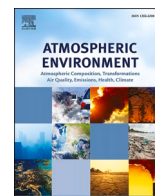




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Real-world emissions of carbonyls from vehicles in an urban tunnel in south China

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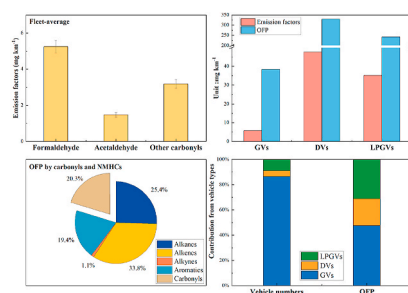
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HIGHLIGHTS

- Real-world vehicular carbonyl emissions were tested in an urban tunnel.
- Formaldehyde was the major species accounting for over half of carbonyl emissions.
- EFs for each fuel type vehicles were obtained via multiple regression analysis.
- Carbonyls could contributed ~20% of the total OFP by vehicular NMHCs and VOCs.
- The OFP of carbonyls from LPGVs was about 5 times that from GVs on average.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbonyls play a vital role in atmospheric photochemistry. Vehicle emission is among most important primary emission sources of carbonyls in urban areas, yet knowledge is quite scarce about real-world emissions of carbonyls with the changing on-road vehicle fleets. In this study, emissions of carbonyls were characterized based on tests in a busy urban tunnel in south China. Emission factor (EF) of carbonyls was measured to be 9.89 ± 0.65 mg km⁻¹ on average, in which formaldehyde alone shared 53.1% with an EF of 5.25 ± 0.35 mg km⁻¹, followed by acetaldehyde with an EF of 1.47 ± 0.13 mg km⁻¹. Glyoxal and methylglyoxal showed identical EFs of 0.18 ± 0.02 mg km⁻¹. Multiple linear regression retrieved total carbonyl EFs of 5.68, 47.71 and 35.09 mg km⁻¹ and ozone formation potentials (OFPs) of 38.4, 329.3, and 242.4 mg km⁻¹ for gasoline, diesel and liquefied

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petroleum gas vehicles (LPGVs), respectively. The unexpectedly high carbonyl emissions from LPGVs were largely attributed to the lack of after-treatment systems or the inefficiency of the after-treatment systems. Among vehicle-emitted non-methane hydrocarbons and carbonyls, carbonyls could contribute ~20% of the total OFPs while they only accounted for 7.1% of the total mass.

1. Introduction

As an important subset of volatile organic compounds (VOCs), carbonyl compounds play key roles in atmospheric chemistry (Carlier et al., 1986; Atkinson et al., 2003; Mellouki et al., 2015; Zhang et al., 2019b). As important ozone precursors, carbonyls could contribute 17–50% of the ozone formation potential (OFP) (Duan et al., 2008; Yuan et al., 2012; Han et al., 2019), and were often found to be among precursors with the highest relative incremental reactivity (RIR) based on observation-based model simulation (Cheng et al., 2010; Yang et al., 2018b). Some carbonyls, such as formaldehyde and acetaldehyde, have been identified as air toxics that are known to cause adverse health effects (World Health Organization, 2000). Carbonyls are substantial contributors to primary organic aerosol (POA) (Jakober et al., 2008), while the uptake of carbonyls on aerosols is also an important pathway for secondary organic aerosol (SOA) formation (Fu et al., 2008; Kawamura et al., 2013; Toda et al., 2014; Mitsuishi et al., 2018; Ling et al., 2020; Xu et al., 2020).

A large fraction of carbonyls occurring in ambient air is secondarily formed via photochemical oxidation of VOCs (Chen et al., 2014; Wang et al., 2015; Yang et al., 2017; de Gouw et al., 2018; Huang et al., 2019a, 2020a), but they can also be emitted primarily from various emission sources, including vehicle exhaust (Tsai et al., 2012; Yao et al., 2015a, b), industrial processes (Kim et al., 2008; Ma et al., 2016), biomass burning (Cerqueira et al., 2013; Yang et al., 2019), interior finish (Huang et al., 2019b), cooking activities (Huang et al., 2011; Cheng et al., 2016) and biogenic emissions (Seco et al., 2007; Sindelarova et al., 2014). Among them, vehicular exhaust is an important source of carbonyls particularly in urban areas. As reported by Chen et al. (2014), vehicle emission could account for 16.6–26.6% of carbonyls in urban Beijing. However, studies on vehicular emissions of VOCs were focused more on non-methane hydrocarbons (NMHCs) than on these carbonyls, and characterizing vehicular emissions of carbonyls would benefit a comprehensive assessment of vehicle emission impacts on urban air chemistry.

Vehicular emissions of carbonyls can be measured by chassis and engine dynamometer test (Dong et al., 2014; Zhou et al., 2015) and on-board emission measurements (Yao et al., 2015a, b; Cao et al., 2016). However, results from different studies showed large variations in the percentages of carbonyls in the total vehicle-emitted VOCs either for gasoline vehicles (GVs) and liquefied petroleum gas vehicles (LPGVs) (Adam et al., 2011; Cao et al., 2016; Li et al., 2019), or for diesel vehicles (DVs) (Tsai et al., 2012; Yao et al., 2015b; Mo et al., 2016), partly due to a limited number of vehicles were tested in the studies. Tunnel test is another widely adopted approach to study emissions of carbonyls from vehicles (Sagebiel et al., 1996, 1998, 1998; Zielinska et al., 1996; Fraser et al., 1998; Staehelin et al., 1998; Grosjean et al., 2001; Kean et al., 2001; Kristensson et al., 2004; Martins et al., 2006; Ho et al., 2007; Legreid et al., 2007; Ban-Weiss et al., 2008; Gentner et al., 2013; Nogueira et al., 2015; Zhang et al., 2016). The advantage of this approach is that it gets involved a large number of vehicles, and that results can represent emissions under real-world operation conditions. Although the number of motor vehicles in China has increased rapidly by approximately four times in the last decade (China statistical Yearbook, 2020), the emissions of carbonyls from vehicles are understudied, and till present only two tunnel studies for vehicular carbonyl emissions are available in China: Ho et al. (2007) reported vehicular emissions of mono-carbonyls based on tests in the Shing Mun Tunnel in 2003 in Hong Kong, and Zhang et al. (2016) reported emissions of dicarbonyls based

on tests in the Zhujiang Tunnel in 2014 in Guangzhou.

