy and reliability of the emission factors, five grades for uncertainty in emission factor for VOC emission for China were established by Wei et al. (2011). Based on the evaluation system (Wei et al., 2011) and the characteristics of VOC emission from the printing industry, the emission factors of this study could be classified into three grades (Table 1): II (with process objects >3), III (with process objects <3) and IV (guideline limits). The uncertainties of the emission factors for the three grade were ± 80 %, ± 150 % and ± 300 %, respectively. Due to the limited sample size, the EFs in this study may not represent the reality of VOC emissions from the printing industry exactly. But through a more comprehensive investigation on the use of raw materials in different process from the enterprises and the consideration of the neglected VOC emissions from diluent, along with a thorough literature review, we believe we have improved its reliability.

3.2. Source profiles

3.2.1. Measured source profiles

The VOC source profiles of fugitive and stack emissions for 13 key discharge processes (S1 ~ S13 in Table 2) were obtained from local measurements. Figs. 4 and 5 show the chemical compositions of emissions from the gravure printing and compound processes, and lithography and compound processes, respectively. As shown in Fig. 4, except for the water-based compound process (S7), OVOCs were the dominant chemical group in fugitive and stack emissions from all processes, with a mass

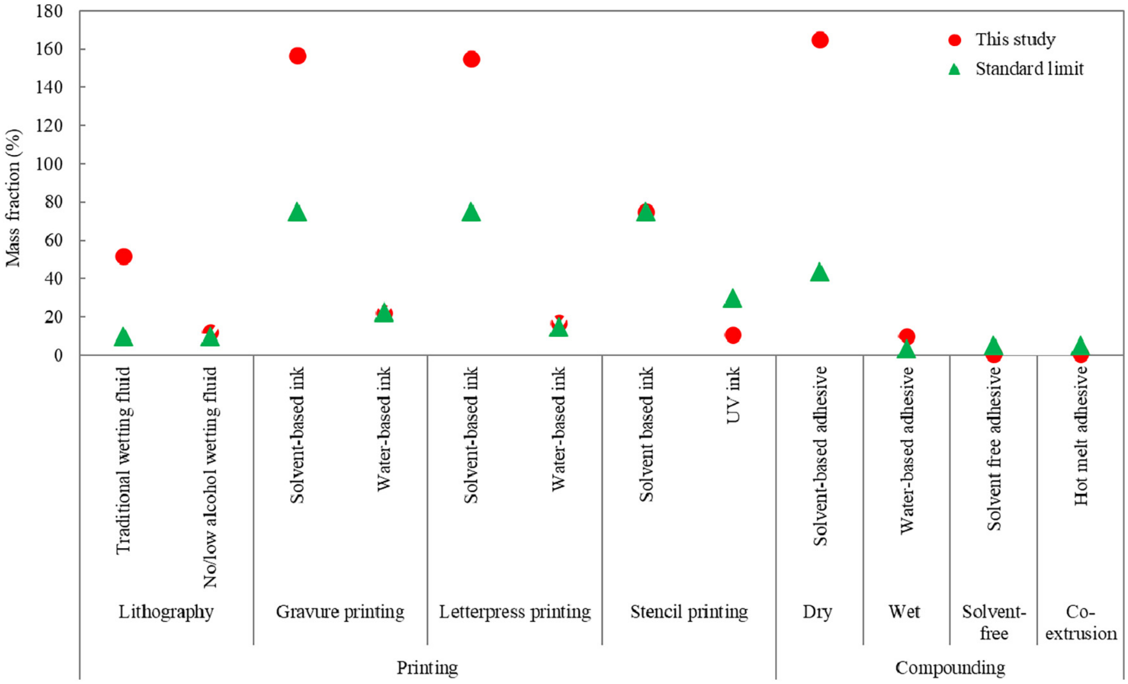

Fig. 3. Comparison of improved printing industry emission factors in this study and the standard limits in China.

