Authors’ responses

A combined file includes: (1) comments from Referees, (2) author's response, (3) author's changes in manuscript, (4) a marked-up manuscript version showing the changes made (using track changes in Word)

Anonymous Referee #1

Received and published: 12 August 2017

In this work, Liu et al. developed an updated VOC emission inventory for the on road vehicles in China for 2015. Particularly, they refined their analysis by using vehicle activity data collected from a large number of GPS records, taking into account VOC evaporation emissions from gasoline vehicles, and including tailpipe IVOC emission estimates. The topic is suitable for the Atmospheric Chemistry and Physics and the technical part of the manuscript is relatively well described. However, there are some technical, editorial, and grammatical issues in the current manuscript that need to be clarified and corrected first. The manuscript is highly suggested to be grammar checked by native English speakers. Considering the results of this work is potentially of great value to the atmospheric modeling community, the reviewer suggests a carefully revised manuscript for publication in ACP.

Response: We have revised as your comments point-by-point. The manuscript was carefully reviewed by two native speakers. We carefully response all technical comments and the provide as much as details and raw data.

Thank you for the help!

Abstract line 27. The authors state in the abstract that the VKT level of "trucks were calculated from reported data by more than 2 million trucks in China". According to the Chinese official statistics, there were ~20 million trucks in China in 2015. That means the authors have collected VKT data of ~10%, which is a quit decent sampling ratio, of all trucks in China. However, relevant results are neither described nor referenced to previous studies in the main text (e.g., Sect. 2.2 and 3.1.2). The reviewer suggests the authors adding the description and discussion paragraphs or sections to introduce the methods and results in detail.

Response: We have added discussion and description on trucks' VKT data in Section 2.2. Those data were purchased from commercial big data platform combining with our survey to get the spatial distribution. The platform is the official service provider for all commercial trucks in China under Ministry of Transportation. Due to the licenses in contract, we could not provide raw data to the third party at this stage. Thus, the details of VKT were not released.

Author's changes in manuscript:

In section 2.2 Vehicle activity:

"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 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."

In section 3.1.2 VKT characteristics of LDPVs:

Table 3 summarized vehicle mileages of trucks in China. VKT of trucks is significantly influenced by vehicle age. The annual mileage of China 0 and China 1 trucks are much lower than vehicles of the same type with better control technologies. Aging of trucks greatly impact their performances due to common overloading seen in China. Several cities have implemented low emission zones to restrict entry of trucks with outdated control technologies.

Table 3 Average annual VKT in China (Km/year)

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

The section heading of Sect. 2.1 is “Vehicle stock and classification”. However, only vehicle classification is described.

Response: The method of getting vehicle stock was discussed in the revised section 2.1. The vehicle stock data was discussed in Section 3.1.

Author's changes in manuscript: The following sentences were added.

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 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.

The classification of vehicles is not very clear to the reviewer and needs more clarification. First, the criterions to distinguish LD, MD, and HD passenger vehicles and to distinguish LD, MD, and HD trucks are not given. Second, if taxis are classified as separate vehicle types, the authors should add a statement previously that LD, MD, and HD passenger vehicles do not include taxis. Third, the reviewer is wondering what kind of vehicles are treated as alternative-fuel vehicles? Electric? Plug-in electric? Hybrid? Internal combustion engine vehicles running on alternative fuels such as CNG/LNG/LPG, methanol, or ethanol? Fourth, the vehicle classification is not consistently used in the manuscript. For example, there are a number of vehicle types in Table S4 and S5 that are not described in Sect. 2.1. "Mini" truck is mentioned line 215, but is not described in Sect. 2.1. The authors classified the vehicles into passenger vehicles (LD, MD, and HD), taxis, buses, trucks (LD, MD, and HD), and motorcycles in Sect. 2.1. However, later in the main text (e.g., Sec. 3.2, Figures 1a, 4c, and 5a, and Table S4 and S5), it seems that they also considered taxis and buses as passenger vehicles. If so, this should be stated in the manuscript, and classification criterions should be clearly provided.

Response: The whole section was rewritten to be clear. The revised section has addressed all concerns from reviewer. The new classification is keeping consistent through the whole manuscript as well as the supporting information. The criterions to distinguish LD, MD, and HD passenger vehicles and to distinguish LD, MD, and HD trucks are given. Second, we provided a statement that LD, MD, and HD passenger vehicles do not include taxis. Third, we discussed what kind of vehicles are treated as alternative-fuel vehicles.

Author's changes in manuscript: The following sentences were added.

“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 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 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 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), heavy-duty gasoline trucks (HDGTs), heavy-duty diesel trucks (HDDTs). For trucks, a light-duty truck refers vehicles with mass less than 3500kg. A medium-duty truck refers to vehicles with mass ranging from 3500kg to 12000kg. A heavy-duty truck refers vehicles of mass more than 12000kg."

Line 143, please double check whether provincial motorcycle population data are provided in China Automotive Industry Yearbook.

Response: The provincial GMs population in 2015 was obtained from the Provincial Statistic Yearbook 2016 of each province.

Author's changes in manuscript: The provincial GMs population in 2015 was obtained from the Provincial Statistic Yearbook 2016 of each province.

Sect. 2.2 and 3.1.2. Except for LDPVs, the authors did not provide any VKT data for all the other vehicle types. The review suggests the authors adding a table to summarize the VKT values of all vehicle types (i.e., LDPV, MDPV, HDPV, Taxi, Bus, LDT, MDT, HDT, and motorcycles) used in this work. The reviewer suggests the authors providing the vehicle population, not the population percentage, by vehicle type (i.e., LDPV, MDPV, HDPV, Taxi, Bus, LDT, MDT, HDT, and motorcycles) and by control technology (i.e., China 0 to 5) in Table 1. The population of motorcycle is missing in Table 1. In addition to Figures 4 and 5, the reviewer suggests the authors providing a table to summarize the VOC/IVOC emissions at the country level by vehicle type (i.e., LDPV, MDPV, HDPV, Taxi, Bus, LDT, MDT, HDT, and motorcycles) and by control technology (i.e., China 0 to 5).

Response:

(1) Beside the VKT data for LDPV, which was discussed in detail, the VKT for all the other vehicle types were summarized in Table 3.

(2) Table 1 was revised and now provides vehicle population instead of percentage.

(3) The population of motorcycle was added into Table 1 and the main text in Sect. 3.1.1.

(4) We also added two tables including detailed tailpipe VOC/IVOC emissions by vehicle type and by control technology. For evaporative emissions, it's not calculated based on the vehicle type or control technology. We could not distribute the total gasoline consumption into these categories. Thus, no such data was provided further than Figure 4.

Author's changes in manuscript:

(1) For VKT:

Table 3 summarized vehicle mileages of other vehicle types in China. VKT of trucks is significantly influenced by vehicle age. The annual mileage of China 0 and China 1 trucks are much lower than vehicles of the same type with better control technologies. Aging of trucks greatly impact their performances due to common overloading seen in China. Several cities have implemented low emission zones to restrict entry of trucks with outdated control technologies.

Table 3 Average annual VKT in China (Km/year)

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

(2) For population:

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.

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		

(3) For motorcycle population:

GMs and non-GM vehicles contributed 34.3% (88,759,010) and 65.7% (169,794,718) respectively (Figure 1) among the 259 million total on-road vehicles in China in the year 2015.

(4) For emissions:

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
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	25.15	41.60	13.52	85.70	6.52	0.03	172.52
MDTs	42.20	12.18	1.16	7.90	0.50	0.01	63.95
HDTs	44.34	10.83	2.47	56.38	5.42	0.21	119.65
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

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
LDPVs	5.07	18.05	1.72	10.11	2.59	0.23	37.76
MDPVs	0.31	0.09	0.02	0.06	0.01	0.00	0.51
HDPVs	0.40	0.28	0.07	0.33	0.26	0.00	1.33
LDTs	1.58	2.66	0.61	18.38	2.82	0.02	26.07
MDTs	2.01	0.77	0.27	4.86	0.60	0.01	8.51
HDTs	1.48	1.19	0.44	27.68	5.25	0.21	36.26
TAs	1.97	5.77	0.49	2.27	0.22	0.01	10.73
BUs	0.02	0.02	0.01	0.02	0.00	0.00	0.07

About emission factors (EFs) of VOC/IVOC, the reviewer has the following suggestions and questions:

(1) Title of Table S4 and S5, indicate these are "tailpipe" VOCs. Please also double check whether the unit is mg/km or g/km

Response: The titles of Table S4 and S5 were revised and the units are all g/km.

Author's changes in manuscript:

Table S4. VOCs tailpipe emission factors used in this study (g/km).

Table S5. IVOCs tailpipe emission factors used in this study (g/km).

(2) Tailpipe EFs of motorcycles are missing in Table S4 and S5. They are not

mentioned in the main text, either.

Response: Tailpipe EFs of motorcycles are added in Table S4. The IVOC emissions were only calculated for non-GMs. We revised the sentences in Sect. 2.3 to clarify this point.

Author's changes in manuscript:

For IVOCs emission factors, a mapping to match US emission certification level to China emission level was built (Table S3). Only the non-GMs were considered for the IVOC emissions evaluation.

Table S4. VOCs tailpipe emission factors used in this study (g/km).

			Passenger vehicles			
	China 0	China 1	China 2	China 3	China 4	China 5
LDGTAs	3.840	1.368	0.963	0.454	0.277	0.257
LDDTAs	0.785	0.071	0.046	0.024	0.016	0.016
LDATAs	3.788	0.433	0.398	0.115	0.066	0.293
LDGPVs	2.685	0.663	0.314	0.191	0.075	0.056
LDDPVs	0.785	0.071	0.046	0.024	0.016	0.016
LDAPVs	2.236	0.236	0.164	0.094	0.062	0.091
MDGBUs	5.144	5.255	1.980	0.869	0.418	0.418
MDDBUs	2.668	0.576	0.351	0.283	0.107	0.054
MDABUs	3.840	3.200	2.860	1.720	1.192	1.192
MDGPVs	3.695	2.567	1.443	0.373	0.107	0.107
MDDPVs	1.493	1.425	0.425	0.364	0.383	0.383
MDAPVs	1.920	1.600	1.430	0.860	0.596	0.596
HDGBUs	5.144	5.255	1.980	0.869	0.418	0.418
HDDBUs	2.668	0.576	0.351	0.283	0.107	0.054
HDABUs	3.840	3.200	2.860	1.720	1.192	1.192
HDGPVs	5.144	5.255	1.980	0.869	0.418	0.418
HDDPVs	2.668	0.576	0.351	0.283	0.107	0.054
HDAPVs	3.840	3.200	2.860	1.720	1.192	1.192
			Trucks			
		China 0	China 1	China 2	China 3	China 4/ 5
	LDGTs	5.391	3.593	2.389	0.637	0.176
	LDDTs	2.267	2.205	1.411	0.384	0.194
Urban road	MDGTs	7.441	7.326	3.268	1.482	0.619
	MDDTs	4.863	1.742	0.455	0.219	0.111
	HDGTs	7.295	7.306	3.249	1.464	0.600

	HDDTs	4.413	0.970	0.562	0.276	0.139
Provincial road	LDGTs	4.040	2.693	1.841	0.530	0.147
	LDDTs	1.699	1.653	1.087	0.320	0.162
	MDGTs	5.577	5.490	2.449	1.111	0.464
	MDDTs	3.645	1.306	0.341	0.164	0.083
	HDGTs	5.467	5.475	2.435	1.097	0.450
	HDDTs	3.308	0.727	0.421	0.207	0.105
National road	LDGTs	4.376	2.916	1.924	0.549	0.152
	LDDTs	1.840	1.790	1.136	0.331	0.167
	MDGTs	6.040	5.946	2.652	1.203	0.503
	MDDTs	3.947	1.414	0.369	0.178	0.090
	HDGTs	5.921	5.930	2.637	1.188	0.487
	HDDTs	3.582	0.787	0.456	0.224	0.113
Freeway	LDGTs	4.119	2.745	1.837	0.536	0.148
	LDDTs	1.732	1.685	1.085	0.323	0.163
	MDGTs	5.685	5.597	2.497	1.132	0.473
	MDDTs	3.716	1.331	0.348	0.168	0.085
	HDGTs	5.574	5.582	2.483	1.118	0.458
	HDDTs	3.372	0.741	0.429	0.211	0.107
County road	LDGTs	7.010	4.673	3.059	0.798	0.221
	LDDTs	2.948	2.868	1.806	0.482	0.243
	MDGTs	9.677	9.527	4.250	1.927	0.805
	MDDTs	6.324	2.266	0.592	0.285	0.145
	HDGTs	9.487	9.501	4.226	1.903	0.780
	HDDTs	5.740	1.261	0.731	0.358	0.181
			GMs			
GMs				1.269		

(3) The vehicle classification in Table S4 and Table S5 is different from the description in Sect. 2.1. For example, LDGTAs, LDDTAs, LDABs, MDGBUs, MDDBUs, MDABs, HDGBUs, HDDBUs, and HDABs, these vehicle types are not mentioned in Sect. 2.1, nor in the results and discussion section. If the study was conducted with more detailed vehicle classification, it should be introduced in the main text.

Response: Yes. The calculation is based on more detailed classification. We have modified Sect. 2.1. Now the vehicle classification is consistent through the whole manuscript.

Author's changes in manuscript:

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 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 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 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), heavy-duty gasoline trucks (HDGTs), heavy-duty diesel trucks (HDDTs). For trucks, a light-duty truck refers vehicles with mass less than 3500kg. A medium-duty truck refers to vehicles with mass ranging from 3500kg to 12000kg. A heavy-duty truck refers vehicles of mass more than 12000kg.

