JOURNAL OF ENVIRONMENTAL SCIENCES 84 (2019) 80–96

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JES JOURNAL OF ENVIRONMENTAL SCIENCES www.jesc.ac.cn

Recent development of a refined multiple air pollutant emission inventory of vehicles in the Central Plains of China

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ARTICLE INFO

Article history:
Received 20 December 2018
Revised 5 April 2019
Accepted 11 April 2019
Available online 22 April 2019

Keywords:
Vehicle
Emission standard
Gridded allocation
Ozone formation potential
Speciated volatile organic compounds

ABSTRACT

Central Plains region of China, represented by Henan Province, is facing serious air pollution problems. Vehicular exhaust emissions had adverse impacts on the atmospheric environment. The first comprehensive and novel vehicle emission inventory for Henan Province using vehicle kilometers traveled, localized emission factors, and activity data at city-level was developed. Furthermore, 3 km × 3 km gridded emission and temporal variations were determined by using localized information. Results show that the total emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), particular matter with aerodynamic diameter < 10 μm (PM_{10}), aerodynamic diameter < 2.5 μm ($PM_{2.5}$), volatile organic compounds (VOCs), VOCs-evaporation and ammonia in 2015 were 9.1, 533.4, 1190.7, 23.7, 21.6, 150.8, 31.5 and 10.4 Gg, respectively, and the emission intensities of the above pollutants were 0.05, 2.7, 6.0, 0.1, 0.1, 0.8, 0.2 and 0.05 g/km, respectively. Vehicles meeting the Primary China 1, China 3 and China 4 contributed 89.1%, 82.7%, 75.3%, 75.5%, 75.5%, 68.2%, 68.4% and 82.3% for SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-evaporation and ammonia emissions, respectively. Zhengzhou, Zhoukou, Nanyang, Luoyang, Shangqiu and Xinyang showed relatively higher emissions and contributed more than 50% of each pollutant. The spatial distribution indicated obvious characteristics of the road network, and high-level emission was concentrated in the downtown areas. Additionally, the ozone formation potential (OFP) based on the estimated speciated VOC emissions was 569.6 Gg in Henan Province. Aliphatic and aromatic hydrocarbons were the main species of VOCs, whereas olefins contributed the largest proportion of OFP, with 42.2%.

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https://doi.org/10.1016/j.jes.2019.04.010

Introduction

The automotive vehicle, a product under rapid economic development, has become an indispensable means of transportation all over the China. Vehicle exhaust emissions such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) are important precursors to particular matter (PM) and ozone (O_3) formation, which directly affect air quality and human health (Requia Jr et al., 2015; Shindell et al., 2011; Sonawane et al., 2012).

Previous research on emission inventories (Fu et al., 2001; Zhang et al., 2009, 2016), receptor source apportionment of $PM_{2.5}$ (aerodynamic diameter < 2.5 μm) and VOCs (Liang et al., 2016; Li et al., 2015, 2016, 2018a), and air quality simulation (Che et al., 2011) found that vehicle exhaust is one of the predominant sources of air pollution. For emission inventories, results have indicated that vehicles contributed 19.8%–36.1%, 7.9%–39.4%, and 9.0%–67.2% of total NO_x, VOCs, and carbon monoxide (CO) emissions in various areas of China, respectively (Fu et al., 2013; Liu et al., 2018a, 2018b; Qi et al., 2017; Qiu et al., 2014; Wu and Xie, 2017; Zheng et al., 2009b; Zhong et al., 2018). Source apportionment research using the positive matrix factorization (PMF) model for ambient VOCs in Beijing and Chongqing indicated that vehicle-related sources were the largest contributor, contributing 57% and 34% to ambient VOCs in these two cities, respectively (Li et al., 2016; Li et al., 2018a). Ambient VOCs were measured using an online system before, during, and after Asia-Pacific Economic Cooperation (APEC) China 2014 in Beijing. The results showed that controlling vehicle emission was the most effective measure to reduce VOCs, and the vehicle source was the most important precursor source likely responsible for the reduction in secondary organic aerosol (SOA) formation (Li et al., 2015). Additionally, the contribution of vehicle exhaust emission to $PM_{2.5}$ formation reached 14.2%–29.0% in some studies using the PMF (Cheng et al., 2015; Geng et al., 2013; Tao et al., 2013; Wang et al., 2015; Wu et al., 2014; Yu et al., 2013), reaching up to 38% in the developed city of Beijing in 2015 (Ji et al., 2018). Moreover, numerous studies of air quality modeling also found that the vehicle source directly affects the concentrations of both $PM_{2.5}$ and O_3 (Che et al., 2011; Kota et al., 2014; Soret et al., 2014; Zhang et al., 2018).

As for receptor source identification and air quality models, a refined emission inventory (as supporting files and input files) directly affects the accuracy of the result. Additionally, detailed information on inventory and emission characteristics can provide strong support for the creation of vehicle control measures. Therefore, vehicle emissions have been a hot research topic and were estimated at national, regional, or city scale by several studies (Lau et al., 2015; Streets et al., 2003).

A large amount of data such as emission factors (EFs), traffic activity, and fleet composition were used to establish emission inventories, which were strongly influenced by local traffic characteristics (Jing et al., 2016). As for traffic activity, monitored traffic flow data and annual statistical vehicle populations can be used to construct vehicle emission inventories at diverse scales. The former is generally used to estimate emission at smaller scale (city or county) for several months or up to one year for major roads. For example, an hourly emission inventory was developed at three typical road links in Beijing in 2013 and was based on traffic flow data for 24 hr, revealing the workday and weekend variations better (Jing et al., 2016). The latest study from Liu et al. (2018c) investigated hourly traffic-flow data each day to develop an emission inventory from February 2014 to January 2015 in Foshan. The latter method is mainly used to establish vehicle emissions without regional and annual restrictions. For example, vehicle emission inventories at the provincial level such as for Hebei (Liu et al., 2018a) and Guangdong Province (Zhong et al., 2018), developed regions like Beijing–Tianjin–Hebei (BTH) (Qi et al., 2017), Pearl River Delta (PRD) (Zheng et al., 2009b), and Yangtze River Delta (YRD) (Fu et al., 2013) or cities (Gong et al., 2017) were estimated based on the statistical populations of vehicles. Several studies have also adopted this approach in developing historical vehicle emissions at provincial or national levels in China (Huo et al., 2014; Sun et al., 2016).

Plentiful research on emission inventories has focused on the developed region in China, while less attention has been given to the Central Plains. It was identified that there were relatively high emission values in this region, taking Henan Province as a representative (Cai and Xie, 2007; Klimont et al., 2009; Zheng et al., 2014). In reality, the population of vehicles in Henan Province maintained a constant growth rate from 0.7 million in 1999 to 21 million in 2015 (NBS, 2016). Also, as the provincial capital and an important transportation center, Zhengzhou ranks sixth in vehicle ownership across cities in China (NBS, 2016), and is one of the cities with serious air pollution problems in China, with an annual average number of haze days exceeding three months (HNESAR, 2016).

Although several national studies have monitored vehicle emissions for Henan Province (Liu et al., 2017, 2018a; Song and Xie, 2006; Tang et al., 2016), some limitations still exist that do not entirely reflect local characteristics. First, significant gaps and large uncertainty of vehicle emissions has been found among different studies, subcategories of sources, or pollutants (Gong et al., 2017). Second, parts of previous studies only included part of the cities in this region, which resulted in underestimation, and the emission characteristics at the city level were still limited (Liu et al., 2018b; Qiu et al., 2014). Third, air quality modeling is still limited due to the low spatial resolution and scarce temporal distribution. Additionally, there are no studies regarding the speciated vehicle VOC emissions in this region to understand ozone formation potentials (OFPs). Considering the above points, it is critical to establish a comprehensive and updated vehicle emission inventory of high temporal–spatial resolution based on the localized correction of EFs and updated activity data for Henan Province.

In this paper, we developed a 2015-based vehicle emission inventory for Henan Province with 3 km × 3 km grid resolution, including the pollutants sulfur dioxide (SO_2), NO_x, CO, PM_{10} (aerodynamic diameter < 10 μm), $PM_{2.5}$, VOCs, VOCs-evaporation (VOCs-Evap) and ammonia (NH_3). We analyzed the composition of fleet types, emission characteristics of vehicles from different emission standards, fuel types, and cities in Sections 2.1 and 2.2, and characterized the spatial distribution and daily, weekly, and monthly variations of

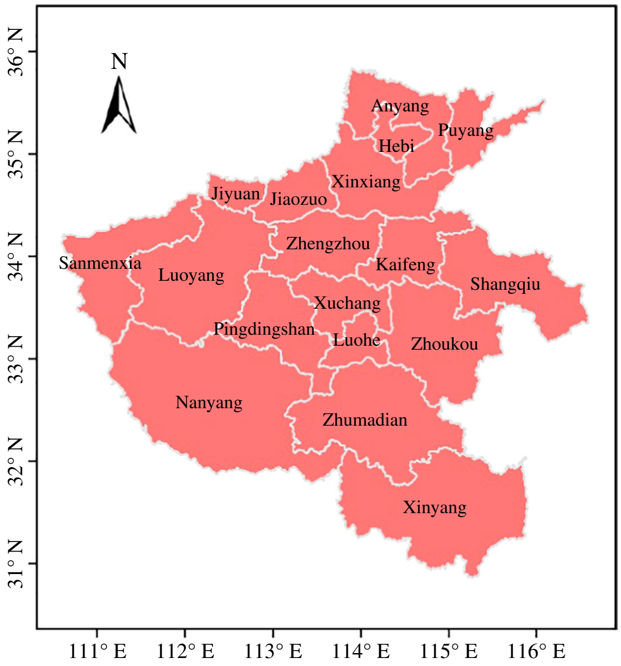

Fig. 1 – Henan Province and its location.