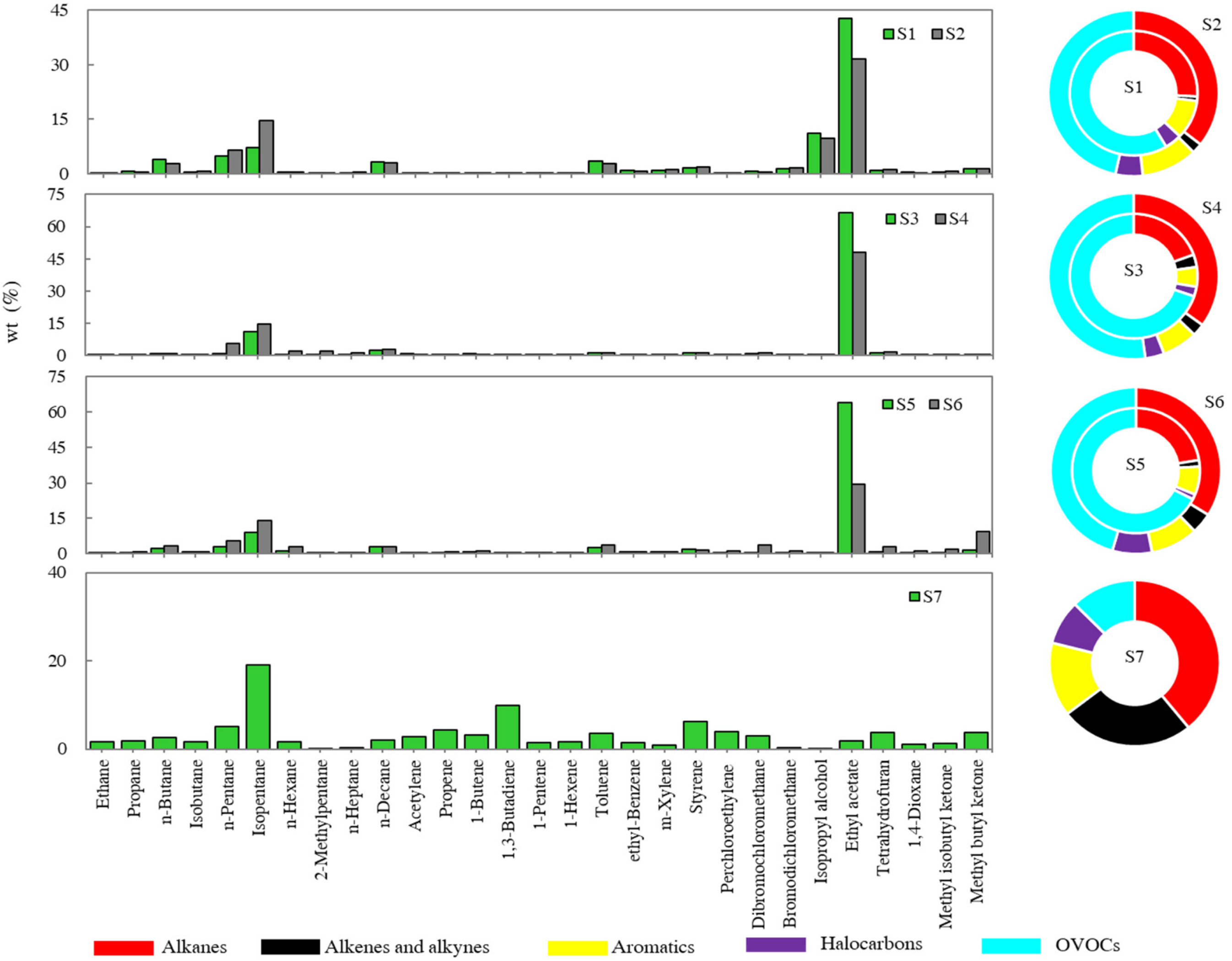

Fig. 4. The VOC composition and chemical groups of the gravure printing and compound processes. S1 and S2 are fugitive and stack emissions for solvent-based gravure printing, respectively; S3 and S4 are fugitive and stack emissions for solvent-based compound process, respectively; S5 and S6 are fugitive and stack emissions for water-based gravure printing, respectively; S7 is fugitive emissions for water-based compound process.

fraction range of 45.5–69.8 %. The characteristics of the VOC chemical groups changed after passing through the pollution collection and end treatment system. Compared to fugitive emissions, the proportion of OVOCs was lower in stack emissions, while the proportion of alkanes was higher. Although the proportion of chemical groups had changed after end treatment, OVOCs and alkanes still were the main chemical groups in stack emissions. For specific VOC species, fugitive emissions from solvent-based printing and compound process seem similar in spite of having different processes, and the same happen with stack emissions. Ethyl acetate, a raw material and thinner commonly used in solvent-based inks and adhesives, was the most common VOC species from the above two processes. It accounted for 43–66 % and 32–48 % of the total mass of VOCs in fugitive and stack emissions, respectively. Ethyl acetate, one of the common constituents of some alcoholic inks and diluents, was also the main VOCs species in the measured water-based gravure printing process. The proportion of ethyl acetate in the emissions decreased after the treatment system (Table 2) from the above three processes, but was still the dominant VOC species. The water-based compound process was very different from the other processes, with emissions comprised of alkanes (39.1 %), alkenes and alkynes (25.4 %), aromatics (14.4 %), OVOCs (12.4 %), and halocarbons (8.6 %). Isopentane, 1,3-butadiene, and styrene were the three most commonly occurring species, together accounting for 35.3 % of all VOCs. The species characteristics were closely related to the composition of the raw materials used in the process (e.g., the large amounts of 1,3-butadiene and styrene were due to the use of water-based styrene-butadiene polymer adhesives). Note that ethanol, as key raw material and thinner of the water-based or alcoholic ink in gravure printing process, was not measured in this work because of limitations of our testing methods.

