

# An updated emission inventory of vehicular VOCs/IVOCs in China

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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  
15 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 a mass of local  
measurement data. Activity data for light-duty vehicles are derived from trajectories of more than 70  
thousand cars for one year. The annual mileages of trucks are calculated from reported data by more  
20 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  
refuelling, hot soak, diurnal and running loss, are taken into account. Results show that the total  
vehicular VOCs emissions in China are 4.21 Tg (with a 95% confidence interval ranges from 2.90-6.54  
Tg) and the IVOCs emissions are 200.37 Gg in 2015. VTEs are still the predominant contributor, while  
25 VEEs are responsible for 39.20% of VOCs emission. 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 among three of the highest,  
with a respective contribution of 10.66%, 8.85% and 6.54% to the total amounts of VOCs from vehicles.  
30 97 VOC species are analyzed in this VOCs emission inventory. I-pentane, toluene and formaldehyde  
are found to be the most abundant species in China's vehicular VOCs emissions. The estimated IVOCs

is another ‘inconvenient truth’, concluding that precursors emission for secondary organic aerosol (SOA) from vehicles are much larger than previous estimation.

## 35 **1 Introduction**

China is one of those countries most threatened by simultaneous pollution of PM<sub>2.5</sub> (particulate matter with aerodynamic diameters of less than 2.5µm) 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 shown that secondary organic aerosols (SOA) is of significant proportion in ambient PM<sub>2.5</sub> mass of Chinese cities (*Cui et al., 2015*). Intermediate-volatility organic compounds (IVOCs) is a series of compounds with effective saturate concentration between 10<sup>3</sup>-10<sup>6</sup>µg/m<sup>3</sup>, 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 . 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 widely attended 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 VOCs emissions and ambient VOCs 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 national level. Cai and Xie (*2009*) reported VOCs 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; Cai et al., 2007; Bo et al., 2008; Liu et al., 2008; Wei et al., 2008; Zhang et al., 2009; Cao et al., 2011; Li et al., 2014; Zheng et al., 2014*). The complete summary of existing studies on vehicular VOCs emission inventory and their respective performance were shown in Table S1 in the Supporting Information. These existing emission inventories have greatly improved our understanding on VOCs emissions. It is worth noting that all studies mentioned above targeted emission inventory prior to 2010 while omitting IVOCs

60 impacts. Considering the dramatic increase of vehicle population, establishment of new emission inventories is of urgent priority. However, multiple key factors changed in the last ten years that require updated methods and data.

First of all, dominant VOCs emission processes of vehicles may switch. Compared with vehicular tailpipe emissions (VTEs), vehicular evaporation emissions (VEEs) have recently been reported to be  
65 nonnegligible contributors to ambient VOCs concentrations (*Yamada et al., 2013; Liu et al., 2015*). VOCs emissions evaporate from gasoline fuelled vehicles consistently regardless of their status of refuelling, 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 ambient atmosphere. Depending on the vehicle's status, evaporative emissions come in four varieties: refuelling loss, running  
70 loss, hot soak loss and diurnal loss. Local emission factors 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. Short  
75 of systematic and integrated analytical methods limit the progress for measurement and quantification of IVOCs (*Goldstein et al., 2007; Jathar et al., 2014*). Therefore, few studies successfully provided emission measurement results of IVOCs. To our knowledge, IVOCs 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  
80 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 purpose without any personal information. This data covered 30 provinces in China, which  
85 would significantly improve our understanding on vehicle usage, and better 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 number, which presume that all vehicles were operated locally, although an acceptable assumption for household vehicles, is irrelevant  
90 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 the corresponding emission intensity of the road type. This method greatly improved NO<sub>x</sub> and PM emission estimation for long-distance inter-province or inter-city cargo transportation (*Yang et al., 2015a*).

95 Imperfections in comprehensiveness and accuracy of estimation can also be improved by using local emission factors and speciation published recently (*Liu et al., 2009; Liu et al., 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 U.S. 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.  
100 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 provides vehicle population data classified by vehicle type (e.g., light-duty passenger vehicles, heavy-duty trucks). More detailed vehicle population  
105 data by fuel type and 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 are solved each and individually based on  
110 scientific calculating methodologies, big data and abundant local emission measurements. IVOCs emission factors used are derived from studies in the US by matching corresponding vehicle emission categories of the two countries, as no local IVOCs emission factors were reported.

## 2 Methodology and data

### 2.1 Vehicle stock and classification

115 In total, 25 types of on-road vehicles were considered in this study, including passenger vehicles, trucks  
and 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  
120 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  
125 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 6000mm and ridership no more  
than 9. Medium-duty refers to vehicles of length less than 6000mm and ridership between 10-19.  
Heavy-duty refers to vehicles of length no less than 6000mm or ridership is no less than 20. These  
130 vehicles were further classified by control technologies (i.e., 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),  
135 heavy-duty gasoline trucks (HDGTs), and heavy-duty diesel trucks (HDDTs). For trucks, a light-duty  
truck refers vehicles of mass less than 3500kg. A medium-duty truck refers to vehicles o with mass  
ranging from 3500kg to 12000kg. A heavy-duty truck refers vehicles of mass more than 12000kg.

Detailed provincial population data of all vehicles excluding GMs in 2015 was obtained by complete  
statistical survey conducted by the Vehicle Emission Control Center (VECC) of China's Ministry of

140 Environmental Protection (MEP), which could be considered highly accurate. The provincial GMs population in 2015 was obtained from the Provincial Statistic Yearbook 2016 of each province.

## 2.2 Vehicle activity

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

145 The provincial annual vehicle kilometers traveled (VKT) data of light-duty passenger vehicles (LDPVs), which was the majority in the fleet and thus had the largest impact on the emission inventory, was acquired by processing and analyzing the big data of GPS records (71059 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  
150 introduced in detail in our previous study (*Yang et al., 2015a*).

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

The average mileage for trucks were obtained from a commercial source with data feeding of more than 2 million trucks, mainly commercial vehicles installed with either the GPS or China Bei Dou  
155 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 VOCs emissions at the provincial level were divided into three parts for calculation,  
160 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 IVOCs emission calculation was similar to that of the  
165 VOCs, while only the tailpipe exhaust for non-GMs was taken into consideration. For IVOCs emission

factors, Zhao et al. (2015, 2016) reported a series of measurements for gasoline and diesel vehicles (Table S2 and Table S3). Details were introduced below.

