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Evaporation process dominates vehicular NMVOC emissions in China with enlarged contribution from 1990 to 2016

Liu Yan[1] ⓘ, Bo Zheng[2,*] ⓘ, Guannan Geng[3] ⓘ, Chaopeng Hong[2], Dan Tong[1] and Qiang Zhang[1]

[1] Ministry of Education Key Laboratory for Earth System Modeling, Department of Earth System Science, Tsinghua University, Beijing, People's Republic of China
[2] Institute of Environment and Ecology, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen, People's Republic of China
[3] State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, People's Republic of China
* Author to whom any correspondence should be addressed.

E-mail: bozheng@sz.tsinghua.edu.cn

Abstract

Non-methane volatile organic compounds (NMVOC) are important precursors of ozone and secondary organic aerosols in PM$_{2.5}$ (particulate matter with aerodynamic diameters smaller than 2.5 μm), both of which cause severe climate, ecosystem, and human health damages. As one of the major anthropogenic sources, onroad vehicles are subject to relatively large errors and uncertainties in the estimation of NMVOC emissions due to complicated methods and parameters involved and a lack of comprehensive evaluation of influencing factors. Here, based on our previous work with necessary improvement, we estimate China's vehicular NMVOC emissions by county and by month during 1990–2016 with a consideration of meteorological influence on the spatial-temporal dynamics of emission factors. Our estimate suggests that vehicular NMVOC emissions in China have peaked around 2008 and then declined up to 2016 with an enlarged contribution of the evaporative process to vehicular NMVOC emissions. Vehicular NMVOC emissions have been dominated by the evaporative process at present. Meteorological factors alter spatial-temporal distributions of NMVOC emissions, especially evaporative emissions, which are enhanced in South China and in summer. Emissions and ozone formation potential of the major chemical groups (i.e. Alkenes, Aromatics, and Alkanes) also increase substantially due to meteorological influences. Our analysis suggests that mitigation strategies for vehicle pollutions should be designed based on a sophisticated emission inventory accounting for the meteorological impact on emission factors to correct the potential underestimation of NMVOC emissions, especially those from the evaporative process.

1. Introduction

Driven by the rapid growth of economy and population, China, whose vehicle ownership reaches 185.7 million in 2016, has become the second-largest vehicle market worldwide (National Bureau of Statistics, IRF 2019). Although promotion of new energy vehicles has been strengthened, the majority of existing vehicles in China are still gasoline and diesel oil vehicles nowadays, which will exhaust air pollutants like non-methane volatile organic compounds (NMVOC) while running, starting, and parking (Liu *et al* 2017, Liang *et al* 2019). Studies on PM$_{2.5}$ (particulate matter with aerodynamic diameters smaller than 2.5 μm) pollution suggested that NMVOC contributed to the formation of secondary organic aerosols which made up a significant proportion of ambient PM$_{2.5}$ mass in Chinese cities (Huang *et al* 2014, Cui *et al* 2015). Recent studies also confirmed that ozone production in China's cities follows

the VOC-limited regime (Geng *et al* 2008, Shao *et al* 2009). Both $PM_{2.5}$ and ozone pollution could cause severe climate, ecosystem, and human health damages (Avnery *et al* 2011, Butler and Huybers 2013, Furlong and Klimentidis 2020, Wang *et al* 2020, Kim *et al* 2021).

NMVOC emissions from road transport in China have been estimated in various emission inventories. Due to the lack of vehicle statistics at county or city level, previous studies usually estimate vehicular NMVOC emissions in China at province level (Streets *et al* 2003, Cai and Xie 2007, Zhang *et al* 2009, Saikawa *et al* 2011, Tang *et al* 2011, Lang *et al* 2014, Tang *et al* 2016, Wu *et al* 2016, Liu *et al* 2017), in which meteorological conditions (e.g. temperature and relative humidity) that could significantly influence vehicular evaporative NMVOC emissions, were not taken into consideration in emission estimation and therefore may lead to misrepresentation of total vehicular NMVOC emissions in China (Wu *et al* 2017). Accounting for the availability of dynamic traffic flow data, near-real-time vehicle emission inventory was able to be built using a transient vehicle emission model (Zheng 2016). Although this method improved both spatial and temporal resolutions of vehicle emission inventory in China, numerous parameters and mass data required by this method hamper its application in nationwide vehicle emission estimation. Such a high-resolution method is only suitable for cities such as Beijing, Foshan, and Chengdu which have detailed traffic data (Huo *et al* 2009, Wang *et al* 2009, Liu *et al* 2018, Zhou *et al* 2019). Besides, chemically resolved emissions of NMVOC species, as well as ozone formation potential (OFP), are also urgently needed for tailored air pollution control measures in China (Li *et al* 2019a). Previous efforts have been made to estimate speciated NMVOC emissions for China based on chemical source profiles (Zhang *et al* 2009, Li *et al* 2014, 2019b, Wu and Xie 2017). However, speciated emissions and OFPs were rarely calculated in vehicular emissions inventories that consider the effect of meteorological conditions on NMVOC emission factors.