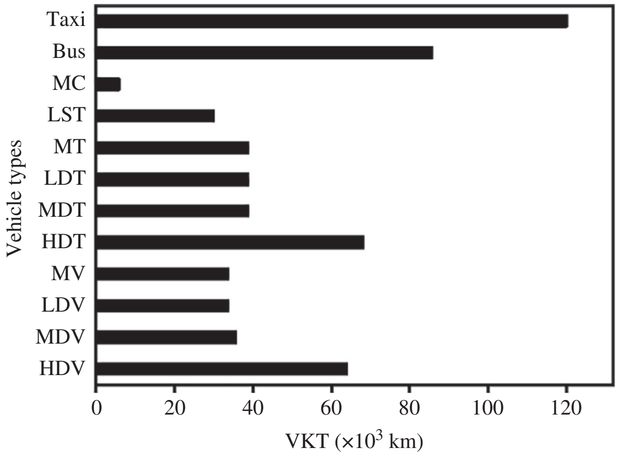

Fig. 2 – Annual averaged vehicle kilometer of travel (VKT) used in this study. MC: motorcycle; LST: low-speed truck; MT: mini duty truck; LDT: light duty truck; MDT: medium duty truck; HDT: heave duty truck; MV: mini duty vehicle; LDV: light duty vehicle; MDV: medium duty vehicle; HDV: heave duty vehicle.

vehicle emissions in Section 2.3. Speciated VOCs emissions were also estimated, and the contributions that fleet type and species for VOCs and OFPs are discussed in Section 2.5. This study aims to improve the knowledge regarding vehicle emission characteristics in central China as well as provide data to support refined management of vehicles in the future.

1. Data and methods

1.1. Study domain

Henan Province is located in east central China between the latitudes of 31°23′N and 36°22′N and longitudes of 110°21′E and 116°39′E. It includes 18 prefecture-level cities (Zhengzhou, the capital city, Kaifeng, Luoyang, Pingdingshan, Anyang, Hebi, Xinxiang, Jiaozuo, Puyang, Xuchang, Luohe, Sanmenxia, Shangqiu, Zhoukou, Zhumadian, Nanyang, Xinyang, and Jiyuan), as indicated in Fig. 1. Also, it is one of the vital economic provinces of China, covering 1.7% of the domestic land territory while accounting for 5.4% of the total national gross domestic product (GDP) in 2015. The population was estimated at 94.8 million in 2015, representing 6.7% of the Chinese population, ranking third among the provinces (NBS, 2016). Additionally, the vehicle population in Henan Province was estimated to have increased from 0.7 million in 1999 to 21 million in 2015 with an annual growth rate of 28%, ranking sixth in China, only below the provinces of Shandong, Zhejiang, Jiangsu, Hebei, and Guangdong in 2015 (NBS, 2016). Serving as a central transportation hub, the length of operational railways reached 5296 km while that of motorways was up to 250,548 km (NBS, 2016), ranking seventh and third in China in 2015, respectively.

1.2. Emission estimation

A bottom-up approach was implemented to develop an inventory of vehicular emissions with activity data at the city level (Zheng et al., 2014; Wang et al., 2015; MEP, 2002). The magnitudes of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-Evap, and NH_3 emissions were calculated based on EFs and vehicle kilometers of travel (VKT) using the following Eq. (1):

$$E_{a,b} = \sum_z \sum_y \left(P_{a,z,y} \times EF_{a,z,y,b} \times VKT_{a,z}\right) \times 10^{-6} \tag{1}$$

where, a represents each city, b is the targeted pollutant, z is the type of vehicle, y is the national vehicular emission limit standards (Pre-China 1, China 1, China 2, China 3, China 4, and China 5 emission standards), E (Mg) represents emission; P is the vehicle population, VKT (km) is the annual average vehicle kilometers of travel, EF (g/km) is the emission factor. The type of vehicle (z) in this study includes 9 diesel vehicles, i.e., heavy-duty diesel vehicles (HDDV), heavy-duty diesel trucks (HDDT), medium-duty diesel trucks (MDDT), medium-duty diesel vehicles (MDDV), light-duty diesel trucks (LDDT), mini-diesel trucks (MDT), low-speed diesel trucks (LSDT), diesel buses (DB), and diesel taxis (DT), 11 gasoline vehicles, i.e., heavy-duty gasoline vehicles (HDGV), medium-duty gasoline trucks (MDGT), medium-duty gasoline vehicles (MDGV), light-duty gasoline trucks (LDGT), mini-gasoline trucks (MGT), light-duty gasoline vehicles (LDGV), mini-gasoline vehicles (MGV), normal gasoline motorcycles (NGMC) (NTCAS, 2001; MEP, 2002), light gasoline motorcycles (LGMC), gasoline taxis (GT), and gasoline buses (GB), and 4 other fuel type vehicles, i.e., other-fuel buses (OFB), other-fuel taxis (OFT), other-fuel heavy-duty vehicles (OFHDV), and other-fuel medium-duty vehicles (OFMDV); other fuels mainly include liquefied petroleum gas (LPG), liquefied natural gas (LNG), and compressed natural gas (CNG).

Table 1 – Population of different vehicle types and corresponding technology distribution in Henan Province for the year of 2015.

Fuel	Vehicle	Population	Emission standard distribution (%)					
			Pre-China 1	China 1	China 2	China 3	China 4	China 5
Gasoline	HDGV	4032	13.5	8.0	23.0	28.5	27.0	–
	MDGT	7130	16.4	18.9	31.3	21.0	12.5	–
	MDGV	7917	11.2	12.0	29.8	22.5	24.4	–
	LDGT	301478	0.2	11.7	8.0	19.7	60.4	–
	MGT	3061	1.1	81.4	11.1	3.9	2.4	–
	LDGV	7728704	0.2	7.0	7.6	16.5	68.7	–
	MGV	232356	2.7	57.0	9.2	13.7	17.4	–
	NGMC	4553669	10.5	28.3	37.7	23.6	–	–
	LGMC	116134	18.0	72.4	4.3	5.3	–	–
	GT	17088	–	–	–	45.3	54.7	–
	GB	378	–	–	12.5	38.5	22.6	26.4
	Subtotal	12971947	4.0	16.1	18.2	18.9	42.8	0.0
Diesel	HDDV	50229	5.5	15.2	9.5	40.0	29.8	–
	HDDT	415592	2.0	5.5	5.3	56.6	30.6	–
	MDDT	70963	2.7	19.5	17.0	43.4	17.5	–
	MDDV	31756	4.6	15.8	13.2	37.2	29.1	–
	LDDT	476548	0.9	5.3	6.4	49.7	37.6	–
	MDT	1907	3.3	64.3	16.0	16.3	–	–
	LSDT	115475	–	–	100.0	–	–	–
	DB	11069	–	–	12.5	38.5	22.6	26.4
	DT	539	–	–	–	100.0	–	–
	Subtotal	1174077	1.6	6.4	16.3	46.0	29.4	0.2
Other fuels	OFT	43928	–	–	–	61.7	38.3	–
	OFB	13811	–	–	12.5	38.5	22.6	26.4
	OFHDV	15846	6.7	19.1	13.7	10.5	19.8	30.2
	OFMDV	3036	5.3	24.2	19.4	11.4	13.2	26.4
	Subtotal	76621	1.6	4.9	5.9	44.9	30.6	12.0
	Total	14222645	3.8	15.2	18.0	21.3	41.6	0.1

Other fuels mainly include compressed natural gas, liquefied natural gas, liquefied petroleum gas and two-fuel for natural gas and liquefied petroleum gas vehicles. -: no data; HDGV: heavy-duty gasoline vehicle; MDGT: medium-duty gasoline truck; MDGV: medium-duty gasoline vehicle; LDGT: light-duty gasoline truck; MGT: mini-gasoline truck; LDGV: light-duty gasoline vehicle; MGV: mini-gasoline vehicle; NGMC: normal gasoline motorcycle; LGMC: light gasoline motorcycle; GT: gasoline taxi; GB: gasoline bus; HDDV: heavy-duty diesel vehicle; HDDT: heavy-duty diesel truck; MDDT: medium-duty diesel truck; MDDV: medium-duty diesel vehicle; LDDT: light-duty diesel truck; MDT: mini-diesel truck; LSDT: low-speed diesel truck; DB: diesel bus; DT: diesel taxi; OFT: other-fuel taxi; OFB: other-fuel bus; OFHDV: other-fuel heavy-duty vehicle; OFMDV: other-fuel medium-duty vehicle; Pre-China 1, China 1, China 2, China 3, China 4, China 5: vehicle pollutant emission limits and measurement methods (different stages).

1.2.1. Handling of vehicle population

Initially, the populations of vehicles with different emission standards corresponding to EFs need to be calculated using the number of newly registered vehicles, survival rate, correction factor, fuel ratio, and the implementation year for each stage of emission standard. The initial vehicle population data and registration numbers of fleet composition for each city were obtained from statistical data at the city level (HNDRC, 2016; HNBS, 2016). The proportions of different fuel types were provided by the department of government, which were used to split the population into primary vehicle types. (HNDEE, 2014; HNDRC, 2016). We assumed that new vehicles registered in a given year were in compliance with the newest emission standard. The population of vehicles with different emission standards was calculated using the number of modified new registration vehicles as follows (Wang et al., 2010):

$$P_{a,z} = \sum_{y=0}^{5} \sum N_{a,z,m,n} \tag{2}$$

where, y is the vehicular emission standard (from Pre-China 1 to China 5); n is the year range during the period the vehicular emission standard y was implemented for vehicle type z in area a; m represents the year when the emission standard China y was started; and N is the number of modified new registration vehicles.