### 2.3.1 Tailpipe emissions from non-truck vehicles

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

$$E_{tailpipe,non-truck,i} = \sum_j \sum_k (EF_{tailpipe,j,k} \times VP_{i,j,k} \times VKT_{i,j}), \quad (1)$$

where  $E_{tailpipe,non-truck,i}$  represents the annual tailpipe emissions from non-truck vehicles in province  $i$  ( $\text{g}\cdot\text{year}^{-1}$ );  $EF_{tailpipe,j,k}$  represents the tailpipe VOCs/IVOCs emission factor of vehicle type  $j$  with  
175 control technology  $k$  ( $\text{g}\cdot\text{km}^{-1}$ );  $VP_{i,j,k}$  represents the registered population of vehicle type  $j$  with control technology  $k$  in province  $i$ ;  $VKT_{i,j}$  represents the annual VKT of vehicle type  $j$  in province  $i$  ( $\text{km}\cdot\text{year}^{-1}$ ).

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-*  
180 *road vehicles (on trial)*) (Table S4).

For IVOCs emission factors, Zhao et al. reported a series of measurements for gasoline and diesel vehicles (Zhao et al.; 2015, 2016). By considering 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  
185 vehicle categories were set up. For the categories lacking measurements (gasoline vehicles before China 1 and all diesel vehicles), emission factors were set identical to China 1 category. For diesel passenger vehicles, the current IVOC emission factors were set identical to the corresponding level of gasoline vehicles. The IVOC emission factors were converted from the original unit of mg/kg-fuel to g/km using fuel economy. For each category, median of emission factors were used for the particular type of  
190 vehicles if more than one available tests are present. Detailed emission factors were 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 inter-city or inter-province cargo transportation, REIB approach instead of the traditional local registration based approach was utilized to calculate truck emissions, as was detailly described in our previous work  
195 (*Yang et al., 2015a*). The provincial tailpipe emissions from freight trucks were estimated by Eq. (2):

$$E_{tailpipe,truck,i} = \sum_j \sum_k \sum_m \left[ \frac{(EF_{tailpipe,j,k} \times VP_{j,k} \times VKT_{j,k} \times DP_{j,m}) \times L_{i,m}}{L_m} \right], \quad (2)$$

where  $E_{tailpipe,truck,i}$  represents the annual tailpipe VOCs/IVOCs emissions from freight trucks in province  $i$  ( $g \cdot year^{-1}$ );  $EF_{tailpipe,j,k}$  represents the tailpipe VOCs/IVOCs emission factor of vehicle type  $j$   
200 with control technology  $k$  ( $g \cdot km^{-1}$ );  $VP_{j,k}$  represents the national population of vehicle type  $j$  with control technology  $k$ ;  $VKT_{j,k}$  represents the annual VKT of vehicle type  $j$  with control technology  $k$  ( $km \cdot year^{-1}$ );  $DP_{j,m}$  represents the distance portion of vehicle type  $j$  running on road type  $m$ ;  $L_{i,m}$  and  $L_m$  represents 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  
205 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 model respectively. Followed by identification of thirty 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  
210 according to the emission rate of each bin that were presented in our previous test results (*Liu et al., 2009*).

For IVOCs emission factors, a mapping to match US emission certification level to China emission level was built (Table S3). Only non-GM vehicles were considered for IVOC emissions evaluation. The emission factors for US fleet were converted for use of Chinas' trucks. As no data were available for  
215 most categories, assumptions had to be made to fill the gap: (1) Medium and heavy-duty trucks had identical emission factors that are 50% higher than light-duty trucks, similar to emission ratios of VOCs

and primary organic aerosol (POA) of the same types. (2) Emission levels of neighboring types were used in the case of lacking data. The final assumption for IVOC emission factors were introduced in Table S5.

### 220 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 emission factors of diurnal and hot soak 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 were summarized in Table S6. The provincial annual diurnal emissions from non-GM gasoline vehicles and GMs were calculated by Eq. (3)-(6) and Eq. (7) respectively. For diurnal emissions, we calculated total parking hours for each parking events 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.

$$230 \quad E_{diur,non-GM,i} = E_{diur,<24,non-GM,i} + E_{diur,24-48,non-GM,i} + E_{diur,>48,non-GM,i}, \quad (3)$$

$$E_{diur,<24,non-GM,i} = [EF_{diur,<24,LDGPPVs} \times (P_{duration,1-24,i} \times T_i - P_{event,1-24,i} \times N_i \times 1)] \times 365 \times VP_{i,non-GM}, \quad (4)$$

$$235 \quad E_{diur,24-48,non-GM,i} = [EF_{diur,<24,LDGPPVs} \times P_{event,24-48,i} \times N_i \times 23 + EF_{diur,24-48,LDGPPVs} \times (P_{duration,24-48,i} \times T_i - P_{event,24-48,i} \times N_i \times 24)] \times 365 \times VP_{i,non-GM}, \quad (5)$$

$$240 \quad E_{diur,>48,non-GM,i} = [EF_{diur,<24,LDGPPVs} \times P_{event,>48,i} \times N_i \times 23 + EF_{diur,24-48,LDGPPVs} \times P_{event,>48,i} \times N_i \times 24 + EF_{diur,48-72,LDGPPVs} \times (P_{duration,>48,i} \times T_i - P_{event,>48,i} \times N_i \times 48)] \times 365 \times VP_{i,non-GM}, \quad (6)$$

where  $E_{diur,non-GM,i}$  represents the total annual diurnal (simultaneous permeation included) emissions from non-GM gasoline vehicles registered in province  $i$  ( $\text{g}\cdot\text{year}^{-1}$ );  $E_{diur,<24,non-GM,i}$ ,  $E_{diur,24-48,non-GM,i}$ ,  $E_{diur,>48,non-GM,i}$  represents the annual diurnal (simultaneous permeation included) emissions occurred respectively in the first day, second day and third day and after of parking ( $\text{g}\cdot\text{year}^{-1}$ );  $EF_{diur,<24,LDGVS}$ ,  $EF_{diur,24-48,LDGVS}$ ,  $EF_{diur,48-72,LDGVS}$  represents the measured diurnal (simultaneous permeation included) emission factors of China 4 LDGVs ( $\text{g}\cdot\text{hour}^{-1}$ ). The evaporative emission control was keeping the same until China 6. Thus, there's no progress on emission reduction since China 1 to China 5 on evaporation. So, the emission factors of China 4 LDGVs could be used for all LDGVs. For the other vehicle types, no data is available from tests and the same EFs with LDGV were used.  $P_{event,1-24,i}$ ,  $P_{event,24-48,i}$ ,  $P_{event,>48,i}$  represents the percentage of parking events that are 1-24 hours, 24-48 hours and above 48 hours;  $P_{duration,1-24,i}$ ,  $P_{duration,24-48,i}$ ,  $P_{duration,>48,i}$  represents the percentage of total parking duration that are between 1 hour and 24 hours, between 24 hours and 48 hours and above 48 hours.  $N_i$  represents the annual average parking events per day per vehicle of province  $i$ .  $T_i$  represents the average parking duration per day per vehicle of province  $i$  (hour).  $VP_{i,non-GM}$  represents the registered population of vehicle excluding motorcycles in province  $i$ .