In this work, a high-resolution NMVOC emission inventory of road transport has been developed, for the first time, by county and by month from 1990 to 2016 in China using a county-level emission estimation approach. Ambient meteorological factors are utilized to simulate spatial-temporal dynamics of emission factors. Vehicle activities with technological and age distributions are reconstructed by county on the base of the fleet turnover model. Based on the new inventory, speciated emissions and OFPs from road transport in China from 1990 to 2016 are estimated based on the updated chemical profiles derived from our previous work.

2. Data and methods

2.1. Process-based emission estimation method

To develop a high-resolution NMVOC emission inventory for road transport in China from 1990 to 2016, we estimated vehicle emissions at county level by exploring geographic differences in key parameters as fully as possible. For a given county, annual NMVOC emissions from vehicles registered in that county were calculated as follows:

$$\text{EMIS}_c = \sum_{\text{state}} \sum_v \sum_f \sum_s \text{Stock}T_c \times \text{Stock}S_{v,f,prov}$$
$$\times \text{VKT}_{v,f} \times \text{FE}_{v,f} \times X_{v,f,s,\text{prov}}$$
$$\times \sum_m EF_{v,f,s,\text{state},c,m} \tag{1}$$

where c represents county and prov represents province where county c is located; m represents month; state represents running, start, and evaporative emission process (here running and start emissions are called exhaust emissions); v represents vehicle category, including motorcycles (MCs), four types of passenger vehicles: heavy-duty buses, medium-duty buses, light-duty buses (LDBs), and mini buses; as well as four types of trucks: heavy-duty trucks, medium-duty trucks (MDTs), light-duty trucks, and mini trucks; f represents fuel type (i.e. gasoline and diesel); s represents vehicle emission standard from stage 0 to stage 4 (corresponding to pre-Euro 1, Euro I, Euro II, Euro III, and Euro IV standards); EMIS_c represents annual NMVOC emissions from vehicles in county c; $\text{Stock}T_c$ is total vehicle population in county c; $\text{Stock}S_{prov,v,f}$ is the proportion of vehicle v using fuel f in total population of province prov; $\text{VKT}_{v,f}$ and $\text{FE}_{v,f}$ is national average vehicle mileage traveled and fuel economy for vehicle v using fuel f; $X_{v,f,s,\text{prov}}$ is technology distribution, which represents the proportion of vehicle v using fuel f and meeting the emission standard s in the total vehicle population of province prov; $EF_{v,f,s,\text{state},c}$ is emission factor under process state of vehicle v using fuel f and meeting emission stage s in county c. Please see supporting information for descriptions of $\text{Stock}S_{v,f,\text{prov}}$, $VKT_{v,f}$, and $FE_{v,f}$.

2.2. Reconstruction of county-level vehicle population

For passenger vehicles and trucks, vehicle population was reconstructed by county using the Gompertz function (Dargay and Gately 1999, Dargay *et al* 2007, Huo and Wang 2012, Zheng *et al* 2014). Vehicle ownership was estimated using historical gross domestic product (GDP) and population data:

$$\text{Stock}T = V^* \times e^{\alpha e^{\beta E}} \times \text{Pop} \tag{2}$$

where V^* represents the saturation level of total vehicles per 1000 people, here V^* was assumed to be 500 vehicles/1000 people according to the moderate vehicle growth scenario for China (Huo and Wang 2012); E represents per-capita GDP calculated by county-level GDP and population obtained from national or regional statistical yearbooks; Pop represents population; α and β are two negative parameters that determine the curve shape, which were derived from province- or city-level regressions following the method in Zheng *et al* (2014). The simulated vehicle population of each county from 1990 to 2016 was compared to statistical data (National Bureau of Statistics 2018) at both provincial and national level, simulation shows good agreement with statistics for most provinces with correlation coefficients (R) greater than 0.9 (table S1 available online at stacks.iop.org/ERL/16/124036/mmedia).

