



## Characteristics and reactivity of volatile organic compounds from non-coal emission sources in China



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### HIGHLIGHTS

- Abundant VOCs species profiles for non-coal sources in China were presented.
- The source profiles developed in our study showed some differences with others reported.
- B/T is a good marker to distinguish some VOCs sources but not all.
- Most of the VOCs emissions from non-coal sources have high air reactivity.

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### ABSTRACT

Volatile organic compounds (VOCs) were sampled from non-coal emission sources including fuel refueling, solvent use, industrial and commercial activities in China, and 62 target species were determined by gas chromatography-mass selective detector (GC-MSD). Based on the results, source profiles were developed and discussed from the aspects of composition characteristics, potential tracers, BTEX (benzene, toluene, ethylbenzene and xylene) diagnostic ratios and chemical reactivity. Compared with vehicle exhausts and liquid fuels, the major components in refueling emissions of liquefied petroleum gas (LPG), gasoline and diesel were alkenes and alkanes. Oppositely, aromatics were the most abundant group in emissions from auto-painting, book binding and plastic producing. Three groups contributed nearly equally in printing and commercial cooking emissions. Acetone in medical producing, chloroform and tetrachloroethylene in wet- and dry-cleaning, as well as TEX in plastic producing etc. were good tracers for the respective sources. BTEX ratios showed that some but not all VOCs sources could be distinguished by B/T, B/E and B/X ratios, while T/E, T/X and E/X ratios were not suitable as diagnostic indicators of different sources. The following reactivity analysis indicated that emissions from gasoline refueling, commercial cooking, auto painting and plastic producing had high atmospheric reactivity, and should be controlled emphatically to prevent ozone pollution, especially when there were large amounts of emissions for them.

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### 1. Introduction

As an important air pollutants in urban atmosphere, VOCs have being attracted more and more concerns in the world. They can chemically interact with oxides of nitrogen and sunlight to form ground-level ozone, and even take part in the formation of haze,

which now is the most serious environmental problem in China with frequent and large-scale occurrences in recent years (Anand et al., 2014; Durkee, 2014). More importantly, they also have detrimental effects on human health by contributing to respiratory illnesses, even being mutagenic or toxic to reproduction and harmful to the unborn (Anand et al., 2014).

Many studies indicated that chemical composition of VOCs emissions varied with different fuels or solvents, regions or countries, and operating sectors or processes (Na et al., 2004; Liu et al., 2008; Zheng et al., 2013; Zhang et al., 2013). The diverse sources

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and complex compositions of VOCs make it difficult to identify major emission sources and estimate the relative importance of each source to ambient VOCs concentrations, consequently hinder us from understanding the formation mechanism of secondary pollutions and devising effective control policies in urban areas seriously. Source profiles are the composition pattern of species emitted from each source category, and so are the fundamental information for developing the speciated pollutants emission inventories used in air quality models, and identifying source contributions via receptor models (Zheng et al., 2009; Marmur et al., 2007). To make sure the accuracy of emission inventories and source contributions, a unique, comprehensive and real-time updated “profile” (composition) is required for each source.

In present China, high population density (19.3% of the world total in 2014, WPS, 2015) and fast growing economy beginning in 1980s, have being accompanied by more and more pollutants emission. For example, the total VOCs emission increased from 13.1 Tg in 1995 to 20.1 Tg in 2005, and possibly would be 25.9 Tg in 2020 as estimated by Wei et al. (2011). Scientists in China have begun in recent years to measure local VOCs profiles for major emission sources, such as vehicular exhaust, gasoline evaporation, coal burning, biomass burning, petrochemical industry and solvent use of painting and printing (Liu et al., 2008; Yuan et al., 2010; Guo et al., 2011; Zhang et al., 2013; Zheng et al., 2013; Wei et al., 2014; Wang et al., 2014). These studies have developed some domestic VOCs source profiles in China, but those profiles are far from enough in covering all the VOCs sources such as LPG refueling, cleaning, plastic producing and book binding etc, or need more supporting evidences, making it hard to establish accurate emission inventories or apportion sources using CMB model in China (Wang et al., 2014a).