Using the LDGV with Standard China 1 in Henan Province as an example, n would range from July 2000 to June 2005, and m represents the year (July 2000) that the emission standard (China 1) was implemented for the LDGV. The sum of the modified new registration numbers for the LDGV during the time period from July 2000 to June 2005 in this region would be a population satisfying Standard China 1, and it is clear that numbers for Pre-China 1 to China 5 could also be calculated by this method. $P_{a,\ z}$ is the total population of LDGV in city a for the target calculation year.

The population for the modified new registrations was calculated as follows (Wang et al., 2010):

$$N_{h,j} = R_h \times S_{h,j-h} \times \frac{\sum_{h=2000}^{h=2015} R_h}{P_{2015}} \tag{3}$$

where, $N_{h,\ j}$ represents the modified new registration vehicle numbers that survived in target year j; R_h represents the numbers of newly registered vehicles in year h (the target year in 2015); $S_{h,\ j-h}$ is the survival rate in the target year with the vehicular age ($j-h$); P_{2015} is the total vehicle population of a certain type in the city for the year of 2015.

In this study, survival rates were supplied by Zachariadis et al. (1995) and simulated using the Weibull distribution by the formula as follows (Huo and Wang, 2012):

$$S_{h,j-h} = \mathrm{EXP}\left[-\left(\frac{(j-h)+c}{T}\right)^{c}\right] \tag{4}$$

where, T and c are two parameters associated with vehicle service life. $T = 32$, $c = 7$ for passenger vehicles, and $T = 25$, $c = 10$ for trucks in Henan Province.

1.2.2. Refined emission factor

The basic emission factors (BEFs) were obtained from the national handbook of vehicle emissions (Technical guidelines on emission inventory development of air pollutants from on-road vehicles (on trial)). The tests were conducted by a research group from Tsinghua University (Hu et al., 2012; Huo et al., 2009; Liu et al., 2017; Wang et al., 2013; Wu et al., 2012), and published by China's Ministry of Environment Protection (MEP, 2015). The BEFs of fleet types in this study are displayed in Appendix A Table S1. The comprehensive and localized EFs were corrected using the actual regional conditions as follows:

$$\mathrm{EF}_{a,b,z,y} = \mathrm{BEF}_{z,y,b} \times \varphi_a \times \gamma_{b,z,y} \times \mu_{b,z,y} \times \theta_z \tag{5}$$

where, $\mathrm{EF}_{a,b,z,y}$ represents the comprehensive EF of pollutant b by vehicle type z with emission standard y in city a; BEF is the baseline EF measured in the standard condition. φ represents the meteorological correction factors (temperature, humidity, and height in each city), and the meteorological condition of the 18 cities in each month of 2015 was provided by HMS, 2016, as shown in Appendix A Table S2. γ and μ are the average velocity correction coefficient and degradation correction coefficient, respectively; θ represents the other emission correction coefficients used in other conditions.

1.2.3. Determination of VKT

Annual VKT is also a significant parameter and can directly influence the estimation of emissions. In this study, it was determined by combining the Statistical yearbook of China (NBS, 2016), previous studies (Zhao, 2014), and a sampling survey of 157,505 sample messages obtained from the Zhengzhou Environmental Protection Bureau (ZZEPB). The annual VKTs used in this study are listed in Fig. 2.

1.3. Refined and optimized temporal–spatial allocation

The regional air quality modeling simulations have exact requirements on the resolution for temporal and spatial allocation. Improved temporal and spatial surrogates will help reduce the uncertainties in developing model-ready input profiles. In this study, the emissions at the city level were allocated to a 3 km × 3 km grid, with the emission of each grid calculated using Eq. (6) (Zheng et al., 2009a):

$$\mathrm{E_{b,w}} = \mathrm{E} \times \frac{\sum\limits_{i}(\mathrm{L_{w,i}} \times \mathrm{Q_{w,i,b}} \times \mathrm{I_{w,i}})}{\sum\limits_{w}\sum\limits_{i}(\mathrm{I_{w,i}} \times \mathrm{I_{w,i,b}} \times \mathrm{I_{w,i}})} \tag{6}$$

where, w and i are grid number and road types, respectively; E is the total emission in grid w; Q, I, and L represent the emission intensity, traffic flow of each vehicle type and road length of different types of roads, respectively.

The road network includes different types of highways, national, provincial, urban, and county roads in Henan Province, which are depicted in Appendix A Fig. S3. The urban roads were further divided into four parts, including ring, artery road, collector, and branch roads. Traffic flows were developed using local investigations and previous research on local conditions (Gong et al., 2017; Zhao, 2014). In this study, sections of spatial distribution were optimized and refined when considering particular situations, for example taxis and buses mainly run through urban roads while low-speed trucks and motorcycles mainly through county roads. Therefore, the vehicle types for taxi, bus, low-speed truck and motorcycle cannot be simply distributed onto the highways as previous studies had done.

In addition, the local traffic flows in six artery roads, six collectors, and six branch roads during weekdays and weekends in Zhengzhou were acquired from an investigation by the ZZEPB in 2015, which were used to improve the daily and weekly temporal variations. The monitoring data of highway traffic flows during one whole year were obtained from the Transportation Department of Henan Province (HNTD, 2017), which were utilized for monthly distribution analysis in this study.

1.4. Speciated VOC emission and its ozone formation potential

The index of OFPs reflects the relative contributions of various types of VOCs to O_3 production, which in turn can identify the key sources and species for O_3 formation.

Various types of VOCs contribute significantly and differently to O_3 due to their different chemical reactivities and reaction mechanisms. The VOC emission profiles were derived by a literature review, and were used to establish the vehicular VOC emission inventory. The VOC emission profiles for gasoline and diesel vehicles with various driving speeds and motorcycles were derived using on-board exhaust tests in Tianjin, Xiamen, and Taiwan (Dai et al., 2013; Mo et al., 2016; Tsai et al., 2012; Wang et al., 2013; Yao et al., 2013). The VOC emission profiles of light duty gasoline vehicles (LDGV), MDV and taxis were obtained following Hsu et al. (2007). The estimation methods of OFPs are shown as follows:

$$\mathrm{OFP} = \sum_{\mathrm{z=1}}^{\mathrm{z}}\sum \mathrm{OFP_z} \tag{7}$$

$$\mathrm{OFP_z} = \sum_{p=1}^{p}\sum E_{z,p} \times \mathrm{MIR_p} \tag{8}$$

where, OFP (Mg) is the total OFP; OFP_z represents the OFP of vehicle type z. $E_{z,p}$ (Mg) is the mass of p th VOC species with

Table 2 – Multi-pollutant emissions of on-road mobile source for Henan Province in 2015.

Fuel	Vehicle	SO_2		NO_x		CO		PM_{10}		$PM_{2.5}$		VOCs		VOCs-Evap		NH_3	
		Emission (Mg)	Percentage (%)	Emission (Gg)	Percentage (%)	Emission (Gg)	Percentage (%)	Emission (Mg)	Percentage (%)	Emission (Mg)	Percentage (%)	Emission (Gg)	Percentage (%)	Emission (Mg)	Percentage (%)	Emission (Mg)	Percentage (%)
Gasoline	HDGV	5.2	0.1	0.7	0.1	9.5	0.8	27.8	0.1	23.9	0.1	0.7	0.4	26.6	0.1	7.2	0.1
	MDGT	5.4	0.1	1.0	0.2	16.9	1.4	35.7	0.2	31.1	0.1	1.3	0.9	36.1	0.1	7.5	0.1
	MDGV	5.7	0.1	0.4	0.1	5.2	0.4	9.1	0.0	8.5	0.0	0.5	0.3	30.2	0.1	7.9	0.1
	LDGT	117.1	1.3	9.9	1.9	127.6	10.7	216.4	0.9	202.1	0.9	12.6	8.4	662.1	2.1	304.3	2.9
	MGT	1.2	0.0	0.3	0.1	4.1	0.3	7.4	0.0	6.4	0.0	0.5	0.3	17.2	0.1	3.1	0.0
	LDGV	2604.6	28.6	38.0	7.1	516.9	43.4	1217.3	5.1	1217.3	5.6	55.6	36.9	13426.0	42.7	6771.9	65.3
	MGV	78.3	0.9	3.3	0.6	53.7	4.5	162.1	0.7	162.1	0.7	5.2	3.5	1008.9	3.2	203.6	2.0
	NGMC	–	–	4.3	0.8	192.4	16.2	351.8	1.5	351.8	1.6	26.6	17.7	15788.0	50.2	2178.0	21.0
	LGMC	–	–	0.1	0.0	4.6	0.4	14.5	0.1	14.5	0.1	2.4	1.6	421.3	1.3	4.9	0.0
	GT	20.6	0.2	0.3	0.1	5.6	0.5	9.3	0.0	9.3	0.0	0.7	0.5	61.7	0.2	53.5	0.5
	GB	0.7	0.0	0.0	0.0	0.2	0.0	1.7	0.0	1.4	0.0	0.0	0.0	1.2	0.0	114.9	1.1
	Subtotal	2838.5	31.2	58.3	10.9	936.6	78.7	2053.4	8.7	2028.6	9.4	106.2	70.4	31479.1	100.0	9656.8	93.2
Diesel	HDDV	450.1	4.9	42.5	8.0	29.6	2.5	2798.4	11.8	2522.3	11.7	2.4	1.6	–	–	54.6	0.5
	HDDT	3975.5	43.7	272.0	51.0	119.5	10.0	12242.0	51.6	11014.6	50.9	17.5	11.6	–	–	482.7	4.7
	MDDT	386.5	4.2	22.2	4.2	12.1	1.0	1564.8	6.6	1409.7	6.5	2.9	1.9	–	–	46.9	0.5
	MDDV	164.7	1.8	5.7	1.1	4.0	0.3	483.9	2.0	439.6	2.0	1.3	0.8	–	–	20.0	0.2
	LDDT	930.6	10.2	86.5	16.2	50.8	4.3	3218.9	13.6	3002.8	13.9	11.2	7.4	–	–	74.5	0.7
	MDT	2.9	0.0	0.5	0.1	0.4	0.0	27.2	0.1	19.2	0.1	0.2	0.1	–	–	0.2	0.0
	LSDT	173.2	1.9	16.8	3.2	12.6	1.1	738.4	3.1	681.6	3.2	4.2	2.8	–	–	13.9	0.1
	DB	174.1	1.9	9.1	1.7	4.6	0.4	373.6	1.6	338.1	1.6	0.3	0.2	–	–	16.2	0.2
	DT	3.2	0.0	0.1	0.0	0.0	0.0	2.6	0.0	1.9	0.0	0.0	0.0	–	–	0.3	0.0
	Subtotal	6260.7	68.8	455.4	85.4	233.5	19.6	21449.8	90.5	19429.9	89.8	40.0	26.5	–	–	709.3	6.8
Other fuels	OFT	–	–	0.3	0.1	3.8	0.3	32.6	0.1	32.6	0.2	0.5	0.4	–	–	–	–
	OFB	–	–	9.1	1.7	7.4	0.6	63.9	0.3	52.0	0.2	1.9	1.3	–	–	–	–
	OFHDV	–	–	10.0	1.9	8.9	0.7	99.2	0.4	85.1	0.4	2.1	1.4		–	–	–
	OFMDV	–	–	0.3	0.1	0.5	0.0	3.5	0.0	3.1	0.0	0.1	0.1	–	–	–	–
	Subtotal	–	–	19.7	3.7	20.7	1.7	199.1	0.8	172.8	0.8	4.6	3.1	–	–	–	–
	Total	9099.3	100	533.4	100	1190.7	100	23702.3	100	21631.2	100	150.8	100	31479.1	100	10366.1	100