Many megacities in the world are confronted with surface ozone pollution (Liu and Wang, 2020), and vehicle emission largely hold accountable for the ozone pollution particularly in urban areas where ozone formation is typically sensitive to VOCs. While vehicle-emitted NMHCs have been identified to contribute substantially to ozone formation potential (Huang et al., 2017, 2020b; Zhang et al., 2019a; Hui et al., 2020; Li et al., 2020a; Wang et al., 2020a, b), the role of vehicle-emitted carbonyls in forming ozone is poorly understood due to the lack of studies to characterize vehicular emission of carbonyls. In fact, previous studies have demonstrated that vehicle emission could significantly contribute to carbonyls in urban air especially during rush hours (Feng et al., 2005; Liu et al., 2015). Moreover, as emission standards and vehicle fleet compositions change dramatically in China in the last decade, it is imperative to understand current status of carbonyl emissions from on-road vehicles under real world driving conditions, which were tested about 10 years ago based on tunnel tests in the Pearl River Delta (PRD) region in south China (Ho et al., 2007; Zhang et al., 2016).

In this study, a field campaign was carried out in the Zhujiang Tunnel during October 13–19, 2019 to characterize emissions of carbonyls from on-road vehicles under real world driving conditions. The fleet average EFs of carbonyls were obtained, and average EFs of carbonyls for GV, DVs and LPGVs were further derived through multiple linear regression. The ozone formation potential of vehicle-emitted carbonyls were compared with those of vehicle-emitted NMHCs.

2. Experimental methods

2.1. Field measurement site

Field measurements were conducted in the Zhujiang Tunnel, which is a densely trafficked underwater tunnel that crosses the Pearl River in the urban area of Guangzhou, China (Zhang et al., 2015). The tunnel is 1238.5 m long with a 721 m flat underwater section and has a total of four lanes of traffic contained in the two separate bores. During sampling, the tunnel's ventilation supply system was turned off, so ventilation was only induced by the flow of traffic through the tunnel and prevailing winds. Two sites were set, 50 m from each end of the flat underwater section, to simultaneously quantify the emissions of gaseous and particulate pollutants from vehicles. The vehicle speed limits in Zhujiang Tunnel are 50 km h⁻¹, and the most typical vehicle speed is 40–50 km h⁻¹. Traffic volumes and compositions of the vehicle fleet travelling through the tunnel were recorded visually with a video camera throughout the campaign. The diurnal variations of total vehicle numbers, as well as numbers of GV, electric vehicles (EVs), DVs and LPGVs were shown in Fig. 1. In this campaign, approximately 36000 vehicles passed through the tunnel each day. On average GV accounted for 75% of the total vehicle numbers, followed by EVs (13%), LPGVs (8%) and DVs (4%) during the whole campaign.

2.2. Sampling and analysis

The concentrations of carbon dioxide (CO₂), carbon monoxide (CO), nitrous oxide (NO_x), sulfur dioxide (SO₂) at the inlet and outlet stations were continuously monitored by an optical filter CO₂ analyzer (model 410i, Thermo, USA), gas filter correlation CO analyzer (model 48i, Thermo, USA), a low source chemiluminescence NO–NO₂–NO_x analyzer (model 42i, Thermo, USA) and a high level pulsed fluorescence SO₂

analyzer (model 43i, Thermo, USA), respectively. The wind speed parallel to the tunnel was sensed by a 3-D sonic anemometer (Campbell, USA).

During October 13–19, 2019, carbonyl samples were collected by drawing air through a 2,4-dinitrophenylhydrazine (DNPH)-impregnated silica gel cartridge (Sep-Pak DNPH-Silica Cartridge, Waters, USA) at a constant flow of 2 L min^{-1} during time intervals of 00:00–02:30, 02:30–05:00, 05:00–06:00, 06:00–07:00, 07:00–08:00, 08:00–09:00, 09:00–10:00, 10:00–11:00, 11:00–12:00, 12:00–13:00, 13:00–14:00, 14:00–15:00, 15:00–16:00, 16:00–17:00, 17:00–18:00, 18:00–19:00, 19:00–20:00, 20:00–21:00, 21:00–22:00, 22:00–23:00, 23:00–00:00 in each day. A potassium iodide (KI) oxidant scrubber and a filter were connected before the cartridge to prevent the interference of ozone and remove the particulate matter, respectively. After sampled, each cartridge was wrapped with aluminum foil and zipped in a Teflon bag, and stored in a refrigerator at $4 \text{ }^\circ\text{C}$ until analysis.

Sample treatments and analytical procedures are described elsewhere (Feng et al., 2005; Lv et al., 2009, 2010, 2010; Liu et al., 2013). Briefly, the sampled cartridges were eluted slowly with 2 ml of acetonitrile (ACN) into a 2 ml volumetric flask and then stored in refrigerated conditions at $4 \text{ }^\circ\text{C}$. The carbonyl compounds were measured by the high-performance liquid chromatography (HPLC) system (HP1200, Agilent, USA). A $10 \mu\text{l}$ aliquot was injected into the HPLC system and the analytical conditions were as follows: Agilent SB-C18 reverse column ($250 \text{ mm} \times 4.6 \text{ mm} \times 5 \mu\text{m}$); gradient mobile phase: 60–65% ACN of water solution for 25 min, 65–100% ACN for 3 min, 100% ACN for 7 min, 100–60% ACN for 1 min and then 60% ACN for 7 min; mobile-phase flow rate: 0.8 ml min^{-1} ; detector: ultraviolet detector at 360 nm for monocarbonyls and 420 nm for dicarbonyls.

Except for collecting the carbonyl samples, on 13 October 2019, non-methane hydrocarbons (NMHCs) were sampled simultaneously, by taking air into cleaned and evacuated 2 L stainless steel canisters by a model 910 canister sampler (Xontek Inc., California, USA). After taken the canisters back to the laboratory, they were analyzed by using a preconcentrator (model 7100, Entech Instruments Inc., USA) coupled to an Agilent 5973 N gas chromatograph with a mass selective detector and a flame ionization detector (GC-MSD/FID, Agilent Technologies, USA) as described in Zhang et al. (2013) and Yang et al. (2018a).

