



Source Profiles and Chemical Reactivity of Volatile Organic Compounds from Solvent Use in Shanghai, China

Hongli Wang, Yuezhen Qiao[†], Changhong Chen^{*}, Jun Lu, Haixia Dai, Liping Qiao, Shengrong Lou, Cheng Huang, Li Li, Shengao Jing, Jianping Wu

Shanghai Academy of Environmental Sciences, Shanghai 200233, China

ABSTRACT

Chamber experiments, exhaust collection, and in-situ sampling were employed to study the emission profiles of volatile organic compounds (VOCs) from solvent use, specifically indoor paint, auto paint, furniture paint, and print ink, which are of significant importance with regard to VOC emissions in Shanghai. The results showed that there were some differences among these VOC source profiles of the emissions associated with different solvents. On the one hand, for emissions from imported indoor solvents, ~50% of the total mass concentration was contributed by aromatics, and ~30% by alkanes. On the other hand, VOC source profiles from domestic indoor paint, furniture paint, and auto paint were similar in the sense that aromatics made a much larger contribution to total VOCs, specifically, 98% for domestic indoor paint, 80%–93% for the other two, and C8–C9 aromatics accounted for ~70% of total VOCs. For VOCs from printing, C2–C5 species dominated by more than 50%, followed by C8–C9 aromatics. VOCs from the use of different solvents presented different chemical reactivities. Among the four solvents listed above, the average OH loss rate constant (k -avg) and maximum incremental reactivity (MIR-avg) of VOCs from printing were the lowest, with values of 8.2×10^{-12} cm³/molecule/s and 2.9 g(O₃)/g(VOC), respectively. For VOCs from painting, the average reactivity was twice that of VOCs from printing, and its value of MIR-avg was 4.7–6.3 g(O₃)/g(VOC). There are significant variations in the VOC source profiles related to solvent use in different studies. The representativeness of the solvents studied and the VOC samples collected should thus be more closely examined. The accuracy of VOC source profiles related to solvent use is highly dependent on location and sampling frequency.

Keywords: Solvent use; Volatile organic compounds; Source profile; Chemical reactivity; Shanghai.

INTRODUCTION

Solvent use including painting and printing processes has been recognized as important emission source of volatile organic compounds (VOCs) in the atmosphere (Na *et al.*, 2003; Liu *et al.*, 2008a; Guo *et al.*, 2011; Huang *et al.*, 2011). The anthropogenic emission of VOCs in China was 20.1 Tg in 2005, and the industrial and domestic solvent use was the largest emission source accounting for 28.6% of the total VOCs (Wei *et al.*, 2009, 2011). These VOCs from solvent use result in heavy indoor air pollution (Celebi and Vardar, 2008; Dales *et al.*, 2008; Kim *et al.*, 2012), and especially, 18–40% of them are hazardous and harmful to human health (Wei *et al.*, 2008, 2009). Additionally, most

VOC species emitted from solvent use are very chemically reactive, and hence play an important role in the formation of tropospheric ozone and fine particles (Derwent *et al.*, 2007; Wang *et al.*, 2007).

Solvent use has gained a considerable amount of attention due to its contribution to VOC emissions. According to the VOC emissions inventory by Huang *et al.* (2011), solvent use, i.e., architecture paint and auto paint, accounted for 25% of the total VOC emission of Shanghai in 2007. From the result of the source apportionment by receptor model, solvent use and production contributed around 32% of the measured ambient VOCs during 2007 to 2010 (Cai *et al.*, 2010), while a contribution of 20% was reported by Wang *et al.* (2012) during the 2010 Shanghai World Expo. Apparently, the results obtained by various methods may differ to some extent, particularly those from the source apportionment by receptor model. One important reason for this variation can be attributed to the different VOC source tracers or fingerprints selected, which contributes a large uncertainty in source profiles.

There have been various kinds of VOC source profiles reported in different regions (Watson *et al.*, 2001; Na *et al.*, 2004; Liu *et al.*, 2008b). Liu *et al.* (2008b) studied the main

[†] now at Jiangsu Academy of Environmental Sciences, Nanjing 210036, Jiangsu, China

^{*} Corresponding author.
Tel.: 86-21-64085119; Fax: 86-21-64085119 ext. 2506
E-mail address: chench@saes.sh.cn

industrial VOC source emissions in China, including the emission from vehicle exhaust, gasoline evaporation, industrial coal combustion, biomass burning, and petrochemical industry. Source profiles of VOCs from solvent use processes were also reported in several cities. In Beijing (Yuan *et al.*, 2010), the largest contributing species, toluene and C8 aromatics, accounted for 76% of total VOCs emitted from painting; heavy alkanes and aromatics were dominant in VOCs from printing. In terms of the source profiles of solvent use in Guangzhou, alkanes, especially C6–C8 alkanes, contributed more than 50% of VOCs from solvent use (Wang *et al.*, 2008). While most VOC emissions from solvent use in Seoul were aromatics (96%) (Na and Kim, 2007), the significant variations among VOC source profiles of solvent use reported by different studies may result in large uncertainty in the source apportionment of VOCs and thus further mislead the policy made for city's air pollution control. Therefore, getting the updated local source profiles of VOCs from solvent use is of fundamental importance for air pollution control in any region. In the present study, source profiles of VOCs from solvent use in Shanghai were investigated by examining emissions from architecture paint, auto paint, furniture paint, and printing.

EXPERIMENT AND METHODS

Selection of Paints

The commonly found commercial paints on the market of China are mostly used for architecture, automobile, furniture, antisepticising, printing, and other industrial processes. Among these paints, the architecture paint has been reported as the primary source, providing ~38% of total emissions of the paints, followed by the furniture paints (~22%) and the antiseptic paints (~22%) in China, and auto paints are also one considerable source of VOCs in some areas (Wei *et al.*, 2009). In Shanghai, the total amount of production and sale of paints was 1.5 million tons in 2011, of which architecture paints took up over 50% (<http://www.stats-sh.gov.cn/tjnj/nj12.htm?d1=2012tjnj/C1212.htm>). According to the VOC emissions inventory by Huang *et al.* (2011), architecture paints accounted for 80% of the total VOC emissions from solvent use, followed by those from auto paints and other painting processes.