Fig. 5 shows that the VOC source profiles of lithography varied substantially among the different types of raw materials. For plant- and water-based inks (i.e., S8), alkanes were the dominant components, accounting for 75.0 % of all VOCs. Among them, isopentane, n-pentane, methyl cyclohexane, and 2,2,4-trimethyl pentane were the most abundant species, accounting for 16.2 %, 8.4 %, 5.2 % and 5.1 % of the total emissions, respectively. The source profiles (i.e., S9) after UV photolysis + granular activated carbon adsorption treatment changed only slightly in terms of chemical groups and species characteristics. For solvent-based ink (i.e., S10), OVOCs were the dominant chemical group, accounting for 78.7 % of all VOCs. As the main diluent of ink, isopropyl alcohol was the most abundant species in OVOCs, accounting for 78.1 % of all VOCs. In the source profiles (i.e., S11) obtained after applying the UV photolysis + granular activated carbon adsorption system, the proportion of OVOCs decreased, while that of the alkanes increased. The OVOCs and isopropyl alcohol remained the dominant chemical group and key species,

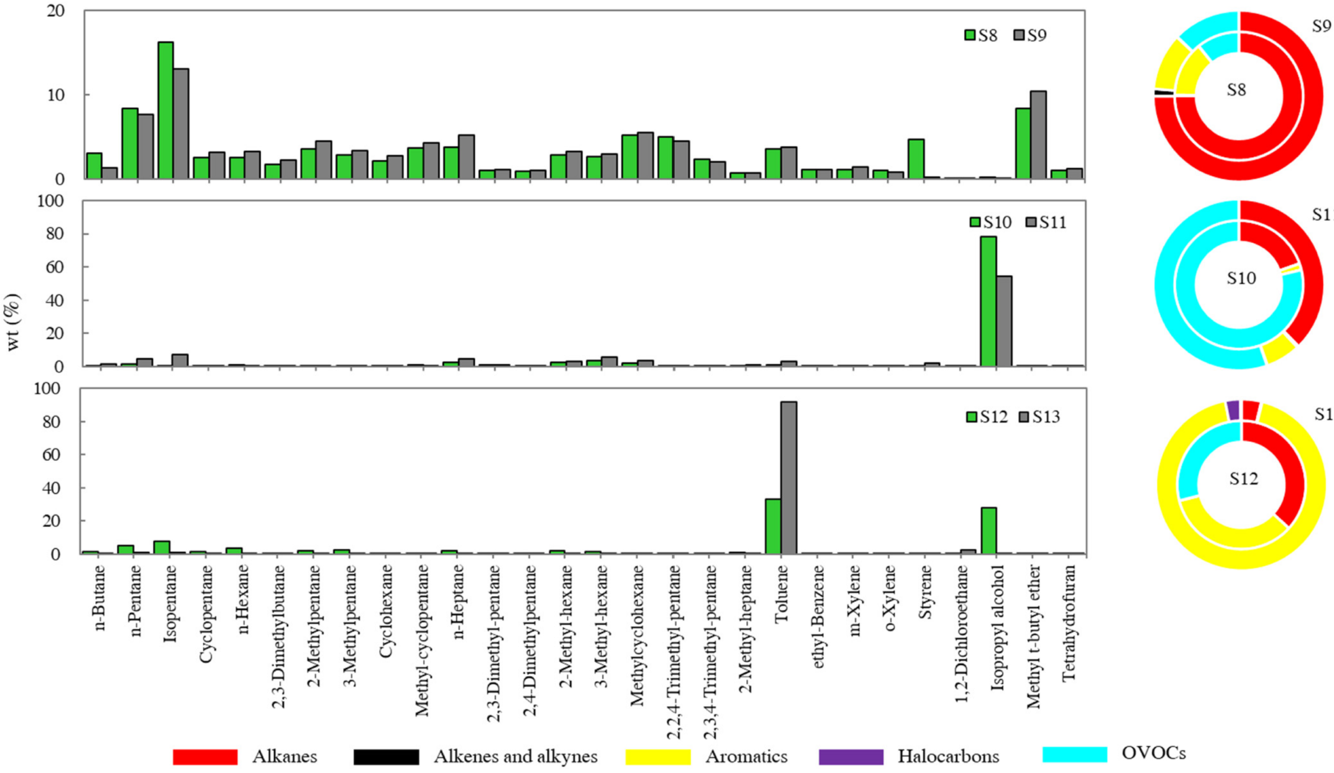

Fig. 5. The VOC composition and chemical groups of the lithography and compound processes. S8 and S9 are fugitive and stack emissions for eco-friendly lithography, respectively; S10 and S11 are fugitive and stack emissions for solvent-based lithography, respectively; S12 is stack emissions for plant-based lithography and S13 is fugitive emissions for solvent-based compound process.

