Environmental Pollution 267 (2020) 115623

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Invited paper

Emissions of nitrogen oxides and volatile organic compounds from liquefied petroleum gas-fueled taxis under idle and cruising modes☆

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A R T I C L E I N F O

Article history:
Received 12 June 2020
Received in revised form
1 September 2020
Accepted 5 September 2020
Available online 9 September 2020

Keywords:
Liquefied petroleum gas (LPG)
Taxis
Emission factor
Volatile organic compounds (VOCs)
Nitrogen oxides (NO_x)
Chassis dynamometer

A B S T R A C T

Liquefied petroleum gas (LPG) as an alternative fuel is increasingly used in mainland China, few reports are however available about emissions from LPG-fueled vehicles. In this study, 26 LPG-fueled taxis in Guangzhou, south China were tested using a chassis dynamometer to obtain their emission factors of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) under idle and cruising (10–60 km h^{-1}) modes. The emission factors of NO_x on average increased with speed from 4.13 g kg-fuel^{-1} at idling to 71.1 g kg-fuel^{-1} at 60 km h^{-1} at a slope of 10.6 g kg-fuel^{-1} per 10 km h^{-1} increase in speed. Alkanes were the most abundant (71.9%) among the VOCs in the exhaust, followed by alkenes (25.2%), ethyne (2.7%), and aromatic species (0.2%). Emission factors of VOCs at idling averaged 8.24 g kg-fuel^{-1}, higher than that of 6.23–7.36 g kg-fuel^{-1} when cruising at 10–60 km h^{-1}, but their ozone formation potentials (OFPs) were lower at idling (15.8 g kg-fuel^{-1}) than under cruising (19.1–23.8 g kg-fuel^{-1}) largely due to higher emission of more reactive alkenes under cruising mode. Emissions of both NOx and VOCs increased significantly with mileages. Measured emission factors of NO_x and reactive VOCs in this study suggested that replacing the gasoline-powered taxis with the LPG-fueled taxis with LPG-gasoline bi-fuel engines and no efficient after-treatment devices would not benefit in reducing the emissions of ozone precursors, and strengthening the emission control for LPG vehicles with dedicated LPG engines and after-treatment converters, as did in Hong Kong, could further benefit in reducing the emission of photochemically active species when using LPG as alternative fuels.

1. Introduction

In urban areas, vehicular emission contributes substantially to ambient nitrogen oxides (NO_x) (Song and Xie, 2006; Mishra et al., 2016; He et al., 2016), volatile organic compounds (VOCs) (Liu et al., 2005; Song and Xie, 2006; Baker et al., 2008; Ho et al., 2009), and particulate matters (PM) (Eiguren-Fernandez et al., 2004; Yu et al., 2013; Liu et al., 2014a; Worton et al., 2014). These primarily emitted pollutants, like NO_x and VOCs, can also react through a series of complex chemical processes in the atmosphere to form secondary ozone (O_3) (National Research Council, 1991) or secondary organic aerosols and nitrate (Gentner et al., 2017). Due to the crucial roles played by vehicular emission in air quality deterioration in megacities (Kirchstetter et al., 1996a, 1996b; Namdeo and Bell, 2005; Chan and Yao, 2008), sustained efforts have been made worldwide to reduce vehicular emissions, particularly in urban areas. Using cleaner fuels is one of the most important ways to lower vehicular emission, and switch from gasoline/diesel oils to liquefied petroleum gas (LPG) for buses and taxis in the public transportation sector is a widely adopted approach (Karamangil,

☆ This paper has been recommended for acceptance by Prof. Pavlos Kassomenos.
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https://doi.org/10.1016/j.envpol.2020.115623
0269-7491/

2007; Myung et al., 2009; Adam et al., 2011; Guo et al., 2011; WLPGA and AEGPL, 2018).

As LPG is mostly composed of less reactive propane and butanes, previous studies have demonstrated that this fuel transition from gasoline/diesel oils to LPG could efficiently reduce emissions of reactive VOCs and particulate matters (Ristovski et al., 2005; Myung et al., 2009, 2012; 2014; Jang et al., 2014). Ristovski et al. (2005) compared the particle emissions between the unleaded petroleum vehicles and LPG vehicles (LPGVs) under different speeds and found that LPGVs were 'cleaner' with 70% fewer particle emissions. Myung et al. (2009) showed that LPGVs had significantly lower emission factors of particulate matters than diesel vehicles without a particulate filter. Myung et al. (2012) pointed out that LPGVs greatly excelled gasoline vehicles with over an order of magnitude lower emission factors of total hydrocarbons, benzene, and PM. Myung et al. (2014) showed that emissions of particle-bound polycyclic aromatic hydrocarbons (PAHs) from LPGVs decreased by over 90% when compared to those from gasoline vehicles. In Hong Kong, remote sensing and on-board measurements confirmed that LPG taxis in contrast to their diesel counterparts could greatly alleviate urban air pollution under real driving conditions (Ning and Chan, 2007; Lau et al., 2011).

While it is quite certain that the transition from gasoline/diesel to LPG would greatly reduce vehicular emissions of reactive VOCs and PM, it might not be so favorable in reducing vehicular emission of NO_x. Early works revealed less NO_x emitted from LPGVs when compared to that from gasoline vehicles (Pulkrabek, 1997), and much lower NO_x emission from LPGVs than from diesel vehicles (Ning and Chan, 2007). Jang et al. (2014) however found LPGVs had slightly higher emissions of NO_x than gasoline vehicles, and Zhang et al. (2013a) demonstrated, by conducting IG195 test for LPG taxis and on-road portable emission measurement system (PEMS) for LPG buses, that heavy-duty LPG buses did not show lower NO_x emissions than diesel buses, and that replacing gasoline/diesel vehicle with LPGVs having no after-treatment equipment would not help mitigate NO_x emissions as expected under real-world driving condition. Some studies further investigated the relationships between NO_x emissions and driving speeds, yet inconsistent results came out possibly due to factors like mileages and performance of catalytic converters (Ning and Chan, 2007; Lau et al., 2011; Chikhi et al., 2014).

In the developing world, with the rapid increase in vehicle population in recent decades, traffic-related air pollution has become a serious environmental health issue, particularly in the urban street microenvironments. For example, in Guangzhou, a megacity in south China's Pearl River Delta region, traffic-related PM, CO and NO_x frequently exceeded the guideline levels of Chinese National Air Quality Standard (Zhang and Mao, 1999; Wei et al., 1999; Fu et al., 2001; Qian et al., 2001) in the late 1990s when vehicles were fueled with gasoline and diesel oils: private vehicles and taxis were then all fueled with gasoline while public buses were fueled with diesel (Chan et al., 2003). High levels of hazardous air pollutants like particle-bound polycyclic aromatic hydrocarbons (PAHs) (Cheng et al., 1996; Bi et al., 2002) and aromatic hydrocarbons (Wang et al., 2002; Chan et al., 2003) in street canyons might pose health risks to commuters (Chan et al., 2002) and pedestrians (Zhao et al., 2004). Facing the deteriorating air quality, Guangzhou pioneered in mainland China to start converting all taxis and public buses to gasoline-LPG bi-fuel vehicles since 2003, following the practice in Hong Kong where dedicated LPGVs were introduced to replace diesel taxis since 1999 (Leung, 2011). Till 2014, the number of LPG-fueled taxis and buses in Guangzhou had reached 34.63 thousand (GZSB, 2015). Although these LPGVs only had a share of 1.38% in the total vehicle population, they accounted for 27% in the on-road vehicle fleet (Zhang et al., 2018a), 47% of the fleet-average vehicle kilometers traveled (VKT) (Zhang et al., 2013a) and consumed 9.2% of total fuel use in Guangzhou(GZSB, 2015). As for VOCs, this fuel conversion in Guangzhou has resulted in the decline of some hazardous air pollutants, like benzene, in ambient air; yet propane and butanes are becoming more and more abundant (Tang et al., 2007, 2008; Zhang et al., 2018a). Source apportionment results based on receptor models also revealed an increasingly important contribution of LPG to ambient VOCs in Guangzhou, such as 8–16% of ambient VOCs shared by LPG in 2004 (Liu et al., 2008) and 25.6% in 2008 (Zhang et al., 2012).

Very recently with the steady decline of ambient $PM_{2.5}$ in China's megacities, surface ozone instead has become an increasingly important air quality problem (Liu et al., 2018). For the control of ozone precursors (NO_x and VOCs), the use of LPG as an alternative fuel for vehicles is becoming a debatable issue. LPGVs may have higher NO_x emissions than conventional vehicles (Murillo et al., 2005; Yang et al., 2007; Zhang et al., 2013a). Meanwhile, results from a tunnel study in Guangzhou showed that LPG-related hydrocarbons, including propane, n-butane, and i-butane, already shared 59% of VOCs in the tunnel exhaust with estimated emission factors of propane over 30 times higher than that in Hong Kong (Zhang et al., 2018a). Although propane and butanes are not so photochemically reactive, high levels of these species accumulated in ambient air may also contribute substantially to ground-level O_3 formation (Blake and Rowland, 1995; Farmer et al., 2011; Lyu et al., 2016).