For MCs, county-level vehicle population was estimated with a simplified method that allocating MC amount from country totals to county based on population.

2.3. Fleet turnover model and technology distribution

Technology distributions were simulated by the fleet turnover model, built upon vehicle age distribution and the implementation year of each emission standard (Zheng *et al* 2014). The fleet turnover model was built as follows:

$$\text{Stock}_{v,y} = \sum_{i=y-t}^{i=y} \text{Sale}_{v,i} \times \text{Surv}_{v,y-i} \qquad (3)$$

$$\text{Surv}_{v,y-i} = \exp\left(-\left((y-i+b)/t\right)^b\right) \qquad (4)$$

where y represents target year ranging from 1990 to 2016 in this work; i represents vehicle registered year; t and b represent parameters of vehicle survival curve associated with vehicle life and survival curve decline rate, respectively, which were derived from the previous study (Huo and Wang 2012) and adjusted with a successive approximation approach; $\text{Stock}_{v,y}$ is vehicle ownership in year y derived from National Bureau of Statistics; $\text{Sale}_{v,i}$ is the number of newly registered vehicles in year i, of which statistical data was available from 2002 to 2016 and data before 2002 was estimated with the back-calculation method developed by Zheng *et al* (2014); $\text{Surv}_{v,y-i}$ is the survival rate of vehicles at age $(y-i)$ in year y.

Based on this fleet turnover model, the number of vehicles registered in each year that could survive in the target year could be modeled and the age distribution could then be simulated. With the implementation year of each emission standard, emission standard that vehicles registered in each year belong to could be identified and the technology distribution was finally determined. As county- or city-level number of newly registered vehicles are not available in

China, the fleet turnover model could only be built at province level. Besides, China's vehicle emission standards are mostly implemented on a provincial basis, so counties within a province share same standard upgrade process. Therefore, we simulated vehicle technology distributions by province and assumed that all counties in one province had the same technology distribution.

2.4. Spatial-temporal dynamic emission factors

Vehicle NMVOC emissions are influenced by meteorological factors such as temperature, humidity, and altitude (Bishop *et al* 2001, Weilenmann *et al* 2009). To account for influences from meteorological conditions, NMVOC emission factors of running, start, and evaporative processes in this work are estimated by county and by month with the following equation (Davis *et al* 2005, Zheng *et al* 2014)

$$\begin{aligned} \text{EF}_{\text{state},v,f,s,\text{vkts}} &= \text{BEF}_{\text{state},v,f,s} \times D_{v,f,s,\text{vkts}} \times \text{Temp}_{\text{state},v,s} \\ &\quad \times \text{RH}_{\text{state},v,s} \times \text{Alt}_{\text{state},v,s}. \end{aligned} \qquad (5)$$

where vkts represents the accumulative mileage groups which are categorized into <80 000, 80 000–160 000, and >160 000 km for passenger vehicles and trucks, <25 000, 25 000–50 000, and >50 000 km for MCs; $\text{EF}_{\text{state},v,s}$ represents NMVOC emission factors; $\text{BEF}_{\text{state},v,f,s}$ is the base emission factor of newly registered vehicles built into the IVE model (International Vehicle Emission model, developed by the International Sustainable Systems Research Center) and constrained by real-world emission factors in China using the method from Zheng *et al* (2014); $D_{v,f,s,\text{vkts}}$ is the deterioration factor used to reflect emission deterioration as the mileage increases, which was derived from the international vehicle emissions (IVE) model. Typically, the base emission factor of newly registered vehicles would decrease with the upgrade of emission standards, while NMVOC emission factors of in-use vehicles would increase as vehicles age (figure S1). $\text{Temp}_{\text{state},v,s}$, $\text{RH}_{\text{state},v,s}$, and $\text{Alt}_{\text{state},v,s}$ are correction factors that reflect the influence of temperature, humidity, and altitude on vehicle emissions, which were generated using the Motor Vehicle Emission Simulator (MOVES) model (developed by the US Environmental Protection Agency). Parameterizations of corrections for temperature, humidity, and altitude in MOVES model please refer to tables S2–S4. To generate meteorological and geographical correction factors, county-level monthly mean temperature and humidity were obtained from the NCEP FNL (Final) Operational Global Analysis (FNL) and Climate Forecast System Reanalysis, both of which are developed by the US National Center for Atmospheric Research. Altitude data were obtained from the Moderate Resolution Imaging Spectroradiometer land use map (Schneider *et al* 2009).