As a very large pollution source in China which is facing notorious air pollution, coal burning always attracts the most attention, but non-coal sources like fossil fuel evaporation (refueling, diurnal and hot-soak), solvent utilization and industrial processes cannot be ignored. After America, China is the second-biggest consumer of LPG and motor gasoline consuming 740.2 and 1615.9 thousand barrels per day respectively in 2010 with the consumption still increasing (EIA, 2015a). During their handling and usage, large amounts of VOCs are discharged. It was reported that approximately 185,000 tons vehicular evaporative emissions were emitted in 2010 in China, among which refueling contributed 67% (Yang et al., 2015). For both solvent utilization and industrial processes, Wei et al. (2011) predicted they would become the highest two VOCs contributors in 2020, emitting 37% (9.40 Tg) and 24% (5.92 Tg) of the total VOCs emissions, respectively, even if China's legislative standards for VOCs emissions would be implemented effectively in the future (Wei et al., 2011). To further enrich the database of VOCs source profiles in China, various non-coal emission sources including refueling of LPG and fossil oils, auto painting, printing, wet and dry cleaning, medicine and plastic producing, as well as commercial cooking in a coastal region of China were investigated, with their potential tracers and BTEX ratios being discussed in our study. Their atmospheric chemical reactivity was also evaluated to give more insights for targeted pollution control.

## 2. Methods

### 2.1. Sample collection

Different volumes of stainless steel canisters (Entech Instrument, Inc., Simi Valley, CA, USA) and Tedlar bags (Dalian Delin Gas Packing Co., Ltd, China) pre-cleaned with high purity nitrogen and evacuated to vacuum were used for sampling. Whole-process samplings were conducted for refueling, auto painting and

printing emissions to explore the general features for them. For the emissions of medical producing, plastic processing and commercial cooking, instantaneous sampling above the exhaust ducts was conducted by the aid of a pump connected to the sampling bags. After sampling, all the samples were delivered to the laboratory for chemical analysis within a week. Detailed information is as follows:

#### 2.1.1. Re-fueling of LPG

In China, a large proportion of residents living in rural places and city villages used the tanked-LPG for cooking instead of coal for convenience and light pollution. The tanks can be used repeatedly by re-inflating. For each sample, refueling emissions were collected from the launch to the stop of inflating operation to a civilian gas tank (capacity, 13.5 kg) using a 400-mL canister in an inflating workshop ( $n = 10$ ;  $T$ , 24.5 °C). Before collecting, the connecting joint between gas inflating and releasing nozzle was wrapped to form a small enclosed space. One side of a tubule was also wrapped in it while the other side was connected to a sampling canister. The average sampling duration was  $9.64 \pm 0.64$  s.

#### 2.1.2. Re-fueling of gasoline and diesel

Oil refueling operations include transferring fuel from bulk storage tanks into transport vehicles like road tankers, road tankers into service station tanks and service station tanks into vehicle tanks. During filling of tanks, vapor evolves as a result of displacement of headspace vapor by the incoming oil, splashing and turbulence. In our study, vapors from storage tanks and vehicle tanks during refueling with gasoline and diesel were collected into 400-mL canisters through inserting sampling intake-tubules into the upper space of the tanks without touching the liquid surface. For gasoline, the vehicles investigated included cars and motorcycles. The sampling number for each refueling source was 10. Rush hours (9:00–10:00 a.m.) were chosen for sampling. The average sampling duration was  $2.57 \pm 0.39$  min for storage tanks, and  $36 \pm 9.61$  s for vehicle tanks. The ambient temperature during the sampling period was 28.3 °C with no strong wind.

#### 2.1.3. Auto painting (automobile painting)

Painting is an important work in car-related industries. In an auto-painting workshop of a typical garage where gasoline-based paints were still the common used paints, 12 canister samples were collected in three different corners far from the door (4 samples for each corner) while 8 bag samples (2.5-L) were collected from the exhaust duct of the auto-painting workshop. There are three most important processes in painting, namely primer painting, finish coat painting and paint-drying with different paints being used (Yuan et al., 2010). With 1L paints for 5 m<sup>2</sup> car surface on average, sampling inside the workshop lasted 108 min covering all the 3 steps of painting with the indoor temperature being 24.9 °C. To avoid influences from other emission sources, all windows in the sampling room were closed and canisters were held near the VOCs sources.

#### 2.1.4. Printing

Sampling was conducted in a comparatively large printing factory that mainly printed books, magazines, newspapers, and packaging materials. The press equipments in the factory were all offset lithographic printing machines. 10 canister samples (2-L) were collected at different sites of the printing room (total area was 4000 square feet), and another 10 were collected in binding room (total area was 5000 square feet) when all the press equipments were operating. The sampling duration was 1 h, during which 20 thousand papers were printed. As the sampling for auto-painting, windows in the sampling rooms were also closed. The average temperature in the printing factory was 23.2 °C.

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