According to the market survey, the present study was carried out on the objects selected as follows: (1) Five brands of indoor paints, two of which were imported, and three brands of furniture paints were selected as the typical paints for the research of VOC source profiles of architecture and furniture paint, respectively; (2) Air samples were collected in automobile painting workshops from different repair service stations to study the VOC source profiles of auto paint; (3) The emission exhaust from a printing company in Shanghai was selected as the object for the investigation of the VOC source profiles for printing ink.

Sampling

Paint for Indoor and Furniture

A commercial environmental chamber (QWH-1000 the Thermo-Humidistat Dew Point Climate chamber) was

employed to simulate the VOC emission from indoor and furniture painting processes. The chamber was sealed with smooth stainless steel as its inner wall and a dimension of 1 m × 1 m × 1 m. Condensation on the surface could be prevented by the very low roughness, which allowed its applicability in the study of VOC emissions from indoor decorative materials and the temperature/humidity equalization treatment for wood and artificial boards or other hazardous emissions from other architecture materials. The chamber was composed of sections for inlets/outlets, mixing unit, sampling unit and unit for adjustment of factors of surroundings. The inner temperature and humidity could be regulated and a specific sampling vent was available, connecting with a SUMMA canister by a Teflon tube.

Following the instruction of the indoor paint, the paint was diluted by 10% water by weight and coated on a glass pane of 50 cm × 50 cm size by three coating layers with a five-minute interval for each coating after homogeneous mixing. The whole painting process was finished in the chamber. Two-hour exposures for natural emission were allowed after the painting process by closing the chamber. The temperature and relative humidity of the chamber was controlled in the ranges of (20 ± 5)°C and (50 ± 5)%, consistent with the condition in the surround environment.

The air sample was collected by a SUMMA canister after two-hour natural volatilization of the painted glass. After air sampling, a three-hour ventilation was carried out by switching on the inlets/outlets for gas washing, preparing for the next experiments.

The sampling process for the furniture paint emissions was similar to that of indoor paint. Varnish, curing agent and diluents were mixed in a ratio of 2:1:1 by weight according to the instructions. The mixed paints were coated on the artificial board and all the subsequent procedures were consistent with those of indoor paints.

To prevent cross contamination, all the painting tools and materials were solely used once. In summary, there were ten VOC samples of indoor paint and six VOC samples of furniture paint collected to investigate VOC source profiles in this study.

Auto Paint

Four different automobile 4S (Sale-Sparepart-Service-Survey) shops in Shanghai were selected for sampling the VOC emissions from paint-spraying and paint-drying processes which were the two most common types of auto painting. The VOC samples were collected right after the paint-spraying process in the specific spray booth. During the paint-spraying period, the aerator was closed for isolating the spray booth from the outside surroundings. The samples of paint-drying process were collected during the drying process after paint-spraying. Four samples were collected for each painting process.

Printing Ink

According to an existing emission inventory, large amounts of VOCs were emitted from printing ink (Huang, *et al.*, 2011). However, the knowledge of its source profiles is still very limited up to now. To better understand the

VOC source profiles from printing ink, two VOC samples from the mixed printing exhausts at a company were collected by SUMMA canisters. This company produces a large amount of production of resins, connection materials, note-printing ink and specific printing ink. The sampling lasted one hour continuously.

Sample Analysis

VOC samples were analyzed by one Gas Chromatograph Mass Spectrometer (GC-MS) system (GC7890A/MSD5975C, Agilent Technologies, USA). Firstly, samples were pumped into a cryogenic pre-concentrator (Entech 7100, USA). The pre-concentrator had a three-stage trapping system. In the first stage, the VOC was adsorbed on glass beads at -150°C and desorbed at 15°C . In the second stage, it was trapped on Tenax at -30°C and desorbed at 180°C . Finally, in the third stage, the VOC was focused on a transfer line at -160°C and heated rapidly to 65°C . Then, the concentrated VOC was injected into the gas chromatograph. The GC oven temperature was initially programmed at -50°C , holding for 2 min, and increasing to 220°C at $6^{\circ}\text{C}/\text{min}$. The VOC compounds were separated on a DB-1 capillary column ($60\text{ m} \times 0.32\text{ mm} \times 1.0\text{ }\mu\text{m}$, Agilent Technology, USA) and quantified using a quadrupole mass spectrometer. The mass spectrometer was operated in SCAN mode and scanned from 20 to 300. The ionization method was electron impacting (EI, 70 eV), and the source temperature was 220°C .

The identification of each compound was based on its retention time and mass spectrum. The PAMS (Photochemical Assessment Monitoring System) standard gas (Spectra, USA) was used to confirm compounds' retention time and identify compounds. In all, 56 species were identified and measured in this study. The sum of these 56 species was defined as "total VOCs" in this text. The target species were quantified by using multipoint external calibration method. To establish calibration curves, a certified gas mixture containing all the target compounds was dynamically diluted with pure nitrogen to five concentration gradients using mass flow controllers. 1,4-difluorobenzene, and chlorobenzene- d_5 were chosen as internal standards in samples. The method detection limits (MDLs) of the various VOC species ranged from less than one to several tens of micrograms per cubic meter (Jia *et al.*, 2009; Qiao *et al.*, 2012).

Estimation of the Reactivity of VOCs from Solvent Use

According to the VOC composition in the emissions from solvent use, the relative contribution from each species to the total VOC reactivity was estimated combining with the constants of OH loss rate (K^{OH}) and the maximum incremental reactivity (MIR). The OH loss rate (L^{OH}) and the ozone formation potential (OFP) of each VOC species was calculated through multiplying its mass concentration by K^{OH} and MIR, and the relative contribution of each species to total L^{OH} and OFP was calculated by the ratio of its L^{OH} and OFP to the total L^{OH} and OFP, respectively. The average K^{OH} and MIR (sign as $k\text{-avg}$ and $\text{MIR}\text{-avg}$) of VOCs from solvent use was obtained through dividing the total L^{OH} and OFP by the total concentration of VOCs. The K^{OH} and MIR adopted in this study were cited in the

literatures (Atkinson and Arey, 2003; Carter, 2008).