VOCs: volatile organic compounds; VOCs-Evap: VOCs-evaporation; NO_x: nitrogen oxides; PM_{10} and $PM_{2.5}$: particular matter with an aerodynamic diameters <10 μm and < 2.5 μm, respectively.

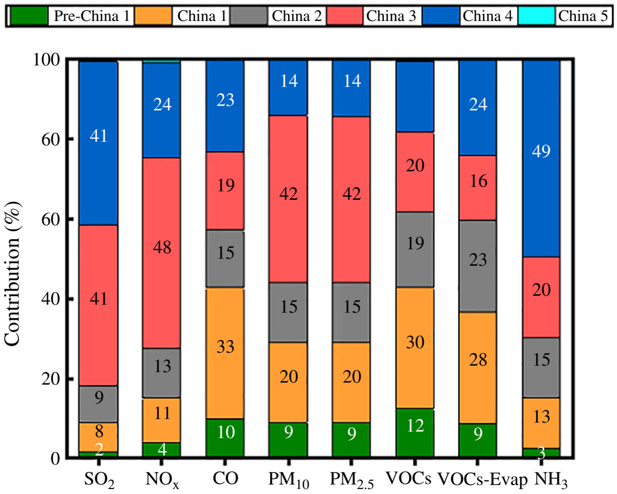

Fig. 3 – Emission contributions of vehicles with different emission standards. The ratio of emission standard composition less than 1% is not marked.

type z; MIR$_p$ (g O$_3$/g VOC) is the corresponding maximum increment reactivity of p th VOC chemical species to generate O$_3$ in a given VOC air mass, which can be found in the literature (Carter, 2008).

2. Results and discussion

2.1. *Traffic composition*

The original population of vehicles acquired from Henan Province Statistics Bureau (HNBS, 2016) included passenger cars, trucks, low-speed trucks, motorcycles, buses, and taxis. In order to develop an emission inventory in combination with the distribution of fuel ratio, the target source category was divided into more extensively detailed fleet compositions as shown in Table 1 and Appendix A Fig. S1. In terms of fuel type, it was demonstrated that gasoline vehicles accounted for 91.2% of the total number of vehicles, with primary fleets of LDGV and NGMC contributing 59.6% and 35.1% to the gasoline vehicle category, respectively. Diesel vehicles accounted for only 8.3% of all vehicles, with the proportionally larger fleets like HDDT and LDDT accounting for 35.4% and 40.6% of diesel vehicles, respectively. The other fuel vehicles accounted for 0.5% of total vehicles, which were mainly obtained from HNTD, 2017. In terms of emission standards in 2015, the regional vehicles of China 4 standard accounted for the largest proportion (41.6%) of the total vehicle fleet, followed by vehicles of China 3 (21.3%), China 2 (18.0%), China 1 (15.2%), Pre-China 1 (3.8%) and China 5 (0.1%).

The fleet compositions of 12 types (HDV, MDV, LDV, DV, HDT, MDT, LDT, DT, MC, LST, taxi, and bus) by city are described in Appendix A Fig. S1. Zhengzhou, the capital city, had a very large population of 2.9 million vehicles, accounting for 20.9% of the total population in Henan Province, and ranked in the top 10 cities of China in 2015 (NSB, 2016), mostly due to the larger number of LDV and LDT types. The second large contributor is the city of Nanyang, and the total vehicle population was 1.6 million, with motorcycles (20.5%) partially determining the fleet composition of the whole vehicle population. The largest ratio of HDT, with population of 0.07 million, was found in Zhoukou, reaching 18.2% of Henan. Additionally, Jiyuan and Hebi had the lowest vehicle populations, most likely due to the fact that they have low permanent populations and occupy the smallest geographic area in Henan Province.

2.2. *2015-based emission inventory and its characteristics*

2.2.1. *Contribution by different vehicle and fuel types*

In 2015, the total vehicle emissions of SO$_2$, NO$_x$, CO, PM$_{10}$, PM$_{2.5}$, VOCs, VOCs-Evap, and NH$_3$ for Henan province were calculated at 9.1, 533.4, 1190.7, 23.7, 21.6, 150.8, 31.5 and 10.4 Gg, respectively. Each pollutant and its contribution from each vehicle type were studied to better understand the emission characteristics and are shown in Table 2.

Although HDDT only accounted for 2.9% of the total population at 0.4 million vehicles, it was the main subcategory for the SO$_2$, NO$_x$, PM$_{10}$, and PM$_{2.5}$ emissions, which were responsible for 43.7%, 51.0%, 51.6%, and 50.9%, respectively. The reason was that the high VKT of HDDT and the high load factor of the diesel engine leads to high temperature and oxygen deficiency in the engine combustion chamber, accelerating the formation of soot. LDDT were the second largest contributor for SO$_2$, NO$_x$, PM$_{10}$, and PM$_{2.5}$ emissions, with a proportion no less than 10%. LDGV, the largest fleet type with a population of 7.7 million vehicles, was the primary source for CO and VOC emissions, accounting for 43.4% and 36.9% of the total emissions, respectively. LDGV were also a major source for VOCs-Evap and NH$_3$ emissions, which accounted for 42.7% and 65.3% of total emissions, respectively. In the past 5 years, the LDGV population has been growing with an annual rate varying between 12.1% and 31.5% (ZSY, 2016). Due to this state of affairs, the Henan government has enacted a policy for the elimination of yellow-label vehicles, which are defined as vehicles with lower emission standard than China 1 for gasoline vehicles and China 3 for diesel vehicles, by the end of 2017 (HNDEE, 2014). However, the elimination rate for these vehicles cannot keep pace with the explosive growth of the vehicle population under the booming economy. Furthermore, NGMC, accounting for 32.0% of the total vehicle fleet, had relatively large contributions to CO, VOCs, VOCs-Evap, and NH$_3$ emissions at 16.2%, 17.7%, 50.2%, and 21.0% of the total emissions, respectively, due to the special topographical features in the region.

In general, results indicated that the gasoline vehicle category was the largest contributor to CO, VOCs, VOCs-Evap, and NH$_3$ emissions, of which LDGV and NGMC were the two major subcategories, while the diesel vehicle category for HDDT and LDDT produced the most SO$_2$, NO$_x$, and PM emissions in this region. Based on these findings, it appears that limiting the LDGV population and non-electric motorcycles would be the most important policies for decreasing CO and VOC emissions, and augmenting the emission standard for HDDT would be effective in reducing the vehicle emissions of NO$_x$ and PM. Other fuel vehicles, like taxis and busses, only accounted for 0.5% of total ownership, thus increasing the use of public transit and reducing

