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Large methane emissions from natural gas vehicles in Chinese cities

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ABSTRACT

In recent years, cities in developing countries have become more and more reliant on natural gas as a clean energy source for transportation to reduce air pollution. In this study, we used street-level measurement of atmospheric CH_4 and CO_2 concentrations to quantify CH_4 emissions from natural gas vehicles (NGVs) in China. These measurements were made in eight cities (Chengdu, Urumqi, Jinan, Nanjing, Lanzhou, Harbin, Guangzhou) with varying sizes of NGV fleet. A traffic CH_4:CO_2 emission ratio (TER) was determined via linear regression of CH_4 versus CO_2 concentration data obtained from each street transect. The TER value was combined with the ratio of NGVs in the street traffic in a mathematical model to obtain the CH_4 emission factor for NGVs. Results show that the TER increases with increasing NGV ratio and decreases with increasing traffic speed. Overall, the NGV CH_4 emission factor in these cities is $0.022 \pm 0.0033\ kg\ m^{-3}$, about 8 times the Intergovernmental Panel on Climate Change (IPCC) default factor for NGVs and is more than 100% higher than the mean NGV tailpipe emission factor found in the published literature. That the overall emission factor is much larger than the tailpipe emission factor indicates that on-road vehicle gas leakage is a widespread problem. A business-as-usual scenario suggests that NGVs may emit $1.23\ Tg\ CH_4\ yr^{-1}$ in 2030, or about 3% of China's current total anthropogenic emission. Our study suggest that curbing the emissions from this sector should be a high priority for global climate mitigation efforts.

1. Introduction

Natural gas (NG) is a relatively clean burning energy source. Compared with other fossil fuels, burning NG results in lower emissions of carbon dioxide and air pollutants for each unit of heat produced (EIA, 1999). In addition, the vehicular NG price is only 28–57% of the gasoline price, based on the equivalent energy content (Ma et al., 2013). These properties have contributed to increasing use of NG as a clean energy source for cities to improve their atmospheric environment (Ma et al., 2013; Reynolds et al., 2011. Martins et al., 2014; Ong et al., 2011; D'Angiola et al., 2010). As of 2016, the global natural gas vehicle (NGV) population reached 23 million, nearly 20 times as in 2000. China now has the world's largest NGV market. Domestic NGV stock in China increased from 2 thousand in 1996 to 5 million in 2016 (Fig. 1).

Methane (CH_4), the main combustible substance in NG, is a strong greenhouse gas with a global warming potential of 28 over a 100-year period, being responsible for 17% of the anthropogenic radiative forcing (Stocker et al., 2013). Emissions inventory reveals that there is an increase of 196 kg of CH_4 emission per vehicle per year because of conversion to NG as the fuel source (Wadud and Khan, 2013). Despite being less than 1% of the total vehicle population, NGVs were responsible for 23% of vehicular CH_4 emissions to the atmosphere in China in 2010 (He et al., 2014). In 2016, five countries (China, Iran, Pakistan, Argentina and India) comprise more than two-thirds of the global NGV fleet (Fig. 1). Emissions regulation and its enforcement in these emerging economies are much less stringent than in developed countries, raising serious concerns regarding the climate consequences of fuel switching.

In this study, we quantify methane emission from NGV fleet in China, a country with the largest NGV population (Fig. 1). Our specific objectives are: (1) to determine the CH_4 emission factor for on-road NGVs and compare it with the IPCC default emission factor and those found in the published studies, (2) to estimate the contribution of on-road fuel leakage to the overall emission, and (3) to project the total national CH_4 emission from the NGV fleet.

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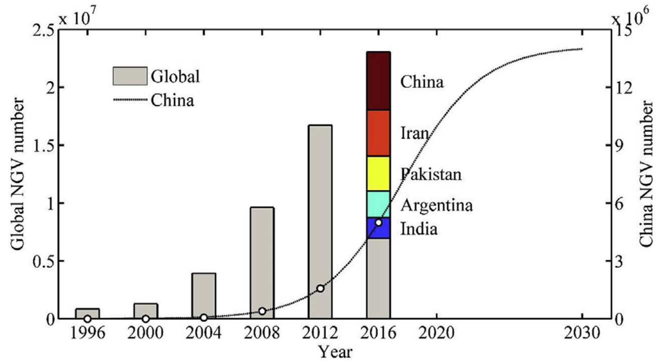

Fig. 1. NGV number as a function of time in China (hollow circle and dotted line) and in the world (bars). The dotted line is a curve ($y = \frac{1.414 \times 10^7}{1 + \exp(-0.370x - 730.5)}$, $P < 0.01$) fitted by the NGV number during 1996–2016.

