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An updated emission inventory of vehicular VOCs and IVOCs in China

Huan Liu[1,2], Hanyang Man[1,2], Hongyang Cui[3], Yanjun Wang[4], Fanyuan Deng[1], Yue Wang[1], Xiaofan Yang[1], Qian Xiao[1], Qiang Zhang[3], Yan Ding[4], and Kebin He[1,2]

[1]State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, 100084, China
[2]State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing, 100084, China
[3]Ministry of Education Key Laboratory for Earth System Modeling, Center for Earth System Science, Tsinghua University, Beijing, 100084, China
[4]Vehicle Emission Control Center (VECC) of the Ministry of Environmental Protection, Beijing, 100084, China

Correspondence to: Huan Liu (liu_env@tsinghua.edu.cn)

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Abstract. Currently, the emission inventory of vehicular volatile organic compounds (VOCs) is one of those with the largest errors and uncertainties due to suboptimal estimation methods and the lack of first-hand basic data. In this study, an updated speciated emission inventory of VOCs and an estimation of intermediate-volatility organic compounds (IVOCs) from vehicles in China at the provincial level for the year of 2015 are developed based on a set of state-of-the-art methods and an abundance of local measurement data. Activity data for light-duty vehicles are derived from trajectories of more than 70 000 cars for 1 year. The annual mileage of trucks are calculated from reported data by more than 2 million trucks in China. The emission profiles are updated using measurement data. Vehicular tailpipe emissions (VTEs) and four types of vehicular evaporation emissions (VEEs), including refueling, hot soak, diurnal and running loss, are taken into account. Results show that the total vehicular VOC emissions in China are 4.21 Tg (with a 95 % confidence interval range from 2.90 to 6.54 Tg) and the IVOC emissions are 200.37 Gg in 2015. VTEs are still the predominant contributor, while VEEs are responsible for 39.20 % of VOC emissions. The control of VEEs is yet to be optimized in China. Among VTEs, passenger vehicles emissions have the largest share (49.86 %), followed by trucks (28.15 %) and motorcycles (21.99 %). Among VEEs, running loss is the largest contributor (81.05 %). For both VTEs and VEEs, Guangdong, Shandong and Jiangsu province are three of the highest, with a respective contribution of 10.66, 8.85 and 6.54 % to the total amounts of VOCs from vehicles. 97 VOC species are analyzed in this VOC emission inventory. *i*-Pentane, toluene and formaldehyde are found to be the most abundant species in China's vehicular VOC emissions. The estimated IVOCs are another "inconvenient truth", concluding that precursor emissions for secondary organic aerosol (SOA) from vehicles are much larger than previously estimated.

1 Introduction

China is one of the countries most threatened by simultaneous pollution of $PM_{2.5}$ (particulate matter with aerodynamic diameters of less than 2.5 µm) and ozone (Sitch et al., 2007; van Donkelaar et al., 2015; Liu et al., 2016). Approximately 1.36 million premature deaths in China were attributed to these two major pollutants in 2010 (Lelieveld et al., 2015). Previous studies have shown that secondary organic aerosols (SOAs) make up a significant proportion of the ambient $PM_{2.5}$ mass in Chinese cities (Cui et al., 2015). Intermediate-volatility organic compounds (IVOCs) are a series of compounds with effective saturated concentration between 10^3 and 10^6 µg m^{-3}, similar to the volatility range of C12–C22 *n*-alkanes (Zhao et al., 2014). Recent studies suggested that both volatile organic compounds (VOCs) and IVOCs contributed to SOA formation with IVOCs being dominant in

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certain regions (Huang et al., 2014; Robinson et al., 2007; Hodzic et al., 2010). Studies on ozone pollution also demonstrated that ozone formation was caused by VOCs in many major Chinese cities (Geng et al., 2008; Shao et al., 2009). VOCs and IVOCs should be monitored for their impacts on air quality and public health.

Previous studies have repeatedly reported that, among the various anthropogenic emission sources in Chinese cities, vehicles were the predominant contributor to both VOC emissions and ambient VOC concentrations (Song et al., 2008; Zheng et al., 2009; Wang et al., 2010; Shao et al., 2011; Cui et al., 2015). A comprehensive and accurate national emission inventory is critical to the design of effective abatement strategies for pollution on a national level. Cai and Xie (2009) reported a VOC emission inventory from on-road vehicles in China during 1980–2005. Several other studies also included vehicles as a part of the transportation section in their comprehensive emission inventories of VOCs (Tonooka et al., 2001; Klimont et al., 2002; Streets et al., 2003; Li et al., 2003, 2014; Cai and Xie, 2007; Bo et al., 2008; Liu et al., 2008; Wei et al., 2008; Zhang et al., 2009; Cao et al., 2011; Zheng et al., 2014). The complete summary of existing studies on the vehicular VOC emission inventory and their respective performance are shown in Table S1 in the Supplement. These existing emission inventories have greatly improved our understanding of VOC emissions. It is worth noting that all studies mentioned above targeted the emission inventory prior to 2010 while omitting IVOC impacts. Considering the dramatic increase in the number of vehicles, the establishment of new emission inventories is of urgent priority. However, multiple key factors changed in the last 10 years that require updated methods and data.

First of all, the dominant VOC emission processes of vehicles may switch. Compared with vehicular tailpipe emissions (VTEs), vehicular evaporation emissions (VEEs) have recently been reported to be non-negligible contributors to ambient VOC concentrations (Yamada et al., 2013; Liu et al., 2015). VOC emissions evaporate from gasoline-fueled vehicles consistently regardless of their status – refueling, running or parked. Vapors generated in VEEs either pass through the equipped carbon canister or permeate elastomers of the vehicle's fuel system before entering the ambient atmosphere. Depending on the vehicle's status, evaporative emissions come in four varieties: refueling loss, running loss, hot soak loss and diurnal loss. Local emission factors (EFs) and profiles of VEEs are necessary for VEEs inventory as they are highly related to local gasoline formula and vehicle controls. In addition, a more sophisticated method is necessary to estimate VEEs. Details are further described in Sect. 2.

Secondly, as dominant precursors of SOA, IVOCs have strong impacts on air quality, global climate and human health. However, there are few studies on IVOCs due to their complex composition. Shortage of systematic and integrated analytical methods limits the progress for measurement and quantification of IVOCs (Goldstein and Galbally, 2007; Jathar et al., 2014). Therefore, few studies successfully provided emission measurement results of IVOCs. To our knowledge, an IVOC emission inventory for China is yet to be reported.

Most importantly, vehicle activity is crucial in total emission estimation, and the big data would greatly reduce uncertainty of the emission inventory in previous studies; these parameters were usually hypothesized based on experiences of other countries or surveys from limited samples (usually less than 2000) (Liu et al., 2007; Yang et al., 2015a). With the development of transportation networking technology, we were able to acquire Global Positioning System (GPS) records of 71 059 cars for research purposes without any personal information. This data covered 30 provinces in China, which would significantly improve our understanding of vehicle usage and improve our estimation with accuracy and comprehensiveness.

In addition, several other new methods and local data are integrated to improve the inventory. Provincial emissions were typically calculated using local registration numbers, which presume that all vehicles were operated locally, although an acceptable assumption for household vehicles is irrelevant for freight trucks. A more comprehensive road-emission-intensity-based (REIB) approach was developed, in which the spatial distribution of emissions were estimated based on the total length of each road type in a province and on the corresponding emission intensity of the road type. This method greatly improved NO_x and PM emission estimation for long-distance inter-province or intercity cargo transportation (Yang et al., 2015a).

Imperfections in comprehensiveness and accuracy of estimation can also be improved by using local emission factors and speciation published recently (Liu et al., 2009, 2015; Yao et al., 2015; Zhang et al., 2015; Cao et al., 2016). Instead of emission factors given by commonly used vehicle emission models developed by the US and Europe – e.g., COPERT, MOVES, MOBILE and IVE – the measured local emission factors offer a relevant and more accurate estimation of local emission levels. Additionally, chemical profiles obtained by experiments in Western countries could not reflect the chemical characteristics of VOCs from vehicles in China accurately. The recent speciation profiles were reported using China's local fuel.

Furthermore, the national statistical data in China only provide vehicle number data classified by vehicle type (e.g., light-duty passenger vehicles, heavy-duty trucks). More detailed vehicle number data by fuel type and emission control technology are required to calculate emissions as they were reported to have distinct influences on emission factors (Huo et al., 2012; Zhang et al., 2015; Cao et al., 2016).

In this study, an updated speciation-based emission inventory of VOCs and an estimation of IVOCs from vehicles in China in 2015 are developed using a set of state-of-the-art methods. The lack of comprehensiveness and accuracy in existing methods is solved for each and individually based on scientific calculating methodologies, big data and abundant local emission measurements. The IVOC emission factors used are derived from studies in the US by matching corresponding vehicle emission categories of the two countries, as no local IVOC emission factors were reported.

2 Methodology and data

2.1 Vehicle stock and classification

In total, 25 types of on-road vehicles were considered in this study, including passenger vehicles, trucks and gasoline motorcycles (GMs). Passenger vehicles were further divided into 18 types: light-duty gasoline passenger vehicles excluding taxies (LDGPVs), light-duty diesel passenger vehicles excluding taxies (LDDPVs), light-duty alternative-fuel passenger vehicles excluding taxies (LDAPVs), medium-duty gasoline passenger vehicles excluding buses (MDGPVs), medium-duty diesel passenger vehicles excluding buses (MDDPVs), medium-duty alternative-fuel passenger vehicles excluding buses (MDAPVs), heavy-duty gasoline passenger vehicles excluding buses (HDGPVs), heavy-duty diesel passenger vehicles excluding buses (HDDPVs), heavy-duty alternative-fuel passenger vehicles excluding buses (HDAPVs), light-duty gasoline taxis (LDGTAs), light-duty diesel taxis (LDDTAs), light-duty alternative-fuel taxis (LDATAs), medium-duty gasoline buses (MDGBUs), medium-duty diesel buses (MDDBUs), medium-duty alternative-fuel buses (MDABUs), heavy-duty gasoline buses (HDGBUs), heavy-duty diesel buses (HDDBUs) and heavy-duty alternative-fuel buses (HDABUs). For passenger vehicles, light-duty refers to vehicles with length less than 6000 mm and ridership of no more than nine. Medium-duty refers to vehicles of length less than 6000 mm and ridership between 10 and 19. Heavy-duty refers to vehicles of length no less than 6000 mm or ridership of no less than 20. These vehicles were further classified by emission control technologies (i.e., the vehicle emission standards China 0, China 1, China 2, China 3, China 4 and above). Alternative-fuel vehicles in this study include compressed natural gas (CNG), liquefied natural gas (LNG) and liquefied petroleum gas (LPG) vehicles.