Passenger vehicles						
	China 0	China 1	China 2	China 3	China 4	China 5
LDGTAs	3.840	1.368	0.963	0.454	0.277	0.257
LDDTAs	0.785	0.071	0.046	0.024	0.016	0.016
LDATAs	3.788	0.433	0.398	0.115	0.066	0.293
LDGPVs	2.685	0.663	0.314	0.191	0.075	0.056
LDDPVs	0.785	0.071	0.046	0.024	0.016	0.016
LDAPVs	2.236	0.236	0.164	0.094	0.062	0.091
MDGBUs	5.144	5.255	1.980	0.869	0.418	0.418
MDDBUs	2.668	0.576	0.351	0.283	0.107	0.054
MDABUs	3.840	3.200	2.860	1.720	1.192	1.192
MDGPVs	3.695	2.567	1.443	0.373	0.107	0.107
MDDPVs	1.493	1.425	0.425	0.364	0.383	0.383
MDAPVs	1.920	1.600	1.430	0.860	0.596	0.596
HDGBUs	5.144	5.255	1.980	0.869	0.418	0.418
HDDBUs	2.668	0.576	0.351	0.283	0.107	0.054
HDABUs	3.840	3.200	2.860	1.720	1.192	1.192
HDGPVs	5.144	5.255	1.980	0.869	0.418	0.418
HDDPVs	2.668	0.576	0.351	0.283	0.107	0.054
HDAPVs	3.840	3.200	2.860	1.720	1.192	1.192

Trucks						
		China 0	China 1	China 2	China 3	China 4/ 5
Urban road	LDGTs	5.391	3.593	2.389	0.637	0.176
	LDDTs	2.267	2.205	1.411	0.384	0.194
	MDGTs	7.441	7.326	3.268	1.482	0.619
	MDDTs	4.863	1.742	0.455	0.219	0.111
	HDGTs	7.295	7.306	3.249	1.464	0.600
	HDDTs	4.413	0.970	0.562	0.276	0.139
Provincial road	LDGTs	4.040	2.693	1.841	0.530	0.147
	LDDTs	1.699	1.653	1.087	0.320	0.162
	MDGTs	5.577	5.490	2.449	1.111	0.464
	MDDTs	3.645	1.306	0.341	0.164	0.083
	HDGTs	5.467	5.475	2.435	1.097	0.450
	HDDTs	3.308	0.727	0.421	0.207	0.105
National road	LDGTs	4.376	2.916	1.924	0.549	0.152
	LDDTs	1.840	1.790	1.136	0.331	0.167
	MDGTs	6.040	5.946	2.652	1.203	0.503
	MDDTs	3.947	1.414	0.369	0.178	0.090

	HDGTs	5.921	5.930	2.637	1.188	0.487
	HDDTs	3.582	0.787	0.456	0.224	0.113
Freeway	LDGTs	4.119	2.745	1.837	0.536	0.148
	LDDTs	1.732	1.685	1.085	0.323	0.163
	MDGTs	5.685	5.597	2.497	1.132	0.473
	MDDTs	3.716	1.331	0.348	0.168	0.085
	HDGTs	5.574	5.582	2.483	1.118	0.458
	HDDTs	3.372	0.741	0.429	0.211	0.107
County road	LDGTs	7.010	4.673	3.059	0.798	0.221
	LDDTs	2.948	2.868	1.806	0.482	0.243
	MDGTs	9.677	9.527	4.250	1.927	0.805
	MDDTs	6.324	2.266	0.592	0.285	0.145
	HDGTs	9.487	9.501	4.226	1.903	0.780
	HDDTs	5.740	1.261	0.731	0.358	0.181
GMs						
GMs				1.269		

Table S5. IVOCs tailpipe emission factors used in this study (g/km).

Passenger vehicles					
	China 0/1	China 2	China 3	China 4	China 5
LDDTAs/LDGTAs/LDGPVs/LDDPVs	**0.09287**[1]	**0.00977**	**0.00809**	**0.00413**	**0.00151**
MDGBUs/MDDBUs/MDGPVs/MDDPVs	**0.01837**	**0.00424**	**0.00532**	**0.00532**	**0.00221**
HDGBUs/HDDBUs/HDGPVs/HDDPVs	**0.01671**	**0.00447**	**0.00447**	**0.02553**	**0.00231**
Trucks					
	China 0/1	China 2	China 3	China 4	China 5
LDGTs	**0.07200**	**0.00266**	**0.00266**	**0.00333**	**0.00272**
LDDTs	0.06072[2]	0.06072	0.06072	0.08574	0.08574
MDGTs	0.10800	0.00399	0.00399	0.00500	0.00409
MDDTs	0.09108	0.09108	**0.09108**	**0.12861**	0.01122
HDGTs	0.10800	0.00399	0.00399	0.00500	0.00409
HDDTs	0.34478	0.34478	0.34478	**0.34478**	**0.01122**

[1] The bold fonts mean that data is from measurements in literature. It is equal to the median of measurements for all samples in this vehicle category.

[2] The non-bold fonts mean that no measurement data is available. The emission factor is derived based on the following assumptions: EF(HD)=EF(MD)=1.5*EF(LD) and EF (control level) = EF (control level±n, where measurement data is available).

(4) The EFs of evaporation are not given. The reviewer suggests adding a table listing EFs of diurnal loss, hot soak, refueling, and running loss by vehicle type (i.e., LDPV, MDPV, HDPV, Taxi, Bus, LDT, MDT, HDT, and motorcycles). Data sources should be provided too

Response: A table including EFs of evaporation is added in supporting information. The data sources are also provided.

Author's changes in manuscript:

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.

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.

$$E_{GMs,i} = EF_{GMs} \times VP_{i,GMs} \times VKT_{i,GMs}\ , \qquad (7)$$

where $E_{GMs,i}$ represents the annual evaporative emissions from GMs registered in province i (g·year^{-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 (ICCT) were utilized (***ICCT, 2012***). $VKT_{i,GMs}$ represents the annual VKT of GMs in province i (km·year^{-1}).

Table S6. Evaporation emission factors used in this study.

		Parking duration	Unite	Emission factors
vehicles	Diurnal	<24 hour	g/hour	0.094[1]
		24-48 hour	g/hour	0.247[1]
		>48 hour	g/hour	0.339[1]
	Hot soak		g/hour	0.083[1]
	Refueling (without control)		g/L	0.848[1]
	Running loss		g/hour	11.6[2]
motorcycle			g/km	0.57[3]

References:

1. Liu, H.; Man, H.; Tschantz, M.; Wu, Y.; He, K.; Hao, J., VOC from Vehicular Evaporation Emissions: Status and Control Strategy. *Environ. Sci. Technol* **2015**, 49, (24), 14424-14431. DOi:10.1021/acs.est.5b04064

2. EPA-420-R-12-027; Development of Evaporative Emissions Calculations for the Motor Vehicle Emissions Simulator MOVES2010; United States Environmental Protection Agency; Washington, DC, **2012**; https://nepis.epa.gov/Exe/ZyPDF.cgi/P100F3ZY.PDF?Dockey=P100F3ZY.PDF

3. 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), 2007; http://www.theicct.org/sites/default/files/publications/twothree_wheelers_2007.pdf

(5) Line 239-248. First, the meanings of T, N, and P in Eqs. (3)-(6) are not provided. Second, besides simply providing the meanings of each variable in Eqs. (3)-(6), the authors are suggested to explain these equations.

Response: The meanings of each parameter were provided now. Some sentences were added above the equations to explain the calculation.

Author's changes in manuscript:

"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."

"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."

"According to the US EPA, hot soak is defined as the evaporative losses that occur within the one-hour period after the engine is shut down (EPA420-R-01-026). If the parking duration is longer than one hour, then the extra vapor losses fall into 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):"

"China is following European control experiences to popularize Stage-II vapor control system in refuelling stations to reduce refuelling loss. 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."

(6) Line 244, 264, why China 4 LDGVs' EFs could be used for all non-motorcycle vehicle types and control technologies?

Response: The EFs were assumed to be the same for China 1 to China. The following description was added.

Author's changes in manuscript:

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.

(7) Eqs. (7), (9), (11), (12). The authors claimed that the units of EFs are g/hour. The reviewer believes that this is not correct.

Response: The equations and the explanation were revised. All the EFs including units were listed in the new Table S6.

Author's changes in manuscript:

Table S6. Evaporation emission factors used in this study.

		Parking duration	Unite	Emission factors
vehicles	Diurnal	<24 hour	g/hour	0.094[1]
		24-48 hour	g/hour	0.247[1]
		>48 hour	g/hour	0.339[1]
	Hot soak		g/hour	0.083[1]
	Refueling (without control)		g/L	0.848[1]
	Running loss		g/hour	11.6[2]
motorcycle			g/km	0.57[3]

Line 290. Is the motor gasoline consumption by province calculated or derived from official statistics? Methods or data sources should be provided.

Response: The gasoline consumption is from statistic data. A sentence was added to provide the method and data source.

Author's changes in manuscript:

"CF_i represents the annual motor gasoline consumption of province i (L·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."

Main text after Sect. 3.2 may need to be polished to make it read like a scientific article.

Response: Two native speakers polished the language of the paper. We also contacted Copernicus Publication copy-editing service. After this manuscript was accepted by ACP, they will polish the language.

The authors are suggested to check citations carefully before submitting the revised manuscript. Examples are: Line 61, change "Cai et al" to "Cai and Xie". Remove "(Cai et al., 2009)" in line 62 Yang et al., 2015 is mentioned several times in the manuscript (e.g., lines 95, 106, 150, 194, 205, 281, etc.). However, there are two references by Yang et al. in 2015. Letters a and b should be added to the year both in the in-text citation as well as in the reference list. Line 163-164, 179-180, "Zhao et al." to "Zhao et al. (2015, 2016)" and remove "(Zhao et al.; 2016; Zhao et al; 2015)" Line 275, 307, "ICCT, 2012" is not in the reference list Line 301, "MOVES, 2010" is not in the reference list Line 326, "Man et al., 2016" is not in the reference list. In the reference list, there are lots of references that are not cited in the main text. Please have them carefully checked before submitting the revised manuscript.

Response: We have checked all the citations. The reference list is match with those cited in the main text now. The Endnote templates from ACP website were used to format all the references. All the problems mentioned above were corrected in this revision.

Minor editorial issues:

Line 121, remove “five”. According the introduction section, it seems that there are six deficiencies, while in Sect. 4, it seems the authors discussed four aspects.

Response: Accepted.

Line 217, “POA” should be defined in the first appearance.

Response: Accepted.

Author's changes in manuscript: “This ratio was similar to the VOCs or primary organic aerosol (POA) emission ratios of heavy/light for trucks.”

Line 257, “GTs”??

Response: Corrected.

Line 324, incomplete sentence

Response: Corrected.

Line 394, “eg.” to “e.g., ”

Response: Corrected.

What is the unit of EFs in Table S3?

Response: Added. The unit is mg/kg-fuel.

The caption of Figure 1 should be self-explained.

Response: Corrected.

Author's changes in manuscript: The caption was revised to "The percentages by vehicle types, fuel types and emission levels of China vehicle fleet".

There are grammatical errors throughout the manuscript. I strongly suggest a grammar checking by native English speaker before submitting the revised manuscript. Examples in the first five pages are: Abstract should be written in the present tense.

Response: A native speaker polished the language of the paper.

Line 41-42. Line 47, remove "the year of"

Response: Accepted.

Line 62, add "during" after "China"

Response: Accepted.

Line 63, "include" to "included", add "a" before "part"

Response: Accepted.

Line 68, "provide" o "provided"

Response: Accepted.

Line 70 remove "trend"

Response: Accepted.

Line 74, "has" to "have", "a non-ignorable contributor" to "non-ignorable contributors"

Response: Accepted.

Line 76 Line 81, "profile" to "profiles"

Response: Accepted.

Line 82, "with" to "to"

Response: Accepted.

Line 83, "were" to "are", "method section" to "Sect. 2"

Response: Accepted.

Line 84, "impact" to "impacts", "atmospheric condition" to "air quality"???

Response: Accepted.

Line 86, "complicate" to "complicated", add "of" after "a series"

Response: Accepted.

Line 90, "measurements" to "measurement", "none of the" to "to our knowledge, there is no", add "for China" at the end of this sentence

Response: Accepted.

Line 98 Line 100, “method” to “methods”

Response: Accepted.

Line 106, “emission” to “emissions were”

Response: Accepted.

Line 109, “common-used” to “commonly-used”

Response: Accepted.

Line 111, “provided” to “provide”, “level” to “levels”

Response: Accepted.

Line 113, “recently” to “recent”.

Response: Accepted.

Line 114, add “furthermore,” at the beginning of the sentence, “provides” to “provide”, “types” to “type”

Response: Accepted.

Line 115, remove “However,”

Response: Accepted.

Line 116-117, change to “More detailed vehicle population data by fuel type and by control technology are required to calculate emissions because they have been reported to . . .”

Response: Accepted.

Line 120, “were” to “are”

Response: Accepted.

Line 121, “were” to “are”

Response: Accepted.

Line 123, “were” to “are”

Response: Accepted.

Line 124, change to “there is no local IVOC emission factor reported”

Response: Accepted.

Anonymous Referee #2

Received and published: 16 August 2017

General Comments: This work developed an updated speciated emission inventory of VOCs and IVOCs from vehicles in China for the year of 2015 based on a set of state-of-the-art methodologies and a mass of local measurement data. The strength of this inventory is that massive GPS records and questionnaire analysis are collected to better characterize the activity level. In addition, in terms of the method, this work improved the emission estimation by including evaporative emission calculation and applying road emission intensity based approach. This well-written and well-structured paper is potentially important and will be valuable in the future for modelling the formation of fine particles and ozone pollution in China. There are a few comments that need to be addressed to improve the paper and make it more accessible to the wide audience of users of the information presented.

Response: Thank you for the comments. We try our best to improve the manuscript based on your comments. The point-by-point response is provided.

Specific Comments:

In the first place, the information need to be made available, for example through the journal with a doi, or through the website of the author's institute.

Response: Accepted. Firstly, instead of providing figures and percentages, we have revised and added several tables to provide the raw data and the emission data. Due to the length limitation, the additional dataset are available upon request.

Author's changes in manuscript:

Table 1 to table 5 and table S1 to table S9 were added to provide information as detail as possible.

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

Table 2 Provincial annual average VKT of LDPVs in China

Table 3 Average annual VKT of trucks in China (Km/year)

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

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

Table S1. Previous studies on emission inventory of VOCs from vehicles in China.

Table S2. Mapping from vehicles in US to China certification level (Gasoline).

Table S3. Mapping from vehicles in US to China certification level (Diesel)

Table S4. VOCs tailpipe emission factors used in this study (g/km).

Table S5. IVOCs tailpipe emission factors used in this study (g/km).

Table S6. Evaporation emission factors used in this study.

Table S7. Characteristics of probability distribution functions for selected key model parameters and input variables included in the uncertainty analysis.

Table S8. Assignments from Real Compounds to Carbon Bond 05 (CB05) Model Species for diesel exhaust, gasoline exhaust and evaporation in China (Gmol).

Table S9. Uncertainty range of emission inventories.

A second recommendation is that speciated emission inventory of VOCs and IVOCs based on prevailing lumped chemical mechanisms like CB05 and SAPRC are suggested to be provided since that this emission database will be mainly used in chemical transport models.

Response: Accepted. A table and discussions were added.

Author's changes in manuscript:

Table S8. Assignments from Real Compounds to Carbon Bond 05 (CB05) Model Species for diesel exhaust, gasoline exhaust and evaporation in China (Gmol).

	Diesel exhaust	Gasoline exhaust	Evaporation
PAR	7.179	39.017	72.452
OLE	0.371	0.994	1.380
TOL	0.217	2.389	0.507
XYL	0.222	1.035	0.189
FORM	4.425	2.700	0.215
ALD2	1.219	1.071	0.095
ETH	0.837	N.D.	0.017
ISOP	N.D.	N.D.	0.012
MEOH	N.D.	N.D.	N.D.
ETOH	N.D.	N.D.	N.D.
ETHA	N.D.	0.882	0.158
IOLE	N.D.	N.D.	2.046
ALDX	0.6852	1.309	0.128
TERP	N.D.	N.D.	N.D.
UNR	1.773	8.276	5.762

There still large uncertainty lies in activity level, emission factor and the estimation method itself. Another recommendation is that uncertainty analysis ought to be conducted and more quantitative results should be provided in Section 3.3.