accounting for 55.3 % and 54.6 % of all VOCs, respectively. For plant-based inks (i.e., S12), the contributions of alkanes, aromatic hydrocarbons, and OVOCs were similar, accounting for 36.8 %, 34.2 %, and 28.9 % of the total, respectively. The alkanes were mainly derived from the plant-based ink itself, and the transformation of other substances after treatment. Aromatics and OVOCs mainly originated from varnishes used after printing, with toluene and isopropanol used as diluents. Aromatics were the main VOCs from the compound process (i.e., S13), accounting for 93.0 % of all VOCs. Toluene acts as a diluent for solvent-based adhesives and accounted for 92.0 % of all VOCs.

3.2.2. Composite source profiles

In order to improve the representation of the source profile of the entire printing industry, a VOC source profile dataset (Table S4) for the printing industry in China was also established by systematically evaluating and integrating our measurements and currently available source profiles. The dataset covered the four major processes of the printing industry (i.e., gravure printing, lithography, letterpress printing, and the compound process) and 145 individual VOC species. Five basic source profiles of gravure printing, including our measurements, were merged to derive composite source profiles, while six source profiles were used for lithography, three for letterpress printing, and only one (i.e., our measurement) for the compound process (due to the lack of a corresponding chemical profile). The VOC chemical groups and top three species of each source profile in the four processes are presented in Figs. S4 and S5.

As shown in Fig. S4, OVOCs accounted for most VOCs from gravure printing according to both our measurements and the available source profiles, accounting for 36–86 % of all VOCs, and were also dominant in the composite profiles, contributing 59.9 % to all VOCs. For lithography, our measurements, and those of Shen et al. (2018), showed that alkanes were dominant, while Xie et al. (2018) and Zhang et al. (2020) reported that OVOCs were most important. In the studies of Zheng et al. (2013) and Liang et al. (2020), alkanes and OVOCs contributed similar amounts. The composite source profiles showed that OVOCs and alkanes were the major VOC groups, accounting for 43.3 % and 34.6 % of all VOCs, respectively. For letterpress printing, aromatics were dominant in the studies of Zheng et al. (2013) and Wu et al. (2020), while Xie et al. (2018) reported that OVOCs were thus the major chemical group. Therefore, aromatics (36.9 %) and OVOCs (33.5 %) were the dominant VOC groups in the composite source profiles. The source components of the compound process were mainly based on our measurements due to a lack of available studies, with OVOCs (31.8 %) being the dominant chemical group followed by aromatics and alkanes (29.3 % and 24.7 %, respectively).

As shown in Fig. S5, ethyl acetate and isopropyl alcohol, which are often used as thinners for gravure inks, were the top two species for gravure printing. Isopentane was another important species. The above three species accounted for 56.1 % of the total VOCs emitted from composite gravure printing. Isopropanol, ethanol and ethyl acetate were the top three species in the composite source profiles of lithography, responsible for 37.1 % of the total VOC emissions. For letterpress printing, toluene, ethyl benzene and isopropyl alcohol were the three main VOC species in the composite chemical profiles. The top three species in the composite compound process (isopentane, 1,3-butadiene, and styrene) were determined directly from our measurements, and accounted for 35.3 % of all VOCs. The differences in the top three species among the different source profiles of each process were mainly related to the raw materials, processes, and end treatment techniques used by the surveyed enterprises. Our study systematically incorporated source profiles to ensure a good representation of the key VOC species emitted from printing processes.

3.3. Emission inventory

The estimated annual emissions of VOC from printing industry in China between 2010 and 2019 are shown in Fig. 6. The total VOC emissions were