2.3. Quality control and quality assurance for carbonyls

Before the sampling, breakthrough tests were conducted by drawing standard mixtures (with concentrations of formaldehyde and acetaldehyde all over $50 \mu\text{g m}^{-3}$) through two cartridges connected in series in the same way as field samples. The concentration of carbonyls (DNPH derivative) detected in the second cartridge had no significant difference from the field blank samples, indicating the collecting efficiencies of carbonyls were nearly 100% without breakthrough.

To prepare calibration standards of the monocarbonyls (as DNPH derivative), two composite stock standard solutions containing 22 monocarbonyl-DNPH derivatives, including Mix 1 ($100 \mu\text{g ml}^{-1}$ in ACN, ChemService, USA), Mix 2 ($100 \mu\text{g ml}^{-1}$ in ACN, ChemService, USA), 2-butanone-DNPH derivative ($100 \mu\text{g ml}^{-1}$ in ACN, Sigma-Aldrich, USA) and methacrolein-DNPH derivative ($100 \mu\text{g ml}^{-1}$ in ACN, Sigma-Aldrich, USA) and concentrations of monocarbonyls-DNPH derivatives in the standard solutions ranged from $0.05 \mu\text{g ml}^{-1}$ to $20.0 \mu\text{g ml}^{-1}$. For calibration standards of the dicarbonyls (DNPH derivative), glyoxal and methylglyoxal (Mix 3, $1000 \mu\text{g ml}^{-1}$ in ACN, AccuStandard, USA) were mixed with $200 \mu\text{g ml}^{-1}$ DNPH under weak acid condition, and then the mixed solution was placed at room temperature for at least 6 h for the complete derivation of glyoxal and methylglyoxal by DNPH, and then after dilution we got the standard solutions with dicarbonyl concentrations ranging from $0.05 \mu\text{g ml}^{-1}$ to $4.0 \mu\text{g ml}^{-1}$. Calibration curves were obtained by running the calibration standards with the HPLC system in the same way as the cartridge extracts and then linearly correlating responses (peak areas) with the amounts of carbonyls ($R^2 > 0.995$). The detail carbonyl species and their method detection limits were also shown in Table S1.

3. Results and discussion

3.1. Emission factors

Among the 23 carbonyls (Table S1) in the standard solutions, only 14 of them were detected in tunnel air samples and the rest of them showed levels below their method detection limits (MDLs). On average total carbonyl concentrations were $27.42 \pm 0.88 \mu\text{g m}^{-3}$ at the inlet and

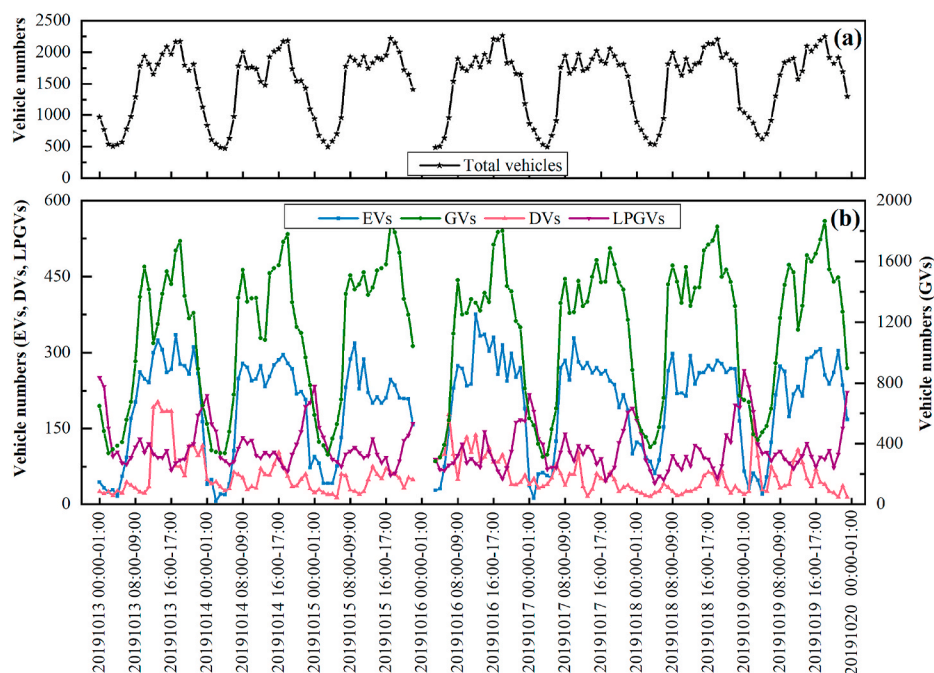


Fig. 1. Time series of traffic volumes of gasoline vehicles (GVs), electric vehicles (EVs), diesel vehicles (DVs) and liquefied petroleum gas vehicles (LPGVs) during the sampling period.

$37.64 \pm 1.12 \mu\text{g m}^{-3}$ at the outlet (Table 1). Acetone was the most abundant species at the inlet with an average concentration of $11.24 \pm 0.43 \mu\text{g m}^{-3}$, followed by formaldehyde ($6.87 \pm 0.20 \mu\text{g m}^{-3}$) and acetaldehyde ($3.59 \pm 0.10 \mu\text{g m}^{-3}$); at the outlet formaldehyde was the most abundant species ($12.57 \pm 0.42 \mu\text{g m}^{-3}$), followed by acetone ($12.21 \pm 0.43 \mu\text{g m}^{-3}$). The average concentrations of glyoxal and methylglyoxal were 0.44 ± 0.03 and $1.23 \pm 0.06 \mu\text{g m}^{-3}$ at the inlet against that of 0.61 ± 0.03 and $1.35 \pm 0.05 \mu\text{g m}^{-3}$ at the outlet, respectively. Differences between inlet-outlet pairs indicates that formaldehyde, acetaldehyde and acetone were the most abundant carbonyls emitted from vehicles, while vehicle emission is an anthropogenic source of glyoxal and methylglyoxal.