2. Methods

2.1. On-road measurements

We determined traffic CH_4:CO_2 emission ratios (TER) in seven cities across China (Fig. 2) from atmospheric CH_4 and CO_2 mixing ratios measured with a gas analyzer installed in a car that traversed on urban streets and traffic tunnels. The actual CH_4:CO_2 emission ratio of NGVs was estimated from a regression model of TER against the NGV ratio or the proportion of NGV in the total on-road vehicles during each measurement.

The seven cities chosen for this study are: Chengdu (CD, longitude 104°01′ E, latitude 30°40′ N) in Sichuan Province, Urumqi (UR, 87°39′ E, 43°47′ N) in Xinjiang Province, Jinan (JN, 117°03′ E, 36°36′N) in Shandong Province, Harbin (HB, 126°46′ E, 45°45′ N) in Heilongjiang Province, Guangzhou (GZ, 113°20′ E 23°10′ N) in Guangdong Province, Nanjing (NJ, 118°48′ E 32°00′ N) in Jiangsu Province, and Lanzhou (LZ, 103°53′ E, 36°03′N) in Gansu Province. Three of these cities (CD, UR, JN) are located in provinces with a large NGV population (187–293 thousand), two (HB, GZ) in provinces with a small population (8–9 thousand), and two (NJ, LZ) in provinces with an intermediate population (42–46 thousand; map in Fig. 2). Detailed motor vehicle statistics for these cities are given in Supplementary Table 1. In some cities (HB, GZ, NJ), NGVs are exclusively used as taxis or for public transportation (buses). In other cities (CD, UR, JN, LZ), 3.4–25.9% of privately-owned cars are also NGVs.

According to the data from the Clean Energy Auto Industry Association of China, about 80% of compressed natural gas (CNG) vehicles were originally manufactured to run on gasoline or diesel as the fuel source and have been modified to use both compressed natural gas and gasoline or diesel (Supplementary Fig. S1). (Gasoline or diesel is used only in cases of emergency, when immediate refueling of natural gas is not possible.) Modification of the engine is carried out at small car shops scattered around the city.

NGVs for public transportation (buses) run on liquefied natural gas (LNG). Over 90% of LNG vehicles use engines built at the factory specifically for LNG. Two video cameras recorded the traffic condition during each observation, and service vehicles (taxis and buses) were tallied from the video recordings. These tallies were adjusted using a city-wide mean proportions of NGVs in the taxi and the bus fleet to obtain the number of NGVs in each street transect observation. Of all the NGVs encountered in our street and traffic tunnel observations, about 8% were buses.

The transportation sector account for 1% (Guangzhou) to 21% (Urumqi) of the total natural gas usage in 2015. Another large consumption sector is industry, accounting for 19% (Harbin and Guangzhou) to 85% (Lanzhou) of the usage. The residential and commercial stationary combustion sector comes as the second, accounting for 6.1% (Lanzhou) to 79.5% (Guangzhou). Other sectors are less than 3%. Usage in the residential sector is primarily cooking. An ANOVA analysis for the city of Nanjing, using traffic speed as a continuous variable and cooking hours (6:00–8:00, 10:30–12:30 and 17:00–19:00) versus non-cooking hours as a class variable, reveals that the difference in the bulk emission factor between cooking and non-cooking hours is not statistically significant ($P = 0.40$). Landfill, wastewater, livestock, fuel and biomass burning are important anthropogenic sources. However, landfill sites, livestock farms, wastewater treatment facilities, and biomass burning were far away from the study street and road transects, and by the time their emission plumes reached the city center, the CH_4 concentration should be sufficiently diluted and uncorrelated with the CO_2 emitted by vehicles; these sources were omitted in our analysis. The transportation sector is the dominant CH_4 emitter, accounting for 52.9% (Guangzhou) to 97.8% (Jinan) of the total NG combustion emission in these cities if the IPCC emission factor of $3.8 \times 10^{-3}\,kg\,m^{-3}$ is used for NGVs. If the emission factor of $0.022\,kg\,m^{-3}$ found in the present study is used, this proportion is even higher, increasing to 86.4% (Guangzhou) to 99.6% (Jinan).