Trucks (or freight trucks) were divided into 6 types: light-duty gasoline trucks (LDGTs), light-duty diesel trucks (LDDTs), medium-duty gasoline trucks (MDGTs), medium-duty diesel trucks (MDDTs), heavy-duty gasoline trucks (HDGTs), and heavy-duty diesel trucks (HDDTs). For trucks, a light-duty truck refers to vehicles of mass less than 3500 kg. A medium-duty truck refers to vehicles of mass ranging from 3500 to 12 000 kg. A heavy-duty truck refers to vehicles of mass more than 12 000 kg.

Detailed provincial data of all the number of vehicles excluding GMs in 2015 were obtained by complete statistical survey conducted by the Vehicle Emission Control Center (VECC) of China's Ministry of Environmental Protection (MEP), which could be considered highly accurate. The provincial number of GMs in 2015 was obtained from the Statistical Yearbook 2016 of each province.

2.2 Vehicle activity

The real-world vehicle activity data used in this study were derived by statistical surveys, field tests and literature review.

The provincial annual vehicle-kilometers-traveled (VKT) data of light-duty passenger vehicles (LDPVs), which were the majority in the fleet and thus had the largest impact on the emission inventory, were acquired by processing and analyzing the big data of GPS records (71 059 cars). Driving frequency of different types of trucks on different kinds of roads (e.g., freeway, national road, provincial road and urban road) was acquired by analysis of survey data from 1060 valid questionnaires, which was introduced in detail in our previous study (Yang et al., 2015a).

In addition, provincial parking characteristics data for evaporative emission calculation, including parking event numbers and parking durations, were also obtained by analysis of the GPS big data.

The average mileage for trucks was obtained from a commercial source with data feeding from more than 2 million trucks, mainly commercial vehicles installed with either the GPS or China's BeiDou Navigation Satellite System (BDS). Location, speed and vehicle-type information are live-fed to the commercial platform. The VKT for each truck category was calculated using the monitored data from the platform.

2.3 Vehicular emission data and estimation

The vehicular VOC emissions at the provincial level were divided into three parts for calculation, including tailpipe emissions from non-truck vehicles (i.e., passenger vehicles, taxis, buses and motorcycles), tailpipe emissions from freight trucks and evaporation emissions from gasoline vehicles. Results from the three parts were summed to yield the total provincial emission amounts in 2015. Emission factors for VOCs used were derived from lab tests, field tests and literature review (MEP, 2015; Zhang et al., 2014; Liu et al., 2015). The IVOC emission calculation was similar to that of the VOCs, while only the tailpipe exhaust for non-GMs was taken into consideration. For IVOC emission factors, Zhao et al. (2015, 2016) reported a series of measurements for gasoline and diesel vehicles (Tables S2 and S3). Details are introduced in the following.

2.3.1 Tailpipe emissions from non-truck vehicles

For a given province, the tailpipe VOC and IVOC emissions from non-truck vehicles were estimated by Eq. (1):

$$E_{\text{tailpipe,non-truck},i} = \sum_j \sum_k \left(\text{EF}_{\text{tailpipe},j,k} \times \text{VP}_{i,j,k} \times \text{VKT}_{i,j}\right), \quad (1)$$

where $E_{\text{tailpipe,non-truck},i}$ represents the annual tailpipe emissions from non-truck vehicles in province i (g yr^{-1}); $\text{EF}_{\text{tailpipe},j,k}$ represents the tailpipe VOC and IVOC emission factor of vehicle-type j with emission control technology k (g km^{-1}); $\text{VP}_{i,j,k}$ represents the registered number of vehicle-type j with emission control technology k in province i; $\text{VKT}_{i,j}$ represents the annual VKT of vehicle-type j in province i (km yr^{-1}).

For VTEs of all vehicles excluding trucks, the emission factors of VOCs obtained by abundant real-world emission tests conducted by our Tsinghua University research group and VECC of China's MEP were adopted ("Technical guidelines on emission inventory development of air pollutants from on-road vehicles (on trial)") (Table S4).

For IVOC emission factors, Zhao et al. (2015, 2016) reported a series of measurements for gasoline and diesel vehicles. By considering the age of a specific vehicle model, after-treatment devices and emission certification standard, each of the tested vehicles was matched to a corresponding category of China emission certification standard (Table S2). Thus, the emission factors for some vehicle categories were set up. For the categories lacking measurements (gasoline vehicles before China 1 and all diesel vehicles), emission factors were set identically to the China 1 category. For diesel passenger vehicles, the current IVOC emission factors were set identically to the corresponding level of gasoline vehicles. The IVOC emission factors were converted from the original unit of mg kg-fuel^{-1} to g km^{-1} using fuel consumption data per km. For each category, the median of emission factors was used for the particular type of vehicles if more than one available test is present. Detailed emission factors are listed in Table S5.

2.3.2 Tailpipe emissions from freight trucks

Considering the fact that the majority of freight trucks are used for long-distance intercity or inter-province cargo transportation, the REIB approach, instead of the traditional local-registration-based approach, was utilized to calculate truck emissions, as was described in detail in our previous work (Yang et al., 2015a). The provincial tailpipe emissions from freight trucks are estimated by Eq. (2):

$$E_{\text{tailpipe, truck},i} = \sum_j \sum_k \sum_m \left[\frac{\left(\text{EF}_{\text{tailpipe},j,k} \times \text{VP}_{j,k} \times \text{VKT}_{j,k} \times \text{DP}_{j,m}\right) \times L_{i,m}}{L_m}\right], \quad (2)$$

where $E_{\text{tailpipe,truck},i}$ represents the annual tailpipe VOC and IVOC emissions from freight trucks in province i (g yr^{-1}); $\text{EF}_{\text{tailpipe},j,k}$ represents the tailpipe VOC and IVOC emission factor of vehicle-type j with emission control technology k (g km^{-1}); $\text{VP}_{j,k}$ represents the national number of vehicle-type j with emission control technology k; and $\text{VKT}_{j,k}$ represents the annual VKT of vehicle-type j with emission control technology k (km yr^{-1}); $\text{DP}_{j,m}$ represents the distance portion of vehicle-type j running on road-type m; $L_{i,m}$ and L_m represent the total length of road-type m in province i and in China respectively (km).

For VTEs of trucks, the operating-mode-bin-based method introduced in our previous study was used to investigate real-world emission factors for VOCs (Yang et al., 2015a). Firstly, second-by-second vehicle-specific power (VSP) and engine stress (ES) data were calculated using GPS records of 16 trucks with equations suggested by the MOVES and IVE models respectively. Followed by identification of 30 operating mode bins based on VSP data and ES data, the time fraction of each bin was given. Finally, the distance-based emission factors for trucks of various types on various roads were calculated according to the emission rate of each bin that was presented in our previous test results (Liu et al., 2009).

For IVOC emission factors, a map to match the US emission certification level to the China emission level was built (Table S3). Only non-GM vehicles were considered for IVOC emissions evaluation. The emission factors for the US fleet were converted for use with China's trucks. As no data were available for most categories, the following assumptions had to be made to fill the gap: (1) medium- and heavy-duty trucks had identical emission factors that were 50 % higher than light-duty trucks, similar to emission ratios of VOCs and primary organic aerosol of the same types; (2) emission levels of neighboring types were used in the case of lacking data. The final assumptions for IVOC emission factors were introduced in Table S5.

2.3.3 Evaporation emissions from gasoline vehicles – diurnal and hot soak

Hot soak and diurnal emissions both occur when vehicles are parked. Diurnal loss is defined as the gasoline vapors that are generated and emitted while vehicles are parked. The diurnal and hot soak emission factors were obtained by a set of Sealed Housing for Evaporative Determination (SHED) tests, as was introduced in our previous study (Liu et al., 2015). The detailed emission factors are summarized in Table S6. The provincial annual diurnal emissions from non-GM gasoline vehicles and GMs are calculated by Eqs. (3)–(6) and (7) respectively. For diurnal emissions, we calculated total parking hours for each parking event and adjust emissions based on how long the vehicle was parked. The first hour for each parking event was treated as the hot soak and was subtracted from the diurnal emissions.