RESULTS AND DISCUSSIONS

Source Profiles of Solvent Use

Three different methods were employed to sample the VOC emissions from different solvent applications in the present study as mentioned above. Because of the complex sampling processes, the concentrations of VOCs were greatly affected by many factors, such as the amount of solvent used, ventilation conditions, and the distance between the sampling site and the emission sources. To characterize the emission source, the information of fingerprint of emission source is much more critical than the absolute value of the emission. Hence, this study was focused on the relative amount of each VOC species in the emissions of solvent use instead of their respective concentrations. In order to compare the VOC composition in different solvent applications, the mass percentage of each VOC species in total VOCs was calculated, as shown in Table 1.

Indoor Paint

VOC source profiles of indoor paints, based on the five most common kinds of indoor paints on the market, were obtained in this study. As shown in Figs. 1(a) and 1(b), the composition of the VOC emissions from two kinds of imported indoor paints was similar to each other, and a similar composition of VOC emissions was also observed for three kinds of domestic indoor paints. However, there was a large difference in VOC composition between imported and domestic indoor paints. In the case of the imported indoor paint, the largest contributing species were aromatics accounting for 51.9% to the mass concentration of all measured VOCs, followed by alkanes (31.8%), and alkenes (16.3%). Among the aromatics, xylenes were the most abundant species, accounting for 11.7% of the total VOCs, followed by toluene (7.9%) and ethylbenzene (7.1%), as shown in Fig. 2. The composition of VOCs from the imported indoor paint was similar to that of the ambient air in Shanghai urban area (Chen *et al.*, 2012; Wang *et al.*, 2013). For the composition of VOC emissions from the domestic indoor paint, aromatics contributed 97.3% of the total VOC mass concentration, and among the aromatics, the largest contributing species were xylenes accounting for 56.3% of total VOCs, followed by ethylbenzene (19.3%) and toluene (1.8%). Obviously, compared to the VOC composition from the imported indoor paint, the VOCs emitted from the domestic indoor paint contained a higher ratio of aromatics, particularly TEX (toluene, ethylbenzene, and xylenes) that contributed more than 80% of the total VOCs. The contribution of xylenes to the VOC emissions of the domestic indoor paint was 56.3%, while a much lower ratio of 11.7% was determined in the imported indoor paint.

Auto Paint

VOC source profiles of auto paint-spraying and paint-drying processes were shown in Figs. 1(c) and 1(d). Aromatics provided the largest contribution to VOCs emitted from auto paint-spraying processes, accounting for 94%, and alkanes

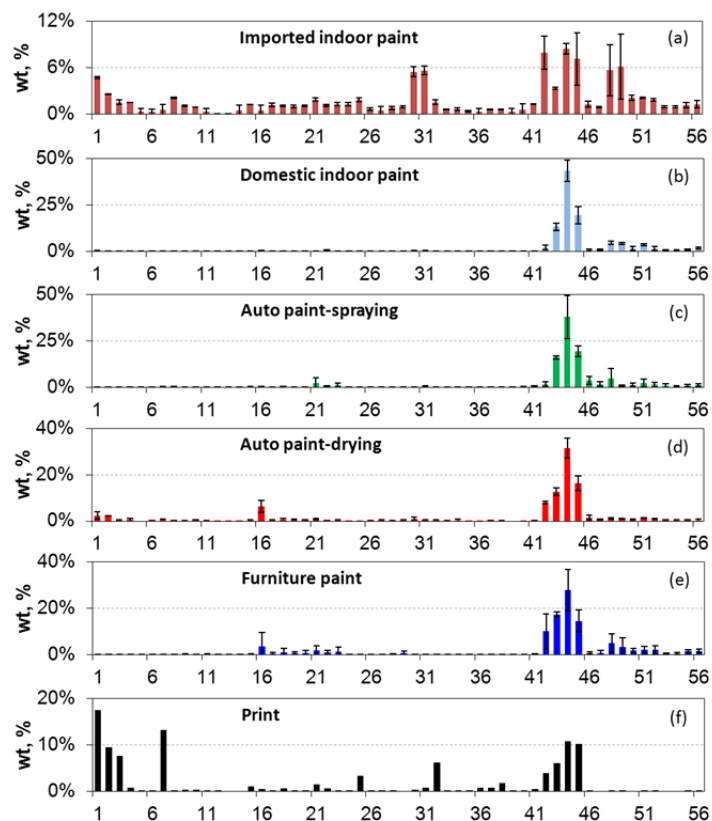
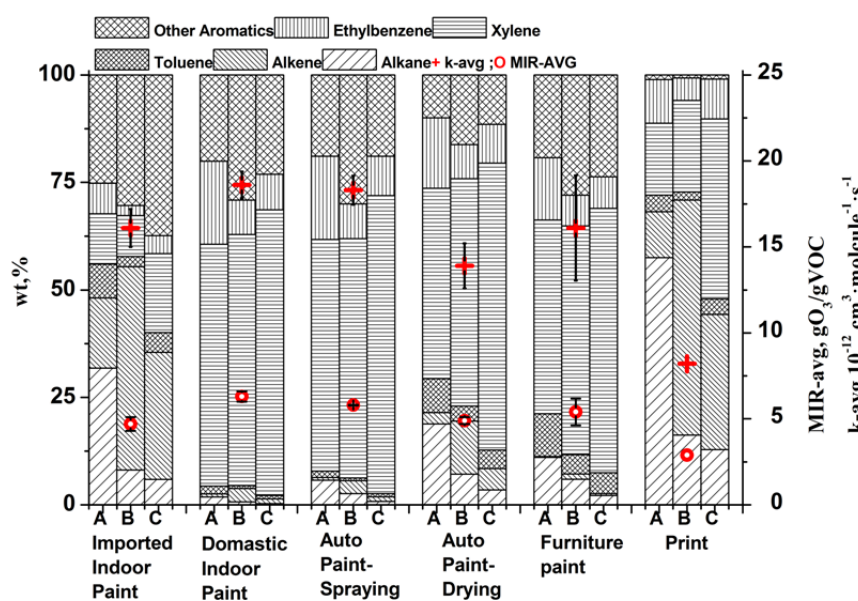
Table 1. Composition of VOCs in different solvent use processes in Shanghai (wt (mean \pm standard deviation), %).