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

$$E_{GMs,i} = EF_{GMs} \times VP_{i,GMs} \times VKT_{i,GMs},$$

(7) where

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

According to the US EPA, hot soak is defined as the evaporative losses within a one-hour period after shutting down of engines (*EPA, 2001*). Any vapor losses occurred after are considered diurnal

emissions. The provincial hot soak emissions for non-GM gasoline vehicles (i.e., LDGPVs, MDGPVs, HDGPVs, LDGTAs, GBUs, LDGTs, MDGTs, HDGTs) were calculated by Eq. (8):

$$270 \quad E_{soak,non-GM,i} = EF_{soak,LDGPVs} \times \left[ (T_i \times 365 \times P_{duration,<1,i}) + (N_i \times 365 \times P_{event,>1,i} \times 1) \right] \times \\ VP_{i,non-GMs} , \quad (8)$$

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

#### 280 **2.3.4 Evaporation emissions from gasoline vehicles- Refuelling**

China follows European countries in popularization of Stage-II vapor control system for reduction of refueling loss in stations. The vehicle refuelling emissions were also measured by our team from SHED tests (*Yang et al, 2015b*). The provincial refuelling emissions from gasoline vehicles were 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.

$$285 \quad E_{refuel,i} = EF_{refuel} \times [(1 - \theta) \times \omega_i + (1 - \omega_i)] \times CF_i , \quad (9)$$

where  $E_{refuel,i}$  represents the annual refuelling emissions from gasoline vehicles in province  $i$  ( $\text{g}\cdot\text{year}^{-1}$ );  $EF_{refuel}$  represents the refuelling emission factor for non-control condition ( $\text{g}\cdot\text{L}^{-1}$ );  $\theta$  represents the average efficiency of the Stage-II vapor control system, which is 82% according to our measurements in Beijing;  $\omega_i$  represents the percentage of filling stations equipped with Stage-II vapor control system 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

of province  $i$  ( $L \cdot year^{-1}$ ), which was retrieved from official statistics (China Energy Statistical Yearbook 2016) and 85% of total gasoline was assumed to be used in on-road vehicles.

### 2.3.5 Evaporation emissions from gasoline vehicles- Running loss

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

$$E_{running,non-GM,i} = EF_{running,LDGVS} \times (24 - T_i) \times 365 \times VP_{i,non-GM} , \quad (10)$$

where  $E_{running,non-GM,i}$  represents the annual running loss emissions from non-GM gasoline vehicles registered in province  $i$  ( $g \cdot year^{-1}$ );  $EF_{running,LDGVS}$  represents the running loss emission factor of LDGVs ( $g \cdot hour^{-1}$ ). 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 of province  $i$  (hour).  $VP_{i,non-GM}$  represents the registered population of vehicle excluding motorcycles in province  $i$ .

### 2.3.6 Uncertainty analysis

The uncertainty for 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 researches on inventory study (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 10000 times to produce the estimate.

### 2.4 Species analysis

The vehicular VOCs emissions speciation was further determined by Eq. (11):

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

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

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The VOCs profiles used in this study for establishment of vehicular VOCs emission inventory were derived from literature review and lab tests. Tailpipe VOCs emissions from gasoline vehicles and diesel vehicles were yielded from corresponding local profiles reported by Yao et al. 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* and *Cao et al, 2016*). For exhaust emissions, the profiles [1, 2]. For vehicle evaporative emissions, a comprehensive species profile was obtained based on results from the 30 cross-over evaporative tests we conducted before (*Man et al., 2016*).

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## 335 **3 Results and discussion**

### **3.1 Activity characteristics of vehicles**

#### **3.1.1 Vehicle population**

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 (Figure 1). LDPVs were the predominant contributor 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%)

340

and buses (BUs, 0.5%). In terms of control technologies, China 3 vehicles accounted for the largest proportion (51.0%) in China's non-GM vehicle fleet, followed by China 4 (22.9%), China 1 (9.5%),  
345 China 2 (7.2%), China 5 vehicles (5.2%) and China 0 (4.2%). China 1, China 2 and China 4 shared  
9.5%, 15.69% and 10.12% respectively in 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.  
350 Table 1 summarized the vehicle population and corresponding proportions classified by fuel types.  
LDPVs, MDPVs and TAs were mainly fuelled by gasoline while HDPVs, LDTs, MDTs, HDTs and  
BUs were primarily fuelled by diesel. Proportion of diesel consuming vehicles increases with vehicle  
weight in both passenger vehicles and freight trucks. Alternative fuels were yet to become mainstream,  
with particularly low proportion in the freight truck fleet, while exceeding 8.7% share in BUs and TAs,  
355 contributed by heavy promotion policies of central and local governments.

### 3.1.2 VKT characteristics

GPS records of 71,059 vehicles operating in 30 provinces from July 1, 2014, to July 1, 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  
360 average VKT of LDPVs in China was  $18,886 \pm 10,469$  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 Figure 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  
365 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 amount of families in both cities own multiple  
vehicles nowadays, causing decrease of annual VKT of individual cars under the circumstances that

370 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 hence smaller annual VKT of vehicles in these two cities. Thirdly, an increase of public transportation usage was observed in Beijing and Shanghai due to the growing traffic jams in peak hours.

Table 3 summarized vehicle mileages of other vehicle types in China. VKT of trucks are significantly  
375 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 control technologies. Aging of trucks in China greatly impact their performances due to common practice of overloading, which expedites integrity loss of the trucks' internal parts. Several cities have implemented low emission zones to restrict entry of trucks with outdated control technologies.

### 380 **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  
385 Beijing and 5.73 in other provinces, yet the annual average parking duration per day per vehicle was similar at 22.21 h and 22.11 h, respectively.

Figure 3(a) and Figure 3(b) shows the distribution of parking events and total parking duration of six time intervals (0-1 h, 1-24 h, 24-48 h, 48-72 h, 72-119.5 h, >119.5 h) in Beijing and other provinces. Parking events of the first two time intervals (<24 h) is of 95.5-98.8% in number, yet only accounted for  
390 51.3-76.8% of total parking duration, indicating that the current VEEs control policy in China where only VOCs evaporated in the first 24 h of parking is 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,  
395 resulting in longer total parking duration. This phenomenon was mainly caused by consistent traffic

control measures implemented in Beijing dating from the 2008 Beijing Olympic Games, in which the number of vehicles allowed within the Fifth Ring Road was strictly limited.