Response: Accepted.

Author's changes in manuscript:

The uncertainty for emission inventory is assessed using a Monte Carlo method. The probability distributions of key model parameters were established with our experimental data, investigation data and literature review (Table S7). Using these assumptions, a Monte Carlo model was run 10000 times to produce the estimate.

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−1 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 uncertainties of detailed categories were listed in Table S9. These confidence ranges are comparable to other bottom-up emission inventories.

Table S7. Characteristics of probability distribution functions for selected key model parameters and input variables included in the uncertainty analysis.

Parameter or variable			Distribution	Standard division	The 95% confidence interval		
					2.5% percentile	50 % percentile	97.5% percentile
Evaporative emission factors	Diurnal emissions (g/hour)	1-24hour	Log-Normal	0.065	0.023	0.077	0.264
		24-48hour	Log-Normal	0.100	0.107	0.229	0.493
		>48hour	Log-Normal	0.085	0.204	0.331	0.536
	Hot Soak (g/hour)		Log-Normal	0.014	0.059	0.082	0.114
	Base Refuelling (g/L)		Log-Normal	0.077	0.707	0.843	1.009
	Running loss (g/hour)		Log-Normal	4.689	5.072	10.712	22.938
	GMs (g/Km)		Log-Normal	0.550	0.086	0.415	1.945
Parking duration per day in Beijing (hour)			Extreme Value	1.1365	19.4652	22.3486	23.8540

Parking duration per day in other provinces (hour)			Extreme Value	0.9919	19.7238	22.2438	23.5538
Percentage of parking events in Beijing	0-1hour		Log-Normal	0.100	0.320	0.475	0.712
	1-24hour		Log-Normal	0.099	0.306	0.460	0.688
	24-48hour		Log-Normal	0.006	0.018	0.027	0.041
	48-72hour		Log-Normal	0.002	0.004	0.007	0.012
	72-119.5hour		Log-Normal	0.002	0.002	0.005	0.010
	>119.5hour		Log-Normal	0.000	0.004	0.004	0.005
Percentage of parking events in other provinces	0-1hour		Log-Normal	0.124	0.352	0.539	0.834
	1-24hour		Log-Normal	0.079	0.290	0.420	0.605
	24-48hour		Log-Normal	0.002	0.007	0.010	0.015
	48-72hour		Log-Normal	0.000	0.002	0.003	0.004
	72-119.5hour		Log-Normal	0.002	0.000	0.002	0.007
	>119.5hour		Log-Normal	0.004	0.000	0.001	0.010
Percentage of parking duration in Beijing	0-1hour		Log-Normal	0.006	0.020	0.029	0.043
	1-24hour		Log-Normal	0.099	0.316	0.471	0.703
	24-48hour		Log-Normal	0.040	0.101	0.162	0.260
	48-72hour		Log-Normal	0.014	0.048	0.071	0.103
	72-119.5hour		Log-Normal	0.020	0.050	0.080	0.127
	>119.5hour		Log-Normal	0.040	0.103	0.163	0.255
Percentage of parking duration in other provinces	0-1hour		Log-Normal	0.020	0.024	0.049	0.101
	1-24hour		Log-Normal	0.121	0.433	0.628	0.902
	24-48hour		Log-Normal	0.184	0.004	0.043	0.468
	48-72hour		Log-Normal	0.010	0.030	0.046	0.069
	72-119.5hour		Log-Normal	0.020	0.022	0.047	0.098
	>119.5hour		Log-Normal	0.020	0.084	0.117	0.161
Tailpipe Emission factors of passenger vehicles	GMs		Log-Normal	0.56	0.52	1.16	2.64
	LDGTAs	China0	Log-Normal	1.694	1.550	3.519	8.045
		China1	Log-Normal	0.599	0.558	1.255	2.839
		China2	Log-Normal	0.418	0.392	0.891	1.968
		China3	Log-Normal	0.200	0.184	0.416	0.957

(g/Km)		China4	Log-Normal	0.121	0.112	0.254	0.582
		China5	Log-Normal	0.114	0.104	0.236	0.543
	LDDTAs	China0	Log-Normal	0.337	0.311	0.726	1.608
		China1	Log-Normal	0.031	0.028	0.065	0.150
		China2	Log-Normal	0.020	0.019	0.042	0.096
		China3	Log-Normal	0.010	0.010	0.022	0.050
		China4	Log-Normal	0.007	0.006	0.015	0.033
		China5	Log-Normal	0.007	0.006	0.015	0.033
	LDATAs	China0	Log-Normal	1.649	1.516	3.471	7.828
		China1	Log-Normal	0.187	0.174	0.396	0.884
		China2	Log-Normal	0.176	0.159	0.367	0.829
		China3	Log-Normal	0.050	0.046	0.105	0.241
		China4	Log-Normal	0.029	0.027	0.060	0.139
		China5	Log-Normal	0.127	0.118	0.270	0.608
	LDGPVs	China0	Log-Normal	1.181	1.105	2.473	5.687
		China1	Log-Normal	0.293	0.269	0.608	1.385
		China2	Log-Normal	0.140	0.127	0.287	0.654
		China3	Log-Normal	0.083	0.077	0.174	0.395
		China4	Log-Normal	0.033	0.031	0.069	0.158
		China5	Log-Normal	0.025	0.023	0.052	0.115
	LDDPVs	China0	Log-Normal	0.337	0.311	0.726	1.608
		China1	Log-Normal	0.031	0.028	0.065	0.150
		China2	Log-Normal	0.020	0.019	0.042	0.096
		China3	Log-Normal	0.010	0.010	0.022	0.050
		China4	Log-Normal	0.007	0.006	0.015	0.033
		China5	Log-Normal	0.007	0.006	0.015	0.033
	LDAPVs	China0	Log-Normal	0.977	0.900	2.071	4.581
		China1	Log-Normal	0.104	0.095	0.217	0.486
		China2	Log-Normal	0.073	0.067	0.151	0.347
		China3	Log-Normal	0.065	0.023	0.077	0.264
		China4	Log-Normal	0.027	0.025	0.056	0.127

	China5	Log-Normal	0.039	0.037	0.084	0.186
MDGBUs	China0	Log-Normal	2.223	2.100	4.741	10.559
	China1	Log-Normal	2.306	2.142	4.859	10.957
	China2	Log-Normal	0.852	0.788	1.805	4.030
	China3	Log-Normal	0.383	0.346	0.791	1.838
	China4	Log-Normal	0.184	0.167	0.380	0.872
	China5	Log-Normal	0.184	0.167	0.380	0.872
MDDBUs	China0	Log-Normal	1.184	1.068	2.424	5.614
	China1	Log-Normal	0.254	0.234	0.532	1.211
	China2	Log-Normal	0.153	0.141	0.323	0.728
	China3	Log-Normal	0.122	0.114	0.260	0.583
	China4	Log-Normal	0.047	0.042	0.098	0.220
	China5	Log-Normal	0.024	0.021	0.050	0.112
MDABUs	China0	Log-Normal	1.694	1.550	3.519	8.045
	China1	Log-Normal	1.415	1.299	2.945	6.765
	China2	Log-Normal	1.256	1.146	2.609	5.943
	China3	Log-Normal	0.745	0.697	1.555	3.567
	China4	Log-Normal	0.517	0.484	1.100	2.460
	China5	Log-Normal	0.517	0.484	1.100	2.460
MDGPVs	China0	Log-Normal	1.623	1.482	3.364	7.725
	China1	Log-Normal	1.123	1.043	2.351	5.301
	China2	Log-Normal	0.628	0.587	1.324	2.993
	China3	Log-Normal	0.165	0.150	0.338	0.779
	China4	Log-Normal	0.047	0.042	0.098	0.220
	China5	Log-Normal	0.047	0.042	0.098	0.220
MDDPVs	China0	Log-Normal	0.663	0.602	1.373	3.165
	China1	Log-Normal	0.612	0.576	1.301	2.933
	China2	Log-Normal	0.185	0.172	0.391	0.873
	China3	Log-Normal	0.160	0.146	0.335	0.764
	China4	Log-Normal	0.166	0.155	0.348	0.797
	China5	Log-Normal	0.166	0.155	0.348	0.797

MDAPVs	China0	Log-Normal	0.834	0.763	1.753	4.002
	China1	Log-Normal	0.704	0.650	1.479	3.335
	China2	Log-Normal	0.628	0.571	1.305	2.985
	China3	Log-Normal	0.376	0.347	0.780	1.797
	China4	Log-Normal	0.259	0.246	0.550	1.245
	China5	Log-Normal	0.259	0.246	0.550	1.245
MDGBUs	China0	Log-Normal	2.223	2.100	4.741	10.559
	China1	Log-Normal	2.306	2.142	4.859	10.957
	China2	Log-Normal	0.852	0.788	1.805	4.030
	China3	Log-Normal	0.383	0.346	0.791	1.838
	China4	Log-Normal	0.184	0.167	0.380	0.872
	China5	Log-Normal	0.184	0.167	0.380	0.872
HDDBUs	China0	Log-Normal	1.184	1.068	2.424	5.614
	China1	Log-Normal	0.254	0.234	0.532	1.211
	China2	Log-Normal	0.153	0.141	0.323	0.728
	China3	Log-Normal	0.122	0.114	0.260	0.583
	China4	Log-Normal	0.047	0.042	0.098	0.220
	China5	Log-Normal	0.024	0.021	0.050	0.112
HDABUs	China0	Log-Normal	1.694	1.550	3.519	8.045
	China1	Log-Normal	1.415	1.299	2.945	6.765
	China2	Log-Normal	1.256	1.146	2.609	5.943
	China3	Log-Normal	0.745	0.697	1.555	3.567
	China4	Log-Normal	0.517	0.484	1.100	2.460
	China5	Log-Normal	0.517	0.484	1.100	2.460
HDGPVs	China0	Log-Normal	2.223	2.100	4.741	10.559
	China1	Log-Normal	2.306	2.142	4.859	10.957
	China2	Log-Normal	0.852	0.788	1.805	4.030
	China3	Log-Normal	0.383	0.346	0.791	1.838
	China4	Log-Normal	0.184	0.167	0.380	0.872
	China5	Log-Normal	0.184	0.167	0.380	0.872
HDDPVs	China0	Log-Normal	1.184	1.068	2.424	5.614

		China1	Log-Normal	0.254	0.234	0.532	1.211
		China2	Log-Normal	0.153	0.141	0.323	0.728
		China3	Log-Normal	0.122	0.114	0.260	0.583
		China4	Log-Normal	0.047	0.042	0.098	0.220
		China5	Log-Normal	0.024	0.021	0.050	0.112
	HDAPVs	China0	Log-Normal	1.694	1.550	3.519	8.045
		China1	Log-Normal	1.415	1.299	2.945	6.765
		China2	Log-Normal	1.256	1.146	2.609	5.943
		China3	Log-Normal	0.745	0.697	1.555	3.567
		China4	Log-Normal	0.517	0.484	1.100	2.460
		China5	Log-Normal	0.517	0.484	1.100	2.460
VKT of passenger vehicles in Beijing (Km)	LDGTAs	China0	Log-Normal	78550	38951	113204	330220
	LDDTAs	China0	Log-Normal	78550	38951	113204	330220
	LDATAs	China0	Log-Normal	78550	38951	113204	330220
	LDGPVs	China0	Log-Normal	7841	3973	11362	33524
	LDDPVs	China0	Log-Normal	7841	3973	11362	33524
	LDAPVs	China0	Log-Normal	7841	3973	11362	33524
	MDGBUs	China0	Log-Normal	4991	40260	50093	59910
	MDDBUs	China0	Log-Normal	4991	40260	50093	59910
	MDABUs	China0	Log-Normal	4991	40260	50093	59910
	MDGPVs	China0	Log-Normal	3143	25009	31310	37380
	MDDPVs	China0	Log-Normal	3143	25009	31310	37380
	MDAPVs	China0	Log-Normal	3143	25009	31310	37380
	HDGBUs	China0	Log-Normal	4991	40260	50093	59910
	HDDBUs	China0	Log-Normal	4991	40260	50093	59910
	HDABUs	China0	Log-Normal	4991	40260	50093	59910
	HDGPVs	China0	Log-Normal	11401	92557	114757	136940
	HDDPVs	China0	Log-Normal	11401	92557	114757	136940
	HDAPVs	China0	Log-Normal	11401	92557	114757	136940
VKT of passenger	LDGTAs	China0	Log-Normal	78325	43077	120437	342273
	LDDTAs	China0	Log-Normal	78325	43077	120437	342273

vehicles in other provinces (Km)	LDATAs	China0	Log-Normal	78325	43077	120437	342273
	LDGPVs	China0	Log-Normal	10796	6013	16571	46419
	LDDPVs	China0	Log-Normal	10796	6013	16571	46419
	LDAPVs	China0	Log-Normal	10796	6013	16571	46419
	MDGBUs	China0	Log-Normal	4991	40260	50093	59910
	MDDBUs	China0	Log-Normal	4991	40260	50093	59910
	MDABUs	China0	Log-Normal	4991	40260	50093	59910
	MDGPVs	China0	Log-Normal	3143	25009	31310	37380
	MDDPVs	China0	Log-Normal	3143	25009	31310	37380
	MDAPVs	China0	Log-Normal	3143	25009	31310	37380
	HDGBUs	China0	Log-Normal	4991	40260	50093	59910
	HDDBUs	China0	Log-Normal	4991	40260	50093	59910
	HDABUs	China0	Log-Normal	4991	40260	50093	59910
	HDGPVs	China0	Log-Normal	11401	92557	114757	136940
	HDDPVs	China0	Log-Normal	11401	92557	114757	136940
	HDAPVs	China0	Log-Normal	11401	92557	114757	136940
Emission factors on Urban road (g/Km)	LDGTs	China0	Normal	2.331	2.172	4.895	10.942
		China1	Normal	1.604	1.454	3.301	7.527
		China2	Normal	1.049	0.962	2.195	5.010
		China3	Normal	0.279	0.259	0.583	1.323
		China4	Normal	0.078	0.070	0.160	0.367
		China5	Normal	0.078	0.070	0.160	0.367
	MDGTs	China0	Normal	3.305	3.014	6.799	15.521
		China1	Normal	3.223	2.942	6.688	15.221
		China2	Normal	1.436	1.319	3.012	6.806
		China3	Normal	0.643	0.601	1.361	3.042
		China4	Normal	0.275	0.249	0.561	1.307
		China5	Normal	0.275	0.249	0.561	1.307
	HDGTs	China0	Normal	3.200	2.955	6.680	15.063
		China1	Normal	3.148	2.944	6.678	14.911
		China2	Normal	1.393	1.346	2.989	6.689

