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Abstract

Reducing emissions of methane (CH_4) in developed regions and urban areas is a practical way to curb the unexpected surge in global CH_4 levels in recent decades. Traffic emissions are among the important anthropogenic CH_4 emission sources in megacities, yet CH_4 emissions from on-road vehicles are less characterized and not well addressed. Based on tunnel tests in an urban tunnel in south China, a real-world emission factor (EF) of CH_4 was measured to be 0.26 $\pm$ 0.03 g·km^{-1} (mean $\pm$95% C.I.) for on-road vehicle fleet which including gasoline vehicles, diesel vehicles, and liquefied petroleum gas vehicles, with an average CH_4/CO_2 mass ratio of 40.6E-5 g·g^{-1}, and CH_4 could account for 1.3% of vehicle CO_2-equivalent emissions. Using the measured CH_4/CO_2 ratio and available automobile CO_2 emission estimates, traffic CH_4 emissions in 2014 could have reached 333 Gg and represented 0.6% of total anthropogenic CH_4 emissions in China, approximately four times the previous reported value of 79 Gg. Our results indicate that improving energy efficiency would have co-benefits for reducing traffic emissions of CH_4, as observed EFs of CH_4 are positively correlated with that of CO_2, and over 90% of traffic CH_4 emissions in China could be avoided if the traffic CH_4/CO_2 ratio can be an order of magnitude lower as previously observed in a tunnel in Switzerland.

Introduction

Methane (CH_4) is the second-largest contributor to anthropogenic greenhouse forcing after carbon dioxide (CO_2) [1, 2]. Apart from trapping the infrared energy emitted by the Earth and its atmosphere, CH_4 has extra warming effects affecting the abundance of other greenhouse gases, such as ozone (O_3), water vapor (H_2O), and CO_2 [3]; becoming involved in gas-aerosol interactions [4]; and absorbing energy from the Sun at shorter wavelengths [5]. The radiative forcing attributed to methane emissions is very likely to be almost twice as large as that from its change in concentration [1]. Data from NOAA observation stations show that global mean atmospheric CH_4 started to rise in 2007 after a near-zero growth from 2000–2006 [6], with a sharper increase beginning in 2014 [7]. As a result, the global level of atmospheric methane climbed to 1.87 ppm in March, 2019 [8] from 1.78 ppm in 2006 [7]. Because CH_4 is a potent greenhouse gas with a 20-year Global Warming Potential (GWP_{20}) of 84 and a 100-year Global Warming Potential (GWP_{100}) of 28–32 [1, 2, 5], this unexpected surge in global methane levels presents a major challenge, and eliminating emissions of CH_4 would quickly provide a benefit for achieving the goals set out in the Paris Agreement to limit temperature increases to 2 °C or, if possible,

to 1.5 °C above preindustrial levels [9, 10]. In the near term, a weakening of aerosol cooling by a reduction in sulfur dioxide emissions would add to future warming, but can be tempered by reductions in methane emissions [1].

Anthropogenic methane emissions have dominated global methane growth since 1999 [11–13]. Although the drivers behind the surge in atmospheric methane in the recent decades are less well understood and hotly debated, there are limited opportunities to address agricultural emissions or natural emissions, and reducing methane emissions in the energy sector is the most practical option available to control global methane levels. In fact, the fractions of CH_4 emissions from the energy sector reached 44.6% in China in 2014 and 26.5% in the world in 2012 [14, 15]. Considering ethics, social justice and equity issues, developed regions and urban areas should take on more responsibilities in emissions reduction efforts. Non-CO_2 forcers such as methane are emitted alongside CO_2, particularly in the energy and transport sectors, and thus, reducing emissions of CO_2 and CH_4 in urban areas can be largely addressed as co-benefits of curbing emissions of air pollutants from the energy and transport sectors. In fact, CH_4 emissions from fossil fuel industries were found to be 20%–60% higher than previously estimated, implying a greater potential for energy efficiency improvements to mitigate anthropogenic climate forcing [16]. Transport is an important sector in fossil fuel consumption. Anthropogenic CH_4 emissions from mobile sources are estimated to contribute only 0.2%–0.5% to the global CH_4 budget [17, 18], and this sector ranked 12th among all sources in 2017 in the US [19]. However, Nakagawa *et al* [20] revealed that up to 30% of CH_4 could come from automobile exhaust in Nagoya, Japan, suggesting substantial opportunities for reducing CH_4 from traffic emissions in megacities.