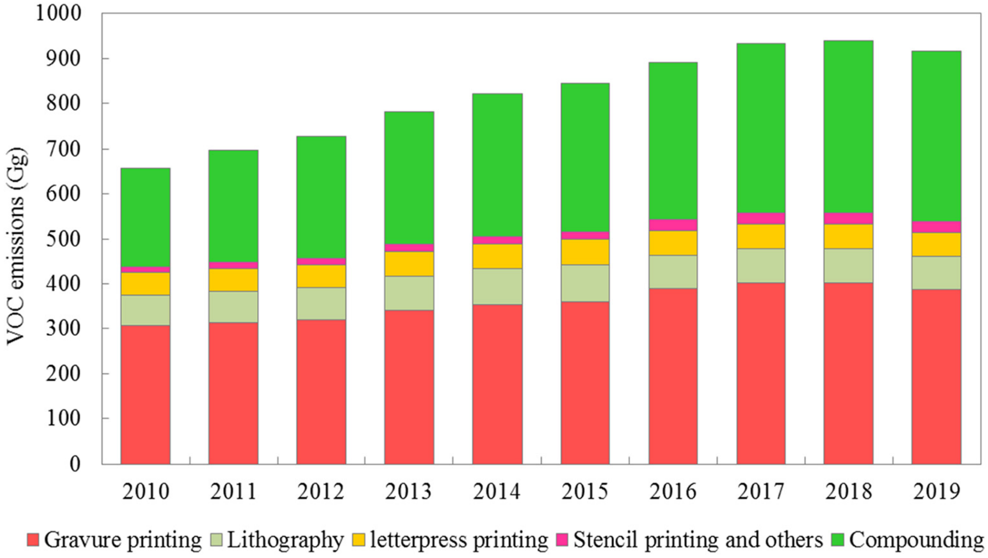

Fig. 6. Annual VOC emissions from the printing industry from 2010 to 2019 in China.

estimated to be 656.4 Gg in 2010, increasing to 916.1 Gg in 2019. VOC emissions were found to continuously increase from 2010 to 2018, reaching their maximum in 2018 at 939.8 Gg, but started to decrease afterwards. The substantial growth is driven by the large demand for ink and adhesive and the absence of effective control measures in the printing industry. In fact, the growth rate of VOC emissions in the printing industry had slowed down rapidly after 2017 from about 4.7 % in 2017 to 0.7 % in 2018, and it was negative in 2019 (−2.5 %), which was closely related to the stricter control measures (Fig. 2). For source characteristics, as shown in Fig. S6, gravure printing and the compound process contributed most of the total VOCs from 2010 to 2019, accounting for 42.3 %–46.7 % and 33.2 %–41.2 %, respectively. Lithographic and letterpress printing contributed 8.0 %–10.6 % and 5.5 %–7.9 %, respectively. Stencil printing and other processes contributed relatively little (< 3 %). Large consumption, along with a certain share of solvent-based raw materials, made gravure printing and compound processes to be the focus of VOC supervision in the printing industry. For gravure printing, the current solvent-based raw material market still accounts for about 80 %, which would bring more VOC emission compared to other printing processes. Regarding compound processes, about 60 % and 30 % of all enterprises are involved in water-based wet compounding and solvent-free compound processes, respectively. Only 10 % is for the solvent-based dry compounding. Therefore, the main reason for its large contribution to VOC emissions was the huge consumption of packaging adhesives.

Provincial VOC emission inventory of the printing industry in 2019 was also compiled based on the output value for package printing. As shown in Fig. 7, Guangdong, Jiangsu, and Zhejiang are the provinces that contribute the most printing industry emissions in China, each accounting for >10 % of the total emissions, and together accounting for 44 %. The development of the printing industry is closely related to the development of the regional economy. Fig. 7 shows the relationship between VOC emissions from printing and gross domestic product (GDP). VOC emissions from the printing industry are also affected by regional imbalances in economic development in China. In general, VOC emissions from the printing industry are mainly concentrated

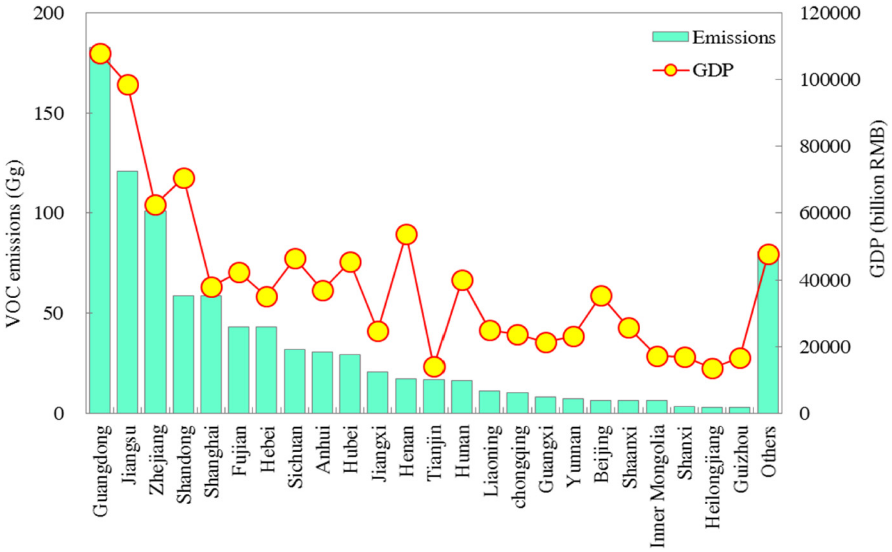

Fig. 7. Provincial VOC emissions from the printing industry in China in 2019.