		China3	Normal	0.639	0.588	1.333	3.062
		China4	Normal	0.261	0.245	0.550	1.242
		China5	Normal	0.259	0.242	0.549	1.237
	LDGTs	China0	Normal	1.774	1.657	3.728	8.432
		China1	Normal	1.182	1.107	2.497	5.675
		China2	Normal	0.802	0.754	1.699	3.831
		China3	Normal	0.233	0.218	0.481	1.120
		China4	Normal	0.064	0.059	0.134	0.302
		China5	Normal	0.064	0.059	0.134	0.302
Emission factors on Provincial road (g/Km)	MDGTs	China0	Normal	0.559	4.572	5.555	6.775
		China1	Normal	2.399	2.225	5.036	11.250
		China2	Normal	1.053	0.997	2.232	5.023
		China3	Normal	0.486	0.453	1.025	2.268
		China4	Normal	0.200	0.190	0.422	0.962
		China5	Normal	0.200	0.190	0.422	0.962
	HDGTs	China0	Normal	2.402	2.198	4.985	11.389
		China1	Normal	2.359	2.185	5.015	11.257
		China2	Normal	1.099	0.983	2.252	5.223
		China3	Normal	0.470	0.446	0.997	2.254
		China4	Normal	0.045	0.368	0.447	0.543
		China5	Normal	0.202	0.181	0.414	0.947
Emission factors on National road (g/Km)	LDGTs	China0	Normal	1.902	1.789	3.988	9.089
		China1	Normal	1.268	1.181	2.697	6.060
		China2	Normal	0.846	0.775	1.764	3.999
		China3	Normal	0.245	0.219	0.500	1.159
		China4	Normal	0.066	0.062	0.138	0.317
		China5	Normal	0.066	0.062	0.138	0.317
	MDGTs	China0	Normal	2.612	2.461	5.547	12.615
		China1	Normal	2.630	2.403	5.400	12.396
		China2	Normal	1.150	1.070	2.424	5.444
		China3	Normal	0.521	0.482	1.100	2.456

		China4	Normal	0.218	0.201	0.461	1.031
		China5	Normal	0.218	0.201	0.461	1.031
	HDGTs	China0	Normal	2.575	2.427	5.430	12.302
		China1	Normal	2.639	2.391	5.387	12.523
		China2	Normal	1.140	1.072	2.415	5.435
		China3	Normal	0.513	0.484	1.088	2.454
		China4	Normal	0.215	0.199	0.446	1.020
		China5	Normal	0.211	0.198	0.449	0.997
Emission factors on Freeway (g/Km)	LDGTs	China0	Normal	1.801	1.691	3.760	8.437
		China1	Normal	1.219	1.121	2.542	5.812
		China2	Normal	0.808	0.746	1.688	3.817
		China3	Normal	0.237	0.214	0.487	1.117
		China4	Normal	0.065	0.059	0.136	0.309
		China5	Normal	0.065	0.059	0.136	0.309
	MDGTs	China0	Normal	2.525	2.249	5.277	11.911
		China1	Normal	2.418	2.264	5.101	11.534
		China2	Normal	1.086	1.003	2.296	5.215
		China3	Normal	0.487	0.457	1.048	2.313
		China4	Normal	0.207	0.188	0.431	0.993
		China5	Normal	0.207	0.188	0.431	0.993
	HDGTs	China0	Normal	2.410	2.242	5.132	11.470
		China1	Normal	2.430	2.327	5.150	11.495
		China2	Normal	1.089	0.992	2.282	5.249
		China3	Normal	0.485	0.456	1.029	2.318
		China4	Normal	0.203	0.182	0.420	0.968
		China5	Normal	0.198	0.187	0.418	0.941
Emission factors on other type roads (g/Km)	LDGTs	China0	Normal	3.066	2.829	6.411	14.780
		China1	Normal	1.994	1.857	4.246	9.496
		China2	Normal	1.370	1.238	2.848	6.525
		China3	Normal	0.352	0.320	0.734	1.679
		China4	Normal	0.094	0.089	0.204	0.452

		China5	Normal	0.094	0.089	0.204	0.452
	MDGTs	China0	Normal	4.173	3.949	8.844	20.062
		China1	Normal	4.122	3.823	8.777	19.875
		China2	Normal	1.900	1.707	3.882	9.052
		China3	Normal	0.839	0.781	1.759	3.984
		China4	Normal	0.357	0.325	0.738	1.709
		China5	Normal	0.357	0.325	0.738	1.709
	HDGTs	China0	Normal	4.102	3.794	8.706	19.494
		China1	Normal	4.055	3.828	8.734	19.229
		China2	Normal	1.847	1.712	3.882	8.921
		China3	Normal	0.843	0.771	1.744	3.988
		China4	Normal	0.336	0.322	0.715	1.600
		China5	Normal	0.344	0.317	0.720	1.632
Emission factors on urban road (g/Km)	LDDTs	China0	Normal	0.990	0.932	2.093	4.717
		China1	Normal	0.966	0.894	2.008	4.547
		China2	Normal	0.615	0.569	1.300	2.913
		China3	Normal	0.168	0.158	0.351	0.787
		China4	Normal	0.085	0.078	0.178	0.406
		China5	Normal	0.085	0.078	0.178	0.406
	MDDTs	China0	Normal	2.125	1.942	4.434	10.110
		China1	Normal	0.772	0.701	1.596	3.656
		China2	Normal	0.200	0.185	0.414	0.940
		China3	Normal	0.096	0.089	0.199	0.457
		China4	Normal	0.048	0.045	0.103	0.233
		China5	Normal	0.048	0.045	0.103	0.233
	HDDTs	China0	Normal	1.900	1.775	4.023	9.117
		China1	Normal	0.417	0.388	0.887	2.006
		China2	Normal	0.245	0.228	0.516	1.154
		China3	Normal	0.121	0.113	0.252	0.576
		China4	Normal	0.060	0.055	0.127	0.286
		China5	Normal	0.060	0.055	0.127	0.286

Emission factors on provincial road (g/Km)	LDDTs	China0	Normal	0.745	0.668	1.552	3.518
		China1	Normal	0.719	0.667	1.515	3.409
		China2	Normal	0.487	0.444	0.998	2.317
		China3	Normal	0.138	0.130	0.294	0.658
		China4	Normal	0.072	0.065	0.148	0.339
		China5	Normal	0.072	0.065	0.148	0.339
	MDDTs	China0	Normal	1.596	1.477	3.328	7.576
		China1	Normal	0.575	0.521	1.211	2.727
		China2	Normal	0.149	0.135	0.311	0.704
		China3	Normal	0.072	0.067	0.151	0.343
		China4	Normal	0.036	0.034	0.077	0.173
		China5	Normal	0.036	0.034	0.077	0.173
	HDDTs	China0	Normal	1.465	1.322	3.030	6.879
		China1	Normal	0.319	0.290	0.665	1.514
		China2	Normal	0.188	0.170	0.385	0.890
		China3	Normal	0.090	0.083	0.190	0.424
		China4	Normal	0.046	0.042	0.096	0.217
		China5	Normal	0.046	0.042	0.096	0.217
Emission factors on national road (g/Km)	LDDTs	China0	Normal	0.810	0.736	1.698	3.816
		China1	Normal	0.765	0.722	1.644	3.692
		China2	Normal	0.494	0.454	1.034	2.351
		China3	Normal	0.143	0.134	0.300	0.693
		China4	Normal	0.074	0.067	0.154	0.352
		China5	Normal	0.074	0.067	0.154	0.352
	MDDTs	China0	Normal	1.730	1.585	3.640	8.182
		China1	Normal	0.611	0.571	1.299	2.923
		China2	Normal	0.161	0.150	0.337	0.764
		China3	Normal	0.078	0.071	0.162	0.369
		China4	Normal	0.040	0.036	0.083	0.190
		China5	Normal	0.040	0.036	0.083	0.190
	HDDTs	China0	Normal	1.570	1.431	3.270	7.502

		China1	Normal	0.345	0.321	0.722	1.635
		China2	Normal	0.202	0.183	0.419	0.953
		China3	Normal	0.099	0.092	0.205	0.465
		China4	Normal	0.050	0.046	0.104	0.236
		China5	Normal	0.050	0.046	0.104	0.236
Emission factors on freeway (g/Km)	LDDTs	China0	Normal	0.757	0.696	1.593	3.582
		China1	Normal	0.715	0.699	1.555	3.487
		China2	Normal	0.480	0.433	0.983	2.249
		China3	Normal	0.141	0.132	0.297	0.677
		China4	Normal	0.071	0.066	0.148	0.336
		China5	Normal	0.071	0.066	0.148	0.336
	MDDTs	China0	Normal	1.639	1.506	3.402	7.777
		China1	Normal	0.582	0.529	1.221	2.748
		China2	Normal	0.153	0.141	0.322	0.724
		China3	Normal	0.073	0.067	0.154	0.345
		China4	Normal	0.037	0.034	0.078	0.175
		China5	Normal	0.037	0.034	0.078	0.175
	HDDTs	China0	Normal	1.456	1.328	3.075	6.925
		China1	Normal	0.324	0.301	0.682	1.522
		China2	Normal	0.187	0.175	0.394	0.892
		China3	Normal	0.021	0.173	0.210	0.255
		China4	Normal	0.046	0.044	0.097	0.219
		China5	Normal	0.046	0.044	0.097	0.219
Emission factors on other type roads (g/Km)	MDDTs	China0	Normal	1.286	1.182	2.694	6.171
		China1	Normal	1.214	1.176	2.635	5.828
		China2	Normal	0.793	0.732	1.667	3.801
		China3	Normal	0.208	0.192	0.439	0.992
		China4	Normal	0.105	0.100	0.224	0.506
		China5	Normal	0.105	0.100	0.224	0.506
		China0	Normal	2.701	2.537	5.734	12.858
		China1	Normal	0.972	0.928	2.080	4.628

	MDDTs	China2	Normal	0.259	0.238	0.546	1.232
		China3	Normal	0.126	0.115	0.260	0.594
		China4	Normal	0.063	0.057	0.134	0.296
		China5	Normal	0.063	0.057	0.134	0.296
	HDDTs	China0	Normal	2.497	2.292	5.314	11.801
		China1	Normal	0.561	0.517	1.170	2.649
		China2	Normal	0.320	0.299	0.675	1.527
		China3	Normal	0.156	0.149	0.328	0.749
		China4	Normal	0.079	0.072	0.165	0.374
		China5	Normal	0.079	0.072	0.165	0.374
VKT of freight trucks (Km)	LDGTs	China0	Normal	2231	17804	22144	26459
		China1	Normal	2231	17804	22144	26459
		China2	Normal	2613	21212	26282	31510
		China3	Normal	2962	23701	29470	35296
		China4	Normal	3412	27517	34137	40880
		China5	Normal	3412	27517	34137	40880
	LDDTs	China0	Normal	1936	15457	19261	23046
		China1	Normal	1936	15457	19261	23046
		China2	Normal	2698	21680	26977	32181
		China3	Normal	3642	29375	36624	43686
		China4	Normal	4564	36264	45280	54401
		China5	Normal	4564	36264	45280	54401
	MDGTs	China0	Normal	3523	28361	35216	42199
		China1	Normal	3523	28361	35216	42199
		China2	Normal	4003	32747	40789	48541
		China3	Normal	4801	38505	47870	57236
		China4	Normal	5273	43193	53491	63709
		China5	Normal	5273	43193	53491	63709
	MDDTs	China0	Normal	2126	17035	21228	25407
		China1	Normal	2126	17035	21228	25407
		China2	Normal	2833	22651	28170	33692

		China3	Normal	3616	29244	36345	43352
		China4	Normal	5998	48661	60346	72179
		China5	Normal	5998	48661	60346	72179
	HDGTs	China0	Normal	2747	22236	27753	33098
		China1	Normal	2747	22236	27753	33098
		China2	Normal	3343	26601	33215	39661
		China3	Normal	4031	32340	40265	48241
		China4	Normal	4524	36913	45806	54752
		China5	Normal	4524	36913	45806	54752
	HDDTs	China0	Normal	2430	19566	24330	29119
		China1	Normal	2430	19566	24330	29119
		China2	Normal	3814	31028	38551	46017
		China3	Normal	6316	51858	64083	76300
		China4	Normal	9863	78952	98396	117258
		China5	Normal	9863	78952	98396	117258
Percentage of driving distance on different type roads	MDG(D)Ts	Urban road	Normal	0.010	0.219	0.239	0.258
		Provincial road	Normal	0.020	0.303	0.342	0.381
		National road	Normal	0.007	0.124	0.137	0.151
		Freeway	Normal	0.014	0.247	0.274	0.301
		others	Normal	0.000	0.007	0.008	0.009
	LDG(D)Ts	Urban road	Normal	0.020	0.314	0.353	0.392
		Provincial road	Normal	0.009	0.167	0.185	0.203
		National road	Normal	0.011	0.196	0.218	0.239
		Freeway	Normal	0.010	0.195	0.215	0.234
		others	Normal	0.001	0.028	0.031	0.034

HDG(D)Ts	Urban road	Normal	0.007	0.128	0.142	0.155
	Provincial road	Normal	0.010	0.173	0.192	0.212
	National road	Normal	0.010	0.226	0.246	0.265
	Freeway	Normal	0.020	0.352	0.391	0.429
	others	Normal	0.002	0.027	0.030	0.033

Table S9. Uncertainty range of emission inventories.

		Unit	Mean	Standard division	C.V	The 95% confidence interval		
						2.5% percentile	50 % percentile	97.5% percentile
Tailpipe emissions	Passenger vehicles tailpipe emissions	Gg	1279.12	252.51	0.20	902.39	1237.21	1891.96
	Trucks tailpipe emissions	Gg	720.89	45.20	0.06	636.52	718.39	816.43
	Motorcycles tailpipe emissions	Gg	562.54	349.17	0.62	158.61	476.40	1444.66
Evaporative emissions	Diurnal emissions (excluding motorcycles)	Gg	138.99	75.27	0.54	56.22	124.26	312.78
	Hot Soak emissions (excluding motorcycles)	Gg	15.75	3.71	0.24	9.70	15.33	24.26
	Refueling emissions	Gg	109.38	7.46	0.07	95.82	108.94	124.92
	Running loss	Gg	1146.18	768.92	0.67	229.90	963.11	3132.67
	Motorcycles evaporation	Gg	251.30	278.70	1.11	29.31	170.14	954.21
Ratio of evaporative emissions versus tailpipe emissions of passenger cars			1.14	0.67	0.5828	0.36	0.98	2.89
Total emissions		Gg	4224.14	943.21	0.22	2897.14	4053.82	6540.95

Technical Corrections:

Section 3.2.2-3.2.4 are too short to be an individual section. I personally think that this part of discussion is not necessarily to be divided into three sections.

Response: Accepted. The original Sect. 3.2.1-3.2.4 were combined to Sect. 3.2. No sub-section was divided.

Supporting Information, Table S4: Some abbreviations of vehicle types (LDGTAs, LDDTAs) ought to be specified.

Response: Corrected. We have modified Sect. 2.1. Now the vehicle classification and abbreviations are consistent through the whole manuscript.

Some in-text citations are missing in the reference list, e.g., MOVES, 2010; ICCT, 2012.

Response: Corrected.