### **3.2 Emissions and implications to policy**

In 2015, China's on-road vehicles emitted 4.21 Tg VOCs in total (Table 4). Figure 4a-f shows the provincial results of vehicular VOCs emissions in 2015. VTEs were still the predominant contributor of total VOCs emission with a proportion of 60.80% (Figure 4a). VEEs, with a contributive share of 39.20% in total emissions, is evidently nonnegligible and should be taken into consideration for future management. Among provinces with large fleets of light-duty vehicles, Guangdong, Shandong and Jiangsu ranked topped the league table, with respective contribution of 10.66%, 8.85% and 6.54% to the total amounts of VOCs emission. A slight difference was observed between the ranking of tailpipe exhaust and evaporation (Figure 4b and 4c), 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 were included in Figure 4a. Refueling was of considerable amount (7.83%) but could be effectively controlled by stage-II systems in service stations. Thus, running loss (81.05%) and diurnal (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 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-4f provided sub-classification for tailpipe exhausts. Passenger vehicles, trucks and motorcycles should all be considered for tailpipe VOCs control, in which their contributions are 49.86%, 28.15% and 21.99% respectively. Tailpipe VOCs of passenger cars were widely recognized and attended, in contrast of motorcycles and trucks that were commonly overlooked. Taxis and buses contributed 12.22% and 2.04% of total tailpipe emissions in public transport. While populations 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 VOCs emissions. While the activity data for LDPVs were greatly improved

in this study for reduced uncertainty, improved data consisting reliable information for LDTs are expected in the future. China 0 vehicles were still a dominant source (35.19%) for tailpipe VOCs emission. Vehicles before China 4 (not included) contributed 94.67% to total tailpipe emissions.

In 2015, China's on-road vehicles emitted 200.37 Gg IVOCs in total (Table 5). Figure 5 provides IVOCs 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 72.4% share in IVOCs emission, was 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 (Figure 6). Detailed emission amounts were provided for 36 species, while others were summed in the category named "others". Toluene, i-pentane, benzene were main species in gasoline exhaust. N-butane, i-pentane, i-butane and propane were the main species in evaporation. Evaporation shared 83.66% of n-butane, i-pentane, and i-butane vehicle emissions. Formaldehyde and acetaldehyde were mostly contributed by diesel tailpipe emissions. Small fraction of unresolved complex mixture (UCM) 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 VOCs emissions, emission intensity by area and emission per vehicles among provinces. As expected, developed regions such as Beijing, Tianjin and Shanghai showed highest emission intensity by area (4500~9000 kg km<sup>-2</sup>), while other provinces range between 13~2700 kg km<sup>-2</sup>. Beijing's lowest emissions per vehicle compared to other regions (14~85 kg per vehicle), indicated its fleet as the cleanest. Thus, mere technological approaches for emission reduction in more developed cities could be increasingly difficult. Alternative approaches such as reduction of population intensity as well as human behavior modification on vehicle operation should be considered as main strategies in these regions.

### 450 3.3 Uncertainty analysis

Inevitable uncertainties are present in VOCs emission inventories due to the use of different input data, including activity characteristics, emission factors and VOCs emission profiles. Total vehicle emissions of VOCs are 4.21 Tg yr<sup>-1</sup> with a 95% confidence interval ranges from 2.90-6.54 Tg. The overall uncertainties in this inventory are estimated at -28.53 to 61.35% for total VOC emissions. The  
455 uncertainties of detailed categories were 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*). Differences could be explained with following reasons. Firstly, vehicle evaporative emission was discretely considered in  
460 detail for the first time, which substantially increased total VOCs. Secondly, data of vehicle usage was derived from big data, which was a survey for 'live' vehicles. The VKT used in our estimation is based on vehicle age, causing lowered estimation for emissions of older vehicles. LDTs, China 0 and China 1 vehicles have substantial shares in total emission, yet data of these vehicles had the largest uncertainty according to the author's experience. Improved activity data of these vehicles would further reduce  
465 uncertainty in our inventory

Evaluation of the uncertainty in IVOC emission inventory is indeed challenging as the majority of them comes from emission factors. IVOC measurements are recently more and more recognized, global attention could be expected to better existing emission factor database, considerably reducing uncertainty.

470 The uncertainty of species profile was significant in exhausts and negligible in evaporation. Reason being 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 specie profile. Achieving minimal uncertainty of the species based inventory for evaporation. The uncertainty  
475 for exhaust species was mainly 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, VOCs 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 on building accurate species profiles for exhaust.

#### 480 **4 Conclusion**

The merits 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 statistic, first-hand evaporation data and REIB framework to account inter-provinces transportation for trucks. The total VOCs emissions from on-road vehicles in China were about 4.21 Tg in 2015.

485 Emission factors for running loss of evaporation are urgently needed to better 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 unconvincing.

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

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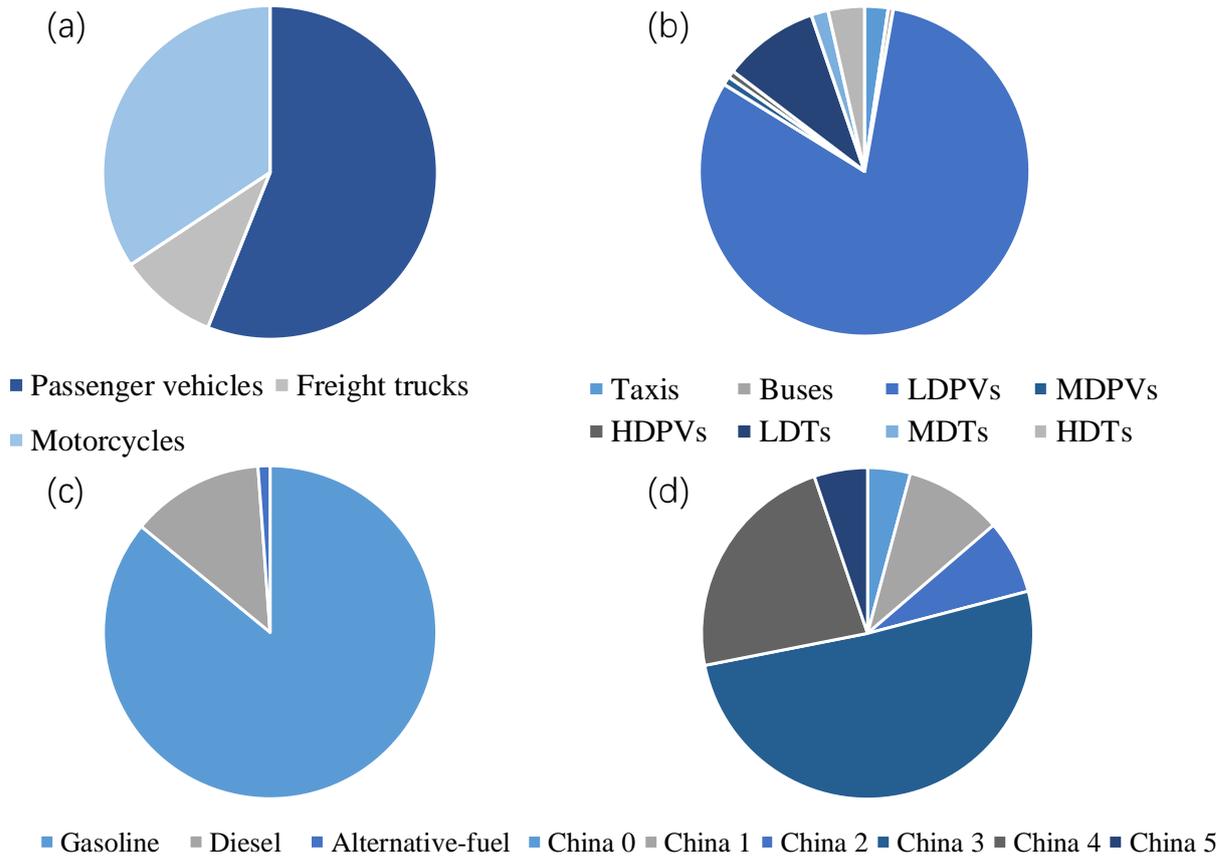
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620 **Figure 1. The percentages by vehicle types, fuel types and emission levels of China vehicle fleet.**