China has become the world's largest manufacturer and consumption market of motor vehicles; thus traffic emissions of CH_4 are of great concern, particularly in urban areas. However, as CH_4 is an unregulated pollutant, there are few studies concern its emissions from vehicle exhausts [21]. Emission factors (EFs) along with traffic statistics are needed to estimate traffic emissions of CH_4 [22], yet EFs of specific vehicle types are mainly derived from European or US vehicle emission databases for compiling China's CH_4 emission inventory using mobile source emission models such as the Computer Programme to Calculate Emissions from Road Transport (COPERT) [21, 23–26], MOBILE [27, 28], International Vehicle Emissions (IVE) [29] and MOtor Vehicle Emission Simulator (MOVES) [30]. Due to differences in technological levels, emission control standards and driving conditions, simply borrowing EFs from developed countries will lead to significant uncertainty when estimating emissions in China [31, 32]. There are only a few tests available that provide EFs for light-duty gasoline vehicles, gasoline vehicles, compressed natural gas vehicles, natural gas vehicles and passenger cars, respectively [33–35], or obtain the CH_4/CO_2 ratio for a whole fleet [22, 36–39].

Tunnel tests have proven to be a useful method to estimate real-world fleet-wide EFs [22, 40]. In this study, we conducted tests in 2014 in the underwater Zhujiang Tunnel with a daily traffic flow of approximately 40,000 vehicles in urban Guangzhou, South China, to provide a real-world EF of CH_4 for an on-road fleet for the first time, to recheck the status of traffic CH_4 emissions in China and to assess the potential of reducing CH_4 emissions in the transport sector.

Methods

Field work

Zhujiang Tunnel is an underwater tunnel crossing the Pearl River in west-central Guangzhou. Located in a relatively populated area, the traffic load in the tunnel reaches ~40,000 vehicles per day. The tunnel is 1238.5 m long with a 721 m flat underwater section. It has two bores with two lanes in each bore. A schematic diagram of the tunnel was given by Liu *et al* [41] and Zhang *et al* [42]. During our test, the ventilation system of the tunnel was not operated. Zhujiang Tunnel has a speed limit of 50 km h^{-1}, and the traffic speed during our sampling (data from Guangzhou Transportation Administration Bureau, personal communication) ranged from 20 km·h^{-1} to 47 km·h^{-1}, averaging 35 $\pm$ 5 km·h^{-1} (95% confidence intervals, 95% C.I.). Detailed descriptions of the Zhujiang Tunnel can be found in the supporting information text S1 is available online at stacks.iop.org/ERC/2/061005/mmedia.

Sampling was conducted from June 25th to July 1st in 2014. We placed the two sampling stations (outlet station and inlet station) 50 m from the ends of the 721-meter flat underwater section. Determined emission factors for air pollutants and halocarbons based on tunnel tests are reported elsewhere [41–45]. CO_2 and CH_4 in this study were measured using the same batch of air samples collected by canisters for the analysis of volatile organic compounds (VOCs). The VOC samples were collected during intervals of 02:00–3:00, 07:00–8:00, 08:00–9:00, 09:00–10:00, 10:30–11:30, 14:00–15:00, 17:00–18:00, 18:00–19:00, and 19:00–20:00 on June 25th, 26th, 28th and 29th. Two more samples were collected on June 27th (02:00–3:00) and June 30th (4:00–5:00) as supplements. The air samples were collected into pre-evacuated 2-liter electro-polished stainless steel canisters using a Model 910 pressurized canister sampler (Xonteck, Inc., Fremont, CA, USA). The sampler was set at a

constant flow rate of 66.7 ml min^{-1} to guarantee that the air pressure of each canister was approximately 2 atm after sampling for 60 min. More details about quality control and quality assurance of canister samples were offered in Zhang *et al* [42].