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 ~~the imperfection of~~ 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~~, with a target~~ for the year of 2015, ~~were~~are developed based on a set of state-of-the-art methods and a mass of local measurement data. A~~The a~~ctivity data for light-duty vehicles ~~were~~are derived from trajectories of more than 70 thousand cars for~~for~~ one year. The annual mileages of trucks are~~were~~ calculated from reported data by more than 2 million trucks in China. The emission profiles ~~were~~ are updated using measurement data. ~~Not only vehicular~~ Vehicular tailpipe emissions (VTEs) and four types of vehicular evaporation emissions (VEEs~~)~~)~~but also four kinds of vehicular evaporation emissions (VEEs)~~, including refuelling, hot soak, diurnal and running loss, ~~were~~ are taken into account. ~~The r~~Results show~~ed~~ that the total vehicular VOCs emissions in China ~~were~~ are 4.21 Tg (with a 95% confidence interval ranges from 2.90-6.54 Tg) and

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the IVOCs emissions ~~were~~ are ~~121. 23~~200.37 Gg in ~~the year of~~ 2015. VTEs ~~were~~ are still the predominant contributor, while~~but~~ VEEs ~~were~~ are ~~already~~ responsible for 39.20% of VOCs emission. ~~Since~~The control of~~on~~ VEES is yet to be optimized in China. ~~has a much less strict control standard, it should be paid much more attention to still needs to be enhanced in China.~~ Among VTEs, passenger vehicles ~~contributed most~~emissions have the largest share (49.86%), followed by trucks (28.15%) and motorcycles (21.99%). Among VEEs, running loss ~~was~~ is the largest contributor (81.05%). For both VTEs and VEEs, Guangdong, Shandong and Jiangsu province ~~took~~ take ~~the first three spots~~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. ~~Totally,~~ 97 VOC species ~~were~~ are ~~analysed~~analyzed in this VOCs emission inventory. I-pentane, toluene and formaldehyde ~~were~~ are found to be the most abundant species in China's vehicular VOCs emissions. The estimated IVOCs is another 'inconvenient truth', ~~providing insights~~ concluding that ~~the~~ precursors emission for~~of~~ secondary organic aerosol (SOA) from vehicles ~~were~~ are much larger than ~~more than~~ ~~the~~ previous estimation.

1 Introduction

China is one of those countries ~~that are facing severe threats of~~ most threatened by simultaneous pollution of $PM_{2.5}$ (particulate matter with aerodynamic diameters of less than 2.5μm) and ozone. ~~pollution. at the same time~~ (***Sitch et al., 2007; van Donkelaar et al., 2015; Liu et al., 2016***). ~~In the year of 2010, nearly~~ ~~a~~Approximately 1.36 million premature deaths in China were attributed to these two major ~~types of chief~~ pollutants in 2010~~.(~~. (***Lelieveld et al., 2015~~; Liu et al., 2015~~***). Previous studies shown that secondary organic aerosols (SOA) is of significant proportion in ambient $PM_{2.5}$ mass of Chinese cities ~~Studies on $PM_{2.5}$ pollution indicated that secondary organic aerosols (SOA) accounted for a significant proportion of ambient $PM_{2.5}$ mass in Chinese cities~~ (***Cui et al., 2015***). ~~i~~Intermediate-volatility organic compounds ~~(~~ (IVOCs) is a series of compounds with effective saturate concentration between 10^3-10^6μg/m^3, ~~corresponding~~similar to the volatility range of C12-C22 n-alkanes (***Zhao et al., 2014***). Recent studies suggested that both volatile organic compounds (VOCs) and intermediate-volatility organic compounds (IVOCs) contributed to SOA formation , with =IVOC being dominant in certain regions (***Huang et al., 2014; Robinson et al., 2007; Hodzic et al., 2010***).IVOCs is a series of compounds with effective saturate concentration between 10^3-10^6μg/m^3, corresponding to the volatility range of C12-C22 n-alkanes (***Zhao et al., 2014***). In some regions, IVOC could be dominant (***Huang et al., 2014; Robinson et al., 2007; Hodzic et al., 2010***). Studies on ozone pollution also demonstrated that ozone formation was caused controlled by VOCs in many major Chinese cities (***Tie, et al., 2006; Geng et al., 2008; Shao et al., 2009***). VOCs and IVOCs should be widely attended for their impacts on air quality and public health. Undoubtedly, to achieve better air quality and to reduce the health hazards resulted from air pollution in China, VOCs and IVOCs should be paid great attention to.

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 foron pollution control at the countryon national level. Cai and Xie (***2009***) et al reported VOCs emission inventory from on-road vehicles in China during 1980-2005 (***Cai et al., 2009***). 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; 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 impacts. ~~We noticed that all the studies above provide~~d ~~emission inventory before 2010 and none of the previous studies took IVOCs into consideration.~~ Considering the dramatic~~al~~ increase ~~trend~~ of vehicle population, ~~it is extremely urgent to~~ establishment of new emission inventories is of urgent priority. However, ~~there were~~ multiple key factors changed in the last ten years that~~which~~ require updated~~new~~ methods and data.

First of all, dominant VOCs emission processes of vehicles may switch. Compared with vehicular tailpipe emissions (VTEs), vehicular evaporation emissions (VEEs) ~~has~~ have recently been reported ~~proved~~ to be nonnegligible~~a non-ignorable~~ contributors to ~~the~~ ambient VOCs concentrations ~~recently~~ (***Yamada et al., 2013; Liu et al., 2015***). VOCs emissions ~~evaporate~~ evaporate from gasoline fuelled vehicles consistently ~~continually whether they were~~ regardless of their status of refuelling, running or parked. ~~The v~~Vapors generated in VEEs either pass ~~go through~~ through the equipped carbon canister or permeate elastomers of the vehicle's fuel system before entering ~~and eventually into the~~ ambient atmosphere.~~. Besides, they could also permeate through elastomers of the vehicle's fuel system to enter the atmosphere at the same time. According to the state vehicles are in ,~~Depending on the vehicle's status, evaporative emissions come in four varieties: refuelling loss, running loss, hot soak loss and diurnal loss. ~~To include VEEs in the inventory, l~~Local emission factors and profiles of VEEs are necessary for VEEs inventory as ~~because~~ they are highly related ~~with~~ to local gasoline formula and vehicle controls. In addition, a more sophisticated method is necessary to estimate VEEs. ~~The d~~Details ~~were~~ are further described in ~~method~~ ~~S~~Sect~~ion~~. 2.

Secondly, as dominant precursors of ~~a great contributors to the formation of~~ SOA, IVOCs have strong impacts on ~~atmospheric condition~~air quality, global climate and human health. However, there are few studies on IVOCs ~~because of the complicated composition of IVOCs.~~due to their complex composition. ~~For these category components with long chains, s~~Short of systematic and integrated analytical~~sis~~ methods limit~~s~~ the progress for~~of~~ measurement~~ing~~ and quantification of~~ying~~ IVOCs (***Goldstein et al., 2007; Jathar et al., 2014***). Therefore,~~ only~~ few studies successfully provided emission measurement~~s~~ results of IVOCs~~,~~. To our knowledge, ~~there is nonone of the~~ IVOCs emission inventory ~~has been reported~~ for China is yet to be reported.

Most importantly, vehicle activity is crucial in total emission estimation and the big data ~~on vehicle activity~~would ~~may~~ greatly reduce ~~the~~ uncertainty of the emission inventory~~. Vehicle activity is critical to total emission estimation.~~ In previous studies, these parameters were usually ~~from~~ hypothesized~~s~~ based on experiences ~~from~~ 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 ~~achieve~~ Global Positioning System (GPS) records of 71,059 cars for research purpose without any personal information. This data covered 30 provinces in China, which ~~could highly~~would significantly improve our understanding on vehicle usage, and better our estimation with accuracy and comprehensiveness.~~and to perform better in the aspects of comprehensiveness and accuracy.~~

In addition, several other new methods and local data ~~could be~~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 for freight trucks.~~Previous studies usually calculated the provincial emissions using the local registration number, which~~

was based on an assumption that all vehicles were running within the province or city where they registered. However, when it comes to freight trucks, this assumption is unwarranted. 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 NOx and PM emission estimation for long-distance inter-province or inter-city cargo transportation (***Yang et al., 2015a***).Instead of the traditional local registration based approach, we have developed a more reasonable road emission intensity based (REIB) approach. Instead of relying on truck population from local registration database, the spatial distribution of emission inventory iss were based on the total length of each road type in this province and also the emission intensity for this road type. Using this approach, NOx and PM emissions were for greatly improved for long-distance inter-province or inter-city cargo transportation (***Yang et al., 2015a***).

The deficienciesImperfections 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 the 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 provided more reality tooffer a relevant and more accurate estimation of local emission levels. Using cAdditionally, Chemical profiles obtained by experiments in western countries could not reflect the chemical characteristics of VOCs from vehicles in China accurately too. The recently speciation profiles were reported using China's a local fuel.

Furthermore, tThe national statistical data in China only provideprovidess the vehicle population data classified by vehicle types (e.g., light-duty passenger vehicles, heavy-duty trucks). However, mMore -detailed vehicle population data byclassified by fuel types and by control technologies technology awere required to calculate emissions as they were reported to have distinct influences on because these two parametersey have been acknowledged reported to influence emission factors. distinctly (***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, with a target year of in 2015, were are developed using a set of state-of-the-art methods. The five deficiencieslack of in comprehensiveness and accuracy in existing methods mentioned above were are solved one by oneeach and individually based on scientific calculating methodologies, big data and abundant local emission measurements. The IVOCs emission factors used were are derived from US studies in the US by matching corresponding vehicle emission categories of the two countries, as no local IVOCs emission factors were reported.between China and US, because currently there were is no local IVOCs emission factors reported.

2 Methodology and data

2.1 Vehicle stock and classification

In total, 22 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 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 ~~the~~ vehicles with~~whose~~ length ~~is~~ less~~less~~ than 6000mm and ridership~~ is less than or equal to~~no more than 9. M~~A~~ ~~m~~edium-duty~~ vehicle~~ refers to vehicles of~~the~~ length less than 6000mm and ridership between~~among~~ 10-19. H~~A h~~eavy-duty refers to vehicles ~~refers to~~ of~~the~~ length ~~more than or equal to~~no less than 6000mm or ~~the~~ ridership is no less than~~equal to or more than~~ 20. These vehicles were further classified by control technologies (i.e., China 0, China 1, China 2, China 3, China 4 and above). A~~The a~~lternative-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), ~~heavy~~and heavy-duty diesel trucks (HDDTs)~~ and gasoline motorcycles (GMs)~~. For trucks, a light-duty truck refers vehicles of~~to the length less than 6000mm and~~ mass less than ~~4~~3500kg. A medium-duty truck refers to vehicles of~~ the length more than or equal to 6000mm or~~ with mass ranging from~~ from~~ ~~4~~3500kg to 12000kg. A heavy-duty truck refers vehicles of ~~to the truck whose~~ mass~~ is~~ more than 12000kg. ~~For passenger vehicles, light-duty~~

refers to the vehicle whose length is less than 6000mm and ridership is less than or equal to 9. A medium-duty vehicle refers to the length less than 6000mm and ridership among 10-19. A heavy-duty vehicle refers to the length more than or equal to 6000mm or the ridership is equal to or more than 20. For trucks, a light-duty truck refers to the length less than 6000mm and mass less than 4500kg. A medium-duty truck refers to the length more than or equal to 6000mm or mass from 4500kg to 12000kg. A heavy-duty truck refers to the truck whose mass is more than 12000kg. These vehicles were further classified by control technologies (i.e., China 0, China 1, China 2, China 3, China 4 and above).

DTo be specific, the detailed provincial population data of all types of vehicles excluding GMs in 2015 was 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 accurateregarded as accurate as possible. The provincial GMs population in 2015 was obtained from the Provincial China Automotive IndustryStatistic Yearbook 2016 offor each province 2016.

2.2 Vehicle activity

The real-world vehicle activity data used in this study was derived by statistical surveys, field tests and literature review. To be specific, the detailed provincial population data of all types of vehicles excluding GMs in 2015 was obtained by complete statistical survey conducted by the Vehicle Emission Control Center (VECC) of China's Ministry of Environmental Protection (MEP), which could be regarded as accurate as possible. The provincial GMs population in 2015 was obtained from China Automotive Industry Yearbook 2016.

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 ~~analysing~~analyzing the big data of GPS records (71059 cars). D~~The d~~riving frequency of different types of trucks ~~running~~ on different kinds of roads (~~i.g.~~e.g., freeway, national road, provincial road and urban road) was acquired by ~~analysing~~analysis of ~~the~~ survey data from~~of~~ 1060 valid questionnaires, which ~~has been~~was introduced in detail in our previous study (***Yang et al., 2015a***). ~~The annual VKT data for trucks were calculated based on report data from 2 million trucks in China.~~

In addition, ~~for evaporative emission calculation, the~~ provincial parking characteristics data for evaporative emission calculation,~~,~~ including parking events numbers and parking durations, were also obtained by ~~processing and analysing~~ ~~analyzing~~ 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, ~~. This is a big data platform with more than 2 million trucks online. Those trucks,~~ mainly commercial vehicles~~,~~ installed with either the GPS or China Bei Dou System(BDS). Location, speed and vehicle type information are live fed to the commercial platform.~~Both the location, speed and vehicle type information are sending to this commercial platform.~~ The VKT for each truck category was calculated using the monitored~~ing~~ data from the~~is~~ platform.

2.3 Vehicular emission data and estimation

The vehicular VOCs emissions at the provincial level were divided into three parts for calculation~~to be calculated~~, 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 ~~Then~~ the total provincial emission amounts in ~~the year of~~ 2015

~~were obtained by summing these three parts of emissions up~~. E~~The e~~mission factors for VOCs used~~ here~~ were derived from~~by~~ lab tests, field tests and literature review (MEP, 2015; Zhang et al., 2014; Liu et al., 2015). The IVOCs emission calculation was ~~very~~ similar to that of the~~with~~ VOCs, while only the tailpipe exhaust for non-GMs wa~~i~~s taken into consideration. For IVOCs emission factors, Zhao et al. (***2015, 2016***) reported a series of measurements for gasoline and diesel vehicles ~~(***Zhao et al.; 2016; Zhao et al.; 2015***)~~ (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 by Eq. (1):

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

where $E_{tailpipe,non-truck,i}$ represents the annual tailpipe emissions from non-truck vehicles in province *i* (g·year^{-1}); $EF_{tailpipe,j,k}$ represents the tailpipe VOCs/IVOCs emission factor of vehicle type *j* with control technology *k* (g·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* (km·year^{-1}).

For VTEs of~~from~~ all ~~types of~~ 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).