ID	Species	Imported indoor paint	Domestic indoor paint	Auto paint-spraying	Auto paint-drying	Furniture paint	Print
1	ethane	4.7 \pm 0.1	0.2 \pm 0.1	0.1 \pm 0.1	2.4 \pm 1.6	0.1 \pm 0.0	17.4
2	propane	2.6 \pm 0.1	0.1 \pm 0.1	0.0 \pm 0.0	2.2 \pm 0.0	0.0 \pm 0.0	9.4
3	n-butane	1.5 \pm 0.3	0.1 \pm 0.1	0.1 \pm 0.0	0.3 \pm 0.2	0.0 \pm 0.0	7.6
4	i-butane	1.5 \pm 0.0	0.1 \pm 0.0	0.1 \pm 0.1	0.5 \pm 0.6	0.0 \pm 0.0	0.7
5	2-2-dime-butane	0.3 \pm 0.4	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.0
6	2-3-dime-butane	0.3 \pm 0.4	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.1
7	n-pentane	0.5 \pm 0.7	0.0 \pm 0.1	0.2 \pm 0.1	0.5 \pm 0.5	0.0 \pm 0.0	13.1
8	i-pentane	2.1 \pm 0.1	0.1 \pm 0.0	0.3 \pm 0.1	0.2 \pm 0.0	0.0 \pm 0.1	0.2
9	2-me-pentane	1.1 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1	0.1 \pm 0.1	0.3
10	3-me-pentane	0.9 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.3 \pm 0.3	0.0 \pm 0.1	0.2
11	2-3-dime-pentane	0.3 \pm 0.4	0.0 \pm 0.0	0.1 \pm 0.1	0.2 \pm 0.1	0.1 \pm 0.2	0.0
12	2-4-dime-pentane	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.0
13	224-tme-pentane	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.0
14	234-tme-pentane	0.5 \pm 0.6	0.0 \pm 0.0	0.1 \pm 0.1	0.1 \pm 0.1	0.0 \pm 0.0	0.0
15	n-hexane	1.2 \pm 0.0	0.0 \pm 0.1	0.2 \pm 0.2	0.4 \pm 0.4	0.1 \pm 0.1	1.0
16	2-me-hexane	0.5 \pm 0.6	0.1 \pm 0.1	0.4 \pm 0.0	6.3 \pm 2.7	3.4 \pm 5.9	0.5
17	3-me-hexane	1.2 \pm 0.2	0.0 \pm 0.1	0.1 \pm 0.1	0.5 \pm 0.2	0.3 \pm 0.5	0.1
18	n-heptane	1.1 \pm 0.1	0.0 \pm 0.1	0.2 \pm 0.1	0.7 \pm 0.6	1.0 \pm 1.5	0.6
19	2-me-heptane	1.0 \pm 0.2	0.0 \pm 0.1	0.1 \pm 0.0	0.6 \pm 0.2	0.5 \pm 0.8	0.1
20	3-me-heptane	1.1 \pm 0.1	0.0 \pm 0.1	0.1 \pm 0.0	0.5 \pm 0.1	0.7 \pm 1.1	0.0
21	n-octane	1.9 \pm 0.2	0.1 \pm 0.0	2.1 \pm 2.8	1.0 \pm 0.4	1.7 \pm 2.0	1.4
22	n-nonane	1.1 \pm 0.1	0.4 \pm 0.5	0.6 \pm 0.1	0.4 \pm 0.0	0.9 \pm 0.9	0.6
23	n-decane	1.3 \pm 0.2	0.1 \pm 0.1	1.1 \pm 1.1	0.6 \pm 0.1	1.3 \pm 2.0	0.1
24	n-undecane	1.3 \pm 0.2	0.1 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.0	0.0 \pm 0.0	0.2
25	n-dodecane	1.8 \pm 0.3	0.1 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.0	0.0 \pm 0.0	3.3
26	cyclopentane	0.6 \pm 0.1	0.0 \pm 0.0	0.1 \pm 0.1	0.2 \pm 0.2	0.0 \pm 0.0	0.0
27	me-cyclopentane	0.4 \pm 0.6	0.0 \pm 0.0	0.1 \pm 0.1	0.3 \pm 0.3	0.0 \pm 0.0	0.2
28	cyclohexane	0.8 \pm 0.2	0.0 \pm 0.0	0.0 \pm 0.0	0.3 \pm 0.1	0.1 \pm 0.2	0.0
29	me-cyclohexane	0.9 \pm 0.1	0.0 \pm 0.0	0.1 \pm 0.0	0.4 \pm 0.2	0.6 \pm 0.8	0.0
30	ethylene	5.5 \pm 0.6	0.3 \pm 0.1	0.1 \pm 0.0	0.9 \pm 0.7	0.1 \pm 0.0	0.2
31	propene	5.6 \pm 0.6	0.3 \pm 0.0	0.3 \pm 0.3	0.7 \pm 0.0	0.1 \pm 0.1	0.8
32	i-butene	1.5 \pm 0.3	0.1 \pm 0.0	0.0 \pm 0.0	0.3 \pm 0.2	0.0 \pm 0.1	6.1
33	cis-2-butene	0.6 \pm 0.1	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.0
34	trans-2-butene	0.6 \pm 0.1	0.0 \pm 0.0	0.0 \pm 0.0	0.4 \pm 0.5	0.0 \pm 0.0	0.0
35	1-3-butadiene	0.4 \pm 0.1	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.0
36	1-pentene	0.3 \pm 0.4	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	0.8
37	cis-2-pentene	0.6 \pm 0.1	0.0 \pm 0.0	0.1 \pm 0.1	0.1 \pm 0.1	0.0 \pm 0.0	0.8
38	trans-2-pentene	0.6 \pm 0.1	0.0 \pm 0.0	0.0 \pm 0.0	0.1 \pm 0.1	0.0 \pm 0.0	1.8
39	isoprene	0.3 \pm 0.4	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.0 \pm 0.0	0.0
40	1-hexene	0.6 \pm 0.8	0.1 \pm 0.1	0.2 \pm 0.1	0.1 \pm 0.1	0.0 \pm 0.0	0.2
41	benzene	1.3 \pm 0.1	0.1 \pm 0.0	0.4 \pm 0.4	0.3 \pm 0.1	0.2 \pm 0.1	0.5
42	toluene	7.9 \pm 2.1	1.8 \pm 1.3	1.5 \pm 1.3	7.9 \pm 0.6	9.9 \pm 7.5	3.9
43	o-xylene	3.3 \pm 0.1	13.1 \pm 1.8	16.0 \pm 0.8	12.8 \pm 1.5	17.3 \pm 0.9	6.0
44	m&p-xylenes	8.4 \pm 0.7	43.2 \pm 5.7	37.9 \pm 11.7	31.6 \pm 4.3	27.8 \pm 8.9	10.8
45	ethylbenzene	7.1 \pm 3.4	19.3 \pm 4.7	19.4 \pm 3.0	16.4 \pm 3.1	14.5 \pm 4.6	10.1
46	styrene	1.3 \pm 0.4	0.6 \pm 0.4	3.5 \pm 2.1	1.6 \pm 0.9	0.7 \pm 0.3	0.0
47	123-TMB*	0.9 \pm 0.1	0.8 \pm 0.3	1.6 \pm 1.3	0.7 \pm 0.3	0.7 \pm 1.0	0.0
48	124-TMB*	5.7 \pm 3.3	4.5 \pm 1.0	4.8 \pm 5.2	1.2 \pm 0.2	4.9 \pm 4.1	0.0
49	135-TMB*	6.1 \pm 4.2	4.2 \pm 0.4	0.7 \pm 0.4	1.1 \pm 0.2	3.1 \pm 4.1	0.0
50	o-ethyltoluene	2.1 \pm 0.4	1.5 \pm 1.0	1.2 \pm 1.0	0.8 \pm 0.1	1.6 \pm 1.1	0.0
51	m-ethyltoluene	2.1 \pm 0.1	3.5 \pm 0.5	2.3 \pm 2.2	1.3 \pm 0.2	2.0 \pm 1.4	0.1
52	p-ethyltoluene	1.8 \pm 0.1	1.5 \pm 1.1	1.4 \pm 1.2	0.9 \pm 0.1	2.1 \pm 1.7	0.1
53	m-diethylnzene	0.9 \pm 0.1	0.4 \pm 0.2	0.8 \pm 0.9	0.5 \pm 0.1	0.4 \pm 0.3	0.0
54	p-diethylnzene	0.9 \pm 0.1	0.4 \pm 0.2	0.4 \pm 0.4	0.5 \pm 0.1	0.4 \pm 0.4	0.0
55	i-propylbenzene	1.2 \pm 0.4	0.7 \pm 0.5	0.7 \pm 0.8	0.6 \pm 0.0	1.4 \pm 0.7	0.2