in areas with a more dynamic economy, including the Pearl River Delta (Guangdong), Yangtze River Delta (Shanghai, Zhejiang, Jiangsu, and Anhui), and Bohai Rim (Beijing, Tianjin, Hebei, Liaoning, and Shandong). Other regions, such as Henan and Fujian, are responsible for fewer emissions. Midwest China, including Tibet, Gansu, Ningxia, and Qinghai, makes a relatively small contribution to VOC emissions due to the relatively underdeveloped printing industry and small GDP.

3.4. Speciated emission and OFP

A speciated VOC emission inventory for the printing industry in 2019 was also established in this study. As shown in Fig. S7, the dominant VOC group in printing industry emissions was OVOCs, which accounted for 44.9 % of all VOC emissions, followed by alkanes (22.6 %), aromatics (18.6 %), alkenes/alkynes (5.8 %), halocarbons (5.0 %), and others (3.1 %). This indicates that OVOCs are an important chemical group that cannot be ignored in the printing industry. Ethyl acetate (28.9 %), toluene (12.5 %), isopropanol (8.0 %), isopentane (7.6 %), and n-pentane (2.4 %) were the top five VOC species from the printing industry, together accounting for 59.4 % of all VOC emissions. Ethyl acetate, toluene, and isopropanol were mainly derived from raw materials including inks, adhesives, and thinners. OVOCs were the chemical group with the largest emissions in all processes except letterpress printing, for which aromatics were the dominant chemical group. The dominant chemical groups and species in the VOC emissions of the printing industry were closely related to the raw materials, processes, and end treatment facilities, as discussed in Sections 3.2 and 3.3. The VOC speciated emission inventory is shown in Table S5.

The OFPEIs of the VOCs emitted from the printing industry in China are shown in Fig. S8, with values ranging from 1.4 to 2.7 g O_3/g VOCs. Letterpress printing had he largest OFPEI (2.7 g O_3/g), followed by the compound process (2.6 g O_3/g), stencil printing and others (2.1 g O_3/g), lithography (1.6 g O_3/g), and gravure printing (1.4 g O_3/g). Aromatics was the dominant chemical group for letterpress printing, the compound process, and stencil printing and others, accounting for 57.1 %, 45.3 %, and 42.9 % of the total OFPEI, respectively. For other processes, the OFPEI mainly depended on aromatics and OVOCs. Therefore, the control of aromatics and OVOCs will be critical for reducing OFP from the printing industry. The total OFP of VOCs from the printing industry in China in 2019 was 1834.5 Gg. As shown in Fig. 8 a, the contribution of each process to the overall OFP varied in a similar manner to the emissions. Gravure printing and the compound process still contributed most to the total OFP (29.9 % and 53.2 %, respectively), followed by letterpress printing (7.9 %), lithography (6.2), and stencil printing and others (2.8 %). As shown in Fig. 8 b, aromatics (38.8 %) was the dominant chemical group in the total OFP for the printing industry, followed by alkenes/alkynes (24.8) and OVOCs (20.6 %). This was very different from the pattern for emissions, in which OVOC (44.9 %) made the largest contribution, followed by alkanes (22.6 %) and aromatics (18.6 %). The contribution of aromatics and alkenes/alkynes to the overall OFP was significant, mainly due to the high MIR values of these species. Toluene (24.9 %), ethyl acetate (9.1 %), 1,3-butadiene (7.3 %), isopentane (5.5 %), and 1-butene (4.0 %) were the top five active species, and were the key species to control in emissions from the printing industry.

4. Discussions

4.1. Comparison with other studies

The VOC emissions from the printing industry in this study are compared with previous estimates. As shown in Fig. 9, our results were much higher than those reported by Wu et al. (2016), Wu and Xie (2017), Sun