An IRGASON eddy covariance system (Campbell, Inc., USA) with an integrated CO_2 and H_2O open-path gas analyzer and a 3D sonic anemometer was used to measure *in situ* CO_2, wind speed/direction and temperature. During the sampling period, the air temperature recorded by the IRGASON at the outlet station ranged from 27.9°C to 41.5 °C with an average of 34.4 ± 0.3 °C (mean ± 95% C.I.).

Laboratory analysis

CH_4 was analyzed by an Agilent Model 6890 GC equipped with an FID and a packed column (5 A Molecular Sieve 60/80 mesh, 3 m × 1/8 inch). CO_2 was analyzed by the same equipment with a different packed column (10Ft 1/8 2 mm HayeSep Q 80/100 SS). CO_2 was first converted by a Ni-based catalyst to CH_4 and then detected by the FID after separation by the packed column [42]. Both CH_4 and CO_2 were quantified by an external calibration method. Working calibration curves were obtained by diluting ultra-pure CO_2 or CH_4 (>99.999%) to working standards with an Entech Model 4700 high-precision static dilution standards preparation system (Entech Instruments Inc., USA), running each of the working standard for three times and then plotting the average responses against the mixing ratios by linear regression. Re-calibration is needed: (1) if the R^2 for the linear dose-response correlation was below 0.99; and (2) the calibration curve is challenged with a NIST traceable standard (Spectra Gases, 398 ppm for CO_2 and 1.01 ppm for CH_4) each day before the analysis of air samples. If the determined mixing ratio is beyond ±0.5% the labelled value of the NIST traceable standard. The method detection limits (MDLs) of the analysis system for CH_4 and CO_2 are 20 ppb and 3 ppm, respectively.

Results and discussion

Emission factor (EF)

The average EF for vehicles traveling through the tunnel during a time interval T can be calculated in the same way as in previous studies [46, 47]:

$$EF_{fleet} = \frac{\Delta c \times V_{air} \times T \times A}{N \times l}$$

where $\boldsymbol{EF_{fleet}}$ (mg·km^{-1}) is the mean emission factor of CH_4, $\boldsymbol{\Delta c}$ (mg·m^{-3}) is the concentration difference between the inlet and outlet sampling stations inside the tunnel, $\boldsymbol{V_{air}}$ (m·s^{-1}) is the air speed velocity parallel to the tunnel sensed by the 3D sonic anemometer with an average of 3.54 m s^{-1}, $\boldsymbol{A}$ (m^2) is the tunnel cross section with value of 58.2 m^2, $\boldsymbol{N}$ is the monitored traffic count passing the tunnel during the time interval $\boldsymbol{T}$(s) (3600 s in this study), and $\boldsymbol{l}$ (m) is the distance between the two sampling points which is 0.621 km.

The results showed that the EFs of CH_4 measured in the tunnel ranged from 0.048 g·km^{-1} to 0.40 g·km^{-1}, with an average of 0.26 ± 0.03 g·km^{-1} (mean ± 95% C.I.). The time series of EFs of CH_4, as well as those of numbers of three fuel-type vehicles travelling through the tunnel, are shown in figure 1. During the time intervals of 2:00–3:00 or 3:00–4:00, the EFs of CH_4 became significantly lower, corresponding to a relatively lower amount (approximately 40%) of gasoline vehicles. The number of gasoline vehicles in these hours decreased by approximately 80% relative to that in rush hours. A highly significant ($p < 0.01$) correlation was observed between the observed EFs of CH_4 and the percentages of gasoline vehicles (figure S1), implying that gasoline vehicles may dominate CH_4 emissions in the traffic fleet. Chassis dynamometer results from Koike and Odaka [33] have also showed that diesel vehicles have significantly lower emissions of CH_4 than gasoline vehicles. Even though there were fewer diesel vehicles on the weekends, no significant difference in the EFs of CH_4 was found between the workdays and weekends.