The total fleet EFs of carbonyls could be calculated according to the method described in detail by Pierson and Brachaczek (1983) and Pierson et al. (1996):

$$EF_{\text{fleet}} = \frac{(C_{\text{out}} - C_{\text{in}}) \times T \times \nu \times A}{N \times l}, \quad (1)$$

where EF_{fleet} is the average vehicle emission factors in unit of mg km^{-1} , C_{out} and C_{in} represent the mass concentrations of specific carbonyls at the tunnel outlet and inlet in mg m^{-3} , ν is the wind speed parallel to the tunnel in m s^{-1} , A is the tunnel cross section in m^2 ($A = 52.8 \text{ m}^2$), N is the total traffic number passing through the tunnel within the time interval T (s), and l is the distance between the two monitoring sites in km ($l = 0.621 \text{ km}$).

Times series of EFs for some important carbonyls, including formaldehyde, acetaldehyde, glyoxal and methylglyoxal, were presented in Fig. 2. As shown in Table 1, calculated total carbonyl emissions factor was $9.89 \pm 0.65 \text{ mg km}^{-1}$ on average, in which formaldehyde shared 53.1% with an average EF of $5.25 \pm 0.35 \text{ mg km}^{-1}$, and acetaldehyde and acetone ranked the second and the third with EFs of 1.47 ± 0.13 and $1.16 \pm 0.12 \text{ mg km}^{-1}$, respectively. A comparison of the EFs for the five important carbonyl species from this study in the Zhujiang Tunnel in comparison with literature data from other tunnel tests was presented in Table 2, and the comparison of the rest carbonyls was showed in Table S2. The EFs of formaldehyde, acetaldehyde and acetone from this study were lower than that from most of previous studies (Fraser et al., 1998; Kristensson et al., 2004; Ho et al., 2007), but near that from tests in the Tuscarora Mountain Tunnel, Pennsylvania, USA (Grosjean et al., 2001). The EFs of the two dicarbonyls, glyoxal and methylglyoxal, were

Table 1

The average concentrations of carbonyls and their emission factors (EFs) in the measurement of Zhujiang Tunnel.

Carbonyls	Concentration $\pm 95\% \text{CI}$ ($\mu\text{g m}^{-3}$)		EFs $\pm 95\% \text{CI}$ (mg km^{-1})
	Inlet	Outlet	
Formaldehyde	6.87 ± 0.20	12.57 ± 0.42	5.25 ± 0.35
Acetaldehyde	3.59 ± 0.10	5.15 ± 0.18	1.47 ± 0.13
Acetone	11.24 ± 0.43	12.21 ± 0.43	1.16 ± 0.12
Propanal	0.38 ± 0.02	0.65 ± 0.03	0.25 ± 0.02
2-Butanone	1.10 ± 0.09	1.42 ± 0.07	0.33 ± 0.04
Butanal	0.49 ± 0.05	0.54 ± 0.02	0.21 ± 0.02
Benzaldehyde	0.48 ± 0.03	0.73 ± 0.04	0.23 ± 0.02
Pentanal	0.15 ± 0.03	0.38 ± 0.04	0.25 ± 0.06
2,5-Dimethylbenzaldehyde	< MDL ^a	0.33 ± 0.04	0.41 ± 0.03
Octanal	0.17 ± 0.01	0.26 ± 0.01	0.10 ± 0.01
Nonanal	1.19 ± 0.04	1.29 ± 0.04	0.14 ± 0.01
Decanal	0.10 ± 0.01	0.13 ± 0.01	0.04 ± 0.01
Glyoxal	0.44 ± 0.03	0.61 ± 0.03	0.18 ± 0.02
Methylglyoxal	1.23 ± 0.06	1.35 ± 0.05	0.18 ± 0.02
Sum of carbonyls	27.42 ± 0.88	37.64 ± 1.12	9.89 ± 0.65

^a Method detection limits.

all $0.18 \pm 0.02 \text{ mg km}^{-1}$, which were decreased by over 80% when compared to that measured in 2014 in the same tunnel (Zhang et al., 2016). The reduced EFs of carbonyls from on-road vehicles might be largely resulted from the upgrade of the emission standards or exhaust treatment technology in recent years (Kaspar et al., 2003; Granger et al., 2011).

The ratios of formaldehyde/acetaldehyde (F/A) and acetaldehyde/propanal (A/P) were widely used to characterize carbonyls from different sources or in ambient air. Generally, F/A ratios vary from 1 to 2 in urban areas to about 10 in forested areas. Propanal is considered to be associated only with anthropogenic emissions, so A/P ratios would be high in remote areas and low in urban areas. Fig. 3 shows the average F/A and A/P ratios in comparison with those reported previously in vehicle exhaust and in ambient air in China's megacities. The F/A and A/P ratios from this tunnel study were 3.57 and 7.22, respectively. They fall between those from previous tunnel studies (Fraser et al., 1998; Grosjean et al., 2001; Kristensson et al., 2004; Ho et al., 2007), but it is interesting to find that these ratios from all tunnel studies seem to be higher than those tested by chassis and engine dynamometers or from on-board emission measurements (Schauer et al., 1999; Siegl et al., 1999; Adam et al., 2011; Tsai et al., 2012; Zhang et al., 2013; Dong et al., 2014; Yao et al., 2015a, b; Mo et al., 2016; Li et al., 2019), implying this kind of tunnel test for on-road emissions is necessary. As illustrated in Fig. 3 and references therein, the average F/A ratio from this study was slightly higher than those observed in ambient air in China's megacities including Hongkong, Guangzhou, Shanghai and Beijing, probably due to a shorter tropospheric lifetime of formaldehyde than acetaldehyde (Seinfeld and Pandis, 1998) and faster degradation of formaldehyde during photochemical ageing. The average A/P ratio from this study was close to that observed in the megacities (Fig. 3), further demonstrating vehicle emission should be a major primary emission source of carbonyls in urban areas.