Table 1. (continued).

ID	Species	Imported indoor paint	Domestic indoor paint	Auto paint-spraying	Auto paint-drying	Furniture paint	Print
56	n-allylbenzene	1.3 ± 0.5	1.7 ± 0.6	1.1 ± 0.6	0.4 ± 0.4	1.5 ± 0.9	0.2
	No. of samples	6	4	4	4	6	2

*TMB: trimethylbenzene.

**Fig. 1.** VOC source profiles from different processes of solvent use in Shanghai (The ID number of each species was the same as that of Table 1.).**Fig. 2.** Composition of VOCs and their contribution to the OH loss rate (L^{OH}) and the ozone formation potential (OFP) in different solvent use processes in Shanghai (A: wt; B: L^{OH} ; C: OFP).

and alkenes accounted for 5.8% and 0.6%, respectively. The most important aromatic species were xylenes (53.9%) and ethylbenzene (19.4%), and the others accounted for 20.1% of the total measured VOC mass concentration, as indicated in Fig. 2. In terms of the VOC composition from auto paint-drying processes, aromatics took up 78% of the total VOCs, followed by alkanes (18.8%) and alkenes (2.7%), and the TEX species proportioned 7.9%, 16.4% and 44.3%, respectively. Apparently, aromatics were the dominant species in the emissions of auto paint, and especially the contribution of xylenes was more than 50%, followed by ethylbenzene (15–20%). Variations could be observed between the composition of VOCs from auto paint-spraying and paint-drying process. For example, the alkanes' contribution was higher in the VOC emissions from auto paint-drying process and 2-methylhexane accounted for 6.3% of the VOC emissions from the auto paint-drying process, much higher than the auto paint-drying process (0.4%), as listed in Table 1. Additionally, the ratios of ethane and propane were also higher in the VOC emissions from the auto paint-drying process, both accounting for 2.4%, as listed in Table 1. Compared to the auto paint-spraying process, the temperature during the auto paint-drying process was higher, which may have caused the decomposition of the large VOC species into smaller ones.

Furniture Paint

Fig. 1(e) showed the source profile of VOCs emitted from the furniture paint. Similar to VOC source profiles from indoor painting and auto painting, aromatics were dominant species from the furniture paint, and C6–C8 aromatics were also considerable components. The contribution of aromatics was 89% to the total VOC mass concentration, followed by alkanes (11%), and alkenes (0.2%). In the case of species, xylenes, ethylbenzene, and toluene were the three largest contributing species, accounting for 45.1%, 14.5%, and 9.9%, respectively, as indicated in Fig. 2. No appreciable difference could be identified among the composition of VOC emissions from different kinds of furniture paints, as indicated by the small error bar of the percentage of each species in source profiles in Fig. 1(e).

Printing Ink

The composition of VOCs emitted from the printing ink was very different from those of painting mentioned above, and the source profile was shown in Fig. 1(f). From Fig. 1(f), we can get that alkanes and aromatics were both very important in VOC emissions of printing. Alkanes accounted for 57.5% of the total VOCs, followed by aromatics (31.8%) and alkenes (10.7%), as shown in Fig. 2. Ethane (17.4%), n-pentane (13.1%), propane (9.4%), and n-butane (7.6%) were the most abundant alkane species, and 1-butene (6.1%) was the largest contributing alkene species, as listed in Table 1. The TEX species were also important contributors to the total VOCs, accounting for 3.9%, 10.1% and 16.7%, respectively.