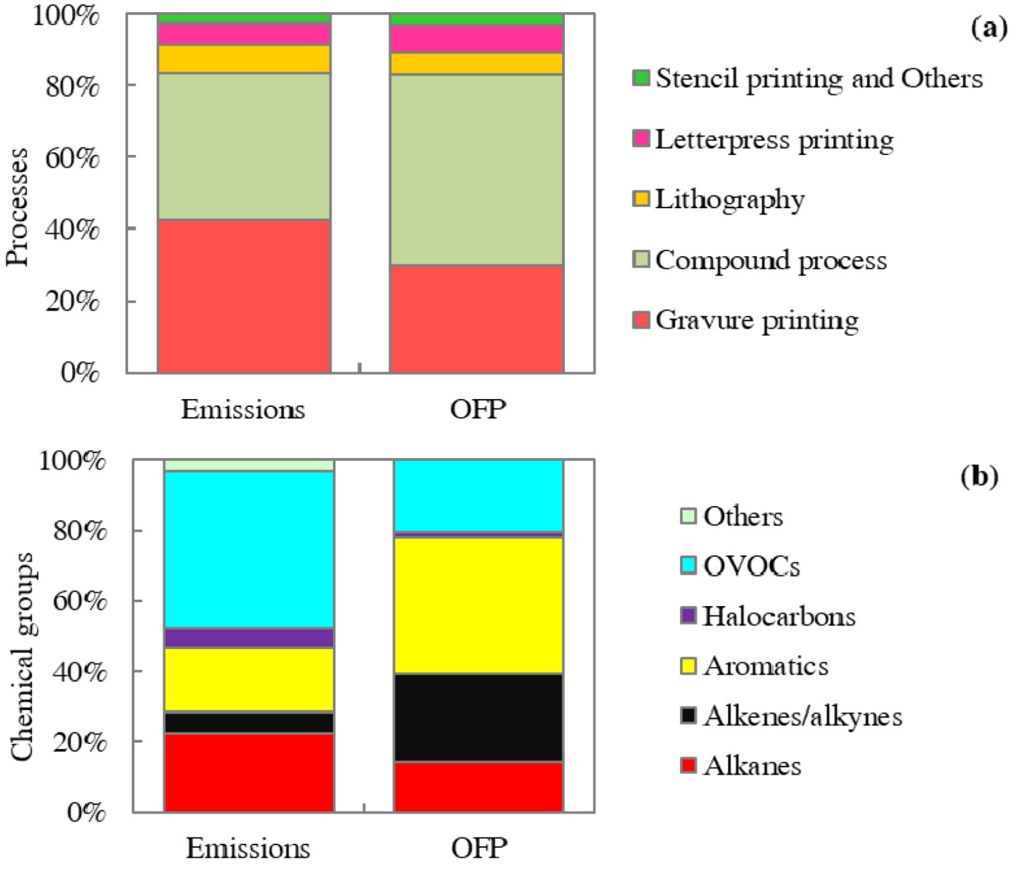

Fig. 8. Source characteristics (a), and total chemical groups (b) of VOC emissions and the OFP from the printing industry in China in 2019.

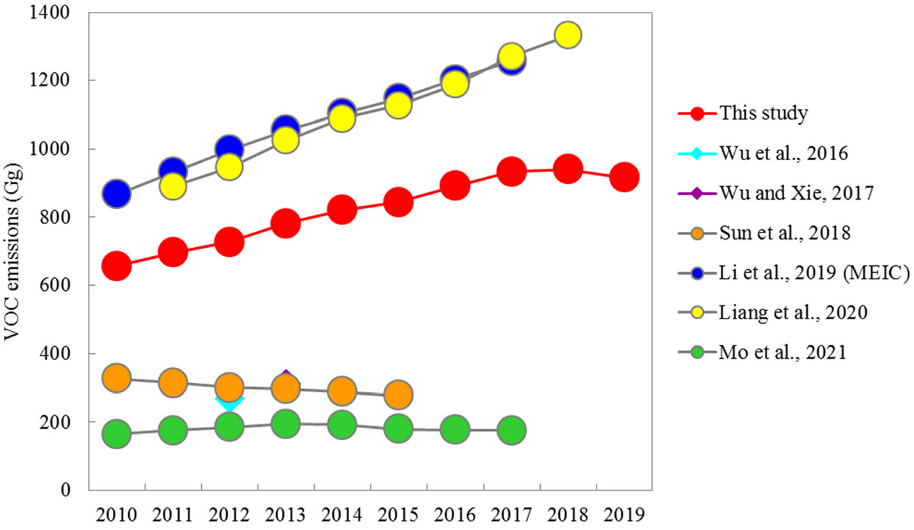

Fig. 9. Comparison of VOC emissions from printing industry between this study and previous estimates.