3.2. Reconstructed EFs for GVs, DVs and LPGVs

Since EVs were free of carbonyl emissions, the fleet average EFs of carbonyls at time interval i , $EF_{\text{fleet}, i}$ could be linked with the average EFs for GVs, DVs and LPGVs as below:

$$EF_{\text{fleet}, i} = \alpha_i \times EF_{\text{GVs}} + \beta_i \times EF_{\text{DVs}} + (1 - \alpha_i - \beta_i) \times EF_{\text{LPGVs}} \quad (2)$$

where α_i and β_i are the number fractions of GVs and DVs in the vehicle fleet during time interval i ; EF_{GVs} , EF_{DVs} and EF_{LPGVs} are the average emission factors for GVs, DVs and LPGVs, respectively. With the fleet average EFs and fleet composition data, average EFs for GVs, DVs and LPGVs could be retrieved through multiple linear regression based on the above equation (2). A comparison between observed and reconstructed fleet average EFs was shown in Fig. S1. The retrieved EFs of individual carbonyls for different fuel type vehicles are presented in Table S3. The results revealed that EFs of carbonyls totalled 5.68, 47.70, 35.09 mg km^{-1} for GVs, DVs, and LPGVs, respectively. As shown in Table 2, all the tunnel studies agreed with each other in the fact that DVs or high-duty vehicles had much higher EFs than GVs or light-duty vehicles, and that formaldehyde was the most abundant carbonyls in vehicle exhaust. EFs of monocarbonyls measured in this study fell in the range of those from tunnel studies in USA (Zielinska et al., 1996; Sagebiel et al., 1996; Grosjean et al., 2001; Kean et al., 2001; Ban-Weiss et al., 2008), but lower than that from tunnel tests in Hong Kong (Ho et al., 2007).

For GVs, the total EFs of monocarbonyls on average was 5.51 mg km^{-1} , in which formaldehyde ($3.06 \pm 0.33 \text{ mg km}^{-1}$), acetone ($0.66 \pm 0.12 \text{ mg km}^{-1}$), acetaldehyde ($0.65 \pm 0.13 \text{ mg km}^{-1}$), 2,5-dimethylbenzaldehyde ($0.31 \pm 0.04 \text{ mg km}^{-1}$) and pentanal ($0.23 \pm 0.07 \text{ mg km}^{-1}$) were the top 5 species. For DVs, the total average EFs of monocarbonyls was 45.42 mg km^{-1} , and the top five species instead were formaldehyde ($22.73 \pm 4.55 \text{ mg km}^{-1}$), acetaldehyde ($6.76 \pm 1.73 \text{ mg km}^{-1}$),

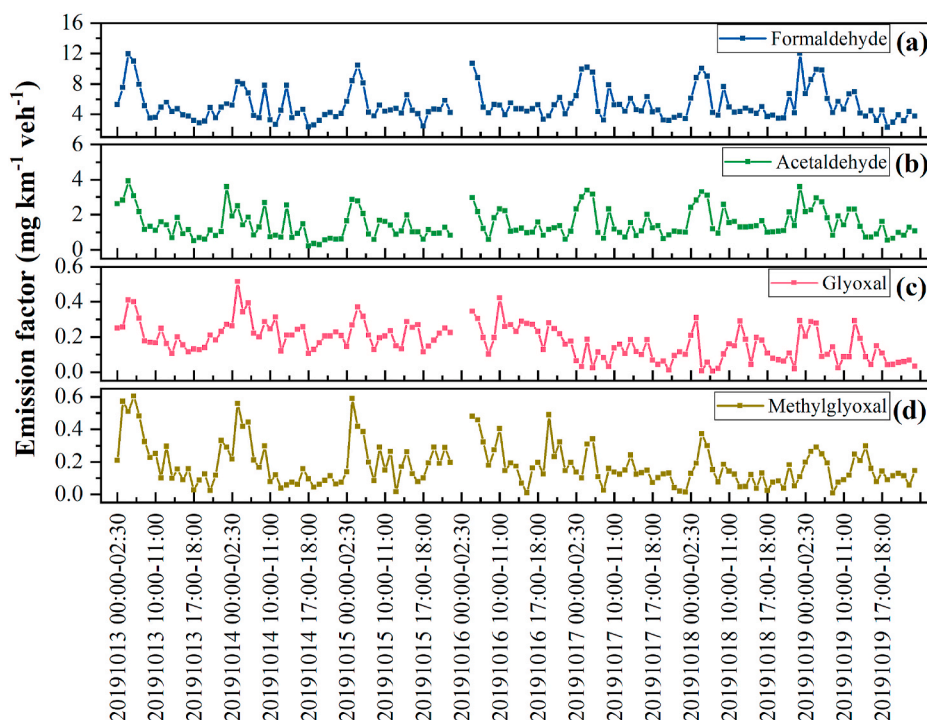


Fig. 2. Times series of emission factors for formaldehyde, acetaldehyde, glyoxal and methylglyoxal.

km^{-1}), acetone ($3.75 \pm 1.71 \text{ mg km}^{-1}$), 2,5-dimethyl-benzaldehyde ($2.44 \pm 0.57 \text{ mg km}^{-1}$) and butanal ($2.41 \pm 0.39 \text{ mg km}^{-1}$). For LPGVs, No EFs of carbonyls from tunnel tests have been reported before. From this study, total EF of monocarbonyls for LPGVs reached 33.92 mg km^{-1} , which was unexpectedly much higher than that of GVs. This result was different from the results reported by Adam et al. (2011) and Zhou et al. (2015) who observed lower EFs from LPGVs than from GVs based on chassis and engine dynamometer testing. This higher EFs of carbonyl for LPGVs than for GVs might be due to, as demonstrated in a previous study (Feng et al., 2020), the lack of after-treatment systems or the inefficiency of the after-treatment systems for LPGVs in Guangzhou. For dicarbonyls, the EFs of glyoxal were 0.10 ± 0.02 , 0.92 ± 0.23 and $0.50 \pm 0.13 \text{ mg km}^{-1}$ for GVs, DVs and LPGVs, while the EFs of methylglyoxal were 0.07 ± 0.02 , 1.36 ± 0.30 and $0.67 \pm 0.17 \text{ mg km}^{-1}$ for GVs, DVs and LPGVs, respectively.