In summary, the source profiles mentioned above could be grouped into three categories. One was the VOC source profile of the imported indoor paint. Its composition of

VOCs was similar to that of ambient air in Shanghai urban area, with a slightly higher contribution of aromatics. The second one was the VOC source profile of the printing. C2–C5 alkanes were the dominant species, and xylenes and ethylbenzene also contributed considerably. The third one was the VOC source profile of the domestic indoor paint, auto paint, and furniture paint. The contribution of aromatics was more than 80% of the total VOCs, and in particular the TEX accounted for more than 90% of the VOCs emitted from the domestic indoor paint and the auto paint.

Reactivity of VOCs Emitted from Solvent Use Indoor Paint

The relative contribution of each VOC group to the total L^{OH} and OFP was shown in Fig. 2. It can be seen from Fig. 2 that the percentage of each group in total L^{OH} and OFP of VOCs from the imported indoor paint was different from that from the domestic indoor paint due to the different relative contribution of each group to the total VOC mass concentration. In VOCs emitted from the imported indoor paint, the largest contributing group to L^{OH} was alkenes, accounting for 47.3%, and the second largest one was aromatics (44.6%), followed by alkanes group (8.1%). Further examination showed that trimethylbenzenes (TMBs) were the primary contributors, accounting for 22.8% to L^{OH} , followed by propene 17.4%, xylenes 9.6%, and ethylene 8.3%. Aromatics were the dominant species for OFP, proportioning 64.5% of total OFP, followed by alkenes (19.5%) and alkanes (5.9%). Xylenes and TMBs were the two largest contributing species of OFP, accounting for 40.6% and 25.2%, respectively. The contribution of ethylbenzene (6.8%), propene (5.1%) and ethylene (3.9%) was also of considerable importance.

In the case of VOC emissions from the domestic indoor paint, aromatics were the most important contributors both for L^{OH} and OFP, and their percentages in L^{OH} and OFP were 96.2% and 98.6%, respectively. Among the aromatics, xylenes were the most abundant species, contributing 56.3% and 58.4% to the total L^{OH} and OFP, respectively, followed by TMBs (17.4% of L^{OH} and 13.8% of OFP) and ethylbenzene (5.5% of L^{OH} and 5.6% of OFP).

Apparently, the largest contributing species of the total reactivity were C8–C9 aromatics for the VOC emissions both from the imported and domestic indoor paint. The contribution of C2–C3 alkenes to the reactivity of VOCs from the imported indoor paint was also very considerable.

Auto Paint

The primary contributor to L^{OH} of the VOC emission from auto paint-spraying was aromatics, contributing 94.4% of the total L^{OH} . Xylenes contributed 55.6% of L^{OH} , followed by TMBs (13.3%), styrene (12.1%), and ethylbenzene (8.1%), as shown in Fig. 2. Compared to the VOC emission from the auto paint-spraying, the contribution of alkenes and alkanes to L^{OH} of VOCs emitted from auto paint-drying was slightly higher, accounting for 12.4% and 7.1%, respectively, and among them, the contribution of C2–C3 alkenes was considerable. Similar to that of auto paint-spraying, xylenes were also the primary source of L^{OH} in the VOC emissions

from auto paint-drying, and the contribution of ethylbenzene (7.9%) and TMBs (7.1%) was also significant. In the case of OFP of VOCs from the auto paint, more than 90% was attributed to the contribution of aromatics, and especially xylenes accounted for more than 65% of the total OFP. In summary, aromatics, especially the C8–C9 aromatic species were the most important contributors to the reactivity of VOCs from the auto paint.

Furniture Paint

Similar to VOCs from the auto paint and the domestic indoor paint, aromatics were also found to be the dominant species for VOC reactivity in the furniture paint, accounting for 92.8% and 97.4% to L^{OH} and OFP, respectively. The other contribution was mostly from alkanes, and the contribution of alkenes could be neglected. Xylenes were the primary species contributing 53.3% and 61.5% of the total L^{OH} and OFP, respectively. TMBs contributed 19.6% and 14.2% to L^{OH} and OFP, followed by ethylbenzene (7.1% of L^{OH} and 7.3% of OFP), as indicated in Fig. 2.

Printing Ink

The relative contribution of each VOC group from the printing ink to L^{OH} and OFP was shown in Fig. 2. Alkenes made the largest contribution to L^{OH} , accounting for 54.7%, and aromatics and alkanes contributed 29.1% and 16.2%, respectively. Considering the contribution of each VOC species to L^{OH} , 1-butene was the primary species and proportioned 27.1%, followed by xylenes (21.3%), pentene (19%), n-pentane (5.5%) and ethylbenzene (5.3%). The major contributors of OFP showed some difference from those of L^{OH} . Aromatics were the most important contributors of OFP, proportioning 55.7%, and alkenes (31.4%) were the second largest sources of OFP, followed by alkanes (12.9%). In the case of VOC species, the contribution of xylenes was the largest, accounting for 41.9% of OFP, followed by 1-butene (18.6%). The contributions of ethylbenzene (9.3%), pentene (7.6%) and pentane (4.6%) were also very considerable. In comparison to VOC emissions from painting, the reactivity of VOCs from printing was mainly attributed to both C8 aromatics and C4–C5 VOCs.

Comparison of the Average Reactivity

The k-avg and MIR-avg of VOCs emitted from different solvent use were calculated as shown in Fig. 2. It can be seen from Fig. 2 that the k-avg of VOCs from the domestic indoor paint and auto paint-spraying were the largest, i.e., $(18.6 \pm 0.8) \times 10^{-12}$ cm³/molecule/s and $(18.3 \pm 0.8) \times 10^{-12}$ cm³/molecule/s, respectively. The second largest k-avg was from the imported indoor paint $((16.1 \pm 1.1) \times 10^{-12}$ cm³/molecule/s) and the furniture paint $((16.1 \pm 3.1) \times 10^{-12}$ cm³/molecule/s). VOCs from the printing ink had the smallest k-avg, 8.2×10^{-12} cm³/molecule/s, which was similar to that in the ambient (Shao *et al.*, 2005; Wang *et al.*, 2013). The k-avg for VOCs from painting obtained in this study was highly similar to that of m,p-xylene (Atkinson and Arey, 2003), and much higher than that in ambient. Apparently, VOCs from painting were more reactive than VOCs in ambient air, and the painting served as one reactive source

of VOCs.