For policy-makers, extensive emission characterization for LPGVs is a prerequisite for taking any further regulatory or control steps. Characterizing emissions of NO_x and VOCs from LPGVs, however, is still far from enough. Schifter et al. (2000) obtained average emission factors of NO_x and hydrocarbons from retrofitted LPGVs under the Mexican official DGN-AA-II-1980 cycle. Chang et al. (2001) measured emission factors of 56 VOCs under the US FTP-75 cycle for vehicles using liquefied petroleum gas as an alternative fuel in Taipei. Guo et al. (2011) conducted chassis dynamometer tests at idling and 4 constant speeds to gain emission factors of individual VOC species from LPG taxis in Hong Kong. Myung et al. (2014) measured the emission factors of BTEX for the LPG-dedicated vehicles in Korea and found they varied greatly under NIER modes. While LPG consumption as alternative fuels is increasing in mainland China, very few reports are available for characterizing emissions from LPGVs except that a few studies reported compositions of VOCs released from LPGVs exhaust at idling (Lu et al., 2003; Wang et al., 2006; Lai et al., 2009).

In this study, emissions of NO_x and VOCs from LPG-fueled vehicles in Guangzhou were tested at idling and different driving speeds using a chassis dynamometer. The study aims to obtain emission factors of NO_x and VOCs for LPG taxis that are needed for assessing the potential impact of LPGV emissions on air quality and to gain source profiles of VOCs from LPGVs, and to explore the influence of speed and mileage on emissions from LPGVs.

2. Methodology

2.1. Descriptions of tested LPGVs

In this study, 26 LPG-fueled taxis were tested on a chassis dynamometer to attain the emission factors of CO_2, CO, NO_x ($NO+NO_2$), and VOCs. These taxis were randomly recruited from the in-use taxis fleet in Guangzhou. The taxis fleet in Guangzhou began to switch from gasoline fueling to LPG fueling in 2003 by retrofitting original gasoline engines to LPG-gasoline bi-fuel engines, and the retrofit for running taxis in Guangzhou was completed by the end of 2006. According to Guangzhou Municipal

Ecological Environment Bureau, annual VKT by taxis was approximately 14×10^4 km veh^{-1} yr^{-1}. As taxis are mandatorily scrapped after running at most 8 years, mileages of running taxis typically span (0–100) $\times 10^4$ km in Guangzhou. The tested vehicles in this study are all LPG-powered taxis with retrofitted bi-fuel engines. They were 1–5 years in vehicle age with mileages ranging (11.0–77.9) $\times 10^4$ km, largely representing the real on-road fleet of in-use LPG-fueled taxis in Guangzhou. The specific data for the selected taxis are listed in Table S1.

2.2. Chassis dynamometer testing system

The selected LPG-fueled taxis were tested using a chassis dynamometer (Foshan Analytical Instrument Co., Ltd, China) inside the campus of the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (GIG-CAS). In this study, exhausts from the taxis were tested at hot idling and six constant speeds of 10 km h^{-1}, 20 km h^{-1}, 30 km h^{-1}, 40 km h^{-1}, 50 km h^{-1}, and 60 km h^{-1}, respectively. The tested taxis were operated by an experienced driver from the local motor vehicle testing center. During a test, the chassis dynamometer control platform gives an accurate reading of parameters including speed and torque. The exhaust from a testing taxi was sampled into a 4L Teflon sampling bag (Delin Gas Packing Co., Ltd, Dalian, China) by an Apex sampling pump (CASELLA CEL, UK) at a flow rate of 0.5 L min^{-1} for 6 min. The air samples were used for the analysis of VOCs, CO, and CO_2. Trace gases including NO_x ($NO+NO_2$) in the exhaust were also directly measured by a flue gas analyzer (F-550, WOHLER, Germany) (Wu et al., 2020).

2.3. Air sample analysis

After statically diluted 1000 times by pure nitrogen, the air samples were analyzed for mixing ratios of VOCs with an automated Preconcentrator (Model 7100, Entech Instruments Inc., CA, USA) coupled with a gas chromatography (Model 6890)-mass selective detector (Model 5973N)/flame ionization detector/electron capture detector (GC-MSD/FID/ECD) (Agilent Technologies, CA, USA) (Wang and Wu, 2008; Zhang et al., 2012, 2013b). VOCs in the samples were identified by their retention times and mass spectra, and were quantified by an external calibration method (Zhang et al., 2012). Method detection limits for the VOCs were all below 50 pptv. CO concentrations were measured using a gas chromatograph (Model 6980, Agilent Technologies, CA, USA) with a flame ionization detector and a packed column (5A molecular sieve, 60/80mesh, 3m $\times$ 1/8 in.) (Zhang et al., 2012). CO_2 was analyzed with an HP 4890D gas chromatograph (Zhang et al., 2018a).

2.4. Quality assurance and quality control

Before sampling the exhaust from the LPG taxis, each sampling bag was cleaned by filling and evacuating pure nitrogen gas repeatedly at least five cycles. To check if the sampling bags were clean, the bags were again filled with humidified pure nitrogen at the end of the filling/evacuating cycles and stored in the laboratory for at least 24h, and then analyzed in the same way as samples. If all the target compounds were not found or were below the method detection limits, the bags were certificated as clean.

2.5. Emission factor

The emission factors for a species $\boldsymbol{p}$ emitted from taxis are calculated using the carbon mass balance method (Kirchstetter et al., 1996a, 1996b; Kean et al., 2000; Martins et al., 2006; Liu et al., 2014b; Deng et al., 2020; Zhang et al., 2020) as:

$$E_P = \frac{\Delta(P)}{\Delta(CO_2) + \Delta(CO) + \Delta(VOCs)} \times W_C \tag{1}$$

Where $\boldsymbol{E}_p$ is the emission factor for species $\boldsymbol{p}$, $\boldsymbol{W_C} = 0.821$ is the weight fraction of carbon in LPG with a propane/butanes ratio of 70:30 (Wang, 2010); $\Delta(\boldsymbol{P})$ is the background-corrected concentration of pollutant $\boldsymbol{p}$, and $\Delta(\boldsymbol{CO_2})$, $\Delta(\boldsymbol{CO})$, and $\Delta(\boldsymbol{VOCs})$ are the background-corrected carbon concentrations for CO_2, CO and VOCs, respectively. For the convenience of comparison with other studies, the fuel-based emission factors in g kg-$fuel^{-1}$ can be converted to mileage-based emission factors in g km^{-1}:

$$E_{P-mileage} = E_{p-fuel} \times G$$

Where G is the LPG consumption in kg per 100 km, and here we assume G = 6.88 kg/100 km (13 L/100 km) according to Guangzhou Municipal Ecological Environment Bureau.

3. Results and discussion

3.1. Emission factors of NO_x (E_{NOx})

As showed in Table 1, the average E_{NOx} at idling and at constant speeds of 10, 20, 30, 40, 50 and 60 km h^{-1} were 4.13 ± 4.21, 19.8 ± 28.5, 31.9 ± 51.2, 34.0 ± 45.9, 44.4 ± 53.1, 61.1 ± 58.5, 71.1 ± 60.4 g kg-$fuel^{-1}$, respectively. Largely due to the lack of after-treatment system or the inefficiency of after-treatment system (He et al., 2019), the tested taxis in this study had the highest E_{NOx} when compared to LPGVs in other studies (Table S2). Chikhi et al. (2014) found the average E_{NOx} of gasoline-LPG vehicles without catalyzers was 4–5 times after treatment. Lyu et al. (2016) also reported that a catalytic converter replacing the project implemented in Hong Kong dramatically reduced NO emissions from LPG-fueled vehicles by 88.6 ± 0.7%.

Fig. 1 shows the changes in E_{NOx} with driving speeds. The average E_{NOx} rose steadily from idling to 60 km h^{-1} with an increase of ~10.6 g kg-$fuel^{-1}$ per 10 km h^{-1} increase in speed. Chikhi et al. (2014) also observed a similar pattern of E_{NOx} versus speed when testing LPGVs without after-treatment devices. However, Lau et al. (2011) revealed a decreasing trend of E_{NOx} with speed for LPGVs with catalytic converters in Hong Kong. Even some LPG-taxis in Guangzhou did equip with catalytic converters, illegal tampering, and malfunction or deterioration of catalytic converters might also result in increasing E_{NOx} with speed, just like vehicles without after-treatment devices (Lau et al., 2011; Chikhi et al., 2014; He et al., 2019).

3.2. Emission factors of VOCs (E_{VOCs})

3.2.1. Emission factors

As shown in Table 1 and Fig. 2, on average the total emission factor of VOCs for the tested LPG taxis at idling was 8.24 g kg-$fuel^{-1}$, higher than those ranging 6.23–7.36 g kg-$fuel^{-1}$ when driving at constant speeds from 10 to 60 km h^{-1}.

In the exhausts, alkanes were the most abundant VOCs (71.9 ± 5.6%), followed by alkenes (25.2 ± 5.4%), while ethyne (2.7 ± 0.5%) and aromatic species (0.2 ± 0.0%) were minor components. These percentages were consistent with those from other studies (Schifter et al., 2000; Chang et al., 2001; Lu et al., 2003; Wang et al., 2006; Adam et al., 2011), except that in Hong Kong where Guo et al. (2011) observed higher emission factors of aromatic species than alkynes.

As the main components of LPG fuel (Schifter et al., 2000), propane and butanes accounted for more than 90% of the alkane

Table 1
Emission factors (kg kg-fuel^{-1} for CO_2, g kg-fuel^{-1} for the others) of CO_2, CO, NO_x, VOC species and OFPs at different speeds for tested vehicles (n = 26).