3.3. Ozone formation potential of VOCs from vehicles exhaust

In this study, 66 species of NMHCs, including 29 alkanes, 19 alkenes, 1 alkyne and 17 aromatics, were also determined on October 13, 2019. As shown in Fig. S2, the composition of NMHCs was similar to that observed in the same Zhujiang tunnel in 2014 (Zhang et al., 2018), but the total EF of NMHCs in this study on average ($122.9 \pm 38.5 \text{ mg km}^{-1}$) was only 27.4% of that in 2014. To compare the ozone formation potential (OFP) of carbonyls with that of NMHCs, OFPs of the carbonyls and NMHCs in vehicle exhaust could be calculated as:

$$OFP = \sum_i EF_i \times MIR_i, \quad (3)$$

where EF_i and MIR_i are the emission factor and the maximum incremental reactivity for species i (Carter, 2009).

As presented in Fig. 4, the total OFP of all measured VOCs from vehicles was 340.3 mg km^{-1} , in which alkenes contributed most (114.9 mg km^{-1}) with a share of 33.8%, followed by alkanes (25.5% , 86.6 mg km^{-1}), carbonyls (20.3% , 69.1 mg km^{-1}) and aromatics (19.4% , 66.1 mg km^{-1}). It is worth noting that carbonyls only accounted for 7.1% of mass in all measured VOCs (NMHCs and carbonyls), but contributed

about one fifth of the total OFPs. Formaldehyde was the major contributor with OFP of 49.63 mg km^{-1} and ranked second among all the detected VOCs. As reported previously, approximately 17–41% of OFP from VOCs was attributed to vehicle exhaust in the urban areas (Cai et al., 2010; An et al., 2017; Huang and Hsieh, 2019; Ma et al., 2019; Li et al., 2020b), carbonyls from vehicles could account for about 3–8% of the total OFP.

With the average emission factor of carbonyls of each fuel type vehicles and the average fleet compositions during our field campaign, the contribution to OFP of carbonyls by GVs, DVs and LPGVs could be further calculated. Results revealed that 47.6% of OFP by carbonyls was attributed to GVs ($33.22 \pm 0.43 \text{ mg km}^{-1}$). This larger share is reasonable since 75% of vehicles travelling through the tunnel during the campaign were GVs although GVs had a smaller average EFs of carbonyls. DVs and LPGVs, contributed 21.2% ($14.81 \pm 1.75 \text{ mg km}^{-1}$) and 31.1% ($21.71 \pm 2.22 \text{ mg km}^{-1}$) of OFP by carbonyls although they only accounted for 4% and 8% in fleet number compositions. In Guangzhou, all LPG-fueled vehicles were taxi cars that had been converted from gasoline-driven to LPG-driven with retrofitted gasoline-LPG bi-fuel engines to reduce the emission of particulate matters and photochemically reactive VOC species. Due to the lack of exhaust treatment systems or the inefficiency of the exhaust treatment systems, while a previous study shows that OFPs by NMHCs from LPGVs were not reduced when compared to GVs (Feng et al., 2020), results from this study further demonstrate that on average the OFP of carbonyls from LPGVs was about 5 times that from GVs, suggesting that poor maintenance of LPGVs would instead increase the emissions of ozone precursors relative to GVs.

4. Conclusions

In this study, we updated the EFs of carbonyls from on-road vehicles by a tunnel test campaign in Zhujiang Tunnel, Guangzhou. Formaldehyde was found to contribute over half of carbonyl emissions with a fleet-average EF of $5.25 \pm 0.35 \text{ mg km}^{-1}$, followed by acetaldehyde ($1.47 \pm 0.13 \text{ mg km}^{-1}$) and acetone ($1.16 \pm 0.12 \text{ mg km}^{-1}$), while glyoxal and methylglyoxal, as the important precursors of SOA, both had

Table 2

Emission factors of carbonyls based on measurements in the Zhujiang Tunnel in comparison with literature data from other tunnel tests.