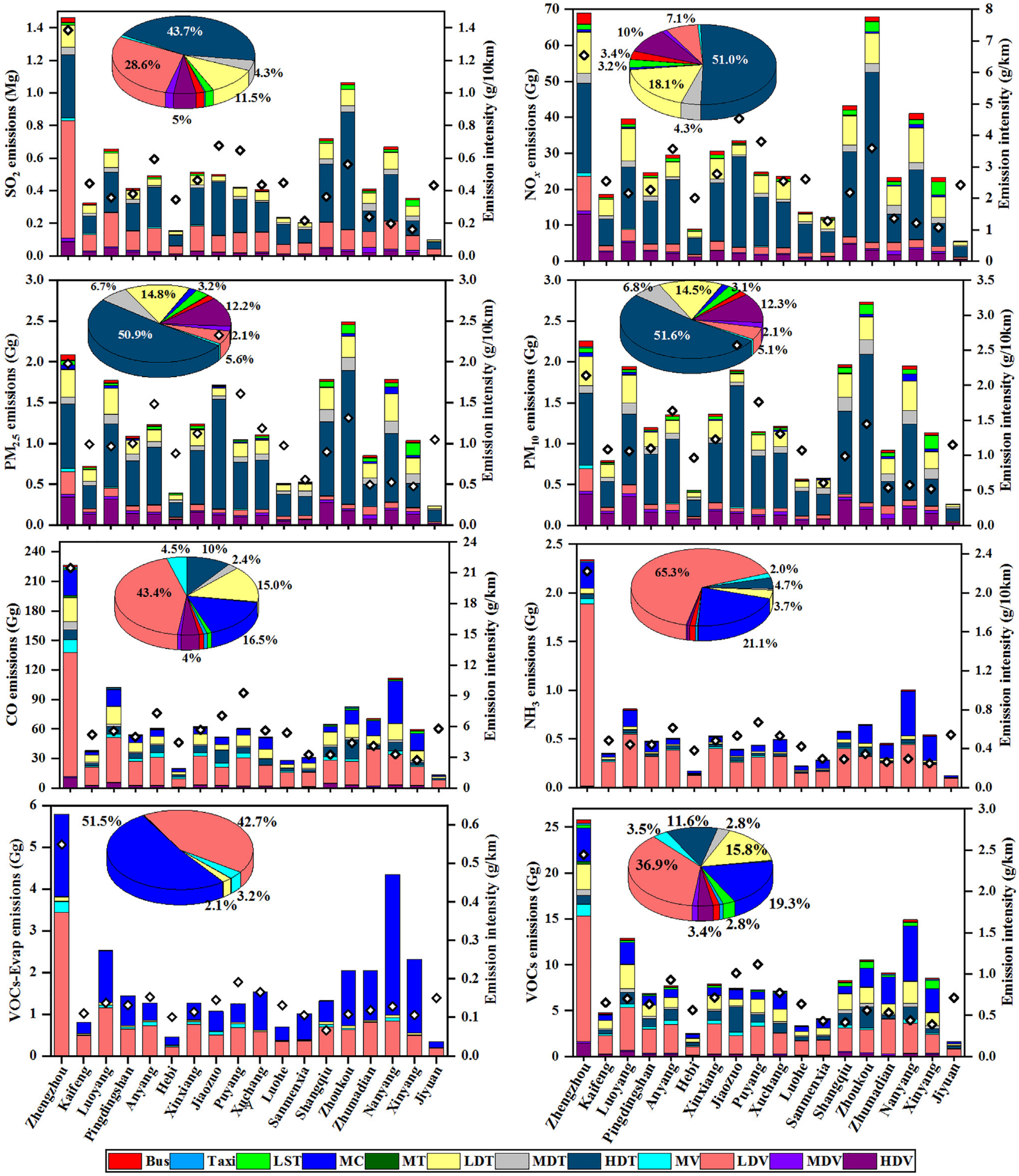

Fig. 4 – Emissions of major fleet types, emission intensity by each city for different pollutants and corresponding emission composition at province level (pie chart), 2015. The ratio of fleet type composition less than 2% is not marked.

private vehicular ownership would also be important measures in reducing pollutant emissions.

2.2.2. *Emission characteristics of different emission standards*

The emission contribution by vehicles of different emission standards is presented in Fig. 3. For each pollutant, the different composition of emission standards was attributed to the main contributive vehicle types. China 4 vehicles, with the largest population (41.6% of the whole vehicle population), contributed 41.0% and 49.4% to SO$_2$ and NH$_3$ emissions, respectively. Vehicles predating China 3 (not shown) contributed 57.3%–61.7% to CO, VOCs, and VOCs-Evap emissions,

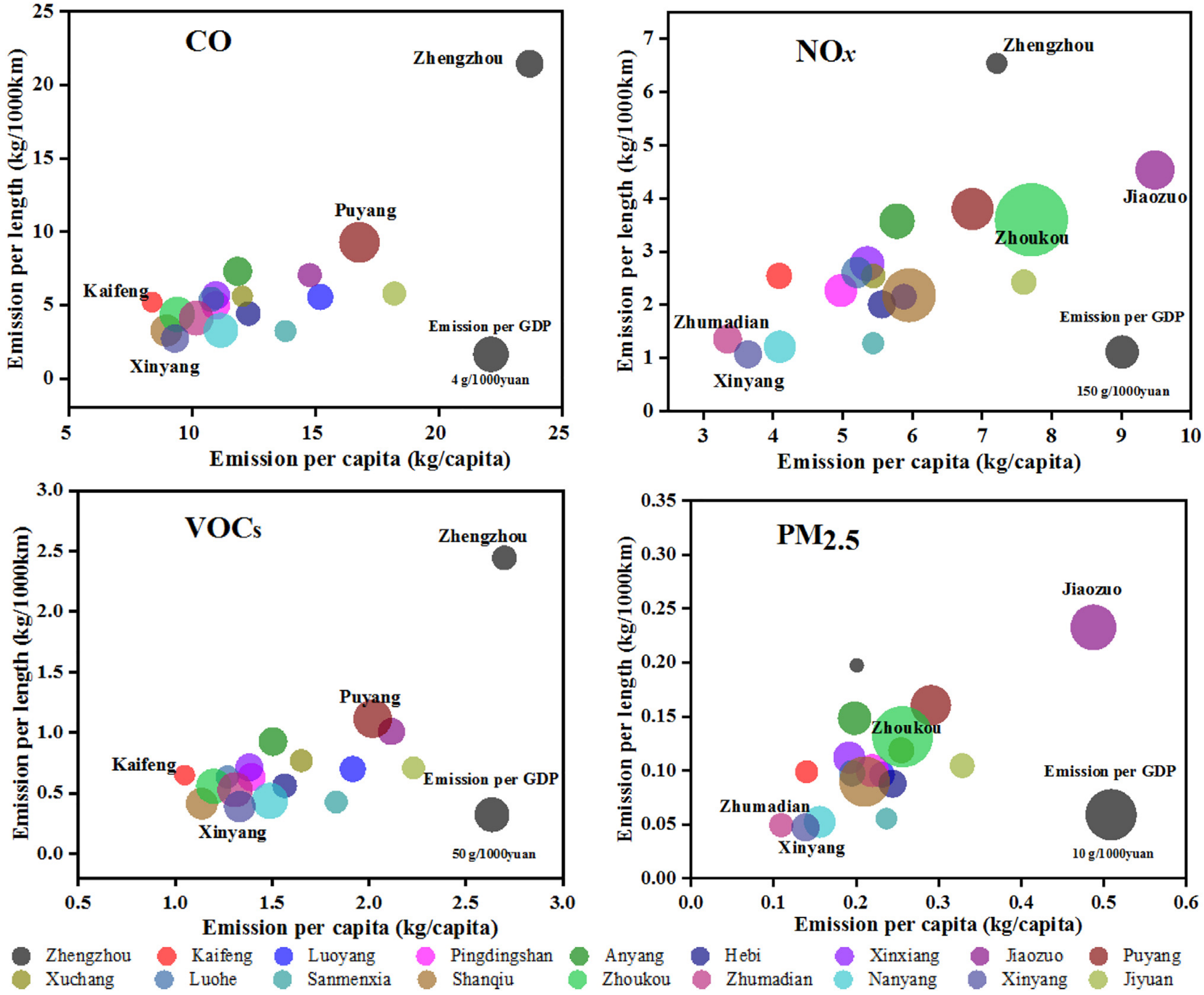

Fig. 5 – Emission intensity, emission per capita and emission per GDP of typical pollutants for each city in Henan Province. GDP: gross domestic product.

influenced by the large EFs and vehicle populations with low emission standards. Vehicles after China 3 largely dominated the NO_x and PM emissions, with contributions between 56.0% and 82.0%. The main reason for this was that among the HDDT, 87.2% of the vehicles were from China 3 to China 5, therefore, in order to reduce the vehicle emissions, it would be necessary to improve the emission standards for passenger cars and hasten the elimination of yellow-label vehicles to a certain extent.

2.2.3. Contribution by city

The municipal emission characteristics among different cities should be taken into consideration in order to receive the benefits of providing information to local policy-makers on future vehicle management. Fig. 4 illustrates the emission contributions of vehicle categories along with corresponding emission intensity (g/km) for each city in 2015. The vehicle fleet compositions in terms of the provincial emission of each pollutant are also provided. Overall, HDT and LDT were the dominant contributors to NO_x and PM emissions, whereas LDV and MC were the dominant contributors to CO and VOCs. Zhengzhou, Zhoukou, Nanyang, Luoyang, Shangqiu, and Xinyang were the top six cities with high emissions in Henan Province (Appendix A Fig. S2), accounting for more than 50% of the total emissions. Conversely, Kaifeng, Hebi, Luohe, and Jiyuan were the four cities with the lowest emissions, accounting for less than 10% for those same pollutants.

Zhengzhou, a city with rapidly developing transportation, housed the largest populations of LDV and LDT, accounting for 38.2% and 13.3% of Henan Province, respectively. The emissions produced in Zhengzhou accounted for 16.1%, 12.9%, 19.0%, 9.5%, 9.6%, 17.1%, 18.4%, and 22.6% of the total SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-Evap, and NH_3, respectively. Nanyang, with the largest proportion of motorcycles, contributed 9.4%, 9.9%, 13.8%, and 9.7% of CO, VOCs, VOCs-Evap, and NH_3 emissions, respectively, and was only lower than Zhengzhou. Zhoukou had the largest HDT population,

Table 3 – Vehicle emissions in different regions or cities.