Compounds	Driving speeds						
	idling	10 km h^{-1}	20 km h^{-1}	30 km h^{-1}	40 km h^{-1}	50 km h^{-1}	60 km h^{-1}
CO_2	2.82 ± 0.291[a]	2.87 ± 0.235	2.89 ± 0.202	2.87 ± 0.219	2.87 ± 0.226	2.84 ± 0.228	2.87 ± 0.211
CO	100 ± 176	76.5 ± 139	63.6 ± 117	74.2 ± 130	75.9 ± 131	91.8 ± 130	74.0 ± 121
NO_x	4.13 ± 4.21	19.8 ± 28.5	31.9 ± 51.2	34.0 ± 45.9	44.4 ± 53.1	61.1 ± 58.5	71.1 ± 60.4
Alkyne							
Ethyne	0.201 ± 0.403	0.215 ± 0.422	0.193 ± 0.418	0.158 ± 0.368	0.142 ± 0.313	0.183 ± 0.383	0.172 ± 0.347
Sub-OFPs	0.187 ± 0.375	0.200 ± 0.393	0.179 ± 0.389	0.147 ± 0.342	0.132 ± 0.391	0.170 ± 0.356	0.160 ± 0.322
Alkenes							
Ethene	0.771 ± 1.30	1.11 ± 1.81	1.18 ± 1.93	1.23 ± 2.16	1.41 ± 2.87	0.953 ± 1.67	1.10 ± 1.75
Propene	0.297 ± 0.457	0.519 ± 0.758	0.515 ± 0.895	0.475 ± 0.893	0.627 ± 1.42	0.577 ± 0.923	0.524 ± 0.851
1-Butene (× 10^3)	39.3 ± 69.4	101 ± 324	43.4 ± 80.3	41.6 ± 81.3	54.5 ± 133.9	42.5 ± 77.5	42.6 ± 74.5
Trans-2-butene (× 10^3)	18.3 ± 46.6	84.2 ± 392	9.55 ± 21.4	8.63 ± 18.9	11.5 ± 28.7	8.33 ± 14.7	8.12 ± 14.9
Sub-total	1.13 ± 1.71	1.82 ± 2.55	1.75 ± 2.84	1.75 ± 3.06	2.10 ± 4.37	1.58 ± 2.49	1.68 ± 2.63
Sub-OFPs	10.8 ± 17.9	17.8 ± 33.4	16.8 ± 28.1	16.7 ± 30.2	20.1 ± 43.0	15.4 ± 26.1	16.1 ± 25.9
Alkanes							
Ethane	0.227 ± 0.319	0.189 ± 0.242	0.167 ± 0.205	0.168 ± 0.213	0.217 ± 0.267	0.201 ± 0.263	0.237 ± 0.385
Propane	4.09 ± 10.6	2.53 ± 3.89	2.46 ± 3.42	2.65 ± 4.32	3.16 ± 4.96	3.09 ± 4.79	2.62 ± 3.53
i-Butane	0.940 ± 2.43	0.558 ± 0.975	0.600 ± 0.853	0.598 ± 0.924	0.686 ± 1.04	0.693 ± 1.08	0.642 ± 0.912
n-Butane	1.63 ± 3.90	0.985 ± 1.73	1.02 ± 1.68	0.912 ± 1.47	1.01 ± 1.68	1.03 ± 1.77	0.977 ± 1.50
i-Pentane (× 10^3)	21.3 ± 38.9	24.8 ± 56.9	23.3 ± 52.2	23.4 ± 57.5	27.7 ± 43.1	28.3 ± 55	25.6 ± 51.4
Sub-total	6.91 ± 17.2	4.29 ± 6.73	4.27 ± 6.08	4.35 ± 6.83	5.10 ± 7.87	5.04 ± 7.79	4.50 ± 6.11
Sub-OFPs	4.83 ± 12.1	2.96 ± 4.94	3.01 ± 4.51	2.98 ± 4.78	3.44 ± 5.45	3.43 ± 5.52	3.11 ± 4.48
Aromatics							
Benzene (× 10^3)	5.70 ± 16.9	2.65 ± 9.91	3.60 ± 9.72	4.29 ± 15.0	5.92 ± 15.3	4.32 ± 12.9	3.29 ± 7.83
Toluene (× 10^3)	4.44 ± 8.99	9.94 ± 35.5	10.6 ± 36.2	11.0 ± 39.9	7.74 ± 21.1	6.64 ± 25.1	9.17 ± 38.8
Sub-total (× 10^3)	10.1 ± 23.0	12.6 ± 45.2	14.2 ± 44.7	15.3 ± 54.5	13.7 ± 35.8	11.0 ± 38.0	12.5 ± 45.6
Sub-OFP (× 10^3)	21.2 ± 46.6	40.4 ± 144	43.7 ± 147	45.5 ± 165	34.1 ± 92.5	28.8 ± 106	37.8 ± 156
Total VOCs	8.24 ± 18.0	6.33 ± 9.24	6.23 ± 8.74	6.27 ± 9.36	7.36 ± 11.3	6.82 ± 10.2	6.36 ± 8.89
Total OFPs	15.8 ± 30.4	21.0 ± 38.8	20.0 ± 33.2	19.8 ± 35.4	23.8 ± 48.8	19.1 ± 32.1	19.4 ± 30.9

[a] Mean±standard deviation.

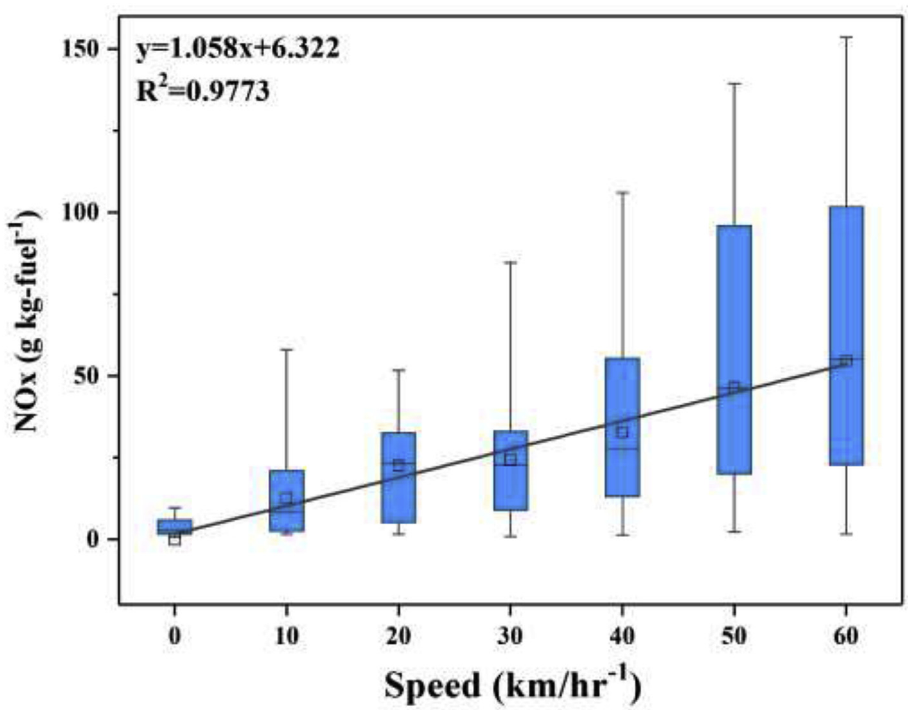

Fig. 1. Changes in average emission factors of NO_x (E_{NOx}) with driving speeds. Box charts present the following order from bottom to the top: 10%, 25%, median (square in the box), average (line in the box), 75%, and 90%. The average emission factors of NO_x at each speed were used to plot the linear regression line.

species and ~70% of total VOCs, as incompletely burned components emitted from the LPG taxis (Tang et al., 2007; Raslavicius et al., 2014). Propane was the dominant VOC species in the exhaust with an average emission factor ranging from 2.46 g kg-fuel^{-1} (20 km h^{-1}) to 4.09 g kg-fuel^{-1} (idling; Table 1), and it alone accounted for 39.5–49.6% of total VOC emissions. If converted to mileage-based emission factors (Table S3), it was approximately twice that reported in Hong Kong (Guo et al., 2011), and more than

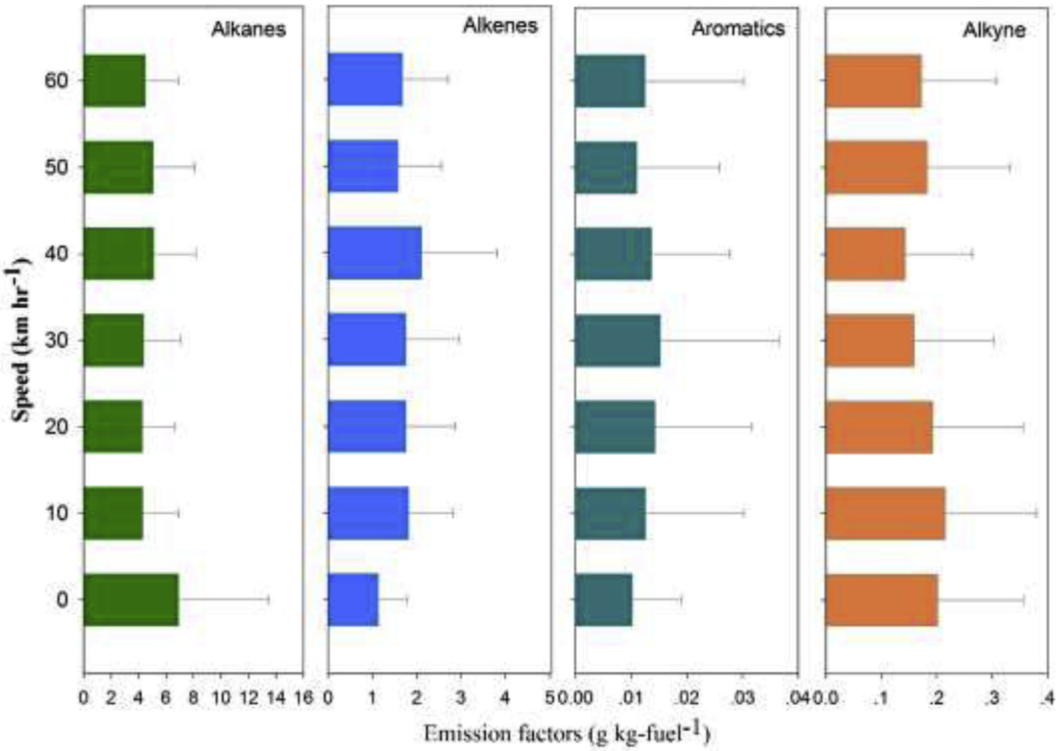

Fig. 2. Emission factors of VOC groups at idling and at constant speeds.