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For IVOCs emission factors, Zhao et al. reported a series of measurements for gasoline and diesel vehicles (***Zhao et al.; 2015, 2016~~; Zhao et al.; 2015~~***). By considering age of a specific vehicle model~~ year~~, after-treatment devices and emission certification standard, each of the tested vehicles was matched to a corresponding~~a~~ category of China emission certification standard (Table S2). Thus, the emission factors for some vehicle categories were set up. For th~~ose~~e categories lacking~~without~~ measurements (gasoline vehicles before China 1 and all diesel vehicles), emission factors were set identical to~~as the same with~~ China 1 category. For diesel passenger vehicles, the current IVOC emission factors were set identical to~~as the same with~~ the corresponding~~same~~ level of gasoline vehicles. The IVOC emission factors were converted from the original unit of mg/kg-fuel to ~~m~~g/km~~,~~ using fuel economy. For each category, median of emission factors were used for the particular type of vehicles if more than one available tests are present~~if there were more than one test available, the median of these emission factors would be used as the emission factor of this type of vehicle~~. D~~ The d~~etailed emission factors were listed in Table S5.

2.3.2 Tailpipe emissions from freight trucks

Considering the fact that the~~a~~ 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 (***Yang et al., 2015a***). The provincial tailpipe emissions from freight trucks were estimated by Eq. (2):

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

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where $E_{tailpipe,truck,i}$ represents the annual tailpipe VOCs/IVOCs emissions from freight trucks in province i (g·year^{-1}); $EF_{tailpipe,j,k}$ represents the tailpipe VOCs/IVOCs emission factor of vehicle type j with control technology k (g·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·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~~from~~ trucks, ~~we used~~ the operating-mode-bin-based method introduced in our previous study were used to investigate ~~the~~ real-world emission factors for VOCs (***Yang et al., 2015a***). Firstly, s~~First, the s~~econd-by-second vehicle-specific power (VSP) and engine stress (ES) data were calculated using ~~the~~ GPS records of 16 trucks ~~and~~with ~~the~~ equations suggested by the MOVES ~~model~~ and IVE model respectively~~, respectively~~. Followed by identification of ~~Then t~~thirty operating mode bins ~~were identified~~ based on ~~the~~ VSP data and~~and~~ ES data, ~~and~~ the time fraction of each bin was given. Finally, the distance-based emission factors for ~~different types of~~ trucks of various types on various roads were ~~running on different kinds of roads were~~ calculated according to the emission rate of each bin that were~~, which was~~ presented in~~based on~~ our previous test results (***Liu et al***., ~~***EST,***~~ ***2009***).

For IVOCs emission factors, a mapping to match US emission certification level to China emission level was built (Table S3). Only ~~the~~ non-GM ~~s~~vehicles were considered for ~~the~~ IVOC emissions evaluation. The emission factors for US fleet were converted ~~to emission factors for~~for use of ~~China trucks~~Chinas' trucks. As ~~There was~~ no data were available for most categories, assumptions had to be made~~. We have to make assumptions~~ to fill the gap: (1~~) Mini and light-duty trucks for the same control level~~

~~shared the same emission factors. (2~~) Medium and heavy-duty trucks had ~~same~~ identical emission factors ~~which was~~that are 50% higher than light-duty trucks,~~. This ratio was~~ similar to emission ratios of~~the~~ VOCs and~~or~~ primary organic aerosol (POA)~~POA emission ratios of heavy/light for trucks~~of the same types. (~~3~~2) Emission levels of neighboring types were used in the case of lacking data.~~For those emission levels without data, the emission factors for neighbour~~neighbor ~~emission level would be used.~~ The final assumption 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 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.

$$E_{diur,non-GM,i} = E_{diur,<24,non-GM,i} + E_{diur,24-48,non-GM,i} + E_{diur,>48,non-GM,i},$$

(3)

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

(4)

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

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

where $E_{diur,non-GM,i}$ represents the total annual diurnal (simultaneous permeation included) emissions from non-GM gasoline vehicles registered in province *i* (g·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 (g·year^{-1}); $EF_{diur,<24,LDGPVs}$, $EF_{diur,24-48,LDGPVs}$, $EF_{diur,48-72,LDGPVs}$ represents the measured diurnal (simultaneous permeation included) emission factors of China 4 LDGVs (g·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 ~~diurnal (simultaneous permeation included)~~evaporative emissions from GMs registered in province *i* (g·year^{-1}); EF_{GMs} represents the evaporative ~~diurnal (simultaneous permeation included)~~ emission factor of GMs (g·~~hour~~km^{-1}~~).~~); For VEEs from GMs, the emission factors given by the International Council on Clean Transportation (ICCT) were utilized (***ICCT, 20~~12~~07***) (Table S6). $VKT_{i,GMs}$ represents the annual VKT of GMs in province *i* (km·year^{-1}).

According to the US EPA, hot soak is defined as the evaporative losses ~~that occur~~ within a~~the~~ one-hour period after shutting down of engines ~~the engine is shut down~~ (***~~EPA420-R-01-026~~EPA, ~~)~~2001)***. Any vapor losses occurred after are considered ~~If the parking duration is longer than one hour, then the extra vapor losses fall into~~ diurnal emissions. The provincial hot soak emissions for non-GM gasoline vehicles (i.e., LDGPVs, MDGPVs, HDGPVs, LDGTAs ~~GTs~~, GBUs, LDGTs, MDGTs, HDGTs) ~~and GMs~~ were calculated by Eq. (8) ~~and Eq. (9) respectively~~:

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

where $E_{soak,non-GM,i}$ represents the annual hot soak (simultaneous permeation included) emissions from non-GM vehicles in province i (g·year^{-1}); $EF_{soak,LDGPVs}$ represents the hot soak (simultaneous permeation included) emission factor of LDPGVs (g·hour^{-1}); T_i represents the ~~annual~~ average parking duration per day per vehicle of province i (hour); N_i represents the ~~annual~~ 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.

~~$E_{soak,GMs,i} = EF_{soak,GMs} \times VP_{i,GMs} \times VKT_{i,GMs}$, (9)~~

~~where $E_{soak,GMs,i}$ represents the annual hot soak (simultaneous permeation included) emissions from GMs registered in province i (g·year^{-1}); $EF_{soak,GMs}$ represents the hot soak (simultaneous permeation included) emission factor of GMs (g·hour^{-1}); For VEEs from GMs, the emission factors given by the International Council on Clean Transportation (ICCT) were utilized (***ICCT, 2012***). $VP_{i,GMs}$ represents the GMs~~

~~population registered in province *i*; $VKT_{i,GMs}$ represents the annual VKT of GMs in province *i* (km·year^{-1}).~~

2.3.4 Evaporation emissions from gasoline vehicles- Refuelling

China follows~~is following~~ European countries~~an control experiences~~ in~~to~~ popularization of~~e~~ Stage-II vapor control system for reduction of refueling loss in ~~in refuelling~~ stations~~to reduce refuelling loss~~. 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. (~~10~~9). The control efficiency and the percentages of gasoline stations equipped with Stage-II systems are the two key factors influencing the final emissions.

$$E_{refuel,i} = EF_{refuel} \times [(1-\theta) \times w_i + (1 - w_i)] \times CF_i \,, \qquad (~~10~~9)$$

where $E_{refuel,i}$ represents the annual refuelling emissions from gasoline vehicles in province *i* (g·year^{-1}); EF_{refuel} represents the refuelling emission factor for non-control condition (g·L^{-1}); θ represents the average efficiency of the Stage-II vapor control system, which is 82%~~in this study~~ according to our measurements in Beijing; ω_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·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.

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2.3.5 Evaporation emissions from gasoline vehicles- Running loss

V~~The v~~ehicle running loss occur during operation of the engine~~happens when the engine is on~~ through the fuel system, not from tailpipe. ~~. However, this emission is not from tailpipe, but from the fuel system.~~ The provincial annual running loss emissions from non-GM gasoline vehicles~~ and GMs~~ were calculated by Eq. (~~11~~10)~~ and Eq. (12) respectively~~:

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

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(~~11~~10)

where $E_{running,non-GM,i}$ represents the annual running loss emissions from non-GM gasoline vehicles registered in province i (g·year^{-1}); $EF_{running,LDGPVs}$ represents the running loss emission factor of LDGVs (g·~~L~~hour^{-1}). The emission factors of running loss were acquired from MOVES model due to the lack of local lab test results (~~***MOVES, 2010***~~***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.

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

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~~(12)~~

~~where $E_{running,GMs,i}$ represents the annual running loss (simultaneous permeation included) emissions from GMs registered in province i (g·year^{-1}); $EF_{running,GMs}$ represents the running loss (simultaneous permeation included) emission factor of GMs (g·hour^{-1}). For VEEs from GMs, the emission factors given by the International Council on Clean Transportation (ICCT) were utilized (***ICCT, 2012***).~~

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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. (~~13~~11):

$$E_{speciated} = E_{tailpipe,gasoline} \times PR_{tailpipe,gasoline} + E_{tailpipe,diesel} \times PR_{tailpipe,diesel} + E_{evap} \times PR_{evap},$$

(~~13~~11)

where $E_{speciated}$ represents the speciated annual VOCs emissions from on-road vehicles registered in province i (g·year^{-1}); $E_{tailpipe,gasoline}$, $E_{tailpipe,diesel}$, and E_{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; $PR_{tailpipe,gasoline}$, $PR_{tailpipe,diesel}$, and PR_{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~~ to generate the speciated~~ for establishment of vehicular VOCs emission inventory were derived from literature review and lab tests. T~~ For t~~ailpipe VOCs emissions from gasoline vehicles and diesel ~~vehicles,~~vehicles were yielded from ~~the ~~corresponding local profiles~~ were~~ 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~~2016***).

3 Results and discussion

3.1 Activity characteristics of vehicles

3.1.1 Vehicle population

~~In the year of 2015, the total population of the 22 types of on-road vehicles in China was 259 million, to which ~~GMs and non-GM vehicles contributed 34.3% (88,759,010) and 65.7% (169,794,718)~~,~~ ~~respectively ~~respectively among the 259 million total on-road vehicles in China in the year 2015 (Figure 1). ~~Among the non-GM vehicles,~~ 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%) 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%), China 2 (7.2%), China 5 vehicles (5.2%) and China 0 (4.2%). ~~This fleet structure varies shifts when adding new vehicles add into to the fleet. For~~

China 1, ~~example, in 2012, the~~ China 2 and China 4 shared~~still occupies~~ 9.5%, 15.69% ~~in total, while China 4 only has~~ 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. ~~Similarly, percentage for China 1 was reduced from 14.92% in 2012 to 9.5% in 2015.~~ In terms of automotive fuels, gasoline was the most common~~widely used~~ fuel for~~of~~ non-GM vehicles in China, with a proportion of 86.0%~~.~~ ~~Comparatively~~;~~,~~ while diesel and alternative fuels were substantially lower~~consumed much less comparatively~~, with ~~a respective~~ proportions of 12.9% and 1.2% respectively. Table 1 ~~lists~~ summarized the vehicle population and corresponding~~the~~ proportions~~ercentage~~ classified by fuel types. LDPVs, MDPVs and TAs were mainly fuelled by gasoline while HDPVs, LDTs, MDTs, HDTs and ~~BUs were~~BUs were primarily fuelled by diesel. Proportion of diesel consuming vehicles increases with vehicle weight in both passenger vehicles and freight trucks.~~For both passenger vehicles and freight trucks, the heavier the vehicles were, the larger the percentage of vehicles using diesel as fuels became.~~ 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, contributed by heavy promotion policies of central and local governments.~~still not a mainstream option, especially in the freight truck fleet. However, the percentages of BUs and TAs using alternative fuel both exceeded 8.7%, which were mainly resulted from the strong promotion of policies made by the central and local governments.~~

3.1.2 VKT characteristics ~~of LDPVs~~

~~In total,~~ GPS records of 71,059 ~~cars running~~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 analyz~~s~~ed to obtain ~~the~~ 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. ~~Table 2 gives the p~~Provincial annual average VKT values with vehicle sample sizes are shown in Table 2.~~,~~ ~~while Figure 2 shows the d~~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 ~~were~~ both are among the ~~highest~~ most developed cities in the world, were much lower than the national average value given by this study.~~.~~ ~~And on~~~~With regard to the time scale, they were~~~~Average VKT were~~Average VKT was also much lower than the corresponding local values given by surveys conducted ~~ten years ago~~ in 2004 (***Liu et al., 2007***). This phenomenon could be explained by ~~was mainly caused by~~ three facts~~reasons~~. Firstly, ~~the~~ 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 ~~which around the same time and near the~~ corresponding local values ~~ten years ago~~ in 2004. A considerable~~ertain~~ amount of families in ~~these two~~both cities own ~~more than one car~~ multiple vehicles nowadays, causing~~leading to the~~ decrease of annual VKT of individual~~each~~ cars under the circumstances that their regular commuting distances have not substantially changed ~~that much~~. Secondly, heavy traffic control policies were enforced in Beijing and Shanghai during recent years, ~~the most stringent traffic control policies ever have been implemented in Beijing and Shanghai in during the past several years,~~ resulting in longer parking duration ~~and~~ 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~~a number of citizens in Beijing and Shanghai have chosen to increase the percentage of traveling by public transportation facing the~~ growing traffic jams in peak hours.

Table 3 summarized vehicle mileages of~~for~~ other vehicle types in China. VKT of~~for~~ trucks are~~is~~ significantly influence~~ds~~ by vehicle age.~~ For China 0 or China 1 trucks, the~~ The annual mileage of China 0 and China 1 trucks ~~a~~were~~is~~ much lower~~ess~~ 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.~~the same type trucks but with better control technologies. On one hand, the overloading of trucks is still very common in China. The deterioration of overall performance of a truck is very serious during its lifetime. On the other hand, several cities have implemented low emission zones to ban the entree of low control technology trucks.~~

3.1.3~~2~~ Vehicle parking characteristics

~~Totally, ~~GPS records including 10~~2,576,888~~3 million continuous parking events and 11~~,465,694,907,444~~ trillion valid seconds~~,~~ were collected and analyzed to ~~get the~~obtain real-world vehicle parking characteristics in China, which have ~~have ~~significant impacts on ~~on~~ VEEs. It was found that~~ the~~ parking characteristics in all~~ the~~ provinces excluding~~ept for~~ Beijing were quite similar. T~~For vehicles in Beijing, t~~he annual average number of parking events per day per vehicle was 3.89 in Beijing and 5.73 in other provinces, yet t~~while t~~he annual average parking duration per day per vehicle was similar at 22.21 h and 22.11 h, respectively. ~~For vehicles in the other provinces, however, these two values were 5.73 and 22.11 h, respectively.~~

Figure 3(a) and Figure 3(b) shows the distribution of parking events and total parking duration of~~in~~ six time intervals~~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~~, respectively~~. Parking events of ~~falling into~~ the first two time intervals (<24 h) is of 95.5-98.8% in number, yet ~~though having~~

percentages of 95.5-98.8% in number, only accounted for 51.3-76.8% of the total parking duration, indicatingshowing 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 next phase of emission control , China 6 emission standard, will enhance the evaporative emission control by adding 48 h duration into consideration. Overall, Beijing was found to have fewer parking events but higher proportionpercentages ofin parking events with duration longer than 1 h, thus resulting in longer total parking duration. This phenomenon was mainly caused by the consistent traffic control measures implemented in Beijing dating from the 2008 Beijing Olympic Games, where the days in which the number of vehicles can be drivenallowed within the Fifth Ring Road waiis strictly limited.