Tunnel		Formaldehyde	Acetaldehyde	Acetone	Glyoxal	Methylglyoxal
Emission factor (mileage-based in mg km⁻¹)						
FortMcHenry Tunnel, Baltimore, USA, 1992 ^a	LDVs ^p	4.31 ± 1.06	1.31 ± 0.31			
	HDVs ^q	32.9 ± 5.88	20.1 ± 1.75			
Tuscarora Mountain Tunnel, Pennsylvania, USA, 1992 ^b	LDVs	3.91 ± 1.38				
	HDVs	27.0 ± 4.54				
Gubrist tunnel, Switzerland, 1993 ^c	GVs	10.5 ± 8.06	2.33 ± 1.12	1.10 ± 1.80	0.17 ± 0.88	0.07 ± 0.91
	DVs	67.8 ± 36.5	14.6 ± 5.09	7.95 ± 8.13	0.00 ± 3.42	0.81 ± 3.53
Tuscarora Mountain Tunnel, Pennsylvania, USA, 1999 ^d	Total fleet	5.41 ± 2.22	2.19 ± 0.68	2.14 ± 0.81	0.18 ± 0.07	0.23 ± 0.17
	LDVs	2.57 ± 1.05	0.64 ± 0.27	1.69 ± 0.73	0.02 ± 0.01	0.03 ± 0.14
	HDVs	6.73 ± 2.04	3.95 ± 0.59	2.49 ± 1.42	0.32 ± 0.03	0.51 ± 0.32
Söderledstunnel, Stockholm, Sweden, 1998–1999 ^e	Total fleet	13 ± 3	3.1 ± 0.8	1.4 ± 0.6		
	LDVs	4.9 ± 4.6	1.4 ± 1.8	1.5 ± 1.8		
	HDVs	215 ± 137	50 ± 35	14 ± 27		
Janio Quadros tunnel, Sao Paulo, Brazil, 2004 ^f	Total fleet	48.4 ± 35.1	45.7 ± 29.1	9.3 ± 4.4		
	Total fleet	18.8 ± 7.5	3.1 ± 0.8	1.4 ± 0.6		
Shing Mun Tunnel, Hong Kong, China, 2003 ^g	NDVs ^r	3.5 ± 5.2	1.4 ± 1.8	1.5 ± 1.8		
	DVs	38.3 ± 6.5	50 ± 35	14 ± 27		
	Total fleet		2.40 ± 0.39	0.78 ± 0.09		
Gubrist tunnel, Switzerland, 2004 ^h	LDVs		2.10 ± 0.60	0.57 ± 0.13		
	HDVs		5.30 ± 6.60	3.94 ± 1.54		
	LDVs	5.7 ± 1.7	7.4 ± 2.7			
Zhujiang Tunnel, Guangzhou, China, 2014 ⁱ	HDVs	28 ± 8	20 ± 16			
	Total fleet				1.18 ± 0.43	0.32 ± 0.26
Zhujiang Tunnel, Guangzhou, China, 2019 ^k	GVs				1.64 ± 1.03	0.17 ± 0.33
	DVs				0.10 ± 3.49	1.68 ± 1.20
	LPGVs				0.58 ± 2.37	0.70 ± 0.66
	Total fleet	5.25 ± 0.35	1.47 ± 0.13	1.16 ± 0.12	0.18 ± 0.02	0.18 ± 0.02
Zhujiang Tunnel, Guangzhou, China, 2019 ^k	GVs	3.06 ± 0.33	0.65 ± 0.13	0.66 ± 0.12	0.10 ± 0.02	0.07 ± 0.02
	DVs	22.7 ± 4.55	6.76 ± 1.73	3.75 ± 1.71	0.92 ± 0.23	1.36 ± 0.30
	LPGs	17.6 ± 2.53	6.72 ± 0.96	4.72 ± 0.95	0.50 ± 0.13	0.67 ± 0.17
	Total fleet	128	29		27	27
Van Nuys Tunnel, Los Angeles, USA, 1993 ^l	LDVs	31.0 ± 4.00	8.00 ± 0.80	4.00 ± 0.50	0.09 ± 0.03	0.75 ± 0.15
	GVs	4.51 ± 7.4	9.92 ± 3.32	6.12 ± 3.05		
	DVs	28.80 ± 1.46		78.7 ± 79.3		
Caldecott tunnel, San Francisco, USA, 1999 ^m	Total fleet	61.2 ± 4.04	17.1 ± 1.53	13.5 ± 1.38	2.06 ± 0.19	2.11 ± 0.25
	GVs	35.7 ± 3.89	7.53 ± 1.48	7.66 ± 1.46	1.22 ± 0.20	0.81 ± 0.26
	DVs	265 ± 53.1	78.9 ± 20.2	43.8 ± 19.9	10.8 ± 2.68	15.9 ± 3.50
Zhujiang Tunnel, Guangzhou, China, 2019 ^k	LDVs	43.0 ± 5.0	11.0 ± 1.0	5.60 ± 0.70	0.12 ± 0.04	1.00 ± 0.20
	LDVs	42.0 ± 3.0	9.0 ± 0.5	6.70 ± 0.40	0.17 ± 0.03	0.91 ± 0.10
	LDVs	16.0 ± 1.4	7.0 ± 0.7	3.50 ± 1.10	0.10 ± 0.02	0.29 ± 0.10
	MDVs ^s /HDVs	129 ± 18.0	46.0 ± 12.0	ND	1.00 ± 0.50	2.60 ± 1.70
Zhujiang Tunnel, Guangzhou, China, 2019 ^k	GVs	48.2 ± 5.25	10.2 ± 2.00	10.4 ± 1.97	1.65 ± 0.26	1.09 ± 0.35
	DVs	312 ± 62.5	92.8 ± 23.8	51.5 ± 23.4	12.7 ± 3.15	18.7 ± 4.11

^a Zielinska et al. (1996).^b Sagebiel et al. (1996).^c Staehelin et al. (1998).^d Grosjean et al. (2001).^e Kristensson et al. (2004).^f Martins et al. (2006).^g Ho et al. (2007).^h Legreid et al. (2007).ⁱ Nogueira et al. (2015).^j Zhang et al. (2016).^k This study.^l Fraser et al. (1998).^m Kean et al. (2001).ⁿ Gentner et al. (2013).^o Ban-Weiss et al. (2008).^p Light-duty vehicles.^q High-duty vehicles.^r Non-diesel Vehicles.^s Medium-duty vehicles.^t The average fuel consumption of vehicles was estimated to be 8.57 L per 100 km (2018 new data analysis on real-world driving and fuel consumption for passenger cars in China, <http://www.icet.org.cn/admin/upload/2019081537990401.pdf>).^u we assumed a gasoline oil density of 0.74 g mL⁻¹ and a diesel oil density of 0.85 g mL⁻¹ (Zhang et al., 2016).