33 times of that reported in Varese, Italy (Adam et al., 2011), but ~28% lower than that reported in Taiwan (Chang et al., 2001).

Ethene accounted for more than 60% alkenes emitted from the LPG taxis. It was the second most abundant VOC species in the exhaust with average emission factors from 0.77 g kg-fuel^{-1} (idling) to 1.41 g kg-fuel^{-1} (40 km h^{-1}; Table 1), accounting for 16.3% of the total emissions of VOCs. This fraction was consistent with that of 16.6% from a study in Varese, Italy (Adam et al., 2011) or 13.4% in Taiwan (Chang et al., 2001), while it was quite lower when compared to that of 0.6% in Hong Kong (Guo et al., 2011). Multi-step aliphatic hydrocarbon oxidation mechanisms proposed by Hautman et al. (1981) suggested that aliphatic hydrocarbons like propane or butanes can pyrolyze into ethene as the main intermediate product in high temperature before they are oxidized to CO or CO_2. Therefore, without or with inefficient after-treatment devices, ethene might escape through the exhaust as an incomplete combustion product.

3.2.2. Influence of driving speeds on E_{VOCs}

Emission factors of the different VOC species from the tested LPG taxis at idling and constant speeds of 10–60 km h^{-1} are shown in Table 1 and Fig. 3. It seemed that emission factors of alkanes and alkenes changed in a different pattern with the increase of speeds. Propane, as an example of alkanes, had a higher emission factor at idling (4.09 g kg-fuel^{-1}), and its emission factors fluctuate from 2.53 g kg-fuel^{-1} (20 km h^{-1}) to 2.62 g kg-fuel^{-1} (60 km h^{-1}) when driving at constant speeds from 10 to 60 km h^{-1}. On the contrary, alkenes had lower emission factors at idling than at cruising. Ethene, as an example, had lower emission factors at idling (0.77 g kg-fuel^{-1}), and its emission factors increased with speed until it reached a peak at 40 km h^{-1} (1.41 g kg-fuel^{-1}), then dropped to 1.10 g kg-fuel^{-1} at 60 km h^{-1}. These findings were not consistent with the results from the study by Guo et al. (2011), who found that emission factors of either dominant VOC species, like propane and ethene, or the total VOCs dropped dramatically from 25 km h^{-1} to 50 km h^{-1}, and then became stable at 70 and 100 km h^{-1}. The different trends may be due to the difference in the combustion performance between gasoline-LPG bi-fuel LPG taxis in our study and dedicated LPG taxis in Hong Kong (Guo et al., 2011; Table S5), as a fuel-air mixture of gasoline-LPG bi-fuel engine is often too lean within the higher ranges of engine's rotational speed, which demonstrates the difficulties of ignition, flame extinction, and incomplete combustion and causes a huge increase in levels of emissions (Raslavicius et al., 2014). Moreover, Lyu et al. (2016) also pointed out that replacement of catalytic converter of LPG-fueled taxis in Hong Kong helped greatly in mitigating VOCs emission, so installation, replacement and maintenance of after-treatment devices are of great importance to reduce emissions.

3.3. Relationships between mileages and emission factors

The testes LPG-fueled taxis had model years from 1 to 5 years, and their mileages ranged (11.0–77.9) × 10^4 km. Average $\boldsymbol{E}_{NOx}$ showed an increasing trend with mileages ($\boldsymbol{p} < 0.05$) (Fig. 4) at a slope of ~0.56 g kg-fuel^{-1} per 10^4 km of mileage. This increasing trend seemed to be more significant ($\boldsymbol{p} < 0.01$) between average $\boldsymbol{E}_{VOCs}$ and mileages. However, if we look further into the relationships between $\boldsymbol{E}_{VOCs}$ and mileages, average $\boldsymbol{E}_{VOCs}$ were found to be less than 2.0 g kg-fuel^{-1} when mileages were below 40 × 10^4 km, while they varied from 0.258 to 30.2 g kg-fuel^{-1} when mileages were above 40 × 10^4 km (Fig. 4). The variation in $\boldsymbol{E}_{VOCs}$ between these two mileage ranges may be affected by the maintenance of the vehicles (Chikhi et al., 2014; Tsai et al., 2017), which can be indicated by the CO/CO_2 ratio (Pierson et al., 1996; Chikhi et al., 2014). For LPG taxis with mileages below 40 × 10^4 km, the CO/CO_2 ratios were less than 5%, indicating good maintenance with lower VOC emissions; while for LPG taxis with mileages above 40 × 10^4 km, the CO/CO_2 ratios varied from 0.010–17.4%, indicating

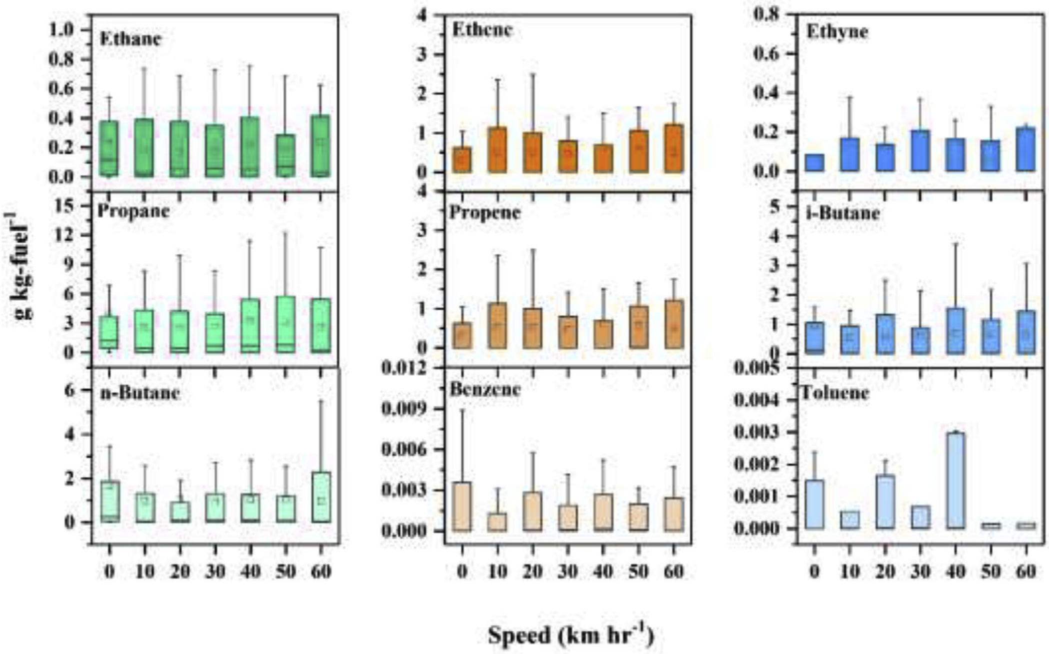

Fig. 3. Average emission factors of individual VOCs at idling and at constant speeds.

bad maintenance for some taxis with higher emissions of VOCs. Tsai et al. (2017) proposed that for gasoline motorcycles above a guaranteed mileage point, the engine and after-treatment system could deteriorate and cause increasing emissions. This study suggests that enhanced maintenance and inspection of LPG taxis at a certain guaranteed mileage, such as 40 × 10^4 km in this study, would greatly reduce emissions of VOCs.

3.4. Environmental implications

LPG is widely recognized for its inherent environmental benefits, replacing a diesel/gasoline engine with an LPG-fueled equivalent would greatly reduce the emission of damaging particulates, reactive organic gases (ozone/SOA precursors) and toxic air pollutants (such as PAHs, benzene, and formaldehyde) in exhaust emission or during refueling. However, as indicated in this study, if engines are not LPG-dedicated and/or with poor exhaust after-treatment, these benefits would be discounted by higher NO_x emission and excessive propane/butanes in the exhaust.