3.2 Emissions and implications to policy

3.2.1 Vehicle VOCs emissions

In the year of 2015, China's on-road vehicles emitted 4.21 Tg VOCs in totally (Table 34). Figure 4a-f shows the provincial results of vehicular VOCs emissions in 2015. VTEs, with a proportion of 60.80%, were still the predominant contributor ofto the total VOCs emission with a proportion of 60.80% amount (Figure 4a), with a proportion of 60.80%. However, VEEs, which were responsible forwith a contributive share of the other 39.20% in totalof emissions remained, is evidently nonnegligible and should be also be paid great attention totaken into consideration forof future management. AmongFor the provinces that ownprovinces with large fleets of light-duty vehicles, eg.e.g. Guangdong, Shandong and Jiangsu were ranked as the top three on thetopped the league table, with a respective contribution of 10.66%, 8.85% and 6.54% to the total amounts of VOCs emission from vehicles. A slight difference was observedThere were was a slightly difference between the ranking of tailpipe exhaust and evaporation (Figure 4b and 4c), which was because caused byof the difference disparity inon vehicle fleets, vehicle parking behaviourbehavior and ambient temperature. Figure 4b provided insights for evaporative emissions offor gasoline vehicles excludingept motorcycles, although but the evaporation from motorcycles were included in Figure 4a. RefuellingRefueling was of considerable amountimportant (7.83%), but it could be controlled immediately but could be effectively controlled by stage-II systems in service stations. Thus, the major main challenge for in the future is will come from running loss (81.05%) and diurnal (10.00%) are the main challenges in future emission control. The new China 6 vehicle emission standard will help to controllingalleviate diurnal emissions, which will target at one of the major evaporative sources. However,yet running loss issues would still presentis still a big issue. EThe current estimation showed shows that it could be the most important part for in current vehicle evaporation for now and especially even more so important in for the future. The calculation of running loss is expected with large uncertainty due to the lack of specific measurement data in China.has the large uncertainty Ssince we don’t have any measurements data in China for running loss, uncertainties for this calculation is the largestit.

Figure 4c-4f provided sub-classification for tailpipe exhausts only. When controlling tailpipe VOCs, passengerPassenger vehicles, trucks and motorcycles should all be considered for tailpipe VOCs control, in which their. Their contributions are 49.86%, 28.15% and 21.99% separatelyrespectively. Tailpipe VOCs of passenger cars were widely recognized and attended, in contrast of We have paid a lot of attention to passenger cars, but maybe too less attention to motorcycles and trucks that were commonly overlooked. TFor public transportation, taxis and buses contributed 12.22%

and 2.04% of total tailpipe emissions~~,~~ in public transport. W~~w~~hile~~ the~~ populations~~ for these categories~~ of motorcycles were~~are~~ merely~~only~~ 2.3% and 0.5%, emissions are considerable ~~. Motorcycles were still important sources ~~in Guangdong, Shandong, Yunnan, Hunan, Guangxi and Fujian, ~~in ~~where~~ the~~ larger motorcycle~~s~~ markets are present~~ were still large~~. LDPVs and LDTs were two dominant subcategories of~~for~~ VOCs emissions. While t~~In this study, t~~he activity data for LDPVs were greatly improved in this study for~~to~~ reduced~~ the~~ uncertainty,~~.~~ improved data consisting reliable information for LDTs are expected in the future.~~ If possible, future studies were~~ are ~~expected to get more detailed information for LDTs.~~ China 0 vehicles were still a dominant source~~still contributed most~~ (35.19%) ~~to the~~ for tailpipe VOCs emission. Vehicles before China 4 (not included) contributed 94.67% to~~of~~ ~~the ~~total tailpipe emissions.

In 2015, China's on-road vehicles emitted 200.37 Gg IVOCs in total~~ly~~ (Table ~~4~~5). Figure 5 provides ~~the~~ IVOCs emissions by province. It should be noted~~ here,~~ that th~~is~~e estimation was entirely based on emission factors from tests in the United States, and is only capable of providing insights of similar magnitude~~. The current estimation can only provide insight of the order of magnitude~~ compared with VOC emission level based on the same input of vehicle activity data. Diesel vehicles ~~contributed~~with 72.4% share in~~to the~~ IVOCs emission,~~ which i~~was higher than gasoline vehicles due to the fact that ~~. This is because the ~~emission factors of trucks are significantly higher than those of ~~the ~~passenger vehicles.

In total, 97 species of evaporation, 30 species of gasoline exhaust and 20 species of diesel exhaust were ~~recognized~~identified (Figure 6). D~~The d~~etailed emission amounts were provided for 36 species, while ~~the ~~others ~~a~~were~~ listed~~ summed in the category named "others". Toluene, i-pentane, benzene ~~a~~were main species ~~from~~in gasoline exhaust. N-butane, i-pentane, i-butane and propane ~~a~~were the main species ~~from~~in evaporation. Evaporation shared 83.66% of ~~vehicle emissions of~~ n-butane, i-pentane, and i-butane vehicle emissions. Formaldehyde and acetaldehyde were mostly contributed by diesel tailpipe emissions. Small~~Our speciation profile for the evaporative emission has a tiny~~ fraction of unresolved complex mixture (UCM) was seen in our speciation profile for evaporative emission. T~~In the future, t~~he speciation profiles for exhaust~~ still needs to be improved~~ of~~for~~ 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.

~~3.2.2 Vehicle IVOCs emissions~~

~~In the year of 2015, China's on-road vehicles emitted 121.23 Gg IVOCs totally. Figure 5 was the IVOCs emissions by provinces. It should be noted here, the estimation was totally~~ entirely ~~based on emission factors from the tests in United States. The current estimation can only provide insight of the order of magnitude compared with VOC emission level based on the same input of vehicle activity data. Diesel vehicles contributed 58.31% to the IVOCs, which is a little bit higher than gasoline vehicles. Passenger vehicles and trucks~~ each ~~contributed half~~ of the total emission.~~to half.~~

~~3.2.3 Speciated vehicular VOCs emission inventory~~

~~In total, 97 species of evaporation, 30 species of gasoline exhaust and 20 species of diesel exhaust were recognized (Figure 6). The detailed emission amounts were provided for 36 species, while the others are listed in~~ the ~~category named "others". Toluene, i-pentane, benzene are main species from gasoline exhaust. N-~~

~~butane, i-pentane, i-butane and propane are main species from evaporation. Evaporation shared 83.66% vehicle emissions of n-butane, i-pentane, and i-butane. Formaldehyde and acealdehydeacetaldehyde were most contributed by diesel tailpipe emissions. Our speciation profile for the evaporative emission has very little percentagea tiny fraction of unresolved complex mixture (UCM). In the future, the speciation profile for exhaust still needs to be improved for both gasoline and diesel vehicles.~~

~~3.2.4 Implication to emission control~~

Figure 7 compared total VOCs emissions, emission intensity by area and emission per vehicles among provinces. As expected, ~~It's not a surprise that the highest emission intensity by area happened in the most~~ developed regions such as~~,~~ ~~eg.~~e.g. Beijing, Tianjin and Shanghai showed highest emission intensity by area (4500~9000 kg km^{-2}), while other provinces range between~~. In the other provinces, the emission intensity varies~~ ranges ~~in~~ 13~2700 kg km^{-2}. ~~So~~~~Therefore, if we only provide league table by total emissions, the readers may be misledading to avoid avoid the importance importance of emission control in such areas other than Beijing, Tianjin and Shanghai.~~ Beijing's ~~has the~~ lowest emissions per vehicle compared to~~with~~ other regions (14~85 kg per vehicle), indicated its fleet as the cleanest. ~~which means the vehicles in Beijing were the cleanest in Beijing.~~ Thus, mere technological approaches for emission reduction in more developed cities could be increasingly difficult. Alternative approaches such as reduction of ~~continuing reducing emissions from these highly developed regions using merely technology would be quite difficult from only technology aspect. The extra efforts from on reducing~~ population intensity~~, to~~ as well as ~~changing~~e human ~~behaviour~~behavior modification on ~~using~~ vehicle operation should be considered as~~s would be considered as the~~ main strategies in~~for~~ these regions.

3.3 Uncertainty analysis

Inevitable uncertainties come into theare 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^{-1} 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 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 wereas in the same order of magnitude of as the comparable to previous estimation and higher than Wei's forecast in 2011 (***Wei et al, 2011***). Following reasons may explain the differencesDifferences could be explained with following reasons. For the first timeFirstly, vehicle evaporative emission was taken into considerationdiscretely considered in detail for the first time, which substantially increased total VOCsadded on the total VOCs. Secondly, the vehicle usage data of vehicle usage was derived from big data, which wasere a survey for those '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 uncertainty in our inventorySo Hence, the emissions from old vehicles were reduced. To further reduce the uncertainty of VOCs emission inventory, activity for LDTs, China 0 and China 1 vehicles should be improved. These categories now still contributed significantly in total emission amount. However, according to due to author's

~~experience, has the usage data of these vehicles had the largest uncertainty among all categories.~~

~~It's very~~E~~ hard challenging to e~~valuation of~~e~~ the uncertainty in~~of~~ IVOC emission inventory is indeed challenging~~. The major uncertainty~~ as the majority of them~~ was~~ comes from~~ the~~ emission factors. ~~Recently,~~ IVOC measurements are recently more and more recognized, global attention could be expected to better existing emission factor database, considerably reducing uncertainty. ~~is getting a lot of attention popular in all around the world recently. The This global attention will help to build up the emission factor database and then reduce the uncertainty greatly.~~

The uncertainty of~~for~~ species profile was significant in~~for~~ exhausts and negligible in~~for~~ evaporation. Reason being that the~~The~~ profile used for evaporation was~~ a~~ reliable and comprehensive, ~~profile one~~ combining vehicle activity, technology contribution in fleet and profiles ~~for~~ in different processes. ~~Thus, the profile for evaporation was representative.~~ In addition, the species of evaporation were predominantly~~almost reached 100% of~~ total hydrocarbons, which ~~providing~~ provided enough resolution for specie profile. Achieving minimal uncertainty~~All the reasons above successfully reduced the uncertainty~~ of the species based inventory for evaporation. The uncertainty for exhaust species ~~were~~ was mainly of~~from~~ 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. ~~even for the individual tests, the recognized species was were too fewless from the VOCs analysis.~~ Thirdly, the species formed from incomplete combustion and ~~from~~ unburned fuel were not understood ~~very~~ well enough, which all present~~ brought~~ difficulties on building accurate species profiles for exhaust.

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4 Conclusion

The advantages merits of this study included: updated vehicle activity data from more than 70,000 cars and 2 million trucks in 30 different provinces, detailed vehicle fleet statistic, the 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.

ETo improve the emission inventory, the emission factors for running loss of evaporation are urgently needed to better the emission inventory. The IVOC emission factors for of all kinds of 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 uncompellingstill weak.

WTo providing provide insights on vehicle emission controls, we suggestsuggests to paying more attention to the reductioningreduce ofthe population density and vehicle usage usagee in highly-developed regions as main approach for emission reduction. Simultaneous alleviation in both traffic congestion and pollutant emission could be seen with these measures. These measures should be work as a "package policy" to deal with both the traffic congestion and also emissions.

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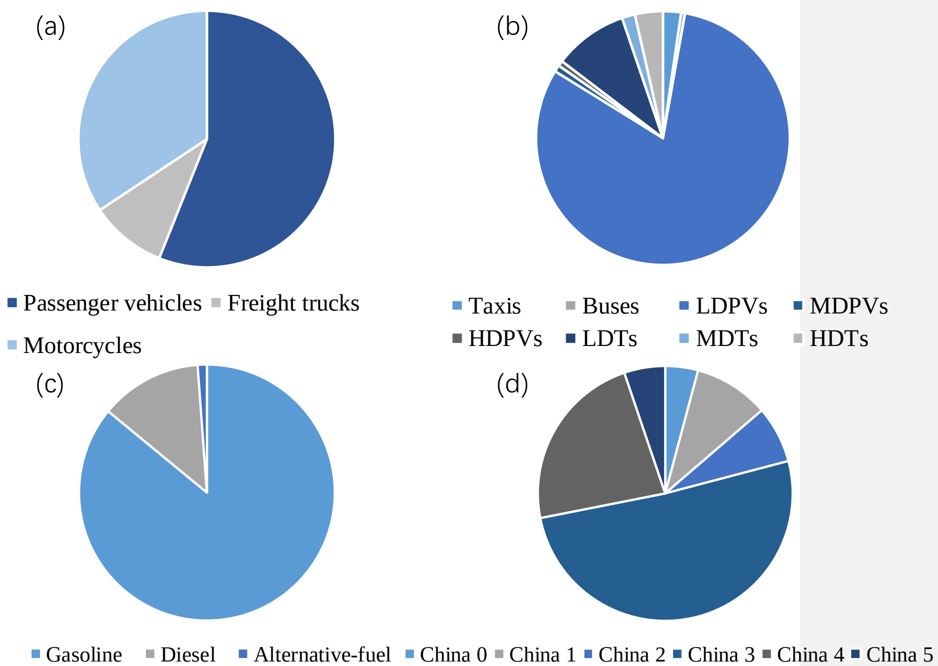


Figure 1. The pPercentages by vehicle types, fuel types and emission levels of China vehicle fleetownerships.