Previous studies in some US cities have demonstrated that leakage from aging distribution pipelines is a large source of urban CH_4 emission (Phillips et al., 2013). Such emission sources can be easily identified from street transect measurements because the instrument would always detect a spike in the CH_4 concentration at the same location. Examination of street transect measurements in all the cities we measured reveals no persistent concentration spikes at the same locations except for busy road intersections. An example is given in Supplementary Fig. S3 for all transect data obtained along Hanzhongmen Avenue-Zhongshan East Road in Nanjing, the longest street transect we measured. Repeated high concentrations occurred at about the 4 km location, which is a busy street intersection, but no stationary concentration spikes were observed elsewhere. In other words, there was no evidence of pipeline leakage, possibly owing to the fact that the distribution pipelines in Chinese cities are relatively new. Therefore, mobile sources in the transportation sector are the dominant CH_4 emitter on roads in this study.

The traffic CH_4:CO_2 emission ratio (TER) was determined with ambient CH_4 and CO_2 mixing ratios measured simultaneously with an ultra-portable $CH_4/CO_2/H_2O$ gas analyzer (model UGGA, Los Gatos Research, Mountain View, CA, USA). The analyzer was checked for drift daily against greenhouse calibration gas mixtures (490.6 ppm for CO_2 and 3.05 ppm for CH_4, supplied by the National Institute of Metrology of China with accuracy of 1%) and moist air generated by a dew-point generator (model LI-610, Licor Inc, Lincoln, NE, dew-point temperature accuracy of 0.01 °C). The analyzer signal drifts were, on average, 0.2% for CO_2, 1.3% for CH_4, and 0.54% for H_2O, between two checks.

The analyzer was installed in a passenger car that moved through chosen urban streets and traffic tunnels. Ambient air was drawn from an inlet port above the car roof through a Teflon tube (0.064 cm outer diameter, length 4.0–7.5 m) into the analyzer, at a flow rate of about $500\,mL\,min^{-1}$. The travel time through the sampling tube (10–18 s) was taken into account in the subsequent data analysis. The air inlet was installed at a height of about 2.5 m above the ground. The sampling frequency was 1 Hz.

Measurements in the chosen cities were carried out by two teams from May to July, in 2016. The sampling took place in at least one traffic tunnel, and 1 to 9 arterial streets in each city, all of which pass through the urban center. A car carrying an analyzer traversed these streets between 05:30 and 22:30 local time. In the case of traffic tunnels, we used the data collected while the car was in the tunnel. In the case of open streets, we used data collected while the car was traveling in the designated street transect. The length of the tunnels and street transects is in the range between 0.4 km and 13 km, and the travel time is in the range of 30 s–60 min. One one-way travel through a tunnel or a street transect is counted as one observation.