$$E_{\text{diur,non-GM},i} = E_{\text{diur},<24,\text{non-GM},i} + E_{\text{diur},24\text{–}48,\text{non-GM},i} + E_{\text{diur},>48,\text{non-GM},i}, \quad (3)$$

$$E_{\text{diur},<24,\text{non-GM},i} = \big[\text{EF}_{\text{diur},<24,\text{LDGPVs}} \times \big(P_{\text{duration},1\text{–}24,i} \times T_i - P_{\text{event},1\text{–}24,i} \times N_i \times 1\big)\big] \times 365 \times \text{VP}_{i,\text{non-GM}}, \quad (4)$$

$$E_{\text{diur},24\text{–}48,\text{non-GM},i} = \big[\text{EF}_{\text{diur},<24,\text{LDGPVs}} \times P_{\text{event},24\text{–}48,i} \times N_i \times 23 + \text{EF}_{\text{diur},24\text{–}48,\text{LDGPVs}} \times \big(P_{\text{duration},24\text{–}48,i} \times T_i - P_{\text{event},24\text{–}48,i} \times N_i \times 24\big)\big] \times 365 \times \text{VP}_{i,\text{non-GM}}, \quad (5)$$

$$E_{\text{diur},>48,\text{non-GM},i} = \big[\text{EF}_{\text{diur},<24,\text{LDGPVs}} \times P_{\text{event},>48,i} \times N_i \times 23 + \text{EF}_{\text{diur},24\text{–}48,\text{LDGPVs}} \times P_{\text{event},>48,i} \times N_i \times 24 + \text{EF}_{\text{diur},48\text{–}72,\text{LDGPVs}} \times \big(P_{\text{duration},>48,i} \times T_i - P_{\text{event},>48,i} \times N_i \times 48\big)\big] \times 365 \times \text{VP}_{i,\text{non-GM}}, \quad (6)$$

where $E_{\text{diur,non-GM},i}$ represents the total annual diurnal (simultaneous permeation included) emissions from non-GM gasoline vehicles registered in province i (g yr^{-1}); $E_{\text{diur},<24,\text{non-GM},i}$, $E_{\text{diur},24\text{–}48,\text{non-GM},i}$, and $E_{\text{diur},>48,\text{non-GM},i}$ represent the annual diurnal (simultaneous permeation included) emissions that occurred respectively in the first day, second day, and third day, and after parking (g yr^{-1}); $\text{EF}_{\text{diur},<24,\text{LDGPVs}}$, $\text{EF}_{\text{diur},24\text{–}48,\text{LDGPVs}}$, and $\text{EF}_{\text{diur},48\text{–}72,\text{LDGPVs}}$ represent the measured diurnal (simultaneous permeation included) emission factors of China 4 LDGVs (g h^{-1}). The evaporative emission control remained the same until China 6. Thus, there is no progress on emission reduction since China 1 to China 5 on evaporation. Therefore, the emission factors of China 4 LDGVs could be used for all LDGVs. For the other vehicle types, no data are available from tests and the same EFs with LDGVs were used. $P_{\text{event},1\text{–}24,i}$, $P_{\text{event},24\text{–}48,i}$, and $P_{\text{event},>48,i}$ represent the percentage of parking events with a duration of 1–24, 24–48 and above 48 h; $P_{\text{duration},1\text{–}24,i}$, $P_{\text{duration},24\text{–}48,i}$, and $P_{\text{duration},>48,i}$ represent the percentage of total parking events with a duration between 1 and 24 h, between 24 and 48 h, and above 48 h. N_i represents the annual average parking events per day per vehicle in province i. T_i represents the average parking duration per day per vehicle in province i (h). $\text{VP}_{i,\text{non-GM}}$ represents the registered number of vehicles excluding motorcycles in province i.

For motorcycles, the calculation of evaporative emissions was simplified. Because the activity data could not support the calculation of diurnal, refueling, hot soak or running loss, we therefore use the following equation to calculate total evaporative emissions for GMs based on the mileage:

$$E_{\text{GMs},i} = \text{EF}_{\text{GMs}} \times \text{VP}_{i,\text{GMs}} \times \text{VKT}_{i,\text{GMs}}, \quad (7)$$

where $E_{\text{GMs},i}$ represents the annual evaporative emissions from GMs registered in province i (g yr^{-1}); EF_{GMs} represents the evaporative emission factor of GMs (g km^{-1}); for VEEs from GMs, the emission factors given by the International Council on Clean Transportation were utilized (ICCT, 2007) (Table S6). $\text{VKT}_{i,\text{GMs}}$ represents the annual VKT of GMs in province i (km yr^{-1}).

According to the US EPA, hot soak is defined as the evaporative losses within a 1 h period after shutting down of engines (EPA, 2001). Any vapor losses that occur after are considered diurnal emissions. The provincial hot soak emissions for non-GM gasoline vehicles (i.e., LDGPVs, MDGPVs, HDGPVs, LDGTAs, GBUs, LDGTs, MDGTs and HDGTs) are calculated by Eq. (8):

$$E_{\text{soak,non-GM},i} = \text{EF}_{\text{soak,LDGPVs}} \times \big[\big(T_i \times 365 \times P_{\text{duration},<1,i}\big) + \big(N_i \times 365 \times P_{\text{event},>1,i} \times 1\big)\big] \times \text{VP}_{i,\text{non-GMs}}, \quad (8)$$

where $E_{\text{soak,non-GM},i}$ represents the annual hot soak (simultaneous permeation included) emissions from non-GM vehicles in province i (g yr^{-1}); $\text{EF}_{\text{soak,LDGPVs}}$ represents the hot soak (simultaneous permeation included) emission factor of LDGPVs (g h^{-1}); T_i represents the average parking duration per day per vehicle in province i (h); N_i represents the average parking events per day per vehicle in province i; $P_{\text{duration},<1,i}$ represents the percentage of total parking duration shorter than 1 h in province i; $P_{\text{event},>1,i}$ represents the percentage of parking events with a duration shorter than 1 h in province i; and $\text{VP}_{i,\text{non-GMs}}$ represents the non-GM gasoline number of vehicles in province i.

2.3.4 Evaporation emissions from gasoline vehicles – refueling

China follows European countries in the popularization of Stage-II vapor control systems for reduction of refueling loss in stations. The vehicle refueling emissions were also measured by our team from SHED tests (Yang et al., 2015b). The provincial refueling emissions from gasoline vehicles are calculated by Eq. (9). The control efficiency and the percentages of gasoline stations equipped with Stage-II systems are the two key factors influencing the final emissions.

$$E_{\text{refuel},i} = \text{EF}_{\text{refuel}} \times [(1-\theta) \times w_i + (1-w_i)] \times \text{CF}_i, \quad (9)$$

where $E_{\text{refuel},i}$ represents the annual refueling emissions from gasoline vehicles in province i (g yr^{-1}); $\text{EF}_{\text{refuel}}$ represents the refueling emission factor for non-control conditions (g L^{-1}); θ represents the average efficiency of the Stage-II vapor control system, which is 82 % according to our measurements in Beijing; ω_i represents the percentage of filling stations equipped with Stage-II vapor control systems in province i – 100 % in Beijing, 90 % in Shanghai and Guangdong, 60 % in Tianjin and Hebei, and 0 % in other provinces in this study according to survey; CF_i represents the annual motor gasoline consumption in province i (L yr^{-1}), which was retrieved from official statistics (Department of Energy Statistics, National Bureau of Statistics, People's Republic of China, 2016) – 85 % of total gasoline consumption was attributed to on-road-vehicle use.

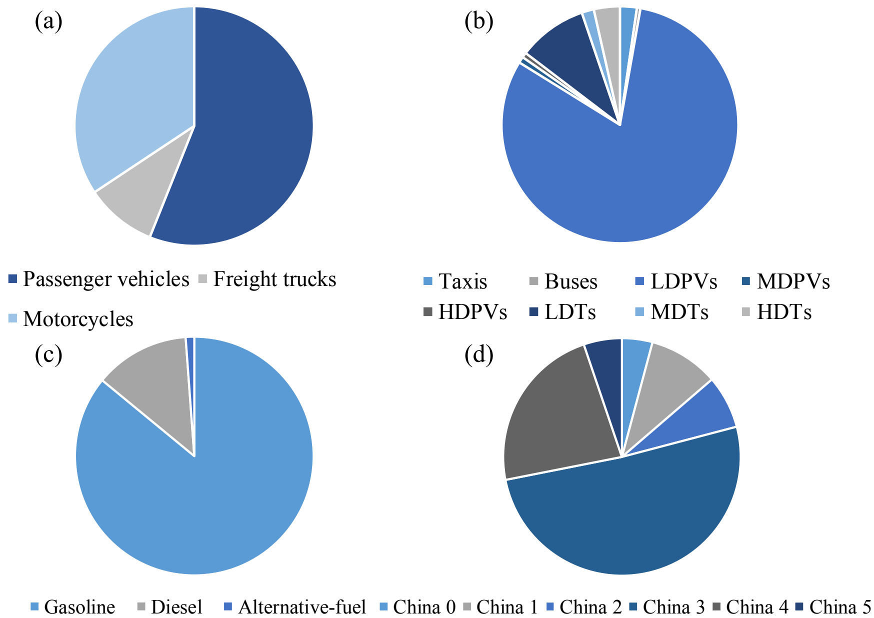

Figure 1. The percentages by vehicle types, fuel types and emission levels of China vehicle fleet.

2.3.5 Evaporation emissions from gasoline vehicles – running loss

Vehicle running loss occurs during operation of the engine through the fuel system – not from tailpipe. The provincial annual running loss emissions from non-GM gasoline vehicles are calculated by Eq. (10):

$$E_{\text{running,non-GM},i} = \text{EF}_{\text{running,LDGPVs}} \times (24 - T_i) \times 365 \times \text{VP}_{i,\text{non-GM}}, \quad (10)$$

where $E_{\text{running,non-GM},i}$ represents the annual running loss emissions from non-GM gasoline vehicles registered in province i (g yr^{-1}); $\text{EF}_{\text{running,LDGPVs}}$ represents the running loss emission factor of LDGVs (g h^{-1}). The emission factors of running loss were acquired from MOVES model due to the lack of local lab-test results (EPA, 2012). T_i represents the average parking duration per day per vehicle in province i (h). $\text{VP}_{i,\text{non-GM}}$ represents the registered number of vehicles excluding motorcycles in province i.

2.3.6 Uncertainty analysis

The uncertainty for the emission inventory is assessed using a Monte Carlo method. This method is an effective and versatile tool for determining uncertainties and has been used widely in previous research on emissions inventory (Zhang et al., 2014; Yang et al., 2015a; Liu et al., 2016; Wang et al., 2008). The probability distributions of key model parameters were established with our experimental data, investigation data and literature review (Table S7) (Zhang et al., 2014; Yang et al., 2015a). Using these assumptions, a Monte Carlo model was run 10 000 times to produce the estimate.