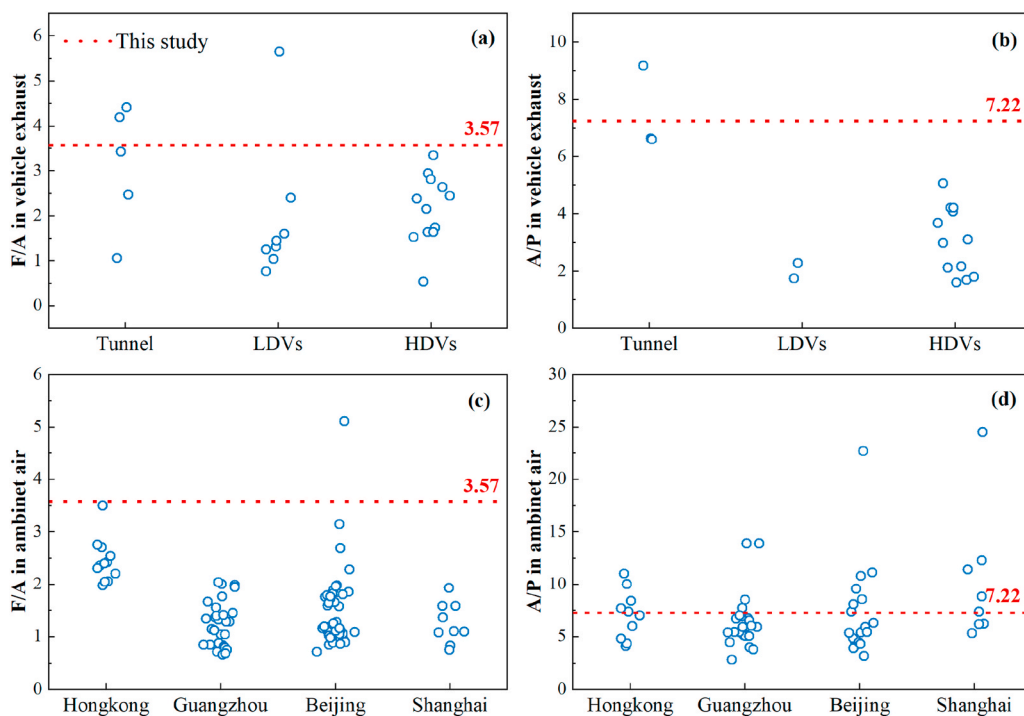


Fig. 3. The ratio of formaldehyde/acetaldehyde (F/A) and acetaldehyde/propanal (A/P) for vehicle emissions and urban observation.

Data from: Tunnel; Fraser et al. (1998), Grosjean et al. (2001), Kristensson et al. (2004), Martins et al. (2006) and Ho et al. (2007);

Light-duty vehicles (LDVs): Adam et al. (2011), Zhang et al. (2013) and Li et al. (2019);

Heavy-duty vehicles (HDVs): Schauer et al. (1999), Siegl et al. (1999), Tsai et al. (2012), Dong et al. (2014), Yao et al. (2015a,b) and Ma et al. (2016);

Hongkong: Lee et al. (2001), Ho et al. (2002), Cheng et al. (2014), Lui et al. (2017) and Yang et al. (2020);

Guangzhou: Feng et al. (2004, 2005, 2007), Yu et al. (2008), Lv et al. (2009, 2010), Wang et al. (2009), Hu et al. (2010), Zhuang et al. (2011), Liu et al. (2013) and Ho et al. (2015);

Beijing: Pang and Mu (2006), Xu et al. (2006), Shao et al. (2009), Xu and Gao (2009), Xu et al. (2010), Wang et al. (2011), Duan et al. (2012), Zhang et al. (2012a,b), Altemose et al. (2015), Ho et al. (2015), He et al. (2016), Rao et al. (2016), Sheng et al. (2018), Yang et al. (2018b), Huang et al. (2020a) and Liu et al. (2020).

Shanghai: Huang et al. (2008, 2009)

and Ho et al. (2015).

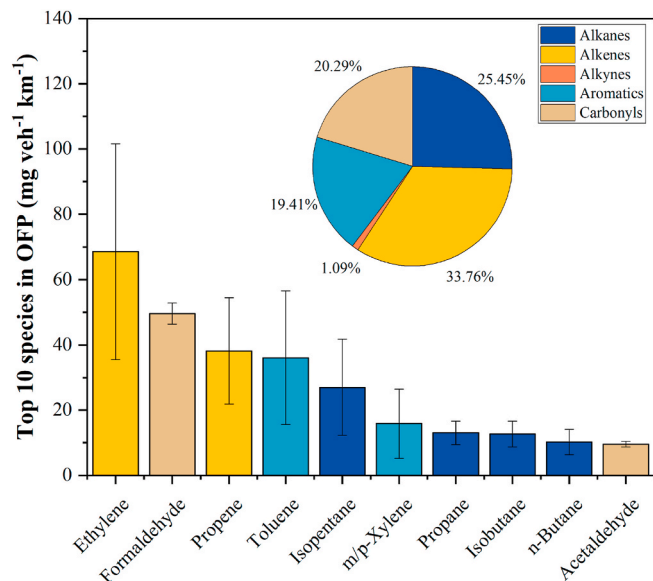


Fig. 4. Contribution percentages by different groups to OFPs (pie chart) and top 10 species among the vehicle-emitted NMHCs and carbonyls in their OFPs (bar charts).

EFs of $0.18 \pm 0.02 \text{ mg km}^{-1}$. Multiple linear regression was used to retrieve the average EFs of carbonyls for GVs, DVs and LPGVs. The EFs of DVs were much higher than that of GVs, which was consistent with results in the previous studies. However, the average EF of total carbonyls for LPGVs were 35.09 mg km^{-1} , which was about 5 times higher than that of GVs. As for the impact on ozone formation, though carbonyls only contributed 7.1% of mass in all measured NMHCs and carbonyls,

they could account for one fifth of the total OFPs by the vehicle-emitted NMHCs and carbonyls. This signals the importance to take carbonyls into consideration in the control of vehicular ozone precursors. Among all the fuel type vehicles, GVs accounted for 47.6% of OFP because 75% of vehicles travelling through the tunnel were GVs; LPGVs contributed 31.1% of OFP with only 8% in fleet, while DVs contributed 21.2% of OFP with only 4% in fleet. The unexpected higher carbonyl emissions from LPGVs suggest installing exhaust treatment systems or improve the performance of exhaust treatment systems for LPGVs will also benefit reducing vehicular emission of carbonyls that are important ozone precursors in urban areas.

CRedit authorship contribution statement

Zhenfeng Wu: Writing – original draft, Methodology, Formal analysis, Writing – review & editing. **Yanli Zhang:** Writing – review & editing. **Chenglei Pei:** Methodology. **Zuzhao Huang:** Methodology. **Yujun Wang:** Methodology. **Yanning Chen:** Methodology. **Jianhong Yan:** Methodology. **Xiaoqing Huang:** Investigation, Data curation. **Shaoyuan Xiao:** Investigation, Data curation. **Shilu Luo:** Investigation, Data curation. **Jianqiang Zeng:** Investigation, Data curation. **Jun Wang:** Investigation, Data curation. **Hua Fang:** Investigation, Data curation. **Runqi Zhang:** Investigation, Data curation. **Sheng Li:** Investigation, Data curation. **Xuwei Fu:** Investigation, Data curation. **Wei Song:** Investigation, Data curation. **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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2021.118491>.

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