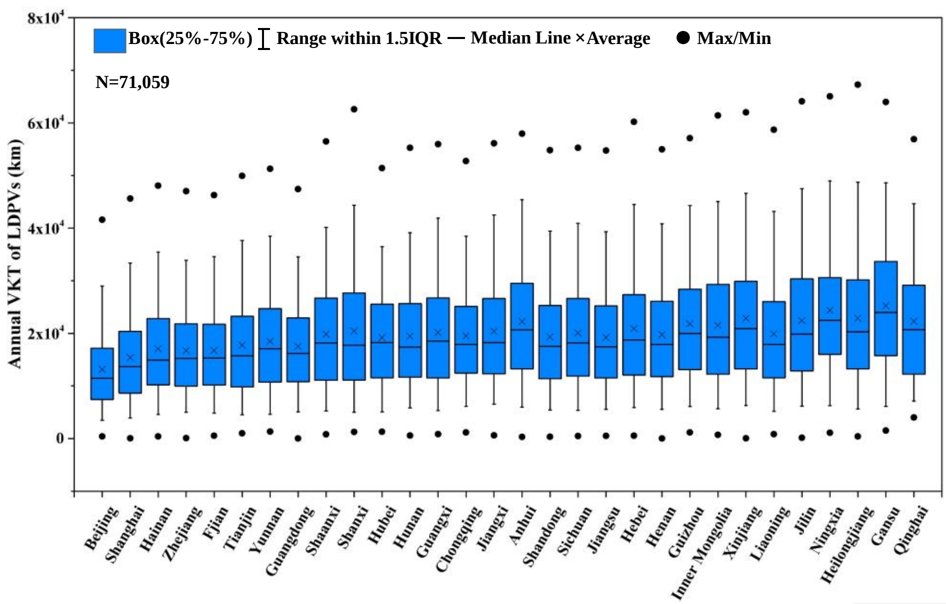
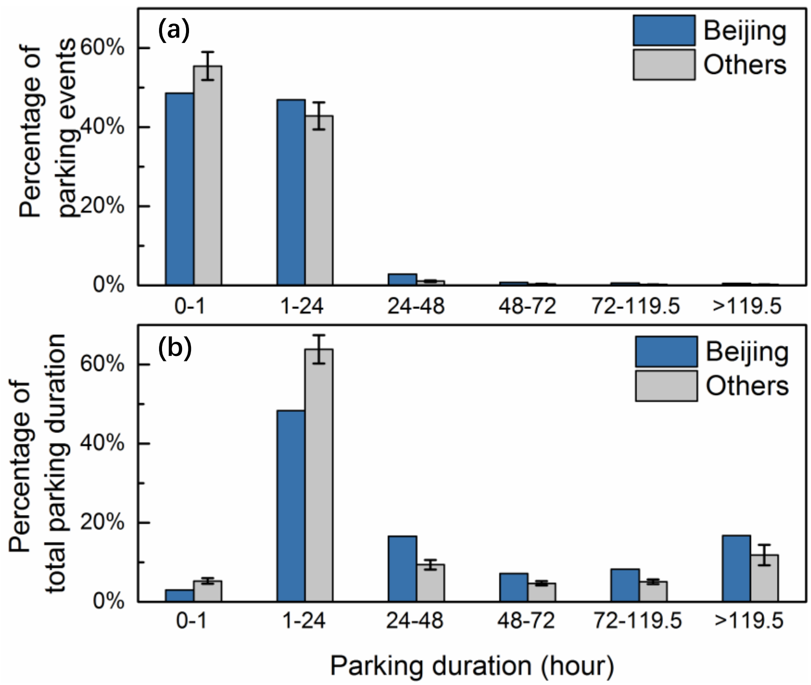


Figure 2. Provincial annual VKT of LDPVs in China.



1560 **Figure 3. Real-world parking duration distribution (a) percentage of parking events, (b) percentage of total parking duration.**

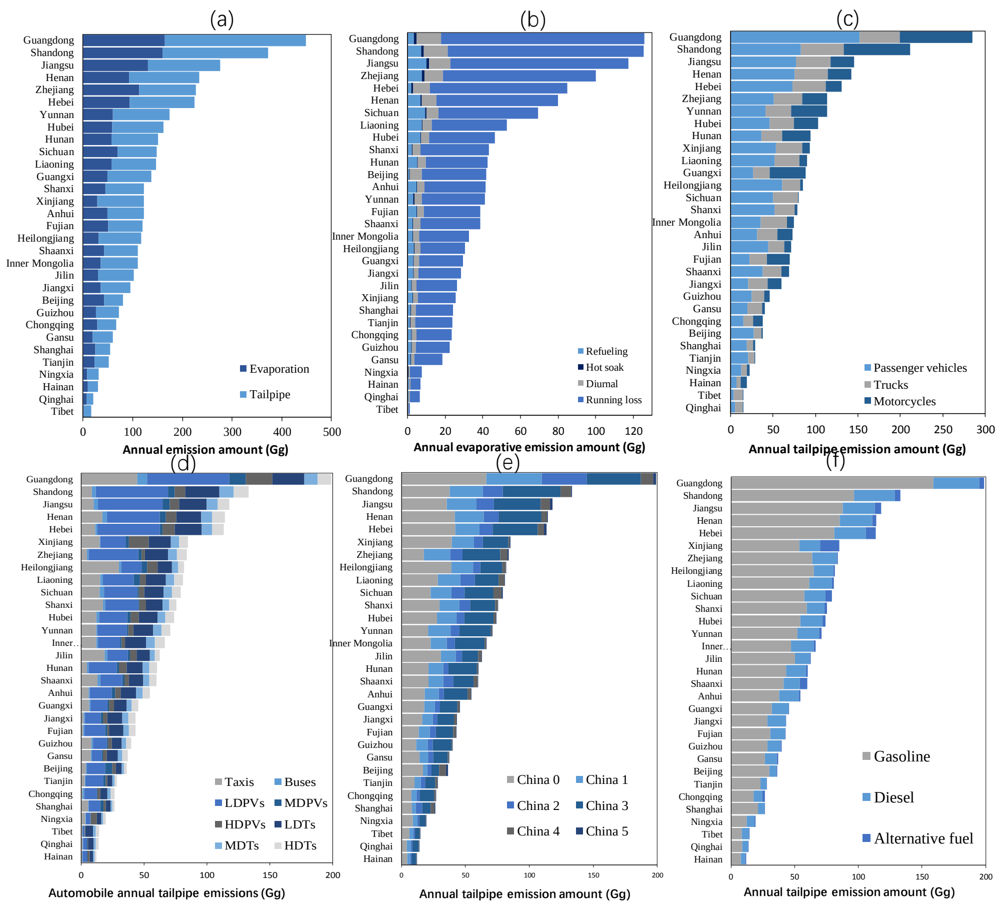


Figure 4. Provincial VOCs emissions from vehicles in 2015 (a) total emission amount classified by emission sources, (b) evaporation emission amount classified by evaporation processes (motorcycles excluded), (c) tailpipe emission amount classified by vehicle types, (d-f) tailpipe emission amounts classified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

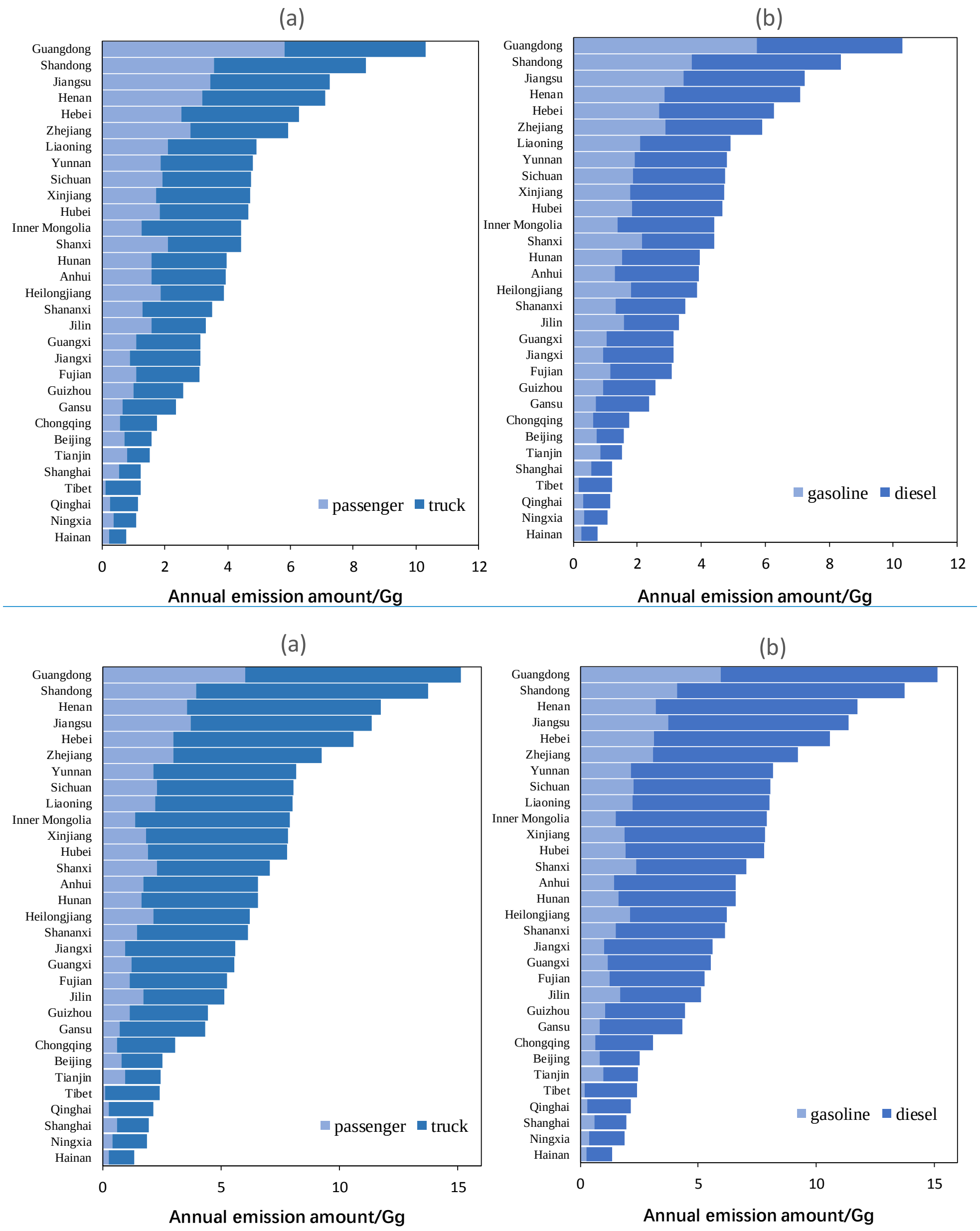


Figure 5. Provincial IVOCs emissions from vehicles in 2015 (a) total emission amount classified by vehicle types, (b) total emission amount classified by emission sources.

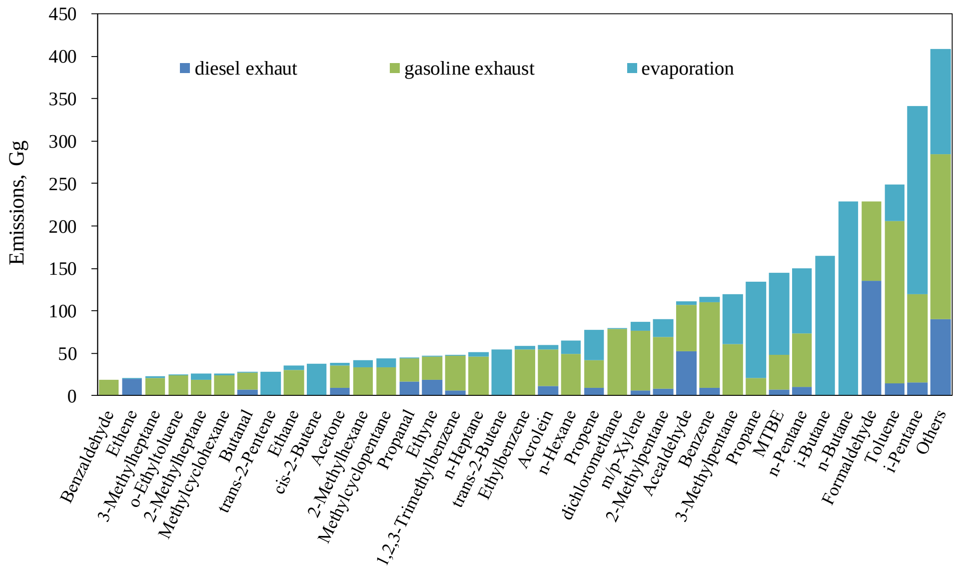


Figure 6. Speciated VOCs components emissions classified by emission source

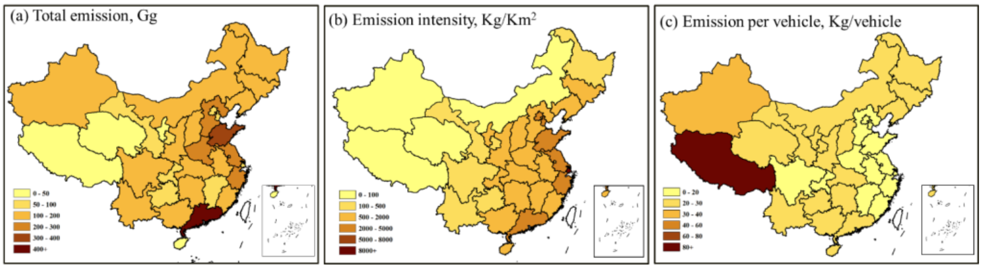
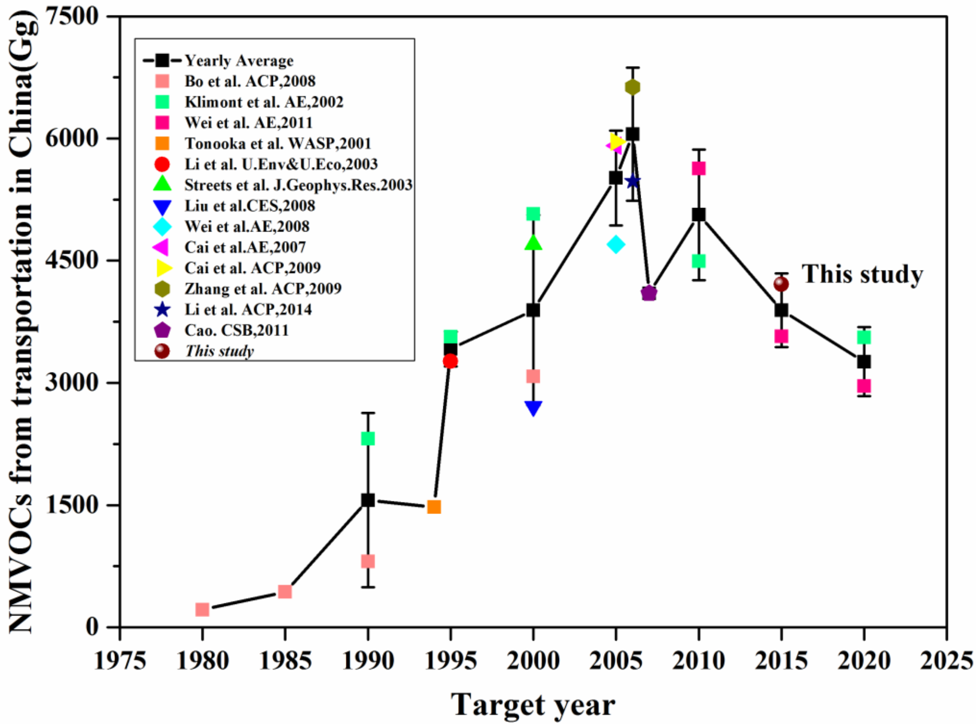


Figure 7. Province based emission analysis (a) total emission amount, (b) emission intensity, (c) emission per vehicles (motorcycles were excluded).



1585 **Figure 8. Comparing this study to previous transportation emission inventories.**

Table 2 Population of different types of vehicles in China in ~~the year of~~ 2015

Vehicle type	Population	Fuel type percentage (%)~~Population 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		

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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

[a] 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
China 0	22160	19270	35196	21231	27716	24372	138000	50000	31300	114800
China 1	22160	19270	35196	21231	27716	24372				
China 2	26335	26964	40766	28140	33226	38485				
China 3	29467	36581	47927	36366	40310	64128				
China 4	34165	45237	53497	60308	45820	98206				
China 5	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
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

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
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