2.4 Species analysis

The vehicular VOC emissions speciation is further determined by Eq. (11):

$$E_{\text{speciated}} = E_{\text{tailpipe,gasoline}} \times \text{PR}_{\text{tailpipe,gasoline}} + E_{\text{tailpipe,diesel}} \times \text{PR}_{\text{tailpipe,diesel}} + E_{\text{evap}} \times \text{PR}_{\text{evap}}, \quad (11)$$

where $E_{\text{speciated}}$ represents the speciated annual VOC emissions from on-road vehicles registered in province i (g yr^{-1}); $E_{\text{tailpipe,gasoline}}$, $E_{\text{tailpipe,diesel}}$, and E_{evap} represent the annual tailpipe VOC emissions from gasoline vehicles (alternative-fuel vehicles included), the annual tailpipe VOC emissions from diesel vehicles and the annual evaporative VOC emissions respectively; $\text{PR}_{\text{tailpipe,gasoline}}$, $\text{PR}_{\text{tailpipe,diesel}}$, and PR_{evap} represent the measured VOC profiles of tailpipe emissions from gasoline vehicles, tailpipe emissions from diesel vehicles and evaporative emissions respectively.

The VOC profiles used in this study for establishment of the vehicular VOC emission inventory were derived from literature review and lab tests. Tailpipe VOC emissions from gasoline vehicles and diesel vehicles were derived from corresponding local profiles reported according to on-board exhaust tests with 18 in-use diesel trucks and 30 in-use light-duty gasoline vehicles in Beijing (Yao et al., 2015; Cao et al., 2016). For vehicle evaporative emissions, a comprehensive species profile was obtained based on results from the 30 crossover evaporative tests we conducted before (Man et al., 2016).

Table 1. Total number of vehicles of different types in China in 2015.

Vehicle type	Total number	Fuel-type percentage (%)			Emission control technology	Total number
		Gasoline	Diesel	Alternative fuels		
LDPVs	137 599 368	97.96	1.15	0.90	China 0	7 062 516
MDPVs	1 428 102	56.53	40.68	2.78	China 1	16 181 788
HDPVs	1 165 836	15.97	75.03	9.00	China 2	12 251 006
LDTs	15 998 479	41.50	58.50	0.00	China 3	86 584 457
MDTs	2 826 881	18.92	81.08	0.00	China 4	38 880 534
HDTs	6 037 719	7.65	92.35	0.00	China 5	8 834 416
TAs	3 910 397	61.89	29.37	8.74		
BUs	827 935	13.76	55.39	30.85		
GMs	88 759 010	100	0	0		

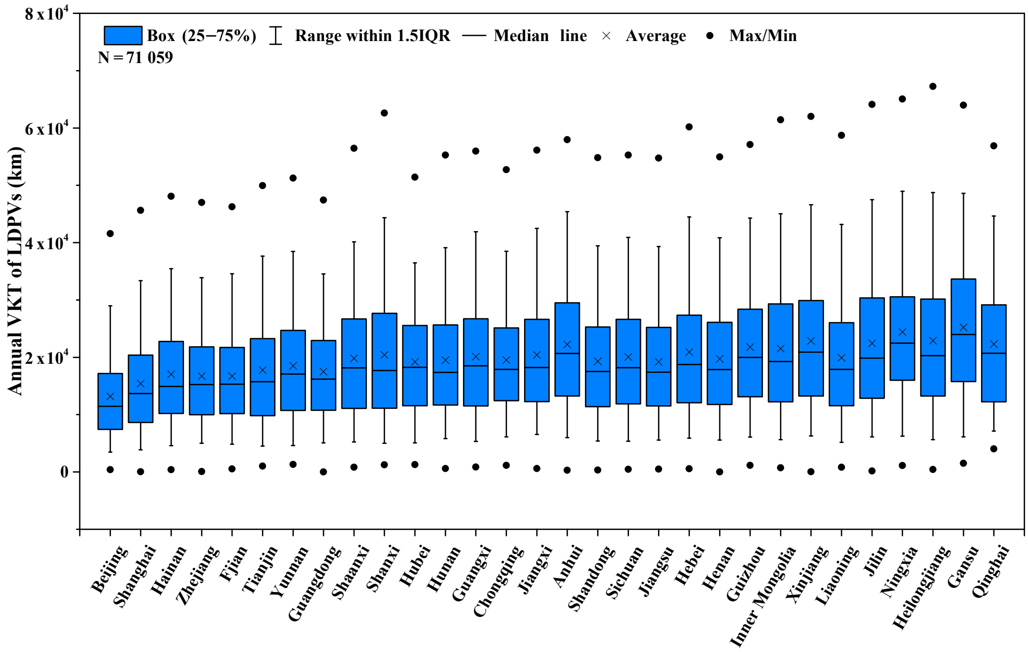

Figure 2. Provincial annual VKT of LDPVs in China.

3 Results and discussion

3.1 Activity characteristics of vehicles

3.1.1 Number of vehicles

GMs and non-GM vehicles contributed 34.3 % (88 759 010) and 65.7 % (169 794 718) respectively among the 259 million total on-road vehicles in China in the year 2015 (Fig. 1). LDPVs were the predominant contributors among non-GM vehicles, with a proportion of 81.0 %, followed by light-duty trucks (LDTs, 9.4 %), heavy-duty trucks (HDTs, 3.6 %), taxis (TAs, 2.3 %), medium-duty trucks (MDTs, 1.7 %), medium-duty passenger vehicles (MDPVs, 0.8 %), heavy-duty passenger vehicles (HDPVs, 0.7 %) and buses (BUs, 0.5 %). In terms of emission control technologies, China 3 vehicles accounted for the largest proportion (51.0 %) of China's non-GM vehicle fleet, followed by China 4 (22.9 %), China 1 (9.5 %), China 2 (7.2 %), China 5 (5.2 %) and China 0 (4.2 %) vehicles. China 1, China 2 and China 4 made up 9.5, 15.69 and 10.12 % respectively of the fleet structure of 2012, which all substantially changed in 2015 with gradual elimination of older vehicles and addition of new ones. In terms of automotive fuels, gasoline was the most common fuel for non-GM vehicles in China, with a proportion of 86.0 %, while diesel and alternative fuels were substantially lower, with proportions of 12.9 and 1.2 % respectively. Table 1 summarized the number of vehicles and the corre-

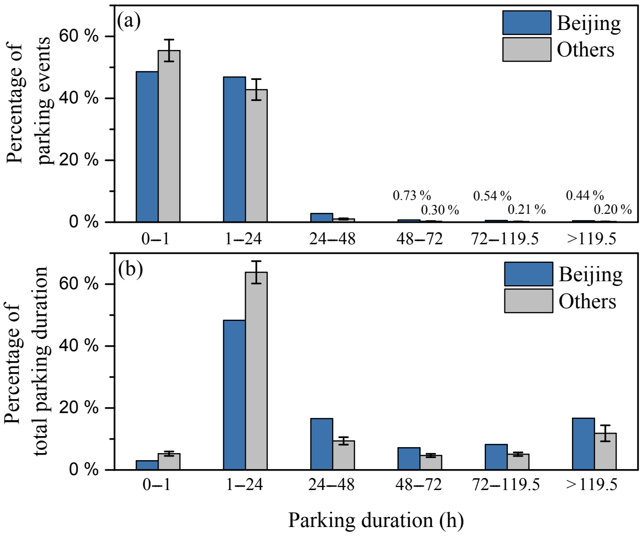

Figure 3. Real-world parking duration distribution: **(a)** percentage of parking events; **(b)** percentage of total parking duration.

sponding proportions classified by fuel types. LDPVs, MDPVs and TAs were mainly fueled by gasoline, while HDPVs, LDTs, MDTs, HDTs and BUs were primarily fueled by diesel. The proportion of diesel-consuming vehicles increases with vehicle weight in both passenger vehicles and freight trucks. Some local governments have released very strong promotion policies for using alternative fuels for BUs and TAs. Therefore, the ratio of alternative fuels for BUs and TAs exceeds 8.7 %.

3.1.2 VKT characteristics

GPS records of 71 059 vehicles operating in 30 provinces from 1 July 2014 to 1 July 2015, including 931 581 667 km driving distances and 1 585 771 787 511 valid seconds, were collected and analyzed to obtain real-world VKT characteristics of LDPVs in different provinces. It was found that the national average VKT of LDPVs in China was 18 886 ± 10 469 km per vehicle per year. Provincial annual average VKT values with vehicle sample sizes are shown in Table 2. Distribution characteristics of annual VKT data in each province are shown in Fig. 2. The annual average VKT data of LDPVs in Beijing and Shanghai, which both are among the most developed cities in the world, were much lower than the national average value given by this study. Average VKT was also much lower than the corresponding local values given by surveys conducted in 2004 (Liu et al., 2007). This phenomenon could be explained by three facts. Firstly, per capita ownership of cars in Beijing and Shanghai during our sampling periods was much higher than the national average value which was similar to the corresponding local values in 2004. A considerable number of families in both cities own multiple vehicles nowadays, causing a decrease in annual VKT of individual cars under the assumption that their regular commuting distances have not substantially changed. Secondly, heavy traffic control policies were enforced in Beijing and Shanghai during recent years, resulting in longer parking duration and hence a smaller annual VKT of vehicles in these two cities. Thirdly, an increase in public transportation usage was observed in Beijing and Shanghai due to the growing traffic jams during peak hours.

Table 2. Provincial annual average VKT of LDPVs in China.