Two video recorders placed at each end of the traffic tunnel or street

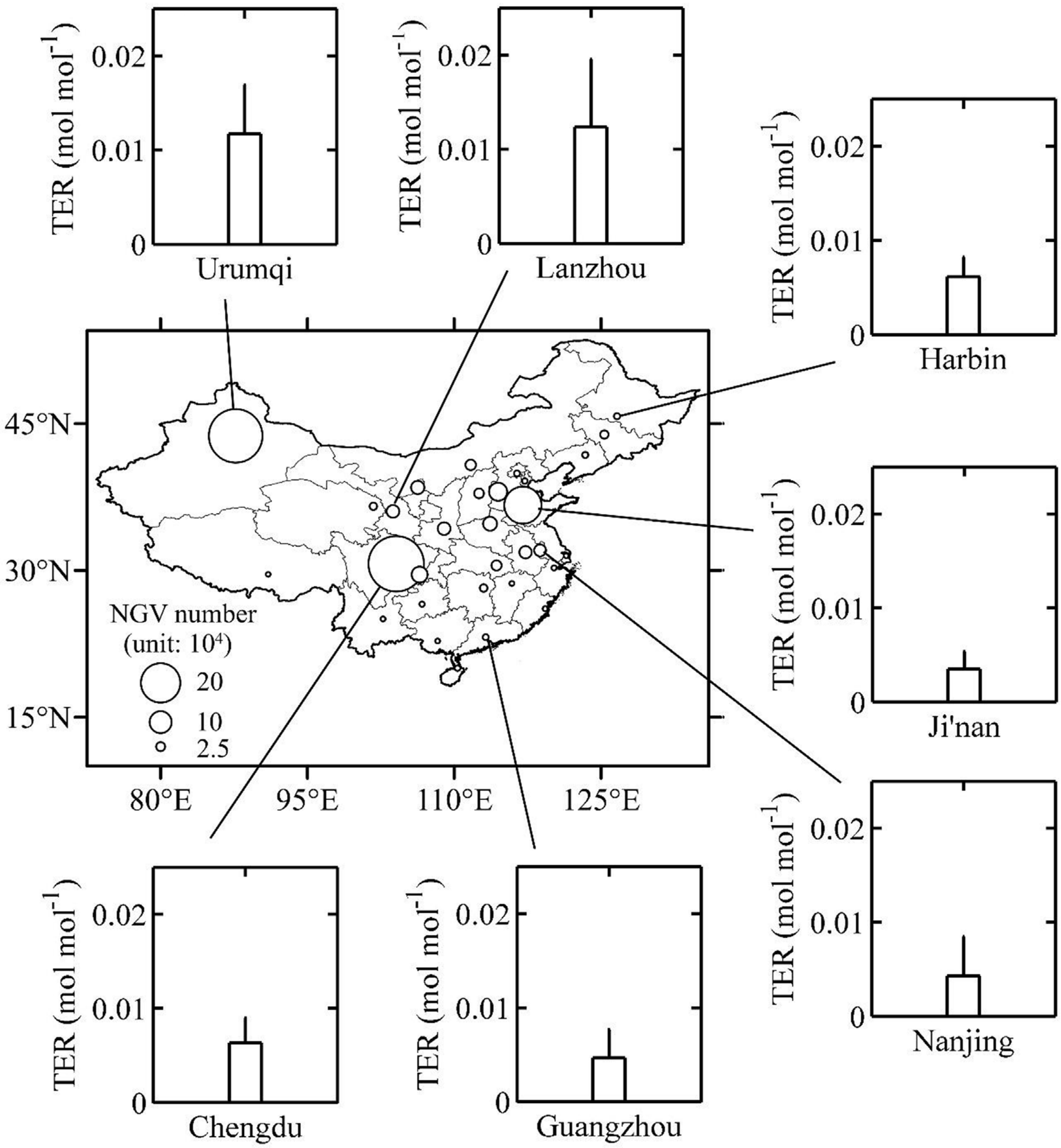

Fig. 2. Regional distribution of NGV population in Mainland China and traffic CH_4:CO_2 emission ratio (TER) in seven cities. Error bars are one standard deviation.

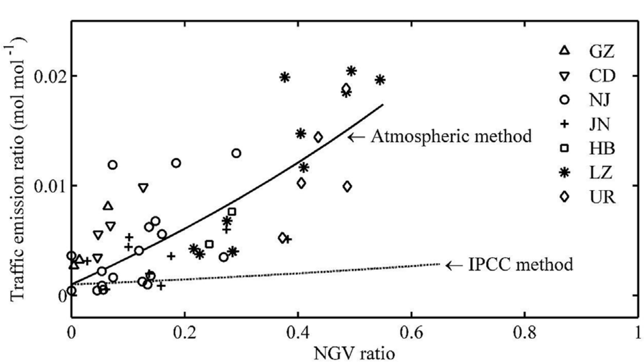

Fig. 3. The relationship between traffic CH_4:CO_2 emission ratio (TER) and the proportion of natural gas vehicles in total on-road vehicles (NGV ratio). City name abbreviations are GZ – Guangzhou, CD – Chengdu, NJ – Nanjing, JN – Jinan, HB – Harbin, LZ – Lanzhou, UR – Urumqi.

segment recorded the traffic condition during each observation. Non-taxi passenger cars, taxis and buses were tallied, and the NGV numbers for the three types of vehicle were modified by multiplying the city-mean proportion of NGVs for each type (Supplementary Table S1). The NGV fraction in each street transect is the ratio of the estimated NGV vehicle number to the total vehicle number observed on that street transect. This street-by-street estimate of NGV fraction is more accurate than simply using the city mean statistics on NGV fractions.

In order to better quantify variabilities of the traffic CH_4 emissions, we carried out more intensive sampling in Nanjing than in the other six cities. A total of 3 street transects and 15 traffic tunnels were used. For short street transects and traffic tunnels, vehicle tallies were based on videos recorded at each end. For long street transects, stationary video recordings cannot accurately reflect inflow and outflow traffic via side streets; for these observations we used traffic information recorded by the dashboard camera on the instrumented car.

Supplementary Fig. S2 shows the statistics of the CH_4 and CO_2 concentrations for the seven cities. Supplementary Figs. S4 and S5 shows examples of 1-Hz CH_4 and CO_2 mixing ratio time series measured along a street transect and through a traffic tunnel in Nanjing. A number of CH_4 concentration spikes are visible in the street data (Supplementary Figs. S4 and S5). Phillips et al. (2013) also observed spiky CH_4 time series on streets in Boston, USA (Phillips et al., 2013). In their study, the high concentrations are hotspots in fixed locations and are caused by leakage of natural gas distribution pipelines. In the present study, the CH_4 spikes were not stationary except for busy street intersections (Supplementary Fig. S3), implying that pipeline leakage was not the cause.