Figure 2 shows a comparison of the EFs of CH_4 for vehicles measured in this study with those previously reported. The average EF of 0.26 ± 0.03 g·km^{-1} from our study is approximately 50 times that of 0.0052 g·km^{-1} for gasoline vehicles measured using chassis dynamometer under New European Driving Cycle (NEDC) conditions [34] and over 20 times higher than that of 0.012 ± 0.003 g·km^{-1} estimated for the US on-road fleet under the Federal Test Procedure −75 (FTP-75) conditions [17]. These large gaps cannot solely be explained by differences in vehicle operating conditions since NEDC and FTP-75 conditions include acceleration and idling modes during which more emissions would typically occur than during the cruising mode for vehicles in the tunnel; instead, they reflect the large gaps in fuel quality, engine performance, exhaust control and vehicle maintenance as well. As an example, during our study on-road vehicles in Guangzhou were mostly composed of Euro 3 (which equals to China 3) (39.4%) and Euro 4 (36.2%), and Euro 1 and Euro 2 still shared a notable proportion of 21.7%. Previous studies demonstrated that CH_4 emissions increase with the age of catalytic convertors [48–51]. Additionally, cars using premium grade gasoline fuel with higher aromatic hydrocarbon content and lower content of saturated hydrocarbons and olefins are found to have lower CH_4 emissions [48],

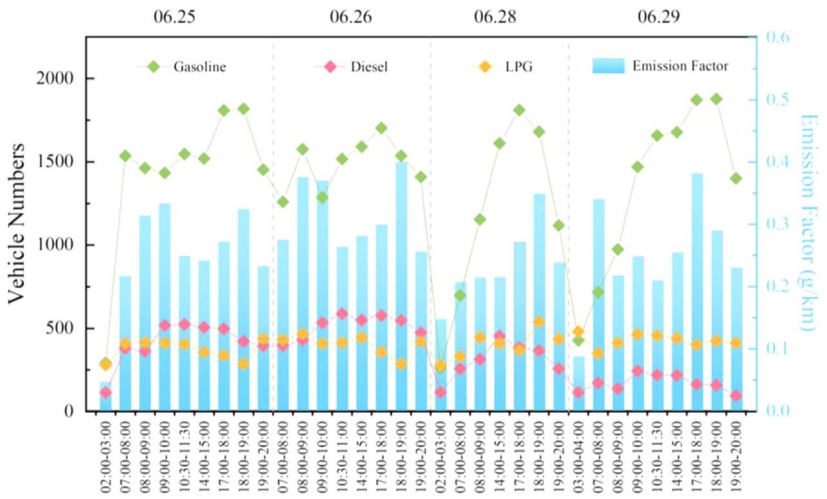

Figure 1. Diurnal variations of the vehicle fleet of gasoline vehicles, diesel vehicles and liquefied petroleum gas (LPG) vehicles and emission factor of CH_4 (Dates 06.25 and 06.26 are weekdays, while 06.28 and 06.29 are weekends).

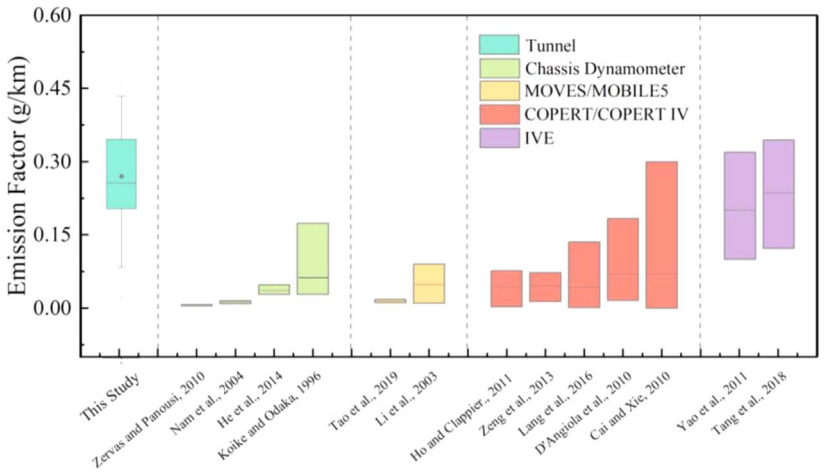

Figure 2. CH_4 emission factor measured in this study compared with previous studies. (a) Data shown in the box plot are the minimum, maximum, first and third quartiles, median, and average values (dark grey diamond); (b) Data in the floating plot are the minimum, maximum and average values; (c) MOVES: MOtor Vehicle Emissions Simulator, COPERT: Computer Programme to Calculate Emissions from Road Transport, IVE: International Vehicle Emissions; (d) Detail information of these studies are provided in table S2.

and the notoriously higher amount of olefins in China' gasoline (a limit of 24% v/v even in the China 6 grade gasoline oil) would probably give rise to the CH_4 emissions from China's gasoline vehicles.