Area		Base year	Emission (Gg)							Reference
			SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOCs	NH_3	
Province	Henan	2015	9.1	533.4	1190.7	23.7	21.6	150.8	10.4	This study
	Henan[b]	2002		168	1271	56		218(NMVOC)		Song and Xie (2006)
		2003	47.3	3450	8513	1533				Zhao et al. (2012)
		2006		385	2100	29	27	410	1.3	Tang et al. (2016)
		2010		445	2300	28	27	415	4	Tang et al. (2016)
	Henan[a]	2010	4.6	209.7	379.3	9.5	6	68.2	2.9	Qiu et al. (2014)
		2011		500	1840			225(NMVOC)		Lang et al. (2014)
	Henan[a]	2012	16	544	987.1	26.5	24.4	168.7(NMVOC)	6.8	Liu et al. (2018b)
		2013		694	1081	43	38	205(HC)		Gong (2017)
		2015		493.3	2278	50.3		276.8(HC)		CVEMAR (2016)
	Hebei	2012		118	102			21.2		Liu et al. (2018a)
	Shandong	2014		513	1723	29.5		234(NMVOC)		Sun et al. (2016)
	Jiangsu	2015	1.9	490	1614.8		16.9	166.3(HC)		Li et al. (2018b)
	Guangdong	2012	6.4	405.7	2470.3	32.6	31	373.6	12.3	Zhong et al. (2018)
	Sichuan[b]	2015	8.5	442.4	1099.1	28.9	26.6	205.9	11.6	Zhou et al. (2018)
Region	BTH[b]	2013	29.5	775.5	1853.8	47.6	46.3	176.2	4	Qi et al. (2017)
	PRD	2010						433		Yin et al. (2015)
	YRD	2010	34.3	691.2		40.2	38.6	515.9	13.2	Fu et al. (2013)
City	Zhengzhou	2015	1.5	69.0	226.5	2.3	2.1	25.8	2.3	This study
		2013	7	106	291	6(PM)		35(HC)		Gong et al. (2017)
	Beijing	2013		105.4	452.1	4.1		21.3(HC)		Jing et al. (2016)
	Tianjin[b]	2013		390			2	25		Qi et al. (2017)
	Shanghai	2012		231	1350		13.5	165		Huo et al. (2014)
	Foshan	2014		2.3	131		1.8	44.6		Liu et al. (2018c)

BTH: Beijing-Tianjin-Hebei; PRD: Pearl River Delta; YRD: Yangtze River Delta; NMVOC: non-methane volatile organic compounds; HC: hydrocarbon.

[a] The study domain only including nine cities of Henan (Xinxiang, Jiyuan, Jiaozuo, Luoyang, Zhengzhou, Kaifeng, Pingdingshan, Xuchang, Luohe) in Liu et al., 2018b and Qiu et al., 2014.

[b] The results in these papers include both on-road and non-road mobile source.

contributing 11.7%, 12.7%, and 11.5% of SO_2, NO_x, and PM, and was also the largest contributor to PM emissions. However, emission intensities were not consistent for each city in Henan Province, with the emission intensities of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-Evap, and NH_3 being 0.05, 2.7, 6.0, 0.1, 0.1, 0.8, 0.2, and 0.05 g/km, respectively. This indicated that higher emissions in the city did not necessarily mean higher emission intensity, except for Zhengzhou. For example, Xinyang and Nanyang had higher emissions, while their emission intensities were lower, which were most likely due to their longer road network lengths.

In order to better understand the emission characteristics at city level, parameters such as road network length, GDP, and permanent resident population were selected to investigate their impacts (HNBS, 2016). The general information and vehicle emission inventories of each city in Henan Province are shown in Appendix A Table S3, and Fig. 5 illustrates the relationships among emission intensities, emissions per capita and emissions per GDP for CO, NO_x, VOCs and $PM_{2.5}$. The emission intensities for each city were found to be inconsistent, depending on the different levels of development and population. For example, a city with a developed system for transportation, such as Zhengzhou, had the highest emission intensities for CO, NO_x, and VOCs. By contrast, cities such as Xinyang and Kaifeng had low emission intensities. Zhumadian and Kaifeng had the lowest emissions per capita, whereas both the emission intensities and emissions per capita for Jiaozuo were higher for $PM_{2.5}$ emissions, which is reasonable since Jiaozuo has a large population of heavy-duty vehicles per capita. Additionally, Zhengzhou and Kaifeng had the lowest emissions per GDP in Henan Province.

2.2.4. Comparison with other inventories

The vehicular emissions of Henan Province from previous studies and vecc-mep (vecc-mep, The Vehicle Emission Control Center of MEP) carried out over the past years, other typical provinces (Hebei, Shandong, Sichuan, Jiangsu, and Guangdong), three major economic zones (YRD, PRD, and BTH) and several cities were summarized and compared (Table 3). The vehicle emission factors of the corresponding studies were obtained to the greatest extent possible, and are listed in Appendix A Table S4.

Compared with the data from vecc-mep in 2016, it was found that the estimation results of NO_x and PM were similar, while the estimation results of CO and VOCs in this study were lower than those in vecc-mep, because small- and medium-sized gasoline engines with non-road mobile sources were considered more in vecc-mep (CVEMAR, 2016). For the previous research performed in Henan province, the discrepancies with the estimated results were mainly due to the differences in the base years, the selection of activity

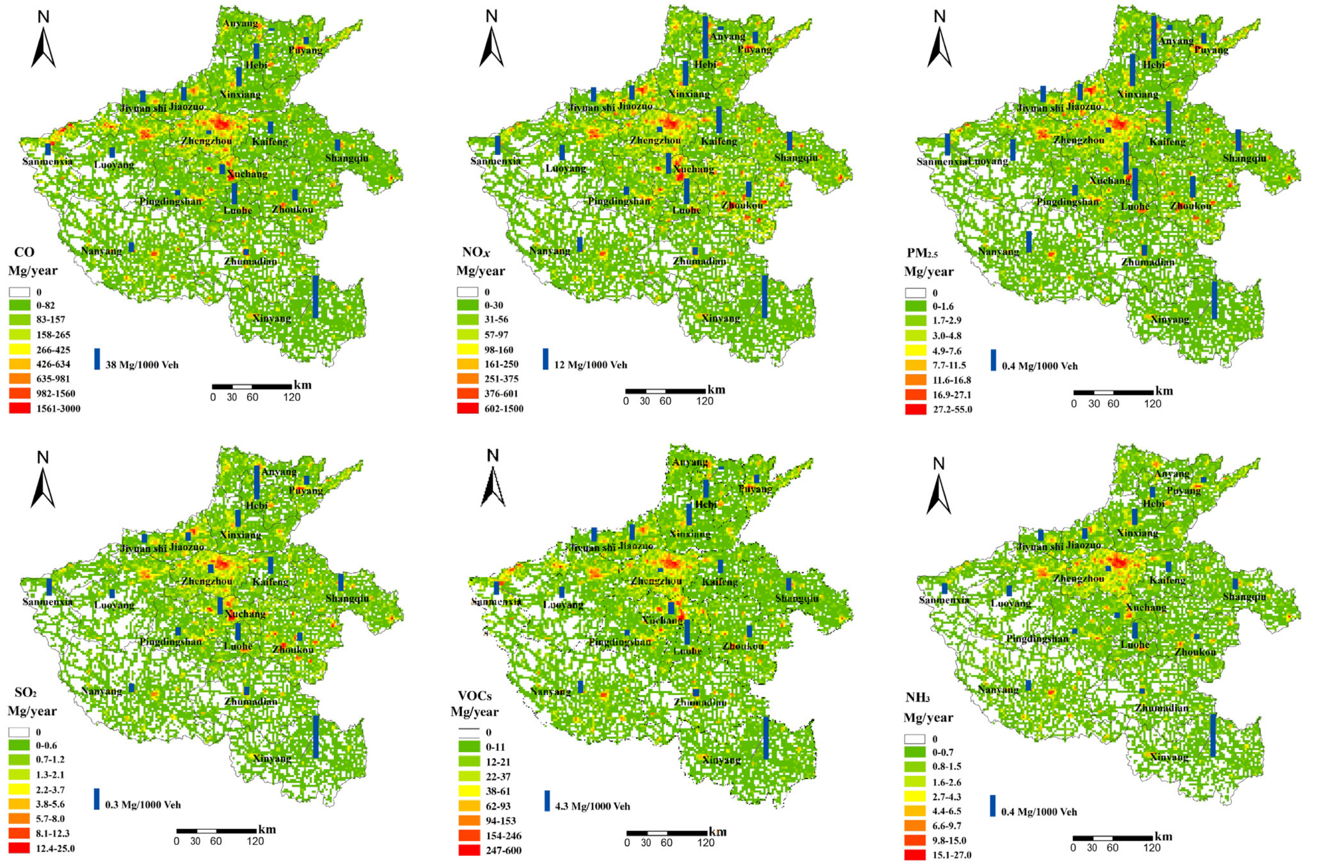

Fig. 6 – Gridded spatial distribution (3 km × 3 km) of pollutants and the emissions per 1000 vehicles (Mg/1000 Veh) by each city in 2015.

levels, and the methods in handling of vehicle populations. First, the surge in vehicle population in recent years has caused previous studies to fail in supporting the latest pollution characteristics. Second, Tang et al. (2016) had based the selection of activity levels on provincial-level activity level estimates for the vehicle pollution emission, and Liu et al. (2018b) and Qiu et al. (2014) adopted activity data by city-level, but only nine cities had been included in Henan province. Third, for population estimation, Song and Xie (2006) did not allocate the base year population to each of the emission standards, while the types of vehicles in the literature (Lang et al., 2014) were not specifically divided by fuel; the approach was too crude and resulted in a greater uncertainty than that by the refined method. Additionally, NH_3 emissions increased due to the introduction of three-way catalytic converters (TWCs) involving the chemistry of selective catalytic reduction (SCR) that controls CO, VOCs, and NO_x emissions (Chang, 2014; Zheng et al., 2012).