2.2. Determination of traffic CH_4:CO_2 emission ratio and NGV CH_4 emission factor

We used the CO_2 and CH_4 mixing ratio time series observed along each street transect or traffic tunnel to determine a TER for that transect or tunnel. This tracer correlation method is commonly used for determining emissions of air pollutants in urban airsheds (Shen et al., 2014; Zimnoch et al., 2010; Wunch et al., 2009). In the present study, the observed variabilities in the CO_2 and CH_4 time series were dominated by the same emission source (street traffic). The TER was taken as the slope between the measured CH_4 and CO_2 mixing ratios by using the geometric mean regression. Supplementary Figs. S4 and S5 shows two examples of the regression. Supplementary Table S2 lists the mean TER for each street transect or traffic tunnel in the seven cities.

The TER is an integrated signal from conventional vehicles and NGVs. Let n_1 and n_2 be the number of NGVs and conventional vehicles, respectively, on a street transect, $E_{m,1}$ and $E_{m,2}$ be their respective CH_4 emission factors (in mol km^{-1}), and $E_{c,1}$ and $E_{c,2}$ be their respective CO_2 emission factors (in mol km^{-1}). The TER can be described by the following equation,

$$\text{TER} = \frac{n_1 E_{m,1} + n_2 E_{m,2}}{n_1 E_{c,1} + n_2 E_{c,2}} \quad (1)$$

This equation can be rearranged to

$$\text{TER} = \frac{fR_1 + (1-f)R_2(E_{c,2}/E_{c,1})}{f + (1-f)(E_{c,2}/E_{c,1})} \quad (2)$$

where $f = n_1/(n_1 + n_2)$ is the NGV fraction, $R_1 = E_{m,1}/E_{c,1}$ is the CH_4:CO_2 emission ratio of NGVs, and $R_2 = E_{m,2}/E_{c,2}$ is the CH_4:CO_2 emission ratio of conventional vehicles. In this equation, TER and f are provided by field observations, and parameters R_2, $E_{c,1}$ and $E_{c,2}$ are determined with the IPCC emission factors, with relatively high accuracy. The only unknown in Equation (2) is R_1.

According to the *IPCC Guidelines for National Greenhouse Gas Inventories*, the CH_4 and CO_2 emission factors for gasoline vehicles are 0.051 mol L^{-1} and 51.1 mol L^{-1}, respectively, giving an emission ratio $R_2 = 9.92 \times 10^{-4}$ mol mol^{-1}. We compared the IPCC default R_2 value with the 5 tunnel observations in Nanjing during which no NGVs were present. The mean TER of these observations (0.00107 ± 0.00143; mean ± 1 standard deviation) is not statistically different from the IPCC default R_2 value ($p = 0.96$). The IPCC CH_4 and CO_2 emission factors for NGVs are 0.00317 kg m^{-3} and 1.93 kg m^{-3}, giving a default emission ratio $R_1 = 0.00451$ mol mol^{-1}. However, the default R_1 is significantly lower than our observations (Fig. 4).

The unknown parameter R_1 in Equation (2) was determined with a nonlinear regression method (Matlab Curve Fitting Toolbox Version 3.4.1). Some street transects were measured more frequently than others (Supplementary Table S2); to avoid uneven representation among the streets, street mean values were used in this regression procedure. We used the IPCC emission ratio of 9.92×10^{-4} mol mol^{-1} for R_2. To convert $E_{c,1}$ and $E_{c,2}$ to values mol km^{-1}, we used the same mileage per cubic meter of natural gas as that per liter of gasoline (Ma et al., 2013). Thus the ratio $E_{c,2}/E_{c,1}$ is 1.164 mol mol^{-1}, implying a 16% reduction in CO_2 emission per km traveled by switching to natural gas. It was slight lower than but not significantly different from the ratio from dual fuel vehicles ($E_{c,2}/E_{c,1}$ = 1.338 and 1.279 mol mol^{-1}, $p = 0.22$), because the fuel economy of gasoline was relatively reduced after the bi-fuel retrofitting (Xie et al., 2011; He et al., 2014).

2.3. Measuring tailpipe emissions

Emissions ratio from the tailpipe were measured for 6 taxis retrofitted to run on natural gas. These vehicles were from a local fleet in Nanjing and ranged in age from 1 to 3 years and in mileage from 50,000 to 300,000 km. For each measurement, 8 L of exhaust gas was collected with a Teflon bag lined with aluminum foil. The bag had been filled with ultra-high purity N_2 gas prior to collection. The collected gas was analyzed on a GC (GC7890B, Agilent Technologies, CA, USA) for CH_4 and CO_2 concentration. The CH_4:CO_2 emissions ratio was determined from the concentrations ratio.