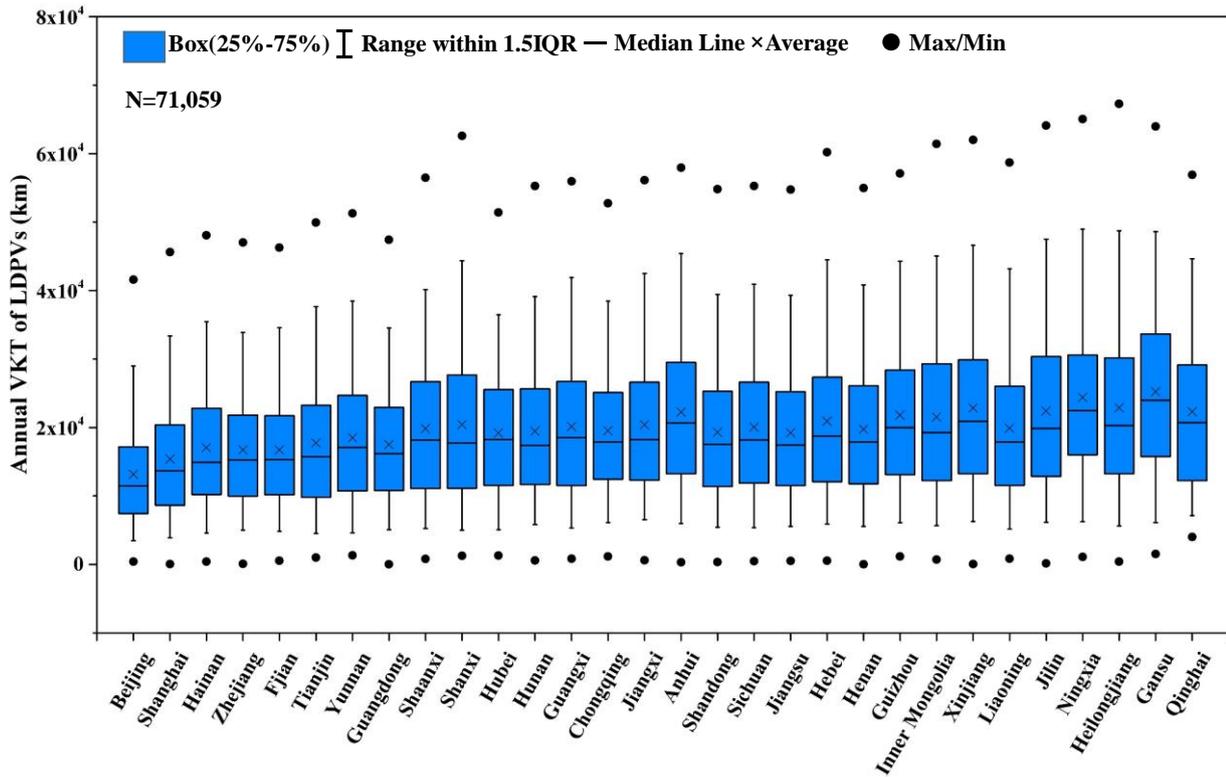
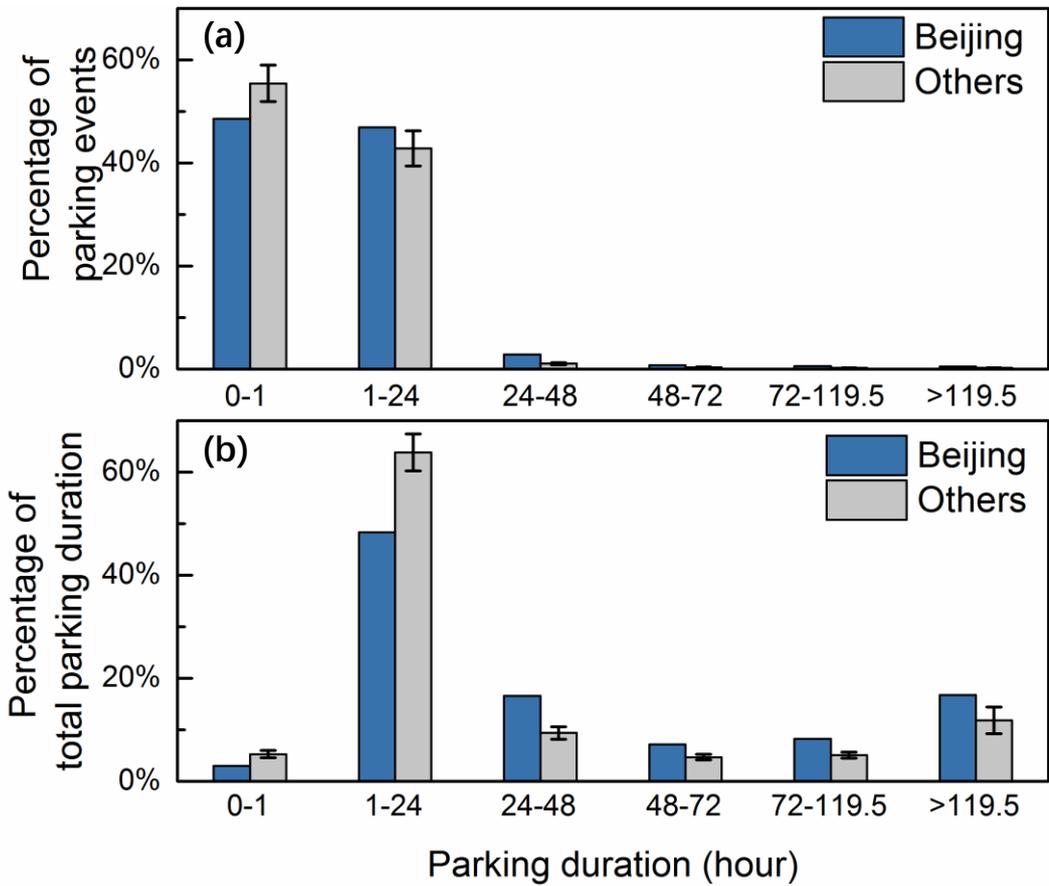