Province*	Vehicle sample size	Annual average VKT (km)
Beijing	2645	13 169 ± 7741
Shanghai	3833	15 389 ± 8972
Hainan	581	16 941 ± 9508
Zhejiang	6356	16 740 ± 8897
Fujian	3059	16 726 ± 8784
Tianjin	772	17 785 ± 10 308
Yunnan	1370	18 609 ± 10 307
Guangdong	16 553	17 503 ± 8952
Shaanxi	1766	19 866 ± 10 964
Shanxi	1225	20 466 ± 12 131
Hubei	976	19 313 ± 9669
Hunan	1320	19 524 ± 10 545
Guangxi	1086	20 251 ± 11 231
Chongqing	1279	19 529 ± 10 022
Jiangxi	903	20 406 ± 10 982
Anhui	1007	22 209 ± 11 744
Shandong	2449	19 333 ± 10 420
Sichuan	1984	20 120 ± 10 959
Jiangsu	5066	19 238 ± 10 331
Hebei	2933	20 915 ± 11 594
Henan	1818	19 759 ± 10 693
Guizhou	746	21 985 ± 11 800
Inner Mongolia	2322	21 660 ± 12 118
Xinjiang	991	22 901 ± 12 122
Liaoning	4049	19 953 ± 11 365
Jilin	1386	22 400 ± 12 630
Ningxia	418	24 345 ± 12 810
Qinghai	171	22 488 ± 12 265
Heilongjiang	1552	23 008 ± 13 102
Gansu	443	25 460 ± 12 659

* There are no VKT data for Tibet, and we used the national average, which was calculated using the data of the other 30 provinces, to represent the annual VKT of Tibet in this study.

Table 3 summarized vehicle mileage of other vehicle types in China. VKT values of trucks are significantly influenced by vehicle age. The annual mileage of China 0 and China 1 trucks were much lower than vehicles of the same type with better emission control technologies. The aging of trucks in China greatly impacts their performances due to the common practice of overloading, which expedites the integrity loss of the trucks' internal parts. Several cities have implemented low emission zones to restrict entry of trucks with outdated emission control technologies.

Table 3. Average annual VKT in China (km yr^{-1}).

	LDGTs	LDDTs	MDGTs	MDDTs	HDGTs	HDDTs	TAs	BUs	MDPVs	LDPVs
China 0	22 160	19 270	35 196	21 231	27 716	24 372	138 000	50 000	31 300	114 800
China 1	22 160	19 270	35 196	21 231	27 716	24 372				
China 2	26 335	26 964	40 766	28 140	33 226	38 485				
China 3	29 467	36 581	47 927	36 366	40 310	64 128				
China 4	34 165	45 237	53 497	60 308	45 820	98 206				
China 5	34 165	45 237	53 497	60 308	45 820	98 206				

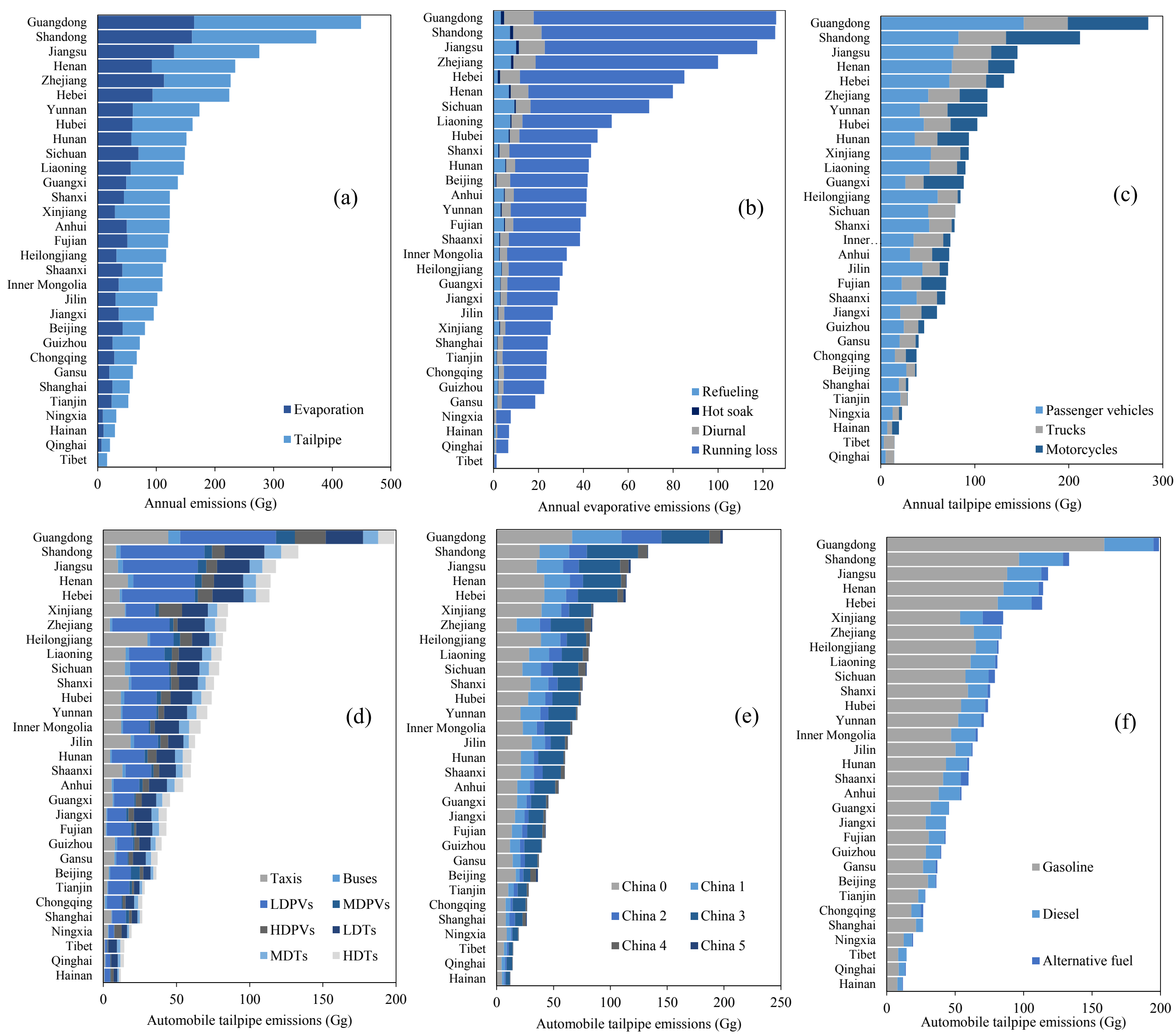

Figure 4. Provincial VOC emissions from vehicles in 2015: **(a)** total emission amount classified by emission sources; **(b)** evaporation emission amount classified by evaporation processes (motorcycles excluded); **(c)** tailpipe emission amount classified by vehicle types; **(d–f)** tailpipe emission amounts classified by detailed categories, emission certification levels and fuel type (motorcycles excluded).

Table 4. VOC tailpipe emissions by vehicle type and by emission control technology in China in 2015 (Gg).

	China 0	China 1	China 2	China 3	China 4	China 5	SUM
LDPVs	173.59	146.09	56.48	240.32	49.09	8.81	674.38
MDPVs	56.73	10.28	7.42	4.88	0.62	0.06	79.98
HDPVs	99.57	22.13	24.31	45.37	5.72	2.12	199.23
LDTs	86.26	109.46	39.44	139.21	12.93	0.56	387.86
MDTs	111.47	18.16	17.61	10.05	0.51	0.01	157.82
HDTs	73.92	17.99	17.91	59.46	5.49	0.22	174.99
TAs	97.44	71.43	50.55	74.33	15.30	2.06	311.12
BUs	5.25	1.65	3.43	1.52	0.09	0.05	11.99
GMs							563.18

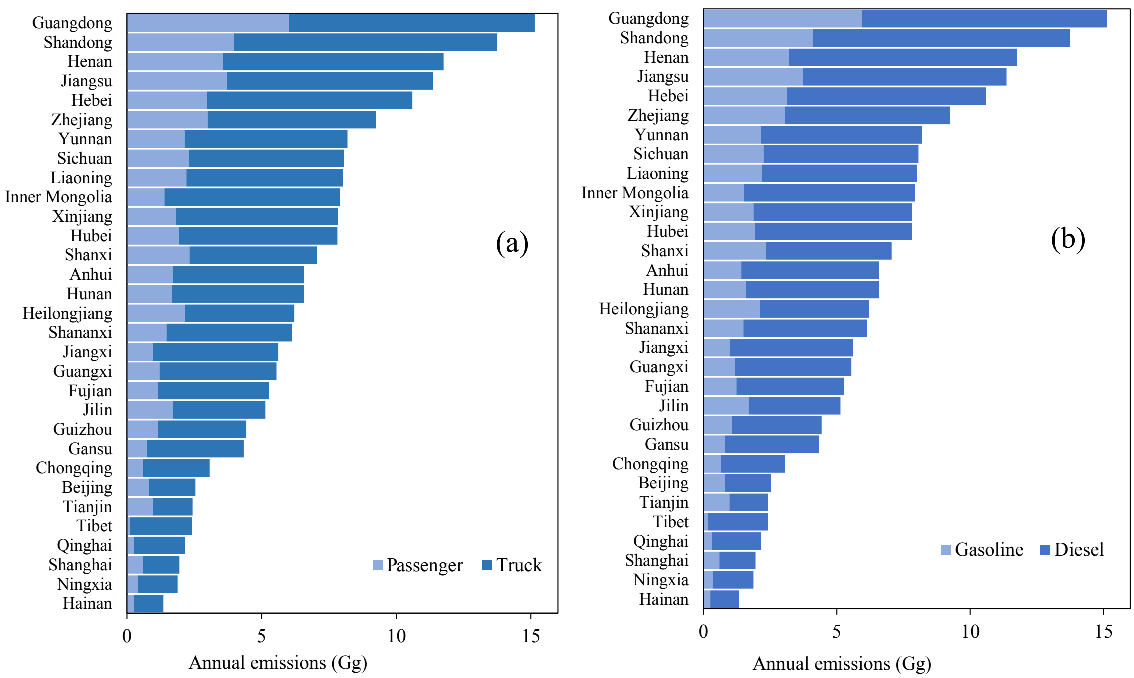

Figure 5. Provincial IVOC emissions from vehicles in 2015: **(a)** total emission amount classified by vehicle types; **(b)** total emission amount classified by emission sources.

3.1.3 Vehicle parking characteristics

GPS records including 103 million continuous parking events and 11 trillion valid seconds were collected and analyzed to obtain real-world vehicle parking characteristics in China, which have significant impacts on VEEs. It was found that parking characteristics in all provinces excluding Beijing were quite similar. The annual average number of parking events per day per vehicle was 3.89 in Beijing and 5.73 in other provinces, yet the annual average parking duration per day per vehicle was similar at 22.21 and 22.11 h respectively.