2.4. Contribution of NGVs to national total anthropogenic CH_4 emission

According to *China Natural Gas Development Report* (NEA et al., 2016), 5 million of NGVs consumed 2.0×10^{10} m^3 of natural gas in 2015, while the total annual consumption in 2015 was 1.93×10^{11} m^3 (NBSC, 2017). Using the above emission factor, we estimate that NGVs emitted 0.44 ± 0.07 Tg of CH_4 to the atmosphere in 2015.

In recent years, the population of NGVs has been increasing steadily (Fig. 1), and it will reach to 10 million and 14 million in 2020 and 2030 (NEA et al., 2016). The CH_4 emission by NGVs will continue to increase. If we assume no changes in Vehicle Miles Traveled (VMT) and fuel consumption per vehicle, we project that in 2020 and 2030, 4.0×10^{10} m^3 and 5.6×10^{10} m^3 of NG will be consumed annually.

2.5. Sources of NGV data

In this study, the global NGV data were obtained from the International Association for Natural Gas Vehicles (http://www.iangv.org/current-ngv-stats). The provincial and municipal NGV data in China were provided by the Clean Energy Industry Association in Zigong City, Sichuan Province.

3. Results

3.1. Traffic CH_4:CO_2 emission ratio

The seven cities we surveyed spanned a wide range of NGV population. Three of the cities (Urumqi, Chengdu, Jinan) are located in provinces with a large NGV population (> 180 thousand), two (Harbin and Guangzhou) are in province with a small population (< 10 thousand), and two (Nanjing and Lanzhou) with an intermediate population (about 50 thousand). NGVs are exclusively used as taxis and for public transportation (buses) in Harbin, Guangzhou and Nanjing. In Urumqi, in addition to NG taxis and buses, 26% of private cars are powered by NG. The proportion of NGVs in the total vehicle population varies from 0.2% to 26% among these cities (Supplementary Table S1).

The TER is an integrated signal that includes contributions from NGVs and conventional vehicles. The mean TER shows a large range of variations among the cities we surveyed. The highest TER was observed in Lanzhou (0.012 ± 0.0072 mol mol^{-1}, mean ± 1 standard deviation), and the lowest in Jinan (0.0035 ± 0.0019 mol mol^{-1}; Fig. 2). The difference between these two cities are significant ($p < 0.01$). The city mean TER is weakly correlated with local NGV population (linear correlation coefficient = 0.82, $p = 0.02$). The street-mean TER shows a strong dependence on the street mean NGV ratio (Fig. 3). The mean TER of the seven cities is 0.0070 ± 0.0036 mol mol^{-1}. This value is an order of magnitude larger than the average emission ratio measured in a traffic tunnel of Zürich (Popa et al., 2014), because of the lower NGV population in Switzerland. But it is generally lower than emission ratios of urban airsheds in China (Shen et al., 2014), Europe (Zimnoch et al.,

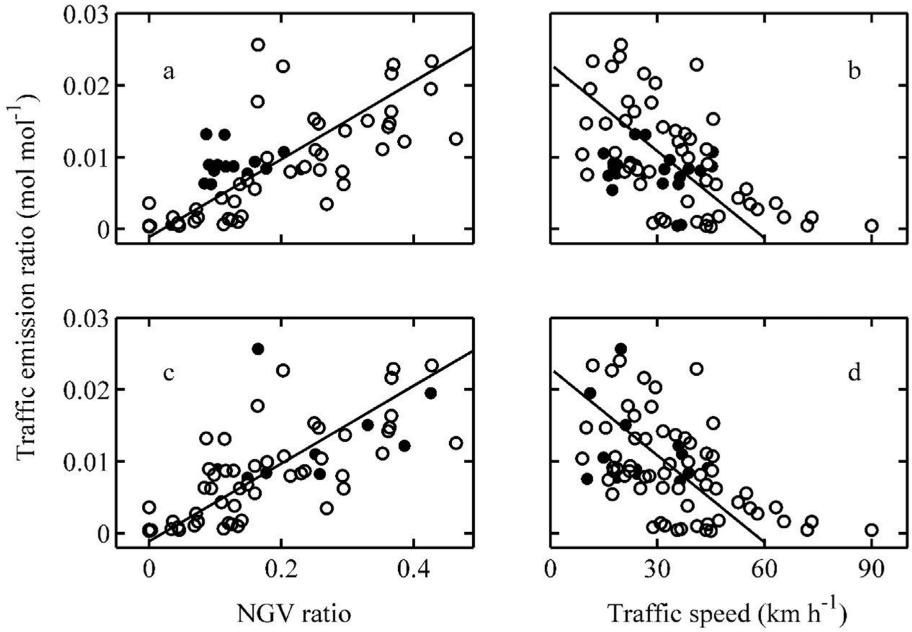

Fig. 4. Relationship between traffic emission ratio (TER) and NGV ratio or traffic speed. Hollow circles in a & b represent non-rush hour and in c & d represent weekday. Solid circles in a & b represent rush hour and in c & d represent weekend.