From the tests in this study, the emission factors (EFs) of NO_x from LPG taxis increased linearly with speeds, therefore at a driving speed of 35 km h^{-1} the EF of NO_x could be estimated as 2.69 g km^{-1}, the average of that at 30 km h^{-1} (34.0 g kg^{-1} or 2.34 g km^{-1}; Table 1) and at 40 km^{-1} (44.4 g kg^{-1} or 3.05 g km^{-1}; Table 1). This average EF of NO_x (2.69 g km^{-1}) at 35 km h^{-1} is about twice of 1.29 g km^{-1} per vehicle for total car fleet at an average speed of ~35 km h^{-1} based on tests in a busy urban tunnel in 2014 in Guangzhou (Zhang et al., 2015). It is worth noting that this study was mainly conducted in 2018, and EF of NO_x from gasoline or diesel cars should have decreased drastically in recent 5 years due to tightened emission limits with the updates of emission standards. A recent test in the same urban tunnel in November 2019 revealed that the average EF of NO_x for car fleet was 0.24 g km^{-1}, far below that of 2.69 g km^{-1} at a driving speed of 35 km h^{-1} as estimated from this study. In fact, this average EF is also much higher that the reported EFs of NO_x ranging 0.010–1.1 g km^{-1} (Table S2) for gasoline vehicles (Ntziachristos and Samaras, 2000; Heeb et al., 2006; Pelkmans and Debal, 2006; André and Rapone, 2009; Adam et al., 2011; Hu et al., 2012; Yang et al., 2018); Although EFs of NO_x for LPG taxis (1.4–4.9 g km^{-1} at 10–60 km h^{-1}) are much lower than those of 10.7–21.8 g km^{-1} reported about 20 years ago for heavy trucks (Wang et al., 1997, 2000; Ramamurthy and Clark, 1999; Prucz et al., 2001), they are even much higher than those of 0.52–1.7 g km^{-1} reported for small or medium trucks when the LPG taxis are driving at 40 km h^{-1} or above (Joumard et al., 2003; André and Rapone, 2009; Chiang et al., 2012; Hu et al., 2012; Chikhi et al., 2014). Therefore, the transition of a gasoline taxi to a taxi with an LPG-gasoline bi-fuel engine and without or with inefficient exhaust after-treatment would not benefit the emission control of NO_x.

LPG is mainly composed of propane and butanes, which are far less reactive and toxic when compared to the constituents of gasoline, therefore replace gasoline-powered taxis with LPG taxis are supposed to lower the ozone formation potentials (OFPs) and hazardous air pollutants in the exhaust. From the study air toxics like aromatic hydrocarbons did become trivial in the exhaust of LPG taxis. As for the OFPs calculated based on the maximum increment reactivity (MIR) method developed by Carter (1994, 2009), they ranged 19.1–23.8 g kg-fuel^{-1}, or 1.3–1.6 g km^{-1}, when cruising at speeds from 10 to 60 km h^{-1}, much higher than that at idling (15.8 g kg-fuel^{-1} or 1.1 g km^{-1}) largely due to higher emission of more reactive alkenes when cruising (Table 1). This is quite different from the results for gasoline vehicles, which have much more emissions of reactive alkenes and aromatics at idling than at cruising (e.g. Zhang et al., 2020). Similarly, if we estimate the EF and the OFP at 35 km h^{-1} as the average of that at 30 km h^{-1} and 40 km h^{-1} (Table 1), LPG taxis at 35 km h^{-1} would have an average EF of VOCs as 6.82 g kg-fuel^{-1} or 0.47 g km^{-1}, similar to that of 0.45 g km^{-1} per vehicle tested in the Zhujiang Tunnel for vehicle fleet in urban Guangzhou (Zhang et al., 2018a). However, the estimated OFPs of VOCs from LPG taxis at 35 km h^{-1}, 21.8 g kg-fuel^{-1} or 1.5 g km^{-1}, was 1.5 times of that 1.0 g km^{-1} reported for vehicle fleet based on the tunnel tests in 2014 (Zhang et al., 2018a). It is also worth noting that based on tests in the same Zhujiang Tunnel, average OFPs of

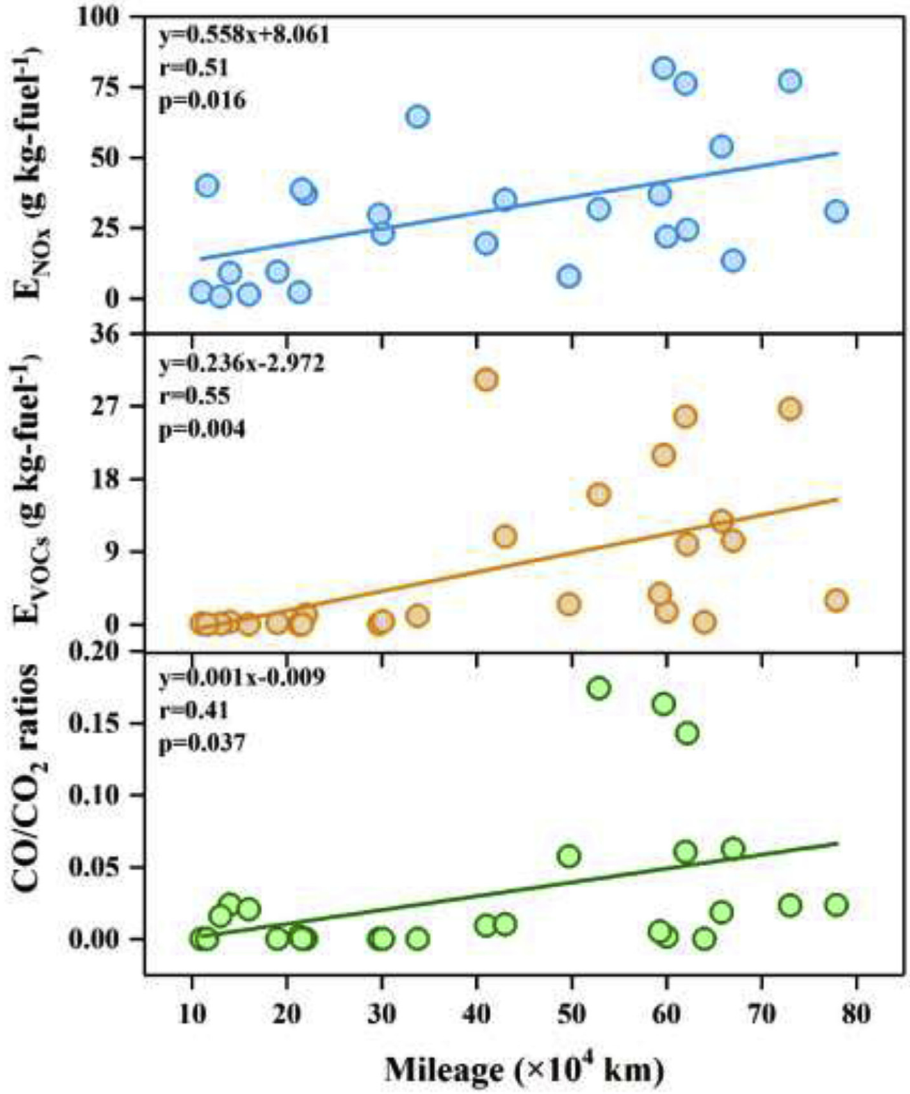

Fig. 4. Correlations of between mileage and NO_x emission factor, VOCs emission factor and CO/CO_2 ratio of LPG-fueled taxis.

VOCs in terms of mg km^{-1} per vehicle decreased by ~50% during 2004–2014, the decrease rate from 2014 to 2018 should be more rapid due to update of fuel quality standards and vehicle emission standards. Therefore, replacing gasoline taxis with the LPG-gasoline bi-fuel taxis without or with inefficient exhaust after-treatment did not show the potentials to reduce reactive VOCs as ozone precursors, largely due to relatively higher emission of reactive alkenes (Table 1). In fact, we can also compare the emissions of reactive VOCs in terms of OFPs with those for on-road vehicles based on tunnel tests in other parts of the world (Table S4). Tested OFPs for the LPG taxis in this study ranged 1.1–1.6 g km^{-1}; they were also much higher when compared to that of 0.15 g km^{-1} per vehicle as observed in the Leopold II Tunnel in Brussels, Belgium for vehicle fleet without LPG cars (Ait-Helal et al., 2015), 0.18 g km^{-1} per vehicle as observed in the Shing Mun Tunnel in Hong Kong (Cui et al., 2018), 0.74 g km^{-1} per vehicle observed in the Hsuehshan tunnel in Taiwan (Liu et al., 2014c), or even 0.44 g km^{-1} per vehicle observed in the Fu Gui Mountain tunnel in Nanjing (Zhang et al., 2018b), and only ~20% lower than that of 1.93 g km^{-1} per vehicle observed in Loma Larga Tunnel in Monterrey, Mexico (Araizaga et al., 2013).

The unsatisfactory performance of the LPG taxis in the emissions of ozone precursors (NO_x and reactive VOCs) is rooted in the fact that the LPG taxis in this study are LPG-gasoline bi-fuel ones without catalytic converters or with inefficient ones. If the LPG-gasoline bi-fuel taxis are ones with dedicated-LPG engines and efficient after-treatment devices as did in Hong Kong, their emission of NO_x and VOCs would be largely reduced (Guo et al., 2011; Lau et al., 2011; Lyu et al., 2016).

4. Conclusions

In this study, 26 LPG-fueled taxis in Guangzhou were tested at idling or cruising at speeds from 10 to 60 km h^{-1} using a chassis dynamometer for their emissions of NO_x and VOCs, both of which are ozone precursors. The results revealed that the $\boldsymbol{E}_{NOx}$ increased with speed steadily from 4.13 ± 4.21 g kg-fuel^{-1} to 71.1 ± 60.4 g kg-fuel^{-1} by approximately 10.6 g kg-fuel^{-1} on average for every 10 km h^{-1} increase in speed; $\boldsymbol{E}_{VOCs}$ of the LPG taxis were higher at idling (8.24 g kg-fuel^{-1}) than when cruising at 10–60 km h^{-1} (6.22–7.36 g kg-fuel^{-1}), but the OFPs at idling (15.8 g kg-fuel^{-1}) was lower than that of 19.1–23.8 g kg-fuel^{-1} when cruising at speeds from 10 to 60 km h^{-1}, largely due to higher emission of more reactive alkenes when cruising. These results also suggested that replacing the gasoline-power taxis with the LPG-powered ones with LPG-gasoline bi-fuel engines and inefficient after-treatment is not a successful approach to reduce the emission of NO_x and reactive VOCs, and therefore would not benefit the surface ozone pollution control, and instead replacing the gasoline-powered taxis with the LPG-ones with dedicated LPG engines and efficient after-treatment converters, as the case in Hong Kong, would greatly lower the emissions of reactive VOCs and NO_x. Our study also revealed that when the $\boldsymbol{E}_{VOCs}$ were found to be less than 2.0 g kg-fuel^{-1} when mileages were below 40×10^4 km, otherwise they ranged 0.258–30.2 g kg-fuel^{-1}, suggesting that enhancing the maintenance and inspection might help curb emissions of VOCs when mileages were above a guaranteed mileage point.