As shown in figure 2 and table S2, our measured EFs of CH_4 for the urban vehicle fleet were also substantially higher than those derived from the COPERT and MOBILE models [17, 21, 23–27, 30], but comparable to those derived from the IVE model [28, 29], which was designed for mobile source emissions of developing countries by researchers at the International Sustainable Systems Research Center and the University of California at Riverside. In contrast to other models that use average speed to represent a driving cycle, the IVE model introduces parameters such as vehicle specific power and engine size to better represent driving conditions [28, 52], making it more suitable for estimating vehicle CH_4 emissions in developing countries.

The CH_4/CO_2 ratio

The CH_4/CO_2 ratio is proven to be a simple and practical way to estimate traffic emissions of CH_4 [17]. In order to better represent the emissions of CH_4 from on-road vehicles, the slope for the correlation of EFs between CH_4 and CO_2 was used as the average CH_4/CO_2 ratio in this study. A significant ($P < 0.01$) positive linear relationship was observed between the EFs of CH_4 and CO_2 measured in the time intervals in the Zhujiang Tunnel (figure 3), with an average CH_4/CO_2 mass ratio of 40.61E-5 $\pm$ 7.21E-5 $g{\cdot}g^{-1}$.

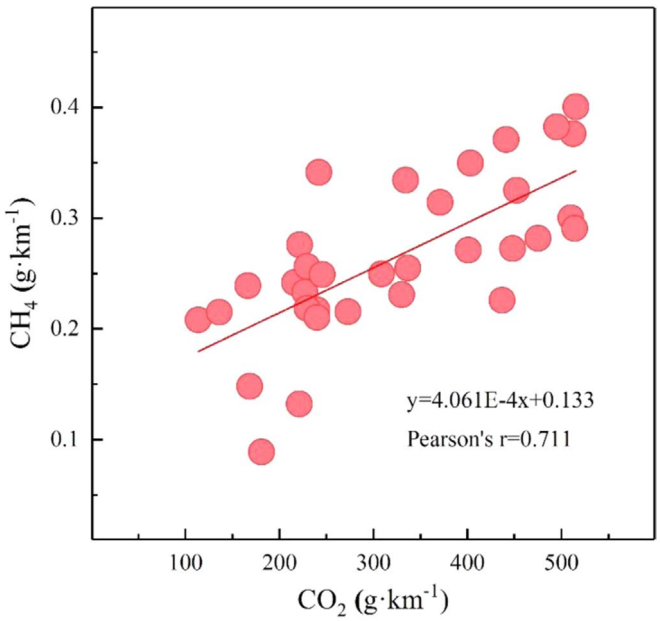

Figure 3. Correlation between emission factors of CH_4 and CO_2 in the Zhujiang Tunnel.

The CH_4/CO_2 ratio in this study is consistent with that of 17.09E-5 to 50.91E-5 obtained in the Nanjing Yangtze River Tunnel in 2014 [39] by using a portable greenhouse gas analyzer to measure the concentration gradient between the inlet and outlet of the tunnel. It is worth noting that gasoline vehicles dominated the traffic composition in the Yangtze River Tunnel during their sampling time. However, tests in the Islisberg Tunnel in Switzerland in 2011 revealed a CH_4/CO_2 ratio of 1.67E-5 $\pm$ 0.07E-5 $g{\cdot}g^{-1}$ [22], which is approximately 25 times lower than the average ratio from this study. Nam *et al* [17] reported a ratio of 15E-5 $\pm$ 4E-5 $g{\cdot}g^{-1}$ for the on-road fleet in the US in 2003 based on chassis dynamometer tests, while Herndon *et al* [36] used a 'Bus Chase' method to obtain a CH_4/CO_2 ($g{\cdot}g^{-1}$) ratio of 21.82E-5 $\pm$ 7.27E-5 for diesel vehicles and 443.64E-5 $\pm$ 65.45E-5 for compressed natural gas (CNG) vehicles. Except for the difference in vehicle size (bus to light-duty vehicle), the equipped oxidation catalyst in cars for emission control may be the main reason for the differences in EFs [36]. Hu *et al* [37] also reported a relatively higher CH_4/CO_2 ($g{\cdot}g^{-1}$) ratio of 254.55E-5 $\pm$ 130.91E-5 for natural gas vehicles (NGV) in China in 2015.