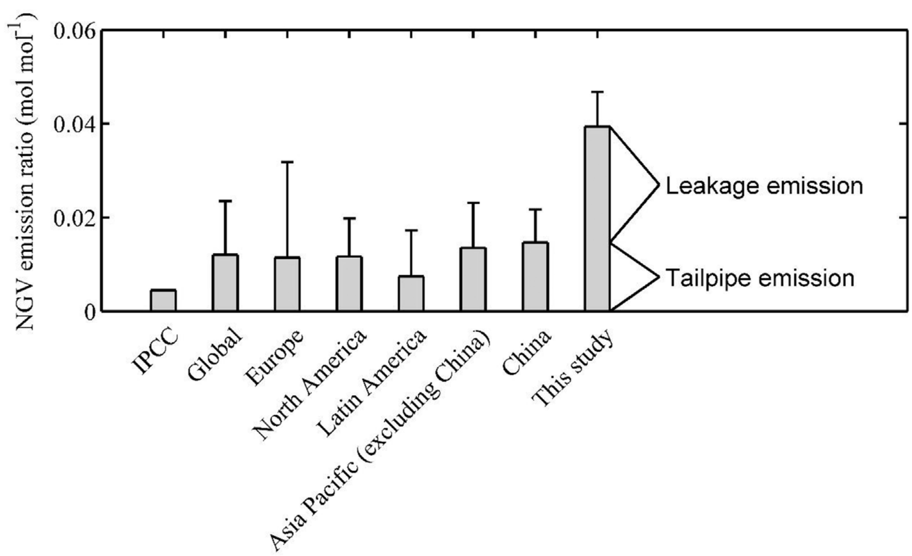

Fig. 5. Comparison of tailpipe CH_4:CO_2 emission ratio for NGVs with the on-road measurement of NGV CH_4:CO_2 emission ratio in this study. Error bars are one standard deviation.

2010) and the USA (Wunch et al., 2009).

The data points in Fig. 3 are ensemble means of a number of observations on single streets or traffic tunnels (Supplementary Table S2). In order to explain traffic emission patterns, we have performed a detailed analysis of the TER values from individual observations in Nanjing (Fig. 4). Here, each data point represents measurement made in one trip along a street transect or through a traffic tunnel. The TER measured in the tunnels (0.0021 ± 0.0020 mol mol^{-1}) is significantly lower than the TER measured on the open streets (0.012 ± 0.0053 mol mol^{-1}, $p < 0.01$). This difference can be explained in part by driving conditions, as the traffic moved much faster in the tunnels (mean speed 49 km h^{-1}) than on the streets (mean speed 27 km h^{-1}). Fuel combustion is more complete so that fuel consumption and emissions are lower when vehicle speed is higher (Vlieger et al., 2000). Putting the two groups of observation together, we find that the TER is gradually reduced with increasing traffic speed (Fig. 4 b & d; linear correlation = −0.52, $p < 0.01$). The highest TER (0.026 mol mol^{-1}) was observed from Hanzhong Gate Avenue to Zhongshan East Road with an NGV ratio of 0.16 of and a traffic speed of 20 km h^{-1}.

Another factor that controls the TER variations is the NGV ratio, or the proportion of NGVs in the total number of vehicles tallied during each observation (Fig. 4 a & c, linear correlation = 0.71, $p < 0.01$). An ANOVA analysis with weekend versus weekday as a class variable and the NGV ratio as a continuous variable (Supplementary Fig. 4a) reveals that there is no difference between weekend versus weekday ($p = 0.84$). Similarly, the difference between rush hours and non-rush hours is also not significant ($p = 0.95$, Fig. 4c). The NGV ratio and traffic speed together explain 58% of the observed variations in the TER in Nanjing.