It is worth noting that we only investigated emissions under idle and cruising modes. We did not test emissions during criteria driving cycles or conducted on-board emission measurements for real-world driving, and we did not further test emissions under the acceleration and deceleration modes, largely due to that we wanted to measure specific VOC species to assess their OFPs. However, as indicated in previous studies (e.g., Lau et al., 2011), emissions of hydrocarbons from LPG taxis under acceleration and deceleration modes might be near that under cruising mode, and the emissions of NO_x under cruising might lay between that under the acceleration and deceleration modes, therefore the conclusion that the studied LPG taxis do not benefit ambient ozone pollution control still stands.

Authorship Contribution Statement

Jingjing Feng: Writing - original draft, Methodology, Formal analysis, Writing - review & editing. Yanli Zhang: Conceptualization, Writing, Methodology, Formal analysis, Writing - review & editing. Wei Song: Investigation, Data curation, Resources. Wei Deng: Investigation, Data curation, Resources. Ming Zhu: Investigation, Data curation, Resources. Zheng Fang: Investigation, Data curation. Yuqing Ye: Investigation, Data curation. Hua Fang: Investigation, Data curation. Zhenfeng Wu: Investigation, Data curation. Scott Lowther: Writing - review & editing. Kelvin C. Jones: Writing - review & editing. Xinming Wang: Project administration, Conceptualization, Writing, Funding acquisition, Supervision- review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (Grant No. 41530641/41673116/41961144029), the National Key Research and Development Program (2016YFC0202204), the Chinese Academy of Sciences (Grant No. QYZDJ-SSW-DQC032/XDA23010303), Youth Innovation Promotion Association, CAS (2017406) and Guangdong Foundation for Program of Science and Technology Research (Grant No. 2017B030314057/2019B121205006).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115623.

References

Adam, T.W., Astorga, C., Clairotte, M., Duane, M., Elsasser, M., Krasenbrink, A., Larsen, B.R., Manfredi, U., Martini, G., Montero, L., Sklorz, M., Zimmermann, R., Perujo, A., 2011. Chemical analysis and ozone formation potential of exhaust from dual-fuel (liquefied petroleum gas/gasoline) light duty vehicles. Atmos. Environ. 45 (17), 2842–2848.

Ait-Helal, W., Beeldens, A., Boonen, E., Borbon, A., Boréave, A., Cazaunau, M., Chen, H., Daële, V., Dupart, Y., Gaimoz, C., Gallus, M., George, C., Grand, N., Grosselin, B., Herrmann, H., Ifang, S., Kurtenbach, R., Maille, M., Marjanovic, I., Mellouki, A., Miet, K., Mothes, F., Poulain, L., Rabe, R., Zapf, P., Kleffmann, J., Doussin, J.F., 2015. On-road measurements of NMVOCs and NO_x: determination of light-duty vehicles emission factors from tunnel studies in Brussels city center. Atmos. Environ. 122, 799–807.

André, M., Rapone, M., 2009. Analysis and modelling of the pollutant emissions from European cars regarding the driving characteristics and test cycles. Atmos. Environ. 43, 986–995.

Araizaga, A.E., Mancilla, Y., Mendoza, A., 2013. Volatile organic compound emissions from light-duty vehicles in monterrey, Mexico: a tunnel study. Int. J. Environ. Res. 7 (2), 277–292.

Baker, A.K., Beyersdorf, A.J., Doezema, L.A., Katzenstein, A., Meinardi, S., Simpson, I.J., Blake, D.R., Rowland, F.S., 2008. Measurements of nonmethane hydrocarbons in 28 United States cities. Atmos. Environ. 42 (1), 170–182.

Bi, X.H., Sheng, G.Y., Peng, P.A., Zhang, Z.Q., Fu, H.M., 2002. Extractable organic matter in PM_{10} from Liwan district of Guangzhou city, PR China. Sci. Total Environ. 300 (1–3), 213–228.

Blake, D.R., Rowland, F.S., 1995. Urban leakage of liquefied petroleum gas and its impact on Mexico City air quality. Science 269, 953–956.

Carter, W.P.L., 1994. Development of ozone reactivity scales for volatile organic compounds. J. Air Waste Manag. Assoc. 44, 881~899.

Carter, W.P.L., 2009. Update Maximum Incremental Reactivity Scale and Hydrocarbon Bin Reactivities for Regulatory Application. California Air Resources Board. Contract 07-339.

Chan, C.Y., Chan, L.Y., Wang, X.M., Liu, Y.M., Lee, S.C., Zou, S.C., Sheng, G.Y., Fu, J.M., 2002. Volatile organic compounds in roadside microenvironments of metropolitan Hong Kong. Atmos. Environ. 36 (12), 2039–2047.

Chan, L.Y., Lau, W.L., Wang, X.M., Tang, J.H., 2003. Preliminary measurements of aromatic VOCs in public transportation modes in Guangzhou, China. Environ. Int. 29 (4), 429–435.

Chan, C.K., Yao, X.H., 2008. Air pollution in mega cities in China. Atmos. Environ. 42 (1), 1–42.

Chang, C.C., Lo, J.G., Wang, J.L., 2001. Assessment of reducing ozone forming potential for vehicles using liquefied petroleum gas as an alternative fuel. Atmos. Environ. 35 (35), 6201–6211.

Cheng, Y., Sheng, G.Y., Fu, J.M., Min, Y.S., 1996. Quantitative analysis of polycyclic aromatic hydrocarbons (PAHs) in aerosol. Environ. Chem. 15 (4), 360–365 (in Chinese).

Chiang, H.L., Lai, Y.M., Chang, S.Y., 2012. Pollutant constituents of exhaust emitted from light-duty diesel vehicles. Atmos. Environ. 47, 399–406.

Chikhi, S., Boughedaoui, M., Kerbachi, R., Joumard, R., 2014. On-board measurement of emissions from liquefied petroleum gas, gasoline and diesel powered passenger cars in Algeria. J. Environ. Sci. (China) 26 (8), 1651–1659.

Cui, L., Liang, X., Fai, K., Gao, Y., Liu, C., Sai, S., Ho, H., Wei, H., Cheng, S., Ming, X., Qiong, B., Huang, Y., Chow, J.C., Watson, J.G., Chen, L.W., 2018. Decrease of VOC emissions from vehicular emissions in Hong Kong from 2003 to 2015: results from a tunnel study. Atmos. Environ. 177, 64–74.

Deng, W., Deng, W., Fang, Z., Wang, Z., Zhu, M., Zhang, Y., Tang, M., Song, W., Lowther, S., Huang, Z., Jones, K., Peng, P.A., Wang, X., 2020. Primary emissions and secondary organic aerosol formation from in-use diesel vehicle exhaust: comparison between idling and cruise mode. Sci. Total Environ. 699, 134357.

Eiguren-Fernandez, A., Miguel, A.H., Froines, J.R., Thurairatnam, S., Avol, E.L., 2004. Seasonal and spatial variation of polycyclic aromatic hydrocarbons in vapor-phase and $PM_{2.5}$ in Southern California urban and rural communities. Aerosol Sci. Technol. 38 (5), 447–455.

Farmer, D.K., Perring, A.E., Wooldridge, P.J., Blake, D.R., Baker, A., Meinardi, S., Huey, L.G., Tanner, D., Vargas, O., Cohen, R.C., 2011. Impact of organic nitrates on urban ozone production. Atmos. Chem. Phys. 11 (9), 4085–4094.

Fu, L.X., Hao, J.M., He, D.Q., He, K.B., Li, P., 2001. Assessment of vehicular pollution in China. J. Air Waste Manag. Assoc. 51, 658–668.

Gentner, D.R., Jathar, S.H., Gordon, T.D., Bahreini, R., Day, D.A., Haddad, I.E., Hayes, P.L., Pieber, S.M., Platt, S.M., Joost, D.G., Goldstein, A.H., Harley, R.A., Jimenez, J.L., Prévôt, A.S.H., Robinson, A.L., 2017. Review of urban secondary organic aerosol formation from gasoline and diesel motor vehicle emissions. Environ. Sci. Technol. 51 (3), 1074–1093.

Guangzhou Statistics Bureau (GZSB), 2015. Guangzhou Statistical Yearbook. China Statistics Press, Beijing (in Chinese).

Guo, H., Zou, S.C., Tsai, W.Y., Chan, L.Y., Blake, D.R., 2011. Emission characteristics of nonmethane hydrocarbons from private cars and taxis at different driving speeds in Hong Kong. Atmos. Environ. 45 (16), 2711–2721.

Hautman, D.J., Dryer, F.L., Schug, K.P., Glassman, I., 1981. A multiple-step overall kinetic mechanism for the oxidation of hydrocarbons. Combust. Sci. Technol. 25 (5–6), 219–235.