Compared with other provinces, such as Shandong Province with a vehicle population of 5.5 million, which is higher than that of Henan, the emission levels were higher in Shandong than in Henan Province except for NO_x (Sun et al., 2016). For this reason, the China 3 emission standards for HDT were implemented in Shandong in 2008, two years before Henan Province imposed them. Meanwhile, the emission factors of NO_x reported by Sun et al. (2016) were all below those in this study. As for the study of Guangdong Province (Zhong et al., 2018), the difference was largely related to the method by which the activity data were collected. Vehicular hourly flows were adopted in the former, whereas statistical populations were used in this study. Moreover, for the LDV of China 3, the VOC emission factor reported by Zhong et al. (2018) was 3.8 times that of this study, which was the reason why the estimation of VOCs was higher than that in this study. The estimation results of mobile sources of Sichuan reported by Zhou et al. (2018) included road mobile sources and non-road mobile sources. By comparing the data of vehicle population and emission factors (NBS, 2016), it was reasonable that the estimation was lower than that in this study.

From the urban point of view, Zhengzhou was listed in order to compare with other cities. Although the vehicle numbers kept increasing, the emission decreased from 2013 to 2015, most likely due to the China 4 emission standard being implemented for most fleet types since July of 2013. Strict vehicle emission standards in Beijing (Jing et al., 2016), Tianjin (Qi et al., 2017), and Shanghai (Huo et al., 2014) were implemented early, leading the emission factors to be lower than in Zhengzhou. But the emissions of vehicles were higher than in Zhengzhou due to heavy traffic with high vehicle population in these developed areas. Overall, the estimation results could certify the validity of this study to some extent.

2.3. Characteristics of the temporal and spatial distribution

2.3.1. Spatial distribution

A 3 km × 3 km gridded spatial distribution of vehicle emissions based on the updated and optimized methods in Section 1.3 was established. As indicated in Fig. 6, the spatial distribution had the following characteristics: (1) there was a significant difference in emissions among cities; (2) the peak emission areas were concentrated on the central part in each city due to the common characteristics of dense traffic network, heavy traffic flow and high emission intensities in urban areas; (3) the spatial distribution of emissions was linear, consistent with the combination of several road type allocations, especially in the cities with low emissions which are significantly affected by the traffic flows of highways and national roads, and the emission characteristics of line sources are more obvious.

From the perspective of pollutants, the spatial distribution also showed discrepancies due to the different contributions to each pollutant by fleet type. For instance, the CO and VOC emissions were mainly concentrated on light-duty vehicles, thus, concentrating higher emissions in the city centers, ring roads and highways. At the same time, vehicular NO_x and PM emissions were mainly from heavy-duty vehicles, with higher emissions concentrated in ring roads and highways. These discrepancies resulted in emissions of CO and VOCs in the central part of city, where a large number of light-duty vehicles are driven, being significantly higher than those of NO_x and PM, while NO_x and PM emissions in the ring roads and highways, where there are higher traffic flows of heavy-duty vehicles, were significantly higher than CO and VOCs. Overall in Zhengzhou, the chief transportation hub, vehicle emissions are at high values, while the surrounding cities like Jiaozuo, Xuchang, and Luoyang, have emissions that were not low for all pollutants.

2.3.2. Temporal variation

The average monthly traffic volume as a temporal surrogate was analyzed using the data of daily average traffic flow for 12 months among 437 toll stations on highways provided by the Henan Provincial Department of transportation (HNDT), and is shown in Fig. 7a. The hourly traffic flow data for heavy and light vehicles for six arterial roads, six collector roads, and six branch roads during a full week in the downtown area of Zhengzhou were obtained from the Environmental Protection Monitoring Center Station of Zhengzhou (EPMZZ), with the weekly and daily variations displayed in Fig. 7b and c, respectively.

Overall, the characteristics of monthly variation were more gradual than weekly and daily ones. Passenger traffic and freight are two prominent factors affecting the traffic flows on highways. Freight was greatly affected by the weather, thus resulting in reduced traffic flows for winter with bad weather. Passenger traffic was closely related to personal travel, in particular the amounts of outings increased during the holidays and tourist seasons. Combining the above factors, the traffic flows in February, August and September were higher than the other months, with the lowest traffic flow observed in November.

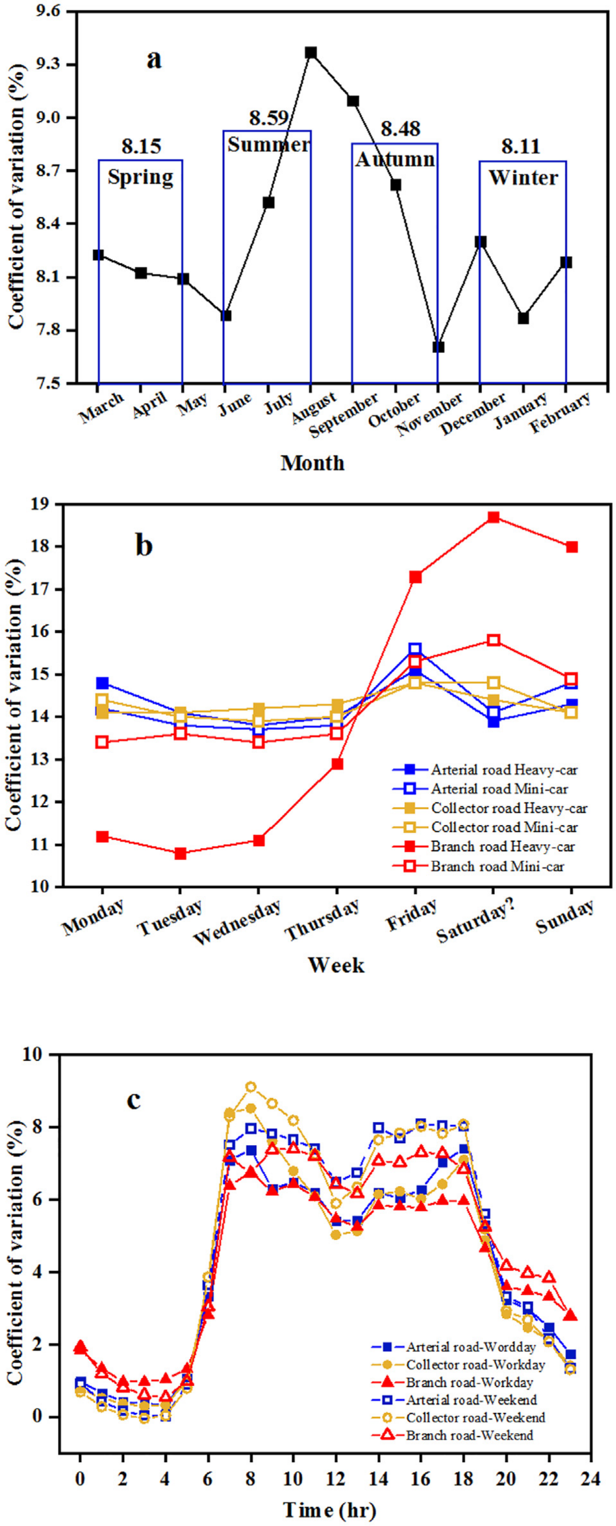

Fig. 7 – (a) Monthly variations of traffic flow for highway, (b) weekly and (c) diurnal variations of traffic flow for urban roads.

As depicted in Fig. 7b, the variation tendencies of arterial roads and collector roads were less than for branch roads. The traffic flow of heavy vehicles fluctuated more than that of

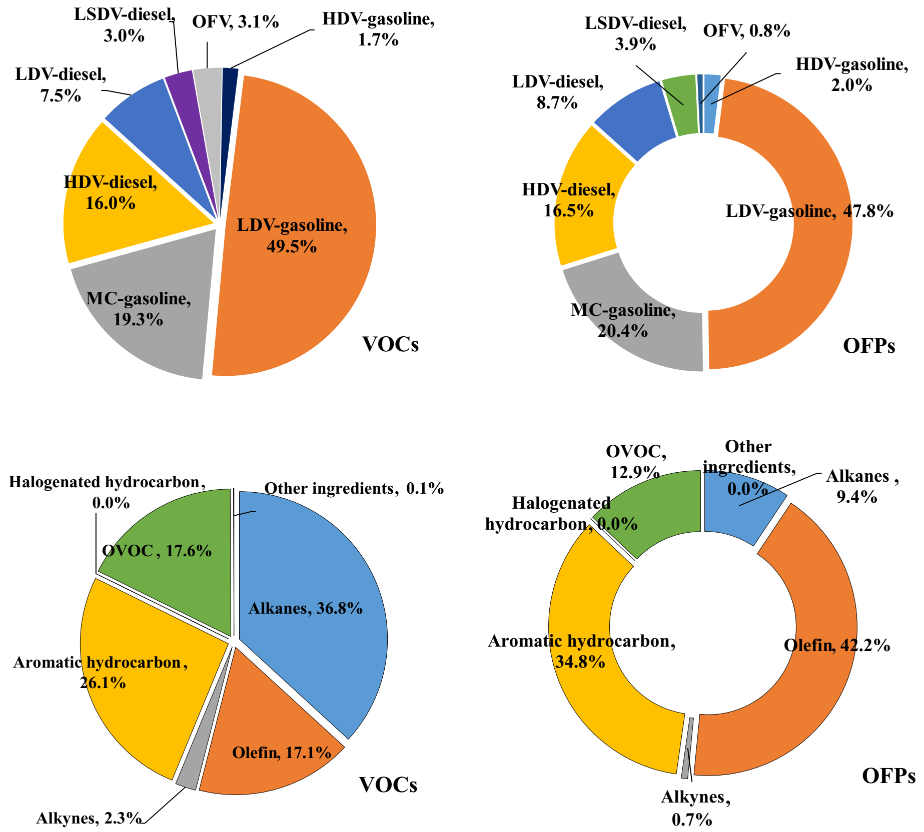

Fig. 8 – Contributions of vehicle types and major speciation to VOCs emission and OFPs (ozone formation potentials). OVOC: other volatile organic compound; LSDV-diesel: low-speed duty vehicle diesel; OFV: other fuel vehicles.

light vehicles because of the greater unpredictability and uncertainty associated with branch roads.