3.2. Methane emission factor for natural gas vehicles

We used a regression model to infer the actual NGV CH_4:CO_2 emission ratio from the TER data (Equation (2)). The model assumes that the CH_4:CO_2 emission ratio of conventional vehicles is known. (We used the IPCC value for this.) It then expresses the average TER observed on a street transect or through a traffic tunnel as a function of the corresponding NGV ratio, with the CH_4:CO_2 emission ratio of NGVs being the only unknown parameter. By applying a curve fitting tool to all the data obtained from the seven cities (Fig. 3), we estimate that the CH_4:CO_2 emission ratio is 0.031 ± 0.0047 mol mol^{-1} (mean ± 95% confidence bounds) for NGVs in China (Fig. 5). This ratio is nearly seven times the ratio obtained from the IPCC default CH_4 and CO_2 emissions factors for NGVs, which is 80% higher than the tailpipe emission ratio of NGVs measured in Nanjing (Supplementary Table S3), and 120% greater than the mean ratio of tailpipe CH_4 and CO_2 emissions found in published studies for NGVs in China (Fig. 5). The difference between the emission ratio measured on-road and the mean tailpipe emission ratio is statistically significant ($p < 0.01$).

Combining this emission ratio with the IPCC CO_2 emission factor of 1.93 kg m^{-3} for NGV, we estimate that the actual emission factor for NGVs is 0.022 ± 0.0033 kg m^{-3}.

In Equations (1) and (2), we do not distinguish between natural gas passenger cars and buses. Because these buses are manufactured as NGVs, it can be argued that they are less likely to experience leakage than natural gas passenger cars. To investigate how this scenario affects the result, we have modified Equation (2) by breaking the NGVs into two groups (taxis and buses). The emission ratio for taxis is still an unknown variable, and the emission ratio for buses was assigned the mean tailpipe value found in the literature for NGVs in China (0.015 ± 0.0071 mol mol^{-1}; Fig. 4). Applying the curve fitting routine yielded a new TER estimate of 0.033 ± 0.0055 mol mol^{-1}, which is slightly higher than the original estimate of 0.031 ± 0.0047 mol mol^{-1}, but the difference between the two estimates is not statistically significant ($p = 0.95$).

4. Discussion and conclusions

Passenger NGVs (private cars and taxis) in China run on compressed natural gas (CNG) as the main fuel source. In China, about 80% of passenger NGV vehicles are retrofitted from gasoline and diesel vehicles to use dual fuels (CNG and gasoline or CNG and diesel) after purchase. (Gasoline or diesel is used only in cases of emergency when refueling of CNG is not possible.) Retrofitted dual-fuel engines have lower combustion efficiencies than single-fuel original engines (Norbeck et al., 1998; Lima et al., 2010). This is one reason for why the tailpipe CH_4:CO_2 emission ratio is greater in China than the IPCC default emission ratio (Fig. 5), although the tailpipe emission ratios in Europe and North America are also greater that the IPCC value.

NGV vehicle conversion and repair are done in small workshops in China. On-road fuel leakage from the modified fuel delivery system is a common problem. Our interviews with taxi drivers and repair mechanics have identified a number of weak points on the vehicle's high-pressure fuel delivery system that have high risks of fuel leakage (Supplementary Fig. S1). That the overall NGV emission ratio is much larger than the tailpipe emission ratio indicates that on-road leakage is a widespread problem. Subtracting the mean tailpipe emission factors (0.010 ± 0.005 kg m^{-3}) from the actual emission factor (0.022 ± 0.0033 kg m^{-3}), we estimate that the leakage emission factor is 0.012 kg m^{-3}.

NGVs in China consumed 2.0×10^{10} m^3 of natural gas in 2015 (NEA et al., 2016), which is only slightly lower than that total consumption by the transportation sector (2.38×10^{10} m^3 in 2015; NBSC, 2017), indicating that NGVs are the dominant consumers in this sector. Using the above emission factor, we estimate that NGVs emitted 0.44 ± 0.07 Tg of CH_4 to the atmosphere in 2015, accounting for 1.0% of anthropogenic CH_4 emissions in China (Kirschke et al., 2013; Peng et al., 2016). In recent years, the population of NGVs has been increasing steadily (Supplementary Fig. S6), and these trends will likely to continue in future years (NDRC, 2014). In this business-as-usual scenario, the CH_4 emission by NGVs will likely reach 1.23 Tg yr^{-1} in 2030, or about 3% of the current national total anthropogenic emission.

One unintended consequence of fuel switching is increase in CH_4 emissions which contribute to global warming. Since the majority of the global NGV fleet is found in emerging economies (including China; Fig. 1) where emission regulations are not as strict as in developed countries, NGV CH4 emissions may be a significant contributor to the global methane budget and curbing the emissions from this sector should be a high priority for global climate mitigation efforts. Our results indicate that tightening emission standards for NGVs should bring clear climate benefits. Elimination of the on-road leakage problem, a low-hanging fruit in climate mitigation efforts, can cut the NG vehicular emission by half according to the data shown in Fig. 5.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.06.007.

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