Using the process-based emission estimation method, a high-resolution NMVOC emission inventory of road transport in China was built based on the simulation of county-level vehicle population and emission factors, which are two key parameters in emission estimation. As other parameters have been simulated as high resolution as possible under existing conditions, their influences on the resolution of emission inventory are limited even they are not simulated at county level.

2.5. NMVOC speciation and OFP

Based on the method of Li *et al* (2014), emissions of individual NMVOC species were developed using an integrated source profile database for China (Li *et al* 2019b). OFP, a widely used indicator of ozone production potential (Song *et al* 2007, Zheng *et al* 2009), was calculated based on maximum incremental reactivity (MIR):

$$\text{OFP}_{f,i,c} = \text{EVOC}_{f,c} \times X_{f,i} \times \text{MIR}_i \qquad (6)$$

where *f*, *i*, and *c* represent vehicle fuel type, chemical species, and county, respectively. OFP is ozone formation potential; EVOC is the total NMVOC emissions; *X* represents the mass fraction for species *i* emitted from the vehicle using fuel *f*, which was derived from the composite profiles from Li *et al* (2019b); MIR is the maximum incremental reactivity factor for species *i* to scale its ozone production potential (Carter 1994, 2018).

3. Results

3.1. Trends in NMVOC emissions from 1990 to 2016

NMVOC emissions of road transport in China are estimated to have increased from 1.8 million tonnes in 1990 to 8.4 million tonnes in 2008 and then decline to 7.1 million tonnes in 2016, as shown in figure 1. The persistent growth during 1990–2000 is due to China's rapid increase in vehicle ownership (figure S2) and the absence of effective control measures. During this period, around 70% of total vehicular NMVOC emissions come from the exhaust process as NMVOC emission factors of the exhaust process are much higher than that of the evaporative process for pre-Euro 1 vehicles. For example, NMVOC emissions from the evaporative process are only 1%–11% of the exhaust emissions for pre-Euro 1 LDBs using gasoline in China (figure S2). Since the first vehicle emission standard was implemented in 2000, the continuously updated vehicle emission standards and retirement of old vehicles (figure S4) offset the emission increase driven by the growth of vehicle ownership and finally lead to the decline in NMVOC emissions of road transport in China after 2008. As vehicle emission standards in China