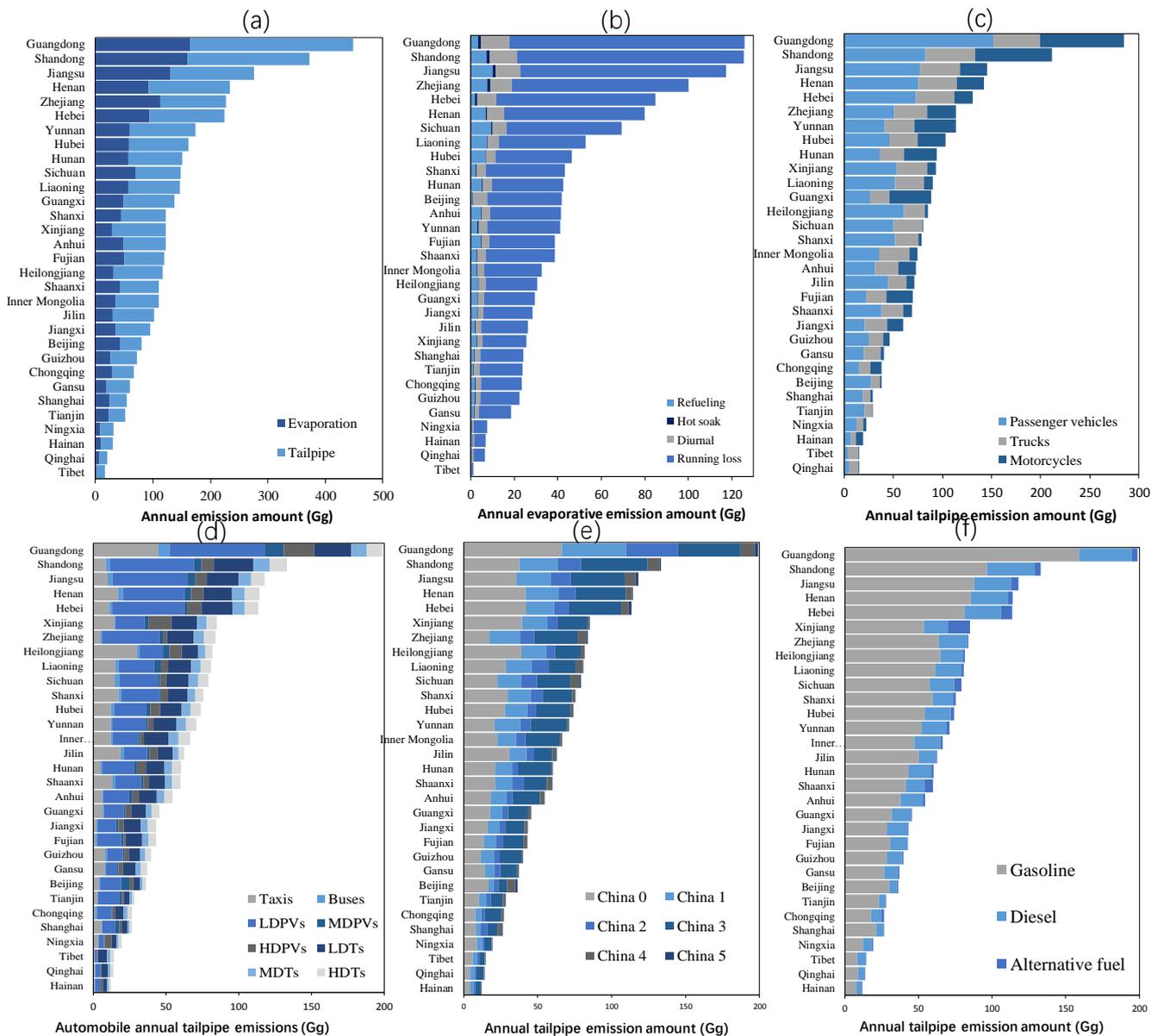