As for daily variation, the tendencies of arterial roads, collector roads, and branch roads were consistent. There appeared to be two obvious peaks in traffic flows, with a morning peak from 8:00 to 10:00 and an evening peak from 17:00 to 19:00, and all lines displayed similar troughs around mealtime (13:00 for lunch). During weekends, the morning peak period appeared 2 hr later while the later peak period appeared 1 hr earlier than weekdays for collector roads. Moreover, the variability around the traffic flow peaks on weekdays was significantly lower than that on weekends, which is consistent with the result shown in Fig. 7b.

2.4. Uncertainties of the emission inventory

The uncertainties in emissions estimation from the activity data and EFs were taken into consideration. For example, the proportions of fleet type distribution in some cities were calculated according to Henan province data surrogates because of incomplete statistics at the city level. The population of each fleet type per emission standard was estimated based on newly registered vehicles and the implementation year for each stage of emission standards, and not all vehicles met the ideal conditions. The fuel ratios for each vehicle type were adopted to separate the vehicle population, whereas the actual fuel vehicle populations were not directly available. In addition, EFs were untested for the local traffic environment, and were modified based on the EFs tested by Tsinghua University research group in this study, whereas VKT numbers were obtained from investigations using a large number of samples.

For spatial distribution, the total emissions based on the provincial-level were allocated to each county by using increasing amounts of secondary industries as an alternative value in the literature (Song and Xie, 2006; Tang et al., 2016) allocated provincial emissions to cities using secondary and tertiary industry data, with passenger and freight turnover used to estimate traffic density. The above method of spatial distribution is bound to increase the uncertainty of spatial distribution. However, in this study, the spatial distributions of emission were calculated based on the city-level data, which was reasonable on the whole compared with previous studies.

Overall, the emissions of vehicles were estimated utilizing the best available data and methods provided in this study. In the future, more perfect methods and data will likely be applied in updating the vehicle emission inventory. Additionally, it is anticipated that through cooperation with governmental departments, future emission inventory results will be more accurate and complete.

2.5. Speciated VOC emissions and the ozone formation potential

In order to better understand the results as shown in Fig. 8, vehicles were reclassified as heavy-duty vehicle gasoline

(HDV-gasoline, including HDGV, MDGV, GB, and MGGT), light-duty vehicle gasoline (LDV-gasoline, including LDGT, LDGV, MGT, MGV, and GT), motorcycle gasoline (MC-gasoline, including NGMC and LGMC), heavy-duty truck diesel (HDT-diesel, including HDDV, HDDT, MDDV, and MDDT), light-duty truck diesel (LDT-diesel, including LDDT and MDT), low-speed truck diesel (LST-diesel, including LSDT, DB, and DT), and other fuel vehicles (OFV, including OFT, OFB, OFHDV, and OFMDV).

Total VOCs and OFP were 150.8 and 569.6 Gg, respectively. The contribution characteristics by fleet type to VOCs and OFPs are shown in Fig. 8. The LDV-gasoline, MC-gasoline, and HDV-diesel were three significant fleet types for vehicular VOC emissions, representing 49.5%, 19.3%, and 16.0%, respectively. Equally important, these constituted 47.8%, 20.4%, and 16.5% of total OFPs, respectively, whereas HDV-gasoline, LDV-diesel, LSDV-diesel, and OFV made up 15.3% of VOCs and OFPs in total. These results indicate that realistically controlling gasoline vehicles for LDV, MC, and diesel vehicles for HDV, while actively promoting other fuel vehicles, may reduce their contribution to VOCs and OFPs. In addition, the conversion of MC-gasoline to electric-powered motorcycles may also reduce contributions for VOCs and OFPs effectively. For species characteristics, as indicated in Fig. 8, the speciation with highest emissions did not necessarily have high OFPs, which is related to the values of MIR for each speciation and therefore, 36.8% of vehicular VOCs were alkanes and accounted for only 9.4% of the OFPs. At the same time, the highest contribution to OFPs was olefins with a proportion of 42.2%, while making up 17.1% of VOC emissions. Aromatic hydrocarbons and OVOC accounted for 26.1% and 17.6% of VOCs, contributing 34.8% and 12.9% to OFPs, respectively.

The top 10 contributing species of VOC and OFP in the different fleet types are shown in Appendix A Tables S5 and S6. For LDV-gasoline, aromatic hydrocarbon species were in the top 10 VOC and OFP species for 4 and 7 places, while ethylene was the largest contributing species with 20.7% to VOC. Alkane and olefin species were in 5 places in the 10 top VOC and OFP species for MC-gasoline, with isopentane and butylene being largest contributing species with 9.8% and 11.8% in VOCs and OFPs, respectively. For HDV-diesel, OVOC occupied a large number of places, with ethylene being the largest contributing species in VOCs and OFPs with 8.8% and 20.3%, respectively.

In recent years, the local government has issued a series of air pollution control measures to reduce $PM_{2.5}$ emissions (HNDEE, 2014). Nevertheless, the slightly increasing trend of ground-level O_3 concentration has introduced a serious challenge in reducing VOC emissions in this region. Faced with this obstacle, local government has not published announcements regarding a government platform to control VOC emissions. VOCs are important precursors for ozone formation, and OFP is an integration of both emission magnitude and reactivity. There was obvious variation between the emission-based and OFP-based key species, and high-emission-contributing species do not necessarily have the same significance in the contribution to OFPs. Therefore, when it is time for the government to create measures for controlling O_3 pollution, it must consider not only reducing VOCs emissions, but also decreasing VOC species with high incremental reactivity emissions.

3. Conclusions

In this study, we developed a 2015-based vehicle emission inventory with 3 km × 3 km spatial resolution based on a bottom-up methodology from city-level activity data for eight kinds of air pollutants, including SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-Evap, and NH_3, in Henan Province. A total of 24 fleet types were considered, which included 9 diesel vehicles, 11 gasoline vehicles, and 4 other fuel type vehicles. The total pollutant emissions of SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, VOCs, VOCs-Evap, and NH_3 were 9.1, 533.4, 1190.7, 23.7, 21.6, 150.8, 31.5, and 10.4 Gg, respectively. Gasoline vehicle categories LDGV and NGMC were responsible for the largest emissions, with 59.6%, 54.5%, 92.8%, and 86.3% contributions to CO, VOCs, VOCs-Evap, and NH_3, respectively; while diesel vehicles emitted the most SO_2, NO_x, and PM emissions, and HDDT and LDDT were the dominant sources for the SO_2, NO_x, PM_{10}, and $PM_{2.5}$ emissions and were responsible for 53.9%, 67.2%, 65.2%, and 64.8%, respectively. In terms of the contributions per different emissions standards, low-emission vehicles for Pre-China 1, China 1, and China 2 made up the largest contribution to CO, VOCs, and VOCs-Evap at more than 57%. For SO_2, NO_x, and NH_3, vehicles above China 3 contributed 82.0%, 72.4%, and 69.8%, respectively.

Vehicle emissions showed a large amount of variation at the city level. Zhengzhou, Zhoukou, Nanyang, Luoyang, Shangqiu, and Xinyang were the top six cities with the major emissions in Henan Province, and accounted for more than 50% of the total emissions for each pollutant, which was most likely due to the larger vehicle populations present in these cities. Although the spatial distribution of vehicle emissions obviously differed among cities, they all had common characteristics in that high levels of pollutants were concentrated in urban cores and were in line with the road network. The daily, weekly, and monthly temporal variations appeared to correspond with personal work schedules. The weekly variation in urban areas showed an obvious workday and weekend effect, the daily variation showed two significant peaks, and the monthly variation showed larger traffic flows in August and September and lower traffic flows in June and October.

The total estimated results of VOCs and OFPs were 150.8 and 569.6 Gg, respectively. LDV-gasoline for primary vehicles contributed 49.5% and 47.8% for VOCs and OFPs, while the MC-gasoline and HDV-diesel accounted 19.3% and 20.4 for VOCs and OFPs, respectively. Alkanes and aromatic hydrocarbons were the main species for VOCs, contributing 36.8% and 26.1%, respectively. However, olefins accounted for the largest proportion in OFPs at 42.2% due to their high MIR.

Overall, the estimated result in this study, based on activity data at city-level, optimal estimation method of vehicle pollutants, and an updated method of high-resolution spatial distribution, was reasonable. Further, the vehicle emission inventory focused on up-to-date work for more comprehensive and real-time activity levels and higher resolution temporal and spatial distributions in this region. Meanwhile, it is hoped that this emission inventory can provide critical data for understanding local vehicular emission characteristics, and provide support for the development of vehicle management and air pollution control in Henan Province.

Acknowledgments

This work was financially supported by the National Key Research and Development Program of China (Nos. 2017YFC0212401 and 2016YFC0201505).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jes.2019.04.010.

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