Vehicle CH_4 emissions estimate in China

In 2014, annual traffic CO_2 emissions in China were 820 Tg while annual traffic CH_4 emissions were estimated to be 79 Gg, and annual anthropogenic CH_4 emissions were 5357 Gg [14]. However, using the CH_4/CO_2 mass ratio of 40.61E-5 $\pm$ 7.21E-5 from this study and China's traffic CO_2 emissions of 820 Tg yr^{-1} in 2014, we could roughly obtain an estimate of 333 Gg yr^{-1} for traffic CH_4 emissions in China in 2014, which is approximately 4 times that reported value of 79 Gg and accounts for approximately 0.6% of the total anthropogenic CH_4 emissions (5357.2 Gg) in China. This percentage is above the upper limit of $<$0.5% estimated by Metz [18], and comparatively higher than the values of $<$0.2% estimated by Nam *et al* [17] and 0.20% compiled for America in 2014[19]. With a GWP100 of 32 [5] and a CH_4/CO_2 ratio of (40.61E-5 $\pm$ 7.21E-5) $g{\cdot}g^{-1}$ for the on-road fleet, CH_4 can account for approximately 1.3% of traffic CO_2-equivalent emissions from our study in China, which is significantly higher than the range of 0.3%–0.4% estimated by Nam *et al* [17] for global CH_4 emissions and the value of 0.08% estimated for the U.S. on-road fleet in 2014 [19]. In upcoming decades, such as in a 20-year horizon, the global warming potential of CH_4 is 84 times that of CO_2 [1], indicating an even greater contribution to climate warming.

Conclusion

As the CH_4/CO_2 mass ratios measured for an urban on-road fleet in this study in the Pearl River Delta were consistent with those obtained in the Yangtze River Delta in 2014 [39], so the much higher CH_4/CO_2 mass ratios might be common for vehicle emissions in China. Using the CH_4/CO_2 mass ratios obtained in this study, we can roughly estimate that on-road vehicles in China might have contributed ~330 Gg CH_4 (~1 Tg CO_2-equivalent) in 2014, or 0.6% of China's total anthropogenic CH_4 emissions. Our results indicate that improving energy

efficiency would have co-benefits for reducing traffic emissions of CH_4, as the observed EFs of CH_4 are positively correlated with those of CO_2. On the other hand, if the traffic CH_4/CO_2 ratio can be lowered to the level of 1.67E-5, as observed in the Islisberg Tunnel [22], over 90% of traffic CH_4 emissions in China could be tempered, suggesting a large potential or space to reduce China's CH_4 emissions in the transport sector.

Why were the EFs of CH_4 or CH_4/CO_2 mass ratios so high for on-road vehicle fleet in China ? The answer is not clear. However, fuel quality, engine performance, exhaust after treatment facilities and/or maintenance might be among the factors inducing more CH_4 emissions. Our previous study in the same tunnel revealed that the global warming potentials associated with refrigerant leakage from on-road vehicles are equal to 1.4% of that of the directly emitted CO_2 [44], and now, we have demonstrated that the emissions of CH_4 from on-road vehicles are equal to 1.3% of the directly emitted CO_2 in terms of the global warming potentials. These findings raises concern about whether we need to consider overall environmental and climatic effects of traffic emissions in formulating emission control policies, such as the emissions of greenhouse gases (CO_2, CH_4 and halocarbons) or secondary formation of ozone, which is also a greenhouse gas that can be formed from photochemical ageing of traffic-emitted VOCs and NO_x.

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Competing interests

The authors declare that they have no competing interests related to the study performed and presented in this paper.

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