mainly limit emission factors from the running process, vehicular NMVOC emission factors during running decrease rapidly with the upgrade of emission standards and almost decrease to the level comparable to evaporative emissions (figure S3). Vehicular NMVOC emissions from the evaporative process have dominated NMVOC emissions of road transport in China since 2005. In 2016, 73% of vehicle NMVOC emissions come from evaporation and the contributions of exhaust emissions drop to 21% and 6%, respectively, for gasoline and diesel oil vehicles.
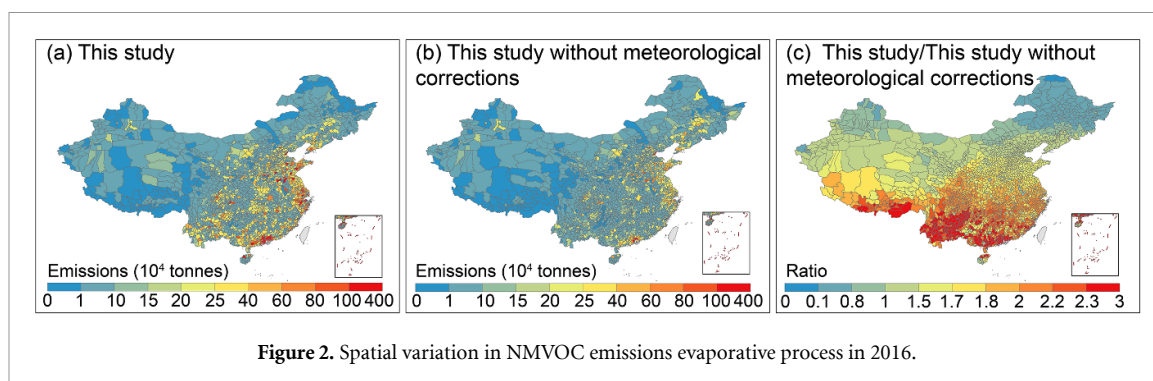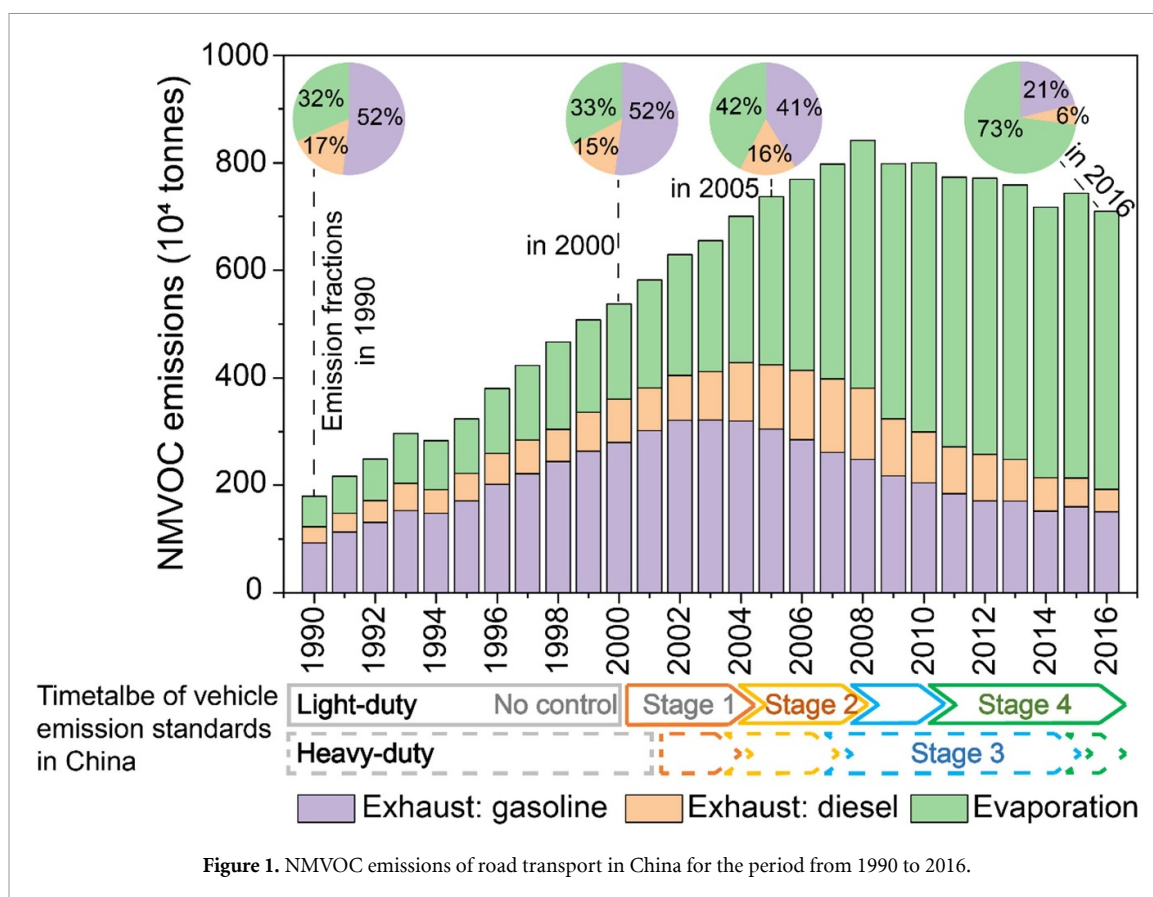
3.2. Spatial-temporal dynamics of NMVOC emissions due to meteorological influences

Meteorological factors reshape the spatial distribution of vehicle NMVOC emissions (figure 2) because evaporative emissions are sensitive to meteorological conditions (figure S5). The spatial pattern of meteorological influence on NMVOC emissions tends to follow the spatial distribution of temperature (figure S6) with high temperatures substantially driving up evaporative emissions. For example, the high temperatures in southeast China can increase emissions three times higher than the estimates based on base emission factors without meteorological corrections. The influence of temperature on NMVOC emissions decreases with ambient temperature declining from south to north in China. In northeast China, NMVOC emissions from the evaporative process are slightly lower than emission estimates based on base emission factors because the weather there is cold. Overall, the spatial variation in meteorological influence on NMVOC emissions suggests that vehicle emission inventories underestimate China's NMVOC emissions if meteorological corrections are not considered.