He, J.J., Wu, L., Mao, H.J., Liu, H.L., Jing, B.Y., Yu, Y., Ren, P.P., Feng, C., Liu, X.H., 2016. Development of a vehicle emission inventory with high temporal-spatial

resolution based on NRT traffic data and its impact on air pollution in Beijing - Part 2: impact of vehicle emission on urban air quality. Atmos. Chem. Phys. 16 (5), 3171–3184.
He, L.Q., Hu, J.G., Yang, L.H.Z., Li, Z.H., Zheng, X., Xie, S.X., Zu, L., Chen, J.H., Li, Y., Wu, Y., 2019. Real-world gaseous emissions of high-mileage taxi fleets in China. Sci. Total Environ. 659, 267–274.
Heeb, N.V., Forss, A.M., Brühlmann, S., Lüscher, R., Saxer, C.J., Hug, P., 2006. Three-way catalyst-induced formation of ammonia—velocity- and acceleration-dependent emission factors. Atmos. Environ. 40, 5986–5997.
Ho, K.F., Lee, S.C., Ho, W.K., Blake, D.R., Cheng, Y., Li, Y.S., Ho, S.S.H., Fung, K., Louie, P.K.K., Park, D., 2009. Vehicular emission of volatile organic compounds (VOCs) from a tunnel study in Hong Kong. Atmos. Chem. Phys. 9 (19), 7491–7504.
Hu, J.N., Wu, Y., Wang, Z.S., Li, Z.H., Zhou, Y., Wang, H.T., Bao, X.F., Hao, J.M., 2012. Real-world fuel efficiency and exhaust emissions of light-duty diesel vehicles and their correlation with road conditions. J. Environ. Sci. 24 (5), 865–874.
Jang, W., Ko, A., Baelk, S.H., Jin, D.Y., Choi, K., Myung, C.L., Park, S., 2014. Study of regulated emissions and nanoparticle characteristics of light-duty direct-injection vehicles fuelled with gasoline and liquefied petroleum gas in the New European Driving Cycle and the Federal Test Procedure 75 driving cycle. Proc IMechE Part D 229, 25–37.
Joumard, R., André, M., Vidon, R., Tassel, P., 2003. Characterizing real unit emissions for light duty goods vehicles. Atmos. Environ. 37, 5217–5225.
Karamangil, M.I., 2007. Development of the auto gas and LPG-powered vehicle sector in Turkey: a statistical case study of the sector for Bursa. Energy Pol. 35 (15), 640–649.
Kean, A.J., Harley, R.A., Littlejohn, D., Kendall, G.R., 2000. On-road measurement of ammonia and other motor vehicle exhaust emissions. Environ. Sci. Technol. 34 (17), 3535–3539.
Kirchstetter, T.W., Singer, B.C., Harley, R.A., Kendall, G.B., Chan, W., 1996a. Impact of oxygenated gasoline use on California light-duty vehicle emissions. Environ. Sci. Technol. 30 (2), 661–670.
Kirchstetter, T.W., Harley, R.A., Kreisberg, N.M., Stolzenburg, M.R., Hering, S.V., 1999b. On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles. Atmos. Environ. 33 (18), 2955–2968.
Lai, C.H., Chang, C.C., Wang, C.H., Shao, M., Zhang, Y.H., Wang, J.L., 2009. Emissions of liquefied petroleum gas (LPG) from motor vehicles. Atmos. Environ. 43 (7), 1456–1463.
Lau, J., Hung, W.T., Cheung, C.S., 2011. On-board gaseous emissions of LPG taxis and estimation of taxi fleet emissions. Sci. Total Environ. 409, 5292–5300.
Leung, V., 2011. Slow diffusion of LPG vehicles in China - lessons from shanghai, Guangzhou, and HongKong. Energy Pol. 39 (6), 3720–3731.
Liu, H., Liu, S., Xue, B., Lv, Z.F., Meng, Z.H., Yang, X.F., Xue, T., Yu, Q., He, K.B., 2018. Ground-level ozone pollution and its health impacts in China. Atmos. Environ. 173, 223–230.
Liu, T.Y., Wang, X.M., Wang, B.G., Ding, X., Deng, W., Lü, S.J., Zhang, Y.L., 2014b. Emission factor of ammonia (NH_3) from on-road vehicles in China: tunnel tests in urban Guangzhou. Environ. Res. Lett. 9 (6), 064027.
Liu, W.T., Chen, S.P., Chang, C.C., Ou-yang, C.F., 2014c. Assessment of carbon monoxide (CO) adjusted non-methane hydrocarbon (NMHC) emissions of a motor fleet – a long tunnel study. Atmos. Environ. 89, 403–414.
Liu, Y., Shao, M., Zhang, J., Fu, L., Lu, S., 2005. Distributions and source apportionment of ambient volatile organic compounds in Beijing city, China. J. Environ. Sci. Health Part A Toxic/Hazardous Subst. Environ. Eng. 40 (10), 1843–1860.
Liu, Y., Shao, M., Lu, S., Chang, C.C., Wang, J.L., Fu, L., 2008. Source apportionment of ambient volatile organic compounds in the Pearl River Delta, China: Part II. Atmos. Environ. 42 (25), 6261–6274.
Liu, Z.R., Hu, B., Liu, Q., Sun, Y., Wang, Y.S., 2014a. Source apportionment of urban fine particle number concentration during summertime in Beijing. Atmos. Environ. 96, 359–369.
Lu, S.H., Bai, Y.H., Zhang, G.H., Ma, J., 2003. Study on the characteristics of VOCs source profiles of vehicle exhaust and gasoline emission. Acta Sci. Nat. Univ. Pekin. 39 (4), 507–511 (in Chinese).
Lyu, X.P., Guo, H., Simpson, I.J., Meinardi, S.L., Peter, K.K., Ling, Z.H., Wang, Y., Liu, M., Luk, C.W.Y., Wang, N., Blake, D.R., 2016. Effectiveness of replacing catalytic converters in LPG-fueled vehicles in Hong Kong. Atmos. Chem. Phys. 16 (10), 6609–6626.
Mishra, R.K., Shukla, A., Parida, M., Pandey, G., 2016. Urban roadside monitoring and prediction of CO, NO_2 and SO_2 dispersion from on-road vehicles in megacity Delhi. Transp. Res. D Transp. Environ. 46, 157–165.
Martins, L.D., Andrade, M.F.D., Freitas, E., Pretto, A., Gatti, L.V., Albuquerque, E.L., Tomaz, E., Guardani, M.L., Martins, M., Junior, O.M.A., 2006. Emission factors for gas-powered vehicles traveling through road tunnels in Sao Paulo, Brazil. Environ. Sci. Technol. 40 (21), 6722–6729.
Murillo, S., Míguez, J.L., Porteiro, J., López González, L.M., Granada, E., Morán, J.C., 2005. LPG: pollutant emission and performance enhancement for spark-ignition four strokes outboard engines. Appl. Therm. Eng. 25 (13), 1882–1893.
Myung, C.L., Lee, H., Choi, K., Lee, Y.J., Parki, S., 2009. Effects of gasoline, diesel, LPG and low-carbon fuels and nanoparticle emission characteristics in light-duty vehicles. Int. J. Automot. Technol. 10 (5), 537–544.
Myung, C.L., Choi, K., Kim, Y.S., Lee, J., Park, S., 2012. Comparative study of regulated and unregulated toxic emissions characteristics from a spark ignition direct injection light-duty vehicle fueled with gasoline and liquid phase LPG (liquefied petroleum gas). Energy 44 (1), 189–196.
Myung, C.L., Ko, A., Lim, Y.S., Kim, S.M., Lee, J., Choi, K., Park, S., 2014. Mobile source air toxic emissions from direct injection spark ignition gasoline and LPG passenger car under various in-use vehicle driving modes in Korea. Fuel Process. Technol. 199, 19–31.
Namdeo, A., Bell, M.C., 2005. Characteristics and health implications of fine and coarse particulates at roadside, urban background and rural sites in UK. Environ. Int. 31, 565–573.
National Research Council, 1991. Rethinking the Ozone Problem in Urban and Regional Air Pollution. National Academy Press, Washington, DC.
Ning, Z., Chan, T.L., 2007. On-road remote sensing of liquefied petroleum gas (LPG) vehicle emissions measurement and emission factors estimation. Atmos. Environ. 41, 9099–9110.
Ntziachristos, L., Samaras, Z., 2000. Speed-dependent representative emission factors for catalyst passenger cars and influencing parameters. Atmos. Environ. 34, 4611–4619.
Pelkmans, L., Debal, P., 2006. Comparison of on-road emissions with emissions measured on chassis dynamometer test cycles. Transport. Res. Part D 11 (4), 233–241.
Pierson, W.R., Gertler, A.W., Robinson, N.F., Sagebiel, J.C., Zielinska, B., Bishop, G.A., Stedman, D.H., Zweidinger, R.B., Ray, W.D., 1996. Real-world automotive emissions – summary of studies in the fort McHenry and tuscarora mountain tunnels. Atmos. Environ. 30 (12), 2233–2256.
Prucz, J.C., Clark, N.N., Gautam, M., Lyons, D.W., 2001. Exhaust emissions from engines of the Detroit Diesel Corporation in transit buses: a decade of trends. Environ. Sci. Technol. 35 (9), 1755–1764.
Pulkrabek, W.W., 1997. Engineering Fundamentals of the Internal Combustion Engine. Prentice-Hall, New York, NY, USA.
Qian, Z.M., Zhang, J.F., Wei, F.S., Wilson, E.W., Chapman, R.S., 2001. Long-term ambient air pollution levels in four Chinese cities: inter-city and intra-city concentration gradients for epidemiological studies. J. Expo. Anal. Environ. Epidemiol. 11 (5), 341–351.
Ramamurthy, R., Clark, N.N., 1999. Atmospheric emissions inventory data for heavy-duty vehicles. Environ. Sci. Technol. 33 (1), 55–62.
Raslavicius, L., Kersys, A., Mockus, S., Kersiene, N., Starevicius, M., 2014. Liquefied petroleum gas (LPG) as a medium-term option in the transition to sustainable fuels and transport. Renew. Sustain. Energy Rev. 32, 513–525.
Ristovski, Z.D., Jayaratne, E.R., Morawska, L., Ayoko, G.A., Lim, M., 2005. Particle and carbon dioxide emissions from passenger vehicles operating on unleaded petrol and LPG fuel. Sci. Total Environ. 345, 93–98.
Schifter, I., Díaz, L., López-Salinas, E., Rodríguez, R., Avalos, S., Guerrero, V., 2000. An evaluation of the lpg vehicles program in the metropolitan area of Mexico city. J. Air Waste Manag. Assoc. 50, 301–309.
Song, X.Y., Xie, S.D., 2006. Development of vehicle emission inventory in China. J. Environ. Sci. 27, 1041–1045 (in Chinese).
Tang, J.H., Chan, L.Y., Chan, C.Y., Li, Y.S., Chang, C.C., Liu, S.C., Wu, D., Li, Y.D., 2007. Characteristics and diurnal variations of NMHCs at urban, suburban, and rural sites in the Pearl River Delta and a remote site in South China. Atmos. Environ. 41 (38), 8620–8632.
Tang, J.H., Chan, L.Y., Chan, C.Y., Li, Y.S., Chang, C.C., Wang, X.M., Zou, S.C., Barletta, B., Blake, D.R., Wu, D., 2008. Implications of changing urban and rural emissions on non-methane hydrocarbons in the Pearl River Delta region of China. Atmos. Environ. 42 (16), 3780–3794.
Tsai, J.H., Huang, P.H., Chiang, H.L., 2017. Air pollutants and toxic emissions of various mileage motorcycles for ECE driving cycles. Atmos. Environ. 153, 126–134.
Wang, B.G., Shao, M., Zhang, Y.H., Lv, W.M., Liu, H.X., Zhang, N., 2006. A study of volatile organic compounds and its emission factor from city vehicles. Res. Environ. Sci. 19, 75–80 (in Chinese).
Wang, W.G., Clark, N.N., Lyons, D.W., Yang, R.M., Gautam, M., Bata, R.M., Loth, J.L., 1997. Emissions comparisons from alternative fuel buses and diesel buses with a chassis dynamometer testing facility. Environ. Sci. Technol. 31 (11), 3132–3137.
Wang, W.G., Lyons, D.W., Clark, N.N., Gautam, M., Norton, P.M., 2000. Emissions from nine heavy trucks fueled by diesel and biodiesel blend without engine modification. Environ. Sci. Technol. 34 (6), 933–939.
Wang, X.M., Sheng, G.Y., Fu, J.M., Chan, C.Y., Lee, S.C., Chan, L.Y., Wang, Z.S., 2002. Urban roadside aromatic hydrocarbons in three cities of the Pearl River Delta, People's Republic of China. Atmos. Environ. 36 (33), 5141–5148.
Wang, X.M., Wu, T., 2008. Release of isoprene and monoterpenes during the aerobic decomposition of orange wastes from laboratory incubation experiments. Environ. Sci. Technol. 42 (9), 3265–3270.
Wang, Y.T., 2010. Research on Prodution and Application of Refinery Liquefied Petroleum Gas Use as Vehicle Fuel. Master Dissertation, Southwest Jiaotong Univesity (in Chinese).
Wei, F., Teng, E., Wu, G., Hu, W., Wilson, W.E., Chapman, R.S., Pau, J.C., Zhang, J., 1999. Ambient concentrations and elemental compositions of PM_{10} and $PM_{2.5}$ in four Chinese cities. Environ. Sci. Technol. 33 (23), 4188–4193.
WLPGA, AEGPL, 2018. Autogas Incentive Policies. World LPG Asso. And European LPG Association. http://www.wlpga.org.
Worton, D.R., Isaacman, G., Gentner, D.R., Dallmann, T.R., Chan, A.W.H., Ruehl, C., Kirchstetter, T.W., Wilson, K.R., Harley, R.A., Goldstein, A.H., 2014. Lubricating oil dominates primary organic aerosol emissions from motor vehicles. Environ. Sci. Technol. 48 (7), 3698–3706.
Wu, Z.F., Zhang, Y.L., He, J.J., Chen, H.Z., Huang, X.L., Wang, Y.J., Yu, X., Yang, W.Q., Zhang, R.Q., Zhu, M., Li, S., Fang, H., Zhang, Z., Wang, X.M., 2020. Dramatic