Figure 2. Provincial annual VKT of LDPVs in China.

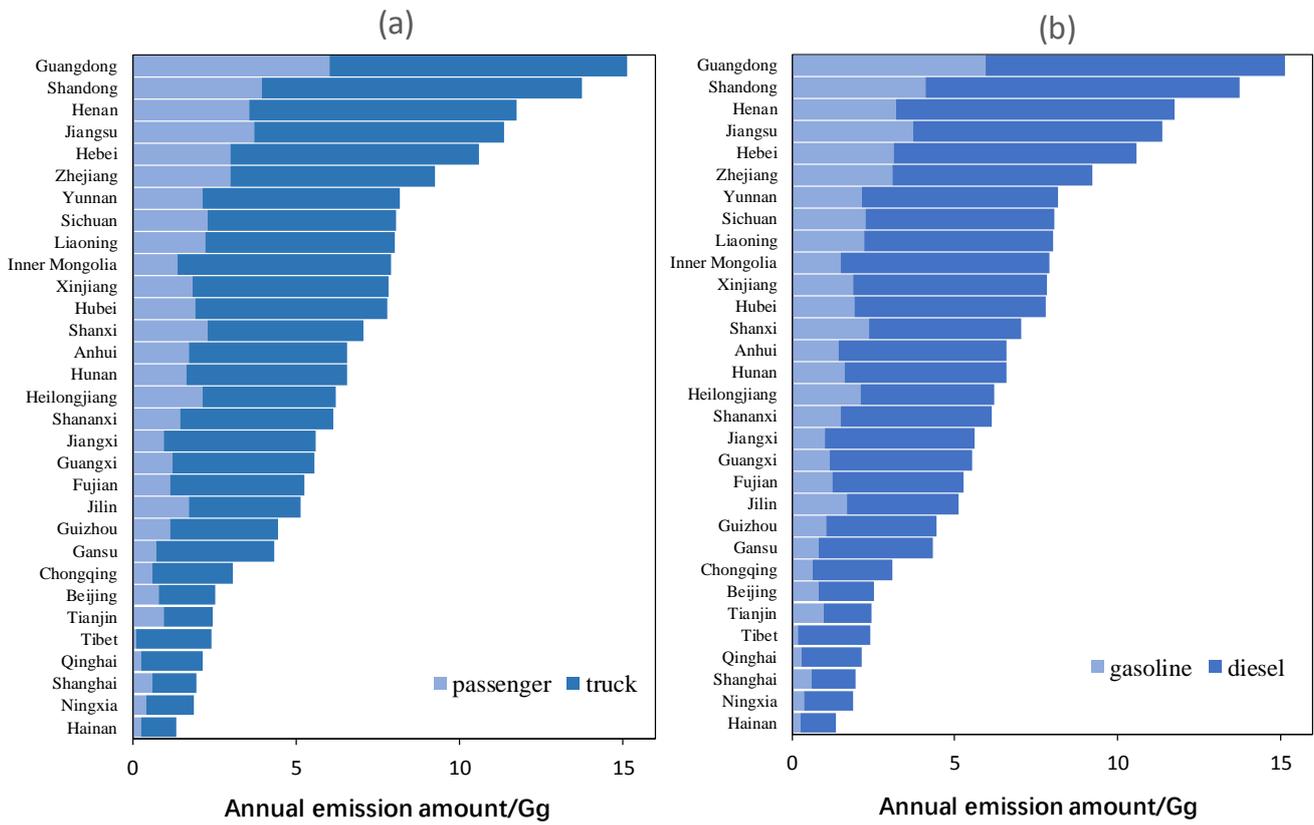


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**Figure 3. Real-world parking duration distribution (a) percentage of parking events, (b) percentage of total parking duration.**

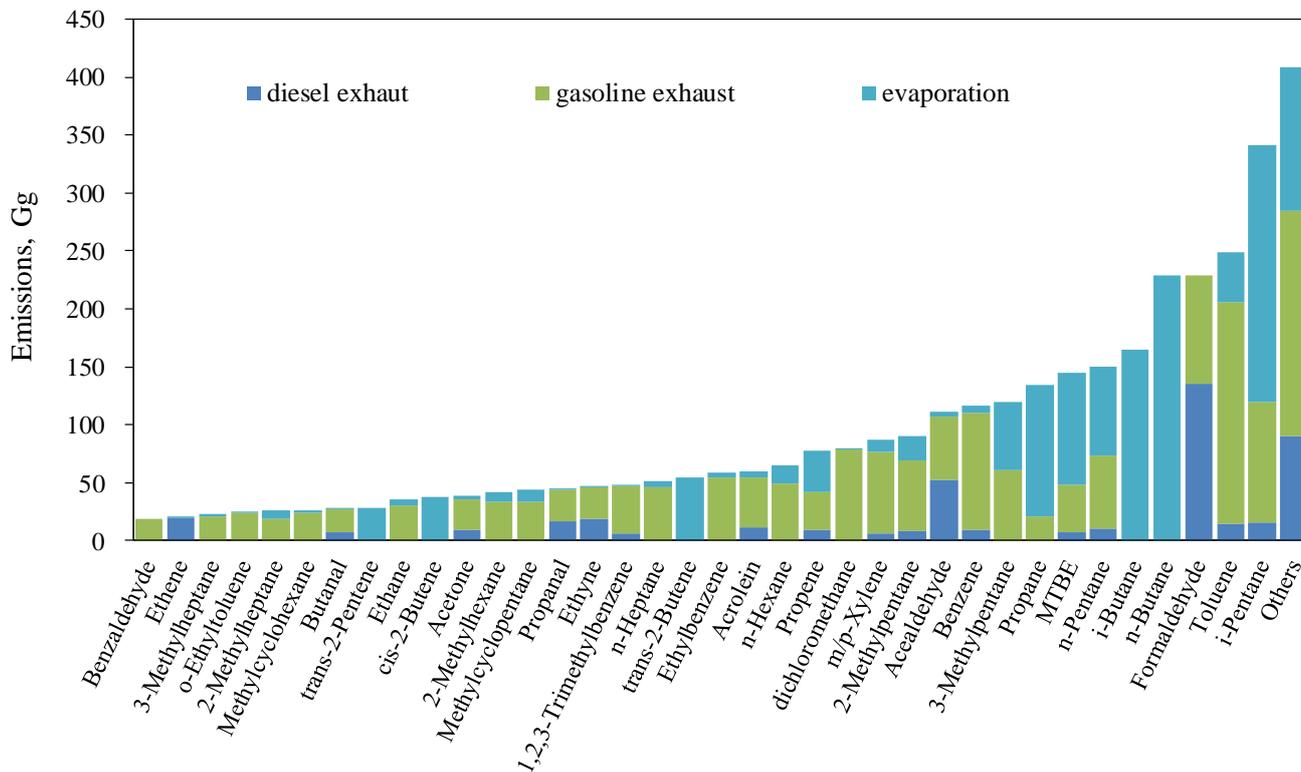


630 **Figure 4. Provincial VOCs emissions from vehicles in 2015 (a) total emission amount classified by emission sources, (b) evaporation emission amount classified by evaporation processes (motorcycles excluded), (c) tailpipe emission amount classified by vehicle types, (d-f) tailpipe emission amounts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



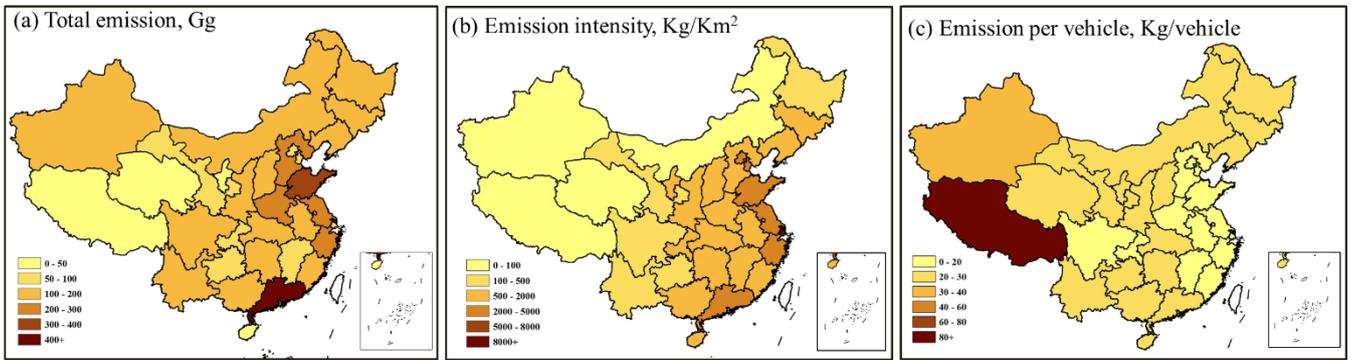
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Figure 5. Provincial IVOCs emissions from vehicles in 2015 (a) total emission amount classified by vehicle types, (b) total emission amount classified by emission sources.