Figure 3a and b shows the distribution of parking events and total parking duration of six time intervals (0–1, 1–24, 24–48, 48–72, 72–119.5, > 119.5 h) in Beijing and other provinces.

About 95.5–98.8 % of parking events have a duration of less than 24 h, but they contribute only 51.3–76.8 % of the total parking duration. This indicates that the current VEEs control policy in China, where only VOCs evaporated in the first 24 h of parking are given a limitation value, cannot effectively cover the majority of VEEs in China. The latest standard of emission control, China 6, will enhance evaporative emission control by adding 48 h duration into consideration. Overall, Beijing was found to have fewer parking events but higher proportions of parking events with duration longer than 1 h, resulting in longer total parking duration. This phenomenon was mainly caused by consistent traffic control measures implemented in Beijing since the 2008 Beijing Olympic Games, in which the number of vehicles allowed within the 5th Ring Road was strictly limited.

Table 5. IVOC tailpipe emissions by vehicle type and by emission control technology in China in 2015 (Gg).

	China 0	China 1	China 2	China 3	China 4	China 5	SUM
LDPVs	5.94	20.58	1.75	10.28	2.71	0.24	5.94
MDPVs	0.32	0.10	0.02	0.07	0.01	0.00	0.32
HDPVs	0.41	0.28	0.07	0.33	0.26	0.00	0.41
LDTs	1.71	2.88	0.70	15.09	3.29	0.02	1.71
MDTs	2.17	0.88	0.30	3.98	0.69	0.00	2.17
HDTs	4.74	4.81	1.87	85.76	16.29	0.02	4.74
TAs	2.32	6.37	0.50	2.30	0.23	0.01	2.32
BUs	0.02	0.02	0.01	0.02	0.00	0.00	0.02

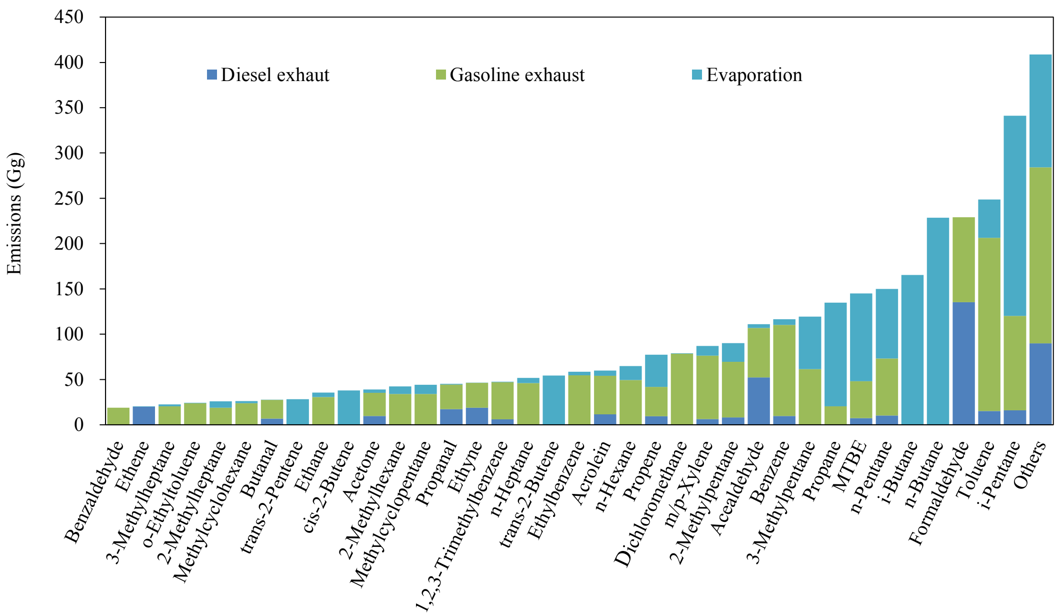

Figure 6. Speciated VOC component emissions classified by emission source.

3.2 Emissions and implications to policy

In 2015, China's on-road vehicles emitted 4.21 Tg of VOCs in total (Table 4). Figure 4a–f show the provincial results of vehicular VOC emissions in 2015. VTEs were still the predominant contributor of total VOC emissions, with a proportion of 60.80 % (Fig. 4a). VEEs, with a contributive share of 39.20 % of total emissions, are evidently non-negligible and should be taken into consideration for future management. Among provinces with large fleets of light-duty vehicles, Guangdong, Shandong and Jiangsu ranked on top of the league table, with a respective contribution of 10.66, 8.85 and 6.54 % to the total amounts of VOC emissions. A slight difference was observed between the ranking of tailpipe exhaust and evaporation (Fig. 4b and c), which was caused by the disparity in vehicle fleets, vehicle parking behavior and ambient temperature. Figure 4b provided insights for evaporative emissions of gasoline vehicles excluding motorcycles; evaporation from motorcycles are included in Fig. 4a. Refueling emissions contributed 7.83 % of the total evaporative emissions. The refueling emissions could be effectively controlled by Stage-II systems in service stations. Thus, running loss (81.05 %) and diurnal loss (10.00 %) are the main challenges in future emission control. The new China 6 vehicle emission standard will alleviate diurnal emissions, yet running loss issues would still be present. Estimation shows that it could be the most important part in current vehicle evaporation and even more so in the future. The calculation of running loss is expected with large uncertainty due to the lack of specific measurement data in China.

Figure 4c–f provided the subclassification for tailpipe exhausts. Passenger vehicles, trucks and motorcycles should all be considered for tailpipe VOC control, in which their contributions are 49.86, 28.15 and 21.99 % respectively. Tailpipe VOCs of passenger cars were widely recognized and monitored, in contrast with motorcycles and trucks that were commonly overlooked. Taxis and buses contributed 12.22 and 2.04 % of total tailpipe emissions in public transport, while

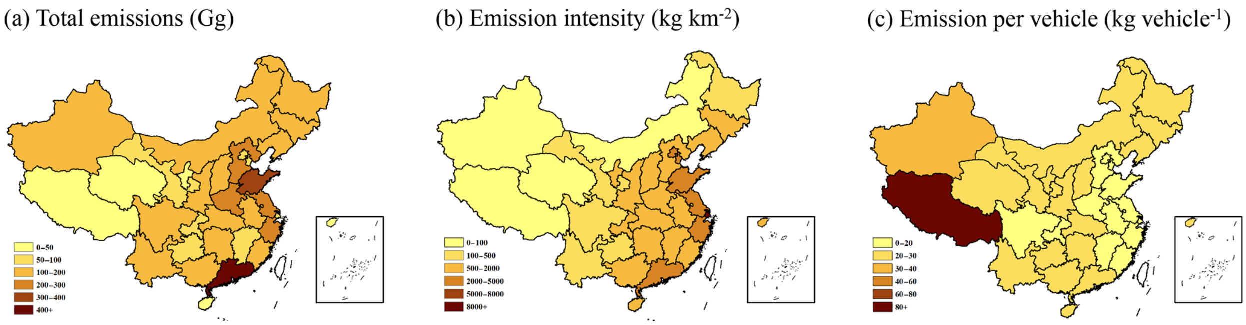

Figure 7. Province-based emission analysis: **(a)** total emission amount; **(b)** emission intensity; **(c)** emission per vehicle (motorcycles were excluded).

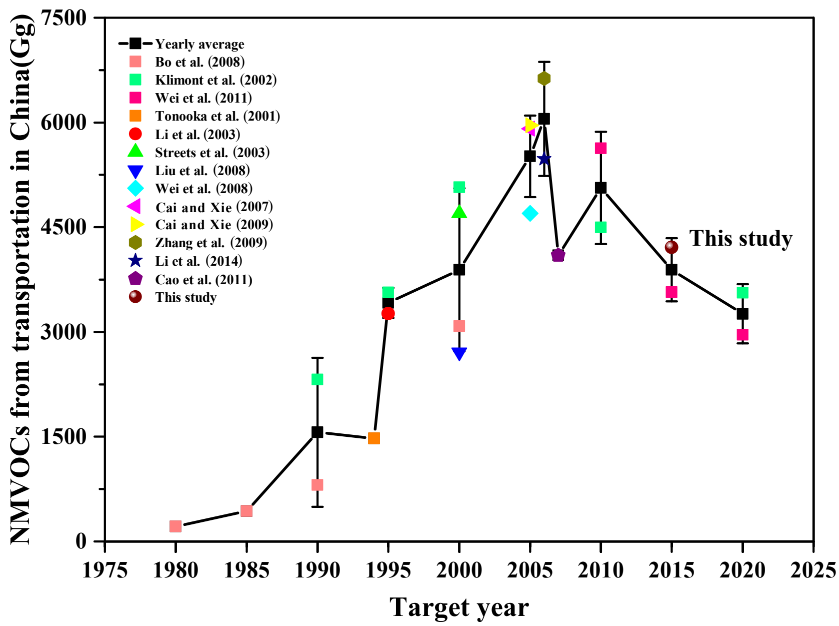

Figure 8. Comparison of this study to previous transportation emission inventories.

the number of motorcycles were merely 2.3 and 0.5 %; emissions are considerable in Guangdong, Shandong, Yunnan, Hunan, Guangxi and Fujian, where larger motorcycle markets are present. LDPVs and LDTs were two dominant subcategories of VOC emissions. While the activity data for LDPVs were greatly improved in this study for reduced uncertainty, improved data consisting of reliable information for LDTs are expected in the future. China 0 vehicles were still a dominant source (35.19 %) for tailpipe VOC emissions. Vehicles before China 4 (not included) contributed 94.67 % to total tailpipe emissions.