The variation trend of MIR-avg among different solvent use was similar to that of k-avg. Generally, the MIR-avg of VOCs emitted from solvent use would be higher if the VOCs emitted from this type of solvent use had higher k-avg. The MIR-avg of VOCs from the domestic indoor paint was the highest, namely (6.3 ± 0.3) g(O₃)/g(VOC), followed by that of auto paint-spraying (5.8 ± 0.02) g(O₃)/g(VOC), furniture paint (5.4 ± 0.8) g(O₃)/g(VOC), auto paint-drying (4.9 ± 0.2) g(O₃)/g(VOC), the imported indoor paint (4.7 ± 0.4) g(O₃)/g(VOC), and printing 2.9 g(O₃)/g(VOC). Compared to the MIR-avg of VOCs from other sources, the MIR-avg of VOCs from the indoor paint, furniture paint and auto paint were higher than that of vehicle exhaust, biomass burning and coal combustion (Yuan *et al.*, 2010). The MIR-avg of VOCs from printing demonstrated a lower value comparing with those from the other abovementioned sources, which was similar to the result in Beijing (Yuan *et al.*, 2010).

Comparison of VOC Source Profiles of Solvent Use in Different Cities

Architecture Paint

VOC source profiles of the solvent use in Shanghai studied in this work was compared with the results from other cities (Na and Kim, 2007; Wang *et al.*, 2008; Yuan *et al.*, 2010). As shown in Fig. 3, the composition of VOCs from architecture paint showed strong location-dependence. The percentages of alkanes in total VOC mass concentration, for example, decreased in the following order Guangzhou (50.2%) > Beijing (27.8%) > Shanghai (16.8%) > Seoul (2.2%). For the contribution of BTEX (benzene and TEX), the percentages could be ranked as follows: Seoul (96.5%) > Beijing (59.9%) > Shanghai (52.7%) > Guangzhou (21.5%). Among the BTEX, toluene displayed the largest location-dependence, contributing 62.4% of total VOCs from the architecture paint in Seoul, which was much higher than those in the other cities. In addition, the proportion of benzene in VOCs from architecture paint was considerable in Beijing, accounting for 7.5%, while its contribution in other cities was negligible. Possible reasons for the observed large difference among the VOC source profiles from the architecture paint in different studies could be considered as following. One was that the definitions of “architecture paint” were not very consistent in different studies. For example, “architecture paint” in this study referred to the indoor paint, and in Yuan *et al.* (2010) study indoor paint or outdoor paint were not differentiated, while it was used to describe the paint-spraying processes in Wang *et al.* (2008) study and the general solvent use in the work of Na and Kim (2007), respectively. Obviously, the different definitions indicated to some extent that the research objects in different studies were not so consistent. The second reason might be the different sampling methods in these studies. For instance, in this study, VOC samples were collected in one environmental chamber used to simulate the VOC emission of the architecture paint, which was not affected by the air in ambient (Wang *et al.*, 2012). The samples in Yuan *et al.* (2010) study were collected in the room of painting. The third reason was that these studies

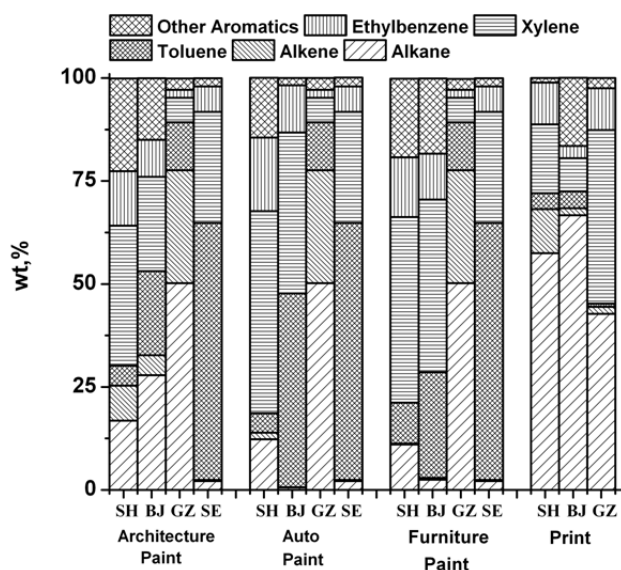


Fig. 3. Comparison of VOC composition from solvent use in different cities.

SH: Shanghai (this study). The result of the architecture paint is the arithmetical average of those of indoor paints. The result of the auto paint is the arithmetical average of paint-spraying and paint-drying processes results.

BJ: Beijing (Yuan *et al.*, 2010).

GZ*: Guangzhou (Wang *et al.*, 2008, in Chinese). All the results of the architecture paint, auto paint and furniture paint are equal to the result of the paint as referred in this reference.

SE*: Seoul (Na and Kim, 2007). All the results of the architecture paint, auto paint and furniture paint are equal to the result of the paint as referred in this reference. No results of printing are found in Seoul.

were carried out over several years, during which the architecture paint on the market might have changed due to incorporation of stricter standards of VOC emissions from solvent use.

Auto Paint

The composition of VOCs from auto paints in different studies was compared in Fig. 3. Owing to the lack of reported results of auto paint in Guangzhou and Seoul, the comparison was focused on the results in Shanghai and Beijing. The result of the auto paint here referred to the average of auto paint-spraying and auto paint-drying in Shanghai. From Fig. 3, large difference can be observed between the composition of VOCs from auto paints in Shanghai and Beijing. Compared to the result in Beijing (Yuan *et al.*, 2010), the proportion of alkanes in VOCs from auto paint in Shanghai was much higher (12.3%), and xylenes were the primary contributors, accounting for 47.1% of total VOCs. While, in Yuan *et al.* (2010) study, the largest contributing species of total VOCs were toluene (47.1%), and xylenes (39.0%). In this study, the contribution of toluene was only 4.7%, much lower than that reported by Yuan *et al.* (2010). The difference between these two studies might result from the sampling conditions and the

different auto painting processes. The samples in this study were collected in the workshop of auto paint-spraying and paint-drying, while VOCs were sampled in the down-wind direction of one automobile manufacturing factory in Yuan *et al.* (2010) study. Compared to Yuan *et al.* (2010) study, VOCs sampling in this study could be more exclusive to emissions of the auto paint, with less influence from other VOC sources.