et al. (2018) and Mo et al. (2021). The most important reason is that the above research did not consider the VOC emissions from solvent products such as adhesives, diluents and wetting fluids used in the printing industry, only estimating the VOC emissions from ink. Compared to the emissions reported in our previous study (Liang et al., 2020) and MEIC (Li et al., 2019), our results were lower. The higher estimation of the above studies were mainly due to the relativity high and universal EFs used in their calculations., without considering the difference of EFs for types of raw materials in various processes. For example, the EFs for all printing process in MEIC (Li et al., 2019) was 540 g VOCs g^{-1} ink, while it was divided into eight EFs according to different printing processes and types of raw materials in this study. Additionally, the EFs for compound process in Liang et al. (2020) was 0.53 g VOCs g^{-1} adhesive, which were 1.65 g VOCs g^{-1} adhesive, 0.10 g VOCs g^{-1} adhesive, 0.01 t g VOCs g^{-1} adhesive and 0.01 g VOCs g^{-1} adhesive for dry compounding, wet compounding, solvent-free compounding and co-extrusion compounding in our study, respectively. The EFs used in this study were refined to different processes and raw materials. Moreover, based on the material balance analysis method, key organic materials such as thinner and adhesive which were often omitted in the previous studies, were also considered in the improved EFs. Therefore, we believe that we have improved the reliability of VOC emission from the printing industry.

4.2. Policy implications

As discussed above, both the results of emissions and OFP show that gravure printing and compound process were the two largest contributors to VOCs from printing industry in China, together accounting for >80 % of total emission and OFP. Therefore, for the scientific and precise emission reduction of VOCs in the printing industry, we could take policy from four aspects. One is to strictly enforce the environmental access of key printing source projects. The projects of water, or UV -based gravure printing, as well as wet and solvent-free compound processes are preferred. Strictly restrict the projects for solvent-based gravure printing and compound process, unless the project is equipped with efficient exhaust collection device and treatment facilities, thus effectively reducing VOCs emissions. The second is to strengthen supervision of key species of the above two key process in the printing industry. Key species control is mainly constrained by emission limits in the emission standards. In *Emission standard of air pollutants for printing industry* (GB 41616–2022) (Ministry of Ecology and Environment (MEE), 2022) in China, specific indicators are set for benzene and its analogies, other species are constrained by non-methane hydrocarbons (NMHC). It is suggested that specific emission limit should be set for other key species commonly used as thinners in the printing industry in the future revision of the standard, such as ethyl acetate and isopropyl alcohol. Third, implement hierarchical and classified management of printing enterprises. The printing industry still lacks effective control measures (Fig. 2) and has great potential for VOC reduction through management control. The printing industry grading standards in the "*Key VOCs Industry Grading Rules of Guangdong Province*"(Department of Ecology and Environment of Guangdong Province, 2021) could be referred to conduct reasonable grading for enterprises. According to the level of the enterprise, strengthen the management and control of low-level controlled enterprises, and promote their transformation and upgrading to high-level controlled enterprises. Finally, increase the research and development of key technologies for source substitution, especially for water-based gravure printing and solvent-free compounding. Specifically, the use of water-based inks on non-absorbent materials for plastic packaging (Wang et al., 2017) and the application bottlenecks of solvent-free compounds on special functional materials (Zuo, 2017) are the current research focus.

5. Conclusions

A long-term VOC emission inventory of the printing industry during 2010–2019 in China were systematically compiled, based on local investigation and measurements. Improved EFs, source profiles, mass and speciated VOC emission inventory, and the OFP were established. Our results showed that the total VOC emissions from the printing industry were estimated to be 656.4 Gg in 2010, increasing to 916.1 Gg in 2019. The total OFP in 2019 were 1834.5 Gg. Gravure printing and the compound process together contributed >80 % to both the total VOC emissions and OFP. Guangdong, Jiangsu, and Zhejiang were the provinces making the largest contributors to total emissions, together accounting for 44 %. The chemical group that accounted for most of the VOCs in emissions was OVOCs (44.9 %), while aromatics (38.9 %) was the dominant chemical group in terms of total OFP. Ethyl acetate, toluene, isopropanol, isopentane, and n-pentane were the top five VOC species in terms of the total emissions, together accounting for 59.4 %. Considering the differences in photochemical reactivity among various VOC species, toluene, ethyl acetate, 1,3-butadiene, isopentane, and 1-butene were the top five species in terms of OFP. We suggest giving priority to the environmental access of water-based and low-VOCs printing projects; controlling key VOCs species in the printing industry through the setting or assessment of indicators in emission standards; implementing hierarchical and classified management of printing enterprises; and increasing the research and development of source substitution technologies.

CRediT authorship contribution statement

Xiaoming Liang: Conceptualization, Writing – original draft, Methodology. **Laiguo Chen:** Supervision, Writing – review & editing. **Ming Liu:** Investigation, Formal analysis. **Haitao Lu:** Investigation. **Qing Lu:** Investigation. **Bo Gao:** Investigation. **Wei Zhao:** Investigation. **Xibo Sun:** Investigation. **Daiqi Ye:** Supervision.

Data availability

Data will be made available on request.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.161295.

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