In 2015, China's on-road vehicles emitted 200.37 Gg of IVOCs in total (Table 5). Figure 5 provides IVOC emissions by province. It should be noted that this estimation was entirely based on emission factors from tests in the United States and is only capable of providing insights of similar magnitude compared with VOC emission level based on the same input of vehicle activity data. Diesel vehicles with a 72.4 % share of IVOC emissions were higher than gasoline vehicles due to the fact that emission factors of trucks are significantly higher than those of passenger vehicles.

In total, 97 species of evaporation, 30 species of gasoline exhaust and 20 species of diesel exhaust were identified (Fig. 6). Detailed emission amounts were provided for 36 species, while others were summed in the category named "others". Toluene, *i*-pentane and benzene were the main species in gasoline exhaust. *n*-Butane, *i*-pentane, *i*-butane and propane were the main species in evaporation. Evaporation made up an 83.66 % share of *n*-butane, *i*-pentane and *i*-butane vehicle emissions. Formaldehyde and acetaldehyde were mostly contributed by diesel tailpipe emissions. A small fraction of unresolved complex mixture was seen in our speciation profile for evaporative emission. The speciation profiles for exhaust of both gasoline and diesel vehicles are expected to be improved in the future. The speciated emission inventory of VOCs based on prevailing lumped chemical mechanism CB05 is provided in Table S8. This emission database could be used in chemical transport models.

Figure 7 compared total VOC emissions, emission intensity by area and emission per vehicles among provinces. As expected, developed regions such as Beijing, Tianjin and Shanghai showed the highest emission intensity by area (4500–9000 $kg\,km^{-2}$), while other provinces range between 13 and 2700 $kg\,km^{-2}$. Beijing's lowest emissions per vehicle compared to other regions (14–85 kg per vehicle) indicated that its fleet was the cleanest. Thus, mere technological approaches for emission reduction in more developed cities could be increasingly difficult. Alternative approaches such as reduction of population density as well as human behavior modification on vehicle operation should be considered as main strategies in these regions.

3.3 Uncertainty analysis

Inevitable uncertainties are present in VOC emission inventories due to the use of different input data, including activity characteristics, emission factors and VOC emission profiles. Total vehicle emissions of VOCs are 4.21 $Tg\,yr^{-1}$, with a 95 % confidence interval range from 2.90 to 6.54 Tg. The overall uncertainties in this inventory are estimated at −28.53 to 61.35 % for total VOC emissions. The uncertainties of detailed categories are listed in Table S9. These confidence ranges are comparable to other bottom-up emission inventories (Bo et al., 2008; Zhao et al., 2011; Yang et al., 2015a).

Figure 8 compared this updated emission inventory with previous studies. Our results were comparable to previous estimation and higher than Wei's forecast in 2011 (Wei et al., 2011). The differences could be explained by the following reasons. Firstly, vehicle evaporative emission was discretely considered in detail for the first time, which substantially increased total VOCs. Secondly, data of vehicle usage were derived from big data, which was a survey for "live" vehicles. The VKT used in our estimation is based on vehicle age, resulting in a lowered estimation of emissions of older vehicles. LDTs, China 0 and China 1 vehicles make up a substantial share of total emissions, yet the data of these vehicles had the largest uncertainty according to the author's experience. Improved activity data of these vehicles would further reduce uncertainty in our inventory.

Evaluation of the uncertainties in the IVOC emission inventory is indeed challenging as the majority of them come from emission factors. As IVOC measurements have become recently more and more recognized, global attention could be expected to improve the existing emission factor database, which would considerably reduce uncertainty.

The uncertainty of the species profile was significant in exhausts and negligible in evaporation. The reason for this is that the profile used for evaporation was reliable and comprehensive, combining vehicle activity, technology contribution in fleet and profiles in different processes. In addition, the species of evaporation were predominantly total hydrocarbons, which provided enough resolution for the species profile. The uncertainty for the exhaust species was mainly composed of three aspects. Firstly, the current profile was based on individual test results and no comprehensive profile was built to represent the fleet average. Secondly, VOC analysis yielded few recognized species regardless of individual or average tests. Thirdly, the species formed from incomplete combustion and unburned fuel were not understood well enough, which all present difficulties in building accurate species profiles for exhaust.

4 Conclusion

The contributions of this study include updated vehicle activity data from more than 70 000 cars and 2 million trucks in 30 different provinces, detailed vehicle fleet statistics, first-hand evaporation data and an REIB framework to account for inter-province transportation for trucks. The total VOC emissions from on-road vehicles in China were about 4.21 Tg in 2015.

Emission factors for running loss of evaporation are urgently needed to improve the emission inventory. IVOC emission factors of all vehicles are urgently needed. The activity data for LDTs and old vehicles should be improved. The species profiles for exhaust, especially for gasoline vehicles, are uncompelling.

We suggest paying more attention to the reduction of population density and vehicle usage in highly developed regions as a main approach for emission reduction. Simultaneous alleviation in both traffic congestion and pollutant emission could be realized with these measures.

Data availability. Datasets of real-world vehicle activity data used in this study are available upon request. Vehicle stock data can be accessed using the Statistical Yearbook for each province edited by Bureau of Statistics of each province in China.

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Competing interests. The authors declare that they have no conflict of interest.

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References

Bo, Y., Cai, H., and Xie, S. D.: Spatial and temporal variation of historical anthropogenic NMVOCs emission inventories in China, Atmos. Chem. Phys., 8, 7297–7316, https://doi.org/10.5194/acp-8-7297-2008, 2008.

Cai, H. and Xie, S. D.: Estimation of vehicular emission inventories in China from 1980 to 2005, Atmos. Environ., 41, 8963–8979, https://doi.org/10.1016/j.atmosenv.2007.08.019, 2007.

Cai, H. and Xie, S. D.: Tempo-spatial variation of emission inventories of speciated volatile organic compounds from on-road vehicles in China, Atmos. Chem. Phys., 9, 6983–7002, https://doi.org/10.5194/acp-9-6983-2009, 2009.

Cao, G. L., Zhang, X. Y., Gong, S. L., An, X. Q., and Wang, Y. Q.: Emission inventories of primary particles and pollutant gases for China, Chinese Sci. Bull., 56, 781–788, https://doi.org/10.1007/s11434-011-4373-7, 2011.

Cao, X. Y., Yao, Z. L., Shen, X. B., Ye, Y., and Jiang, X.: On-road emission characteristics of VOCs from light-duty gasoline vehicles in Beijing, China, Atmos. Environ., 124, 146–155, https://doi.org/10.1016/j.atmosenv.2015.06.019, 2016.

Cui, H. Y., Chen, W. H., Dai, W., Liu, H., Wang, X. M., and He, K. B.: Source apportionment of $PM_{2.5}$ in Guangzhou combining observation data analysis and chemical transport model simulation, Atmos. Environ., 116, 262–271, https://doi.org/10.1016/j.atmosenv.2015.06.054, 2015.

Department of Energy Statistics, National Bureau of Statistics, People's Republic of China: China Energy Statistical Yearbook 2016, China Statistics Press, Beijing, China, 2016.

EPA: Update of Hot Soak Emissions, EPA420-R-01-026 Washington, DC, available at: https://nepis.epa.gov/Exe/ZyPDF.cgi/P10021WT.PDF?Dockey=P10021WT.PDF (last access: 23 October 2017), 2001.

EPA: Development of Evaporative Emissions Calculations for the Motor Vehicle Emissions Simulator MOVES2010, EPA-420-R-12-027, United States Environmental Protection Agency, Washington, DC, available at: https://nepis.epa.gov/Exe/ZyPDF.cgi/P100F3ZY.PDF?Dockey=P100F3ZY.PDF (last access: 23 October 2017), 2012.

Geng, F. H., Tie, X. X., Xu, J. M., Zhou, G. Q., Peng, L., Gao, W., Tang, X., and Zhao, C. S.: Characterizations of ozone, NO_x, and VOCs measured in Shanghai, China, Atmos. Environ., 42, 6873–6883, https://doi.org/10.1016/j.atmosenv.2008.05.045, 2008.

Goldstein, A. H. and Galbally, I. E.: Known and unknown organic constituents in the Earth's atmosphere, Environ. Sci. Technol., 41, 1514–1521, 2007.

Hodzic, A., Jimenez, J. L., Madronich, S., Canagaratna, M. R., DeCarlo, P. F., Kleinman, L., and Fast, J.: Modeling organic aerosols in a megacity: potential contribution of semi-volatile and intermediate volatility primary organic compounds to secondary organic aerosol formation, Atmos. Chem. Phys., 10, 5491–5514, https://doi.org/10.5194/acp-10-5491-2010, 2010.

Huang, R. J., Zhang, Y. L., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y. M., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., and Pévôt, A. S.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218–222, https://doi.org/10.1038/nature13774, 2014.

Huo, H., Yao, Z. L., Zhang, Y. Z., Shen, X. B., Zhang, Q., and He, K. B.: On-board measurements of emissions from diesel trucks in five cities in China, Atmos. Environ., 54, 159–167, https://doi.org/10.1016/j.atmosenv.2012.01.068, 2012.

ICCT: Air Emissions Issues Related to Two and Three-Wheeled Motor Vehicles An Initial Assessment of Current Conditions and Options for Control; International Council on Clean Transportation (ICCT), available at: http://www.theicct.org/sites/default/files/publications/twothree_wheelers_2007.pdf (last access: 23 October 2017), 2007.

Jathar, S. H., Gordon, T. D., Hennigan, C. J., Pye, H. O. T., Pouliotb, G., Adamsa, P. J., Donahuea, N. M., and Robinson, A. L.: Unspeciated organic emissions from combustion sources and their influence on the secondary organic aerosol budget in the United States, P. Natl. Acad. Sci. USA, 111, 10473–10478, https://doi.org/10.1073/pnas.1323740111, 2014.

Klimont, Z., Streets, D. G., Gupta, S., Cofala, J., Fu, L. X., and Ichikawa, Y.: Anthropogenic emissions of non-methane volatile organic compounds in China, Atmos. Environ., 36, 1309–1322, https://doi.org/10.1016/S1352-2310(01)00529-5, 2002.

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525, 367–371, https://doi.org/10.1038/nature15371, 2015.