increase of reactive VOC emission from ships at berth after implementing the fuel switch policy in the Pearl River Delta Emission Control Area. Atmos. Chem. Phys. 20 (4), 1887–1900.
Yang, H.H., Chien, S.M., Cheng, M.T., Peng, C.Y., 2007. Comparative study of regulated and unregulated air pollutant emissions before and after conversion of automobiles from gasoline power to liquefied petroleum gas/gasoline dual-fuel retrofits. Environ. Sci. Technol. 41 (24), 8471–8476.
Yang, W.D., Zhang, Q.Y., Wang, J.L., Zhou, C.Y., Zhang, Y., Pan, Z.R., 2018. Emission characteristics and ozone formation potentials of VOCs from gasoline passenger cars at different driving modes. Atmos. Pollut. Res. 9 (5), 804–813.
Yu, L.D., Wang, G.F., Zhang, R.J., Zhang, L.M., Song, Y., Wu, B.B., Li, X.F., An, K., Chu, J.H., 2013. Characterization and source apportionment of $PM_{2.5}$ in an urban environment in Beijing. Aerosol Air Qual. Res. 13 (2), 574–583.
Zhang, Q.J., Wu, L., Fang, X.Z., Liu, M.Y., Zhang, J., Shao, M., Lu, S.H., Mao, H.J., 2018b. Emission factors of volatile organic compounds (VOCs) based on the detailed vehicle classification in a tunnel study. Sci. Total Environ. 624, 878–886.
Zhang, S.J., Mao, W.F., 1999. Vehicle emission pollution and its effect on human health in Guangzhou. Acta Sci. Nat. Univ. SunYat-sen. 38, 117–120 (in Chinese).
Zhang, S.J., Wu, Y., Liu, H., Wu, X.M., Zhou, Y., Yao, Z.L., Fu, L.X., He, K.B., Hao, J.M., 2013a. Historical evaluation of vehicle emission control in Guangzhou based on a multi-year emission inventory. Atmos. Environ. 76, 32–42.
Zhang, Y.L., Wang, X.M., Blake, D.R., Li, L., Zhang, Z., Wang, S., Guo, H., Lee, F.S.C., Gao, B., Chan, L., Wu, D., Sherwood, R.F., 2012. Aromatic hydrocarbons as ozone precursors before and after outbreak of the 2008 financial crisis in the Pearl River Delta region, south China. J. Geophys. Res. 117, D15306.
Zhang, Y.L., Wang, X.M., Barletta, B., Simpson, I.J., Blake, D.R., Fu, X.X., Zhang, Z., He, Q.F., Liu, T.Y., Zhao, X.Y., Ding, X., 2013b. Source attributions of hazardous aromatic hydrocarbons in urban, suburban and rural areas in the Pearl River Delta (PRD) region. J. Hazard Mater. 250, 403–411.
Zhang, Y.L., Wang, X.M., Li, G.H., Yang, W.Q., Huang, Z.H., Zhang, Z., Huang, X.Y., Deng, W., Liu, T.Y., Huang, Z.Z., Zhang, Z.Y., 2015. Emission factors of fine particles, carbonaceous aerosols and traces gases from road vehicles: recent tests in an urban tunnel in the Pearl River Delta, China. Atmos. Environ. 122, 876–884.
Zhang, Y.L., Yang, W.Q., Simpson, I., Huang, X.Y., Yu, J.Z., Huang, Z.H., Wang, Z.Y., Zhang, Z., Liu, D., Huang, Z.Z., Wang, Y.J., Pei, C.L., Shao, M., Blake, D.R., Zheng, J.Y., Huang, Z.J., Wang, X.M., 2018a. Decadal changes in emissions of volatile organic compounds (VOCs) from on-road vehicles with intensified automobile pollution control: case study in a busy urban tunnel in south China. Environ. Pollut. 233, 806–819.
Zhang, Y.L., Deng, W., Hu, Q.H., Wu, Z.F., Yang, W.Q., Zhang, H.N., Wang, Z.Y., Fang, Z., Zhu, M., Li, S., Song, W., Ding, X., Wang, X.M., 2020. Comparison between idling and cruising gasoline vehicles in primary emissions and secondary organic aerosol formation during photochemical ageing. Sci. Total Environ. 722, 137934.
Zhao, L.R., Wang, X.M., He, Q.S., Wang, H., Sheng, G.Y., Chan, L.Y., Fu, J.M., Blake, D.R., 2004. Exposure to hazardous volatile organic compounds, PM_{10} and CO while walking along streets in urban Guangzhou, China. Atmos. Environ 38 (36), 6177–6184.