Monthly variations in vehicular NMVOC emissions are shown by region in figure 3. Vehicular NMVOC emissions are higher in summer than those in winter because evaporative emissions are more enhanced in hot summer than in cold winter (Bishop *et al* 2001, Weilenmann *et al* 2009). Meteorological factors tend to amplify the seasonal variability in NMVOC emissions, especially over southern China where the temperature is relatively high. Besides, vehicular NMVOC emissions in winter would also be higher in southern China due to the warm meteorological conditions in winter. The analysis of monthly emissions also suggests that vehicular NMVOC emissions could be underestimated, especially in summer, if we do not consider the effect of the meteorological influences on emission factors in our inventory.

3.3. Speciated NMVOC emissions and implications for OFPs

Based on the estimation of vehicular NMVOC emissions from 1990 to 2016, we further calculated speciated NMVOC emissions and their OFPs. Figure 4 shows the emissions and OFPs of each chemical group

Figure 1. NMVOC emissions of road transport in China for the period from 1990 to 2016.

Figure 2. Spatial variation in NMVOC emissions evaporative process in 2016.
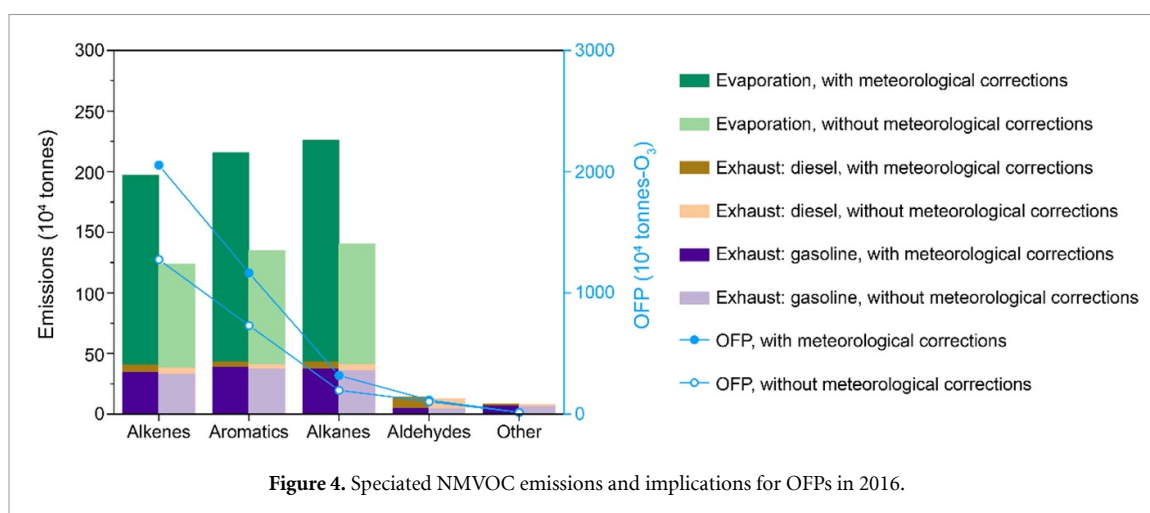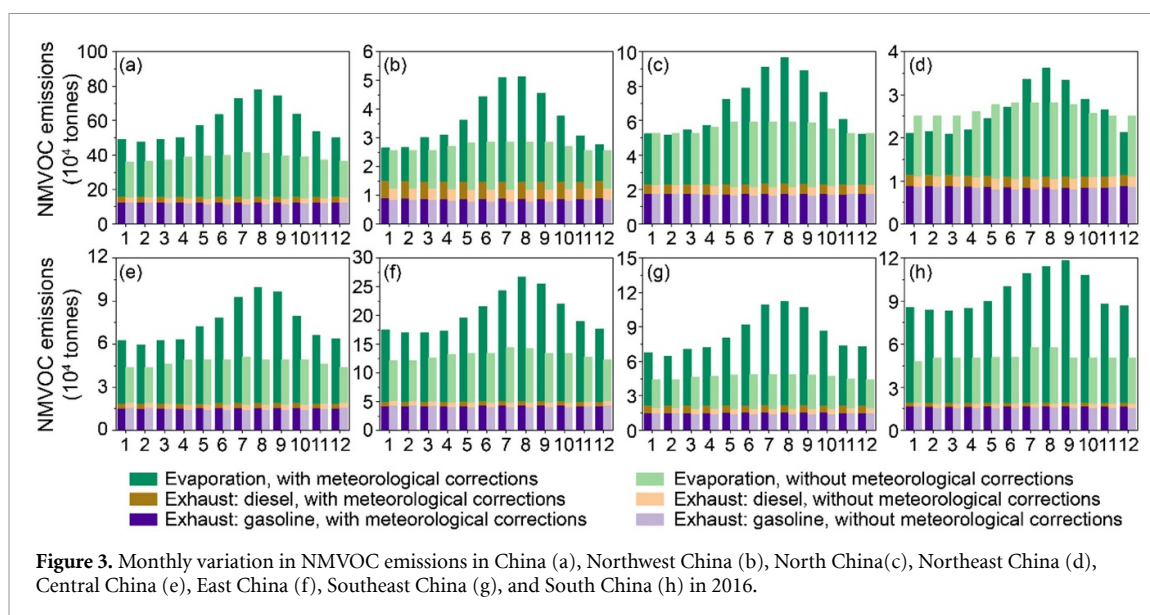
with or without meteorological influences in 2016. Alkenes, Aromatics, and Alkanes are the largest contributors to vehicular NMVOCs emissions and OFPs, and most of them come from the evaporative process. When meteorological influences on NMVOC emissions are considered like in this study, the proportion of evaporative emissions of these three chemical groups could increase up to 80%, and the OFP of these three chemical groups would also increase to around 60% compared to that without meteorological influences.