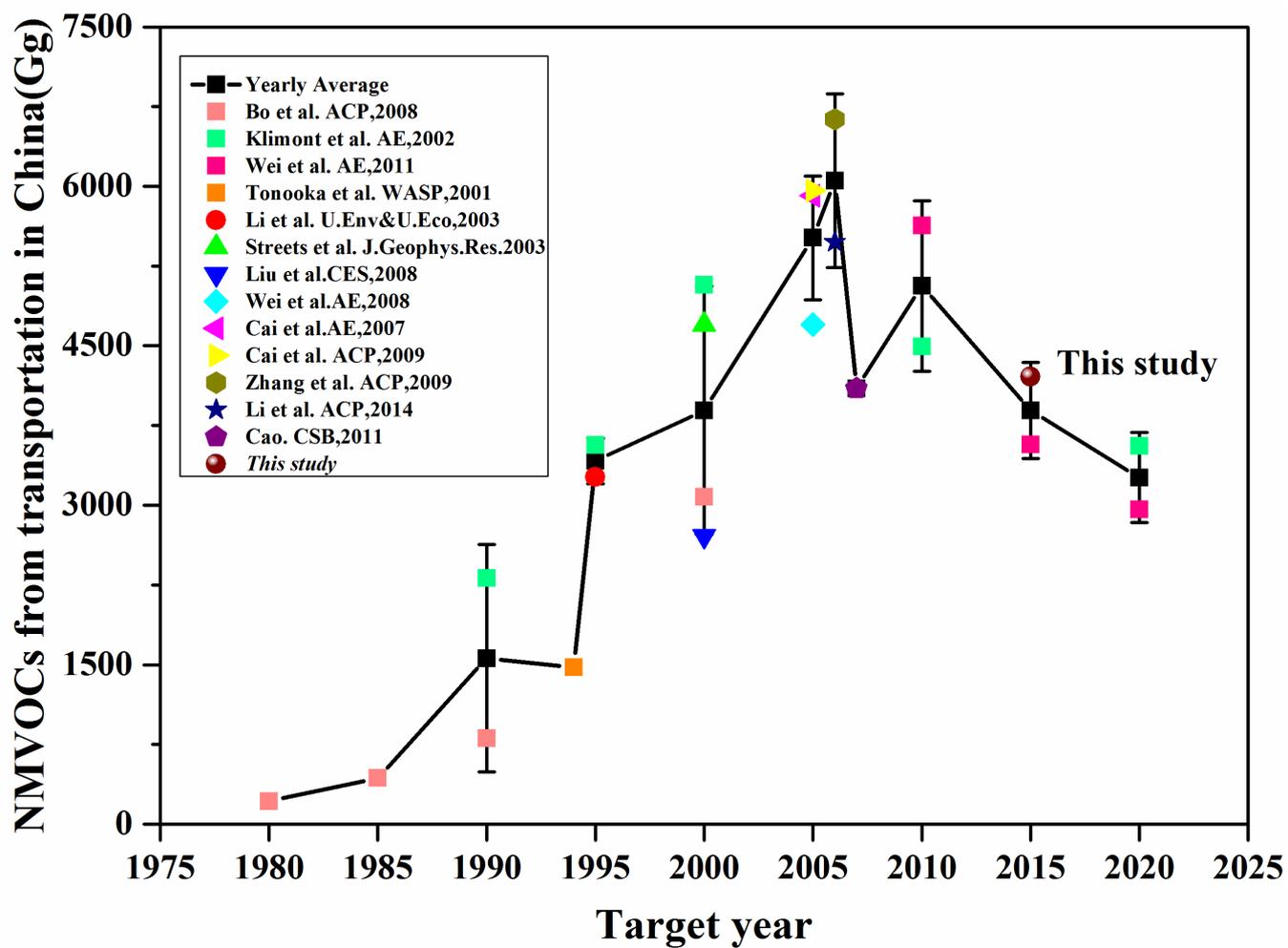


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Figure 6. Speciated VOCs components emissions classified by emission source



645 **Figure 7. Province based emission analysis (a) total emission amount, (b) emission intensity, (c) emission per vehicles (motorcycles were excluded).**



650 Figure 8. Comparing this study to previous transportation emission inventories.

**Table 1 Population of different types of vehicles in China in 2015**

Vehicle type	Population	Fuel type percentage (%)			Control technology	Population
		Gasoline	Diesel	Alternative fuels		
LDPVs	137599368	97.96	1.15	0.90	China 0	7062516
MDPVs	1428102	56.53	40.68	2.78	China 1	16181788
HDPVs	1165836	15.97	75.03	9.00	China 2	12251006
LDTs	15998479	41.50	58.50	0.00	China 3	86584457
MDTs	2826881	18.92	81.08	0.00	China 4	38880534
HDTs	6037719	7.65	92.35	0.00	China 5	8834416
TAs	3910397	61.89	29.37	8.74		
BUs	827935	13.76	55.39	30.85		
GMs	88759010	100	0	0		

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

Province	Vehicle sample size	Annual average VKT (km)
Beijing	2645	13169±7741
Shanghai	3833	15389±8972
Hainan	581	16941±9508
Zhejiang	6356	16740±8897
Fujian	3059	16726±8784
Tianjin	772	17785±10308
Yunnan	1370	18609±10307
Guangdong	16553	17503±8952
Shaanxi	1766	19866±10964
Shanxi	1225	20466±12131
Hubei	976	19313±9669
Hunan	1320	19524±10545
Guangxi	1086	20251±11231
Chongqing	1279	19529±10022
Jiangxi	903	20406±10982
Anhui	1007	22209±11744
Shandong	2449	19333±10420
Sichuan	1984	20120±10959
Jiangsu	5066	19238±10331
Hebei	2933	20915±11594
Henan	1818	19759±10693
Guizhou	746	21985±11800
Inner Mongolia	2322	21660±12118
Xinjiang	991	22901±12122
Liaoning	4049	19953±11365
Jilin	1386	22400±12630
Ningxia	418	24345±12810
Qianghai	171	22488±12265
Heilongjiang	1552	23008±13102
Gansu	443	25460±12659

655 <sup>a</sup>There is 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 Average annual VKT in China (Km/year)**

	LDGTs	LDDTs	MDGTs	MDDTs	HDGTs	HDDTs	TAs	BUs	MDPVs	LDPVs
<b>China 0</b>	22160	19270	35196	21231	27716	24372	138000	50000	31300	114800
<b>China 1</b>	22160	19270	35196	21231	27716	24372				
<b>China 2</b>	26335	26964	40766	28140	33226	38485				
<b>China 3</b>	29467	36581	47927	36366	40310	64128				
<b>China 4</b>	34165	45237	53497	60308	45820	98206				
<b>China 5</b>	34165	45237	53497	60308	45820	98206				

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

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

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

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