Li, M., Zhang, Q., Streets, D. G., He, K. B., Cheng, Y. F., Emmons, L. K., Huo, H., Kang, S. C., Lu, Z., Shao, M., Su, H., Yu, X., and Zhang, Y.: Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms, Atmos. Chem. Phys., 14, 5617–5638, https://doi.org/10.5194/acp-14-5617-2014, 2014.

Li, W., Fu, L. X., Hao, J. M., Ma, H., Li, S. Q., and Hu, W.: Emission inventory of 10 kinds of air pollutants for road traffic vehicles in China, Urban Env. Urban Ecol., 16, 36–38, 2003.

Liu, H., He, K. B., Wang, Q. D., Huo, H., Lents, J., Davis, N., Nikkila, N., Chen, C. H., Osess, M., and He, C. Y.: Comparison of vehicle activity and emission inventory between Beijing and Shanghai, J. Air Waste Manage. Assoc., 57, 1172–1177, https://doi.org/10.3155/1047-3289.57.10.1172, 2007.

Liu, H., He, K. B., Lents, J. M., Wang, Q. D., and Tolvett, S.: Characteristics of diesel truck emission in China based on portable emissions measurement systems, Environ. Sci. Technol., 43, 9507–9511, https://doi.org/10.1021/es902044x, 2009.

Liu, H., Man, H. Y., Tschantz, M., Wu, Y., He, K. B., and Hao, J. M.: VOC from vehicular evaporation emissions: status and control strategy, Environ. Sci. Technol., 49, 14424–14431, https://doi.org/10.1021/acs.est.5b04064, 2015.

Liu, H., Fu, M., Jin, X., Shang, Y., Shindell, D., Faluvegi, G., Shindell, C., and He, K.: Health and climate impacts of ocean-going vessels in East Asia, Nature Climate Change, 6, 1037–1041, https://doi.org/10.1038/nclimate3083, 2016.

Liu, J. F., Zhao, J., Li, T. T., Bai, Y. H., and Liu, Z. R.: Establishment of Chinese anthropogenic source volatile organic compounds emission inventory, China Environ. Sci., 28, 496–500, 2008 (in Chinese).

Man, H. Y., Liu, H., Niu, H., Wang, M., Wang, K., He, K. B., and Hao, J. M.: VOC emissions from the vehicle evaporation process: species and underestimation, 21th International Transport and Air Pollution Conference, Lyon, France, 24–26 May 2016.

MEP: Technical guidelines on emission inventory development of air pollutants from on-road vehicles (on trial); Ministry of Environmental Protection of the People's Republic China, available at: http://www.qhepb.gov.cn/pub/qhepb/xwzx/zqyj/201407/W020140711608241902098.pdf (last access: 23 October 2017), 2015.

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J. R., and Pandis, S. N.: Rethinking Organic Aerosols: Semivolatile Emissions and Photochemical Aging, Science, 315, 1259–1262, https://doi.org/10.1126/science.1133061, 2007.

Shao, M., Zhang, Y. H., Zeng, L. M., Tang, X. Y., Zhang, J., Zhong, L. J., and Wang, B. G.: Ground-level ozone in the Pearl River Delta and the roles of VOC and NO_x in its production, J. Environ. Manage., 90, 512–518, https://doi.org/10.1016/j.jenvman.2007.12.008, 2009.

Shao, M., Huang, D., Gu, D., Lu, S., Chang, C., and Wang, J.: Estimate of anthropogenic halocarbon emission based on measured ratio relative to CO in the Pearl River Delta region, China, Atmos. Chem. Phys., 11, 5011–5025, https://doi.org/10.5194/acp-11-5011-2011, 2011.

Sitch, S., Cox, P. M., Collins, W. J., and Huntingford, C.: Indirect radiative forcing of climate change through ozone effects on the land-carbon sink, Nature, 448, 791–794, https://doi.org/10.1038/nature06059, 2007.

Song, Y., Dai, W., Shao, M., Liu, Y., Lu, S., Kuster, W., and Goldan, P.: Comparison of receptor models for source apportionment of volatile organic compounds in Beijing, China, Environ. Pollut., 156, 174–183, https://doi.org/10.1016/j.envpol.2007.12.014, 2008.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108, 8809, https://doi.org/10.1029/2002JD003093, 2003.

Tonooka, Y., Kannari, A., Higashino, H., and Murano, K.: NMVOCs and CO emission inventory in East Asia, Water Air Soil Poll., 130, 199–204, https://doi.org/10.1023/A:1013890513856, 2001.

van Donkelaar, A., Martin, R. V., Brauer, M., and Boys, B. L.: Use of Satellite Observations for Long-Term Exposure Assessment of Global Concentrations of Fine Particulate Matter, Environ Health Persp., 123, 35, https://doi.org/10.1289/ehp.1408646, 2015.

Wang, H., Chen, C., Huang, C., and Fu, L.: On-road vehicle emission inventory and its uncertainty analysis for Shanghai, China, Sci. Total Environ., 398, 60–67, https://doi.org/10.1016/j.scitotenv.2008.01.038, 2008.

Wang, S. X., Zhao, M., Xing, J., Wu, Y., Zhou, Y., Lei, Y., He, K. B., Fu, L. X., and Hao, J. M.: Quantifying the air pollutants emission reduction during the 2008 Olympic Games in Beijing, Environ. Sci. Technol., 44, 2490–2496, https://doi.org/10.1021/es9028167, 2010.

Wei, W., Wang, S. X., Chatani, S., Klimont, Z., Cofala, J., and Hao, J. M.: Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China, Atmos. Environ., 42, 4976–4988, https://doi.org/10.1016/j.atmosenv.2008.02.044, 2008.

Wei, W., Wang, S. X., Hao, J. M., and Cheng, S. Y.: Projection of anthropogenic volatile organic compounds (VOCs) emissions in China for the period 2010–2020, Atmos. Environ., 45, 6863–6871, https://doi.org/10.1016/j.atmosenv.2011.01.013, 2011.

Yao, Z. L., Wu, B. B., Shen, X. B., Cao, X. Y., Jiang, X., Ye, Y., and He, K. B.: On-road emission characteristics of VOCs from rural vehicles and their ozone formation potential in Beijing, China, Atmos. Environ., 105, 91–96, https://doi.org/10.1016/j.atmosenv.2015.01.054, 2015.

Yamada, H.: Contribution of evaporative emissions from gasoline vehicles toward total VOC emissions in Japan, Sci. Total Environ., 449, 143–149, https://doi.org/10.1016/j.scitotenv.2013.01.045, 2013.

Yang, X. F., Liu, H., Man, H. Y., and He, K. B.: Characterization of road freight transportation and its impact on the national emission inventory in China, Atmos. Chem. Phys., 15, 2105–2118, https://doi.org/10.5194/acp-15-2105-2015, 2015a.

Yang, X. F., Liu, H., Cui, H. Y., Man, H. Y., Fu, M. L., Hao, J. M., and He, K. B.: Vehicular volatile organic compounds losses due to refueling and diurnal process in China: 2010–2050, J. Environ. Sci., 33, 88–96, https://doi.org/10.1016/j.jes.2015.01.012, 2015b.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, https://doi.org/10.5194/acp-9-5131-2009, 2009.

Zhang, S., Wu, Y., Wu, X., Li, M., Ge, Y., Liang, B., Xu, Y., Zhou, Y., Liu, H., and Fu, L.: Historic and future trends of vehicle emissions in Beijing, 1998–2020: A policy assessment for the most stringent vehicle emission control program in China, Atmos. Environ., 89, 216–229, https://doi.org/10.1016/j.atmosenv.2013.12.002, 2014.

Zhang, Y. Z., Yao, Z. L., Shen, X. B., Liu, H., and He, K. B.: Chemical characterization of $PM_{2.5}$ emitted from on-road heavy-duty diesel trucks in China, Atmos. Environ., 122, 885–891, https://doi.org/10.1016/j.atmosenv.2015.07.014, 2015.

Zhao, Y., Nielsen, C. P., Lei, Y., McElroy, M. B., and Hao, J.: Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China, Atmos. Chem. Phys., 11, 2295–2308, https://doi.org/10.5194/acp-11-2295-2011, 2011.

Zhao, Y., Hennigan, C. J., May, A. A., Tkacik, D. S., de Gouw, J. A., Gilman, J. B., Kuster, W. C., Borbon, A., and Robinson, A. L.: Intermediate-volatility organic compounds: a large source of secondary organic aerosol, Environ. Sci. Technol., 48, 13743–13750, https://doi.org/10.1021/es5035188, 2014.

Zhao, Y., Nguyen, N. T., Presto, A. A., Hennigan, C. J., May, A. A., and Robinson, A. L.: Intermediate Volatility Organic Compound Emissions from On-Road Diesel Vehicles: Chemical Composition, Emission Factors, and Estimated Secondary Organic Aerosol Production, Environ. Sci. Technol., 49, 11516–11526, https://doi.org/10.1021/acs.est.5b02841, 2015.

Zhao, Y., Nguyen, N. T., Presto, A. A., Hennigan, C. J., May, A. A., and Robinson, A. L.: Intermediate Volatility Organic Compound Emissions from On-Road Gasoline Vehicles and Small Off-Road Gasoline Engines, Environ. Sci. Technol., 50, 4554–4563, https://doi.org/10.1021/acs.est.5b06247, 2016.

Zheng, B., Huo, H., Zhang, Q., Yao, Z. L., Wang, X. T., Yang, X. F., Liu, H., and He, K. B.: High-resolution mapping of vehicle emissions in China in 2008, Atmos. Chem. Phys., 14, 9787–9805, https://doi.org/10.5194/acp-14-9787-2014, 2014.

Zheng, J. Y., Shao, M., Che, W. W., Zhang, L. J., Zhong, L. J., Zhang, Y. H., and Streets, D.: Speciated VOC emission inventory and spatial patterns of ozone formation potential in the Pearl River Delta, China, Environ. Sci. Technol., 43, 8580–8586, https://doi.org/10.1021/es901688e, 2009.