4. Discussion

Using the method in our previous work with necessary improvement, we develop a county-level monthly emission inventory of NMVOC from road transport in China from 1990 to 2016 considering the meteorological influences for the first time. Our novel emission inventory resolves the long-term historical trends and drivers of NMVOC emissions from road transport in China, better reflecting the meteorological influences on spatial-temporal dynamics of NMVOC emissions, which could benefit both air quality modeling and management.

Here we compared NMVOC emissions estimated in this work to previous studies. Vehicular NMVOC emissions in CEDS (a global emission inventory that combined regional emission inventory in China, latest version 2020), Wu *et al* (2016), and China Mobile Source Environmental Management Annual Report annually published by the Ministry of Ecology and Environment of the People's Republic of China (marked as VECC in figure 5) all peaked around

Figure 3. Monthly variation in NMVOC emissions in China (a), Northwest China (b), North China(c), Northeast China (d), Central China (e), East China (f), Southeast China (g), and South China (h) in 2016.

Figure 4. Speciated NMVOC emissions and implications for OFPs in 2016.

2008 and then started to decline, which are consistent with our work. Emissions in Lang *et al* (2014) also declined during 2008–2016. Continuous growth appeared in China's vehicular NMVOC emissions after 2000 in EDGAR (latest version 5.0), different from this work and other local studies, which may not well capture the emission trend of road transport in China. This persistent growth indicates that the trend of China's vehicular NMVOC emissions in EDGAR was mainly driven by the increase of local vehicle population, while influences of China's upgrading emission standards on vehicular emissions were not well considered. Besides the difference in trend, China's vehicular NMVOC emissions in EDGAR were also lower compared to this work and other local studies. This gap may be due to the underestimation of vehicle emission factors. Differences between EDGAR and local studies indicate that considering more local information and measurement data in the estimation of vehicular emissions in China may help to reduce the gap between global and local inventories.