Furniture Paint

The composition of VOCs from the furniture paint in this study was similar to that of in Beijing (Yuan *et al.*, 2010), except for the relative contribution of toluene, as shown in Fig. 3. The percentage of toluene in VOCs in Beijing was 25.7%, which was only 9.9% in this study. Xylenes accounted for more than 40% of total VOCs from the furniture paint in both studies, and meanwhile the contribution of ethylbenzene was both between 11% and 15%. The samples were collected in the workshop of furniture painting in Yuan *et al.* (2010), similar to the sampling method in this study. The discrepancy of toluene contribution between these two studies needs further investigation.

Printing Ink

Source profiles of the printing ink studied in three megacities of China, namely Beijing (Yuan *et al.*, 2010), Guangzhou (Wang *et al.*, 2008) and Shanghai, were compared in Fig. 3. All these studies indicated that alkanes were the most important species of VOCs emitted from the printing ink, and the percentages of alkanes were 66.7% 57.5% and 42.8% in Beijing, Shanghai and Guangzhou, respectively. However, the dominant alkane species were quite different in these studies. C₂–C₅ alkane species were the most abundant species in this study, and C₈–C₁₁ species were the main alkanes in Yuan *et al.* (2010) study, while in Wang *et al.* (2008) study, 2,4-dimethylpentane was identified as the primary alkane species. Besides the alkane species, the relative contribution of other species differed significantly among these studies. Alkenes accounted for 10.7% of total VOCs from printing, which was much higher than those in Wang *et al.* (2008) and Yuan *et al.* (2010) studies. Moreover, the proportion of xylenes in Wang *et al.* (2008) study was 42.1%, compared to 16.7% in this study and 8.0% in Yuan *et al.* (2010) study.

Uncertainty Analysis

Up to now, there have been numerous studies about the VOC source profiles of solvent use. Generally, aromatics especially C₈–C₉ aromatics were the dominant species of VOCs from painting, while the contribution of alkanes and aromatics shared the highest importance for VOCs from printing equally. However, large uncertainty existed in the detailed VOC species in different studies. Two factors were largely responsible for the observed discrepancy.

One was the representativeness of the solvent studied. Various types of solvents were available on the market with diverse qualities. Therefore, it was very critical to conduct a thorough survey in order to ascertain representative kinds of solvents for study. In addition, due to the rapid economic

development, increasingly stricter emission standard for VOCs from solvent use has been applied in different countries and regions, which has resulted in large discrepancy of VOC source profiles of solvent use studied in different regions or different time. For example, there was large difference between source profiles of imported indoor paint and domestic indoor paint in this study. Thus, the VOC source profiles of solvent use were highly location-dependent and time-dependent.

Second was the representativeness of VOC samples collected from emissions of solvent use. Various solvents were used in numerous industrial processes following different protocols, and the emitted VOC species from each process might vary largely based on their uniqueness, which resulted in different VOC source profiles. For example, in the present study, VOC source profiles of auto paint-spraying and auto paint-drying differed from each other a lot. Therefore, the effect of emission process of solvent use should be thoroughly considered since they would greatly influence the composition of VOCs. Additionally, considering the architecture paint and furniture paint generally being used in open surroundings, special care must be taken during sampling to avoid or to reduce the interference from other VOC sources. A well-characterized chamber, such as that used in this study, was an appropriate way to evaluate the VOC source profiles of solvent use.

CONCLUSIONS

The composition of VOCs from the most important solvent use in Shanghai, including indoor paint, auto paint, furniture paint and print ink, was studied. The local VOC source profiles of solvent use were obtained in the present study. The relative contribution of each VOC species to the total L^{OH} and OFP was calculated, and the average reactivity of VOCs from different solvent use was compared by k-avg and MIR-avg. The VOC source profiles of solvent use obtained here were compared with those published in literatures.

Variations in VOC source profiles were characterized for different solvent use. The composition of VOCs from the imported indoor paint showed considerable similarity with that of ambient air; aromatics contributed ~50% of the total VOC mass concentration, followed by alkanes (~30%) and alkenes (~16%). VOC source profiles from the domestic indoor paint, auto paint and furniture paint were very close. Aromatics were the dominant species, accounting for more than 80% of total VOCs from these three kinds of solvent use. Especially, the relative contribution of aromatics to VOCs from the imported indoor paint was as high as 98%. Alkanes made considerable contribution to the VOCs from auto paint-drying, as the decomposition of large species into small ones resulted from the high temperature. In the case of detailed species, C8–C9 aromatic species were the most abundant species of VOCs from painting, proportioning more than 70%. While, C2–C5 species were more important in the VOC emissions from printing, followed by the C8–C9 species.

Different reactivity of VOCs from solvent use resulted from the different composition of VOCs. According to this

study, the k-avg of VOCs from the domestic indoor paint showed the highest value, followed by those of the imported indoor paint and furniture paint. VOCs from printing displayed the lowest one. The k-avg of VOCs from painting was close to that of m,p-xylene, and twice that of printing. In terms of MIR-avg, VOCs from the domestic indoor paint had the highest MIR-avg, equal to (6.3 ± 0.3) g(O₃)/g(VOC), followed by those of auto paint-spraying, furniture paint, auto paint-drying, and the imported indoor paint sequentially. The MIR-avg of printing was the lowest one, 2.9 g(O₃)/g(VOC). Compared to other VOC sources, the MIR-avg of VOCs from painting was larger than those of vehicle exhaust, biomass burning and coal combustion and that of printing was lower than those of the VOC sources above. Obviously, VOCs from painting were highly reactive in general.

C8–C9 aromatic species were the most important composition of VOCs from painting in different studies, while alkanes and aromatics were both very important for printing emissions. The detailed VOC species from different solvent use showed large uncertainties among different studies. Two considerations should be given, namely the representativeness of the kinds of solvents studied and VOC samples collected. The accuracy of VOC source profiles of solvent use was highly location-dependent and time-dependent, and hence the research needed to be done for each region and the existing VOC source profiles should also be updated in a timely manner. The impact of VOCs from other sources on sampling should be mostly avoided as much as possible. A well-characterized chamber study, such as that employed in this study, could serve as an appropriate approach for the evaluation of VOC source profiles of solvent use.

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