Although the inventory of NMVOC from road transport in China developed in this study was high-resolution, uncertainties still exist in the emission estimation. First of all, national average vehicle mileage traveled and fuel economy were used due to the limitation of data availability, therefore, differences in driving characteristics across counties were not fully considered. Second, the accuracy of emission factors is another key factor that affects the quality of vehicular emission inventories. Local test data has been used as much as possible in this work to constrain base emission factors built into the IVE model, but there are still many counties where local measurements are not available. Emission factors from the IVE model are developed based on testing results in US or European countries and could not accurately reflect the emission characteristics of vehicles in China (Li *et al* 2017). Based on the discussion above, it is necessary to carry out more surveys and tests at county or city level to provide representative local emission factors and vehicle driving characteristics.
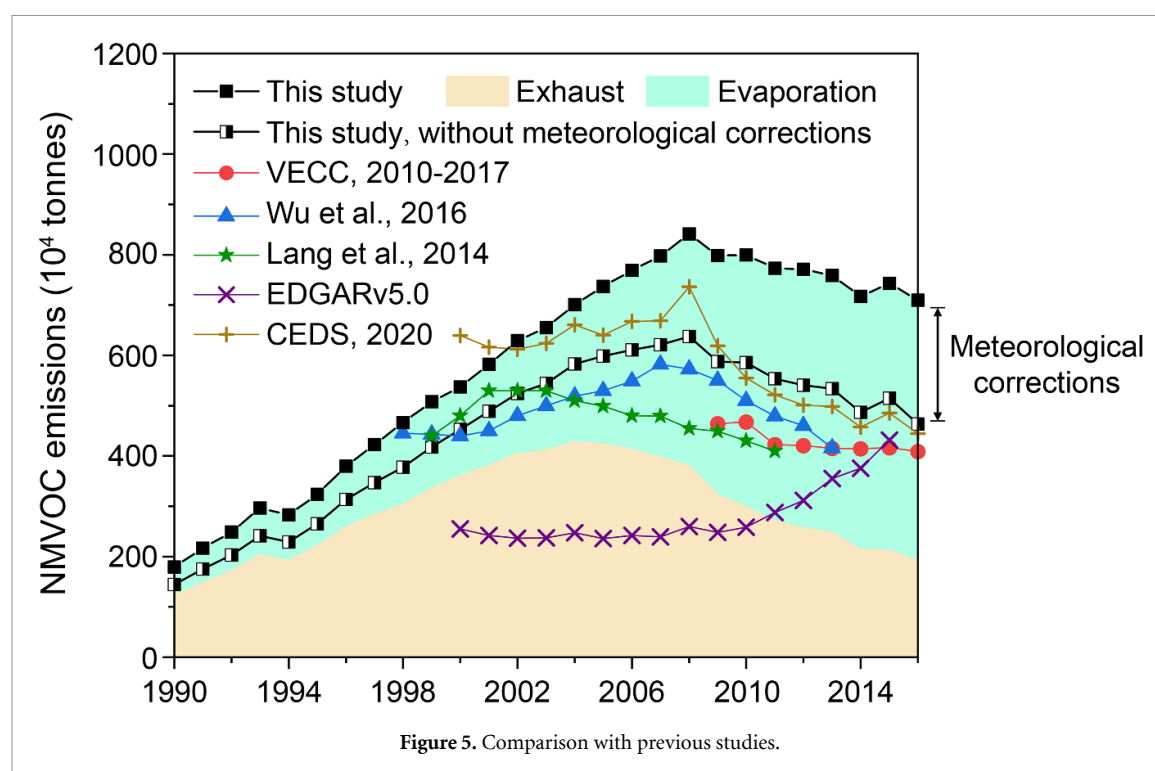
Figure 5. Comparison with previous studies.

Unlike the great achievement gained since 2013 in control of fine PM, ozone pollution in China has become increasingly severe and has not been fully addressed by the government (Gao *et al* 2017, Lu *et al* 2018, Zheng *et al* 2018). Various studies reported that vehicles were the major contributor to anthropogenic emissions of NMVOC in Chinese cities, which were important precursors of ozone formation (Song *et al* 2008, Zheng *et al* 2009, Wang *et al* 2010, Shao *et al* 2011, Cui *et al* 2015). Based on our estimates, the upgrade of emission standards has led to significant emission reductions from the exhaust process since 2004. However, due to the lack of effective control for evaporative emissions, the decrease in total NMVOC emissions from road transport in China was delayed for four years. As NMOVC emission factors from the evaporative process have not effectively been limited like exhaust process in current emission standards, specific limits for the evaporative process need to be considered in the next update of vehicle emission standards. Besides, onboard refueling vapor recovery systems, which are carbon canisters installed in vehicles to capture gasoline vapors evacuated from the gasoline tank and have been used in the US since 2012 to control (Yang *et al* 2015), could also be required to be installed on new vehicles to control evaporative NMVOC emissions.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

ORCID iDs

Liu Yan ⬤ https://orcid.org/0000-0001-9101-5168
Bo Zheng ⬤ https://orcid.org/0000-0001-8344-3445
Guannan Geng ⬤ https://orcid.org/0000-0002-1605-8448

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