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## Measurement report: Insight into greenhouse gas emission characteristics of light-duty vehicles in China in the context of technological innovation

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Abstract. Greenhouse gas (GHG) emissions represent a pivotal driver of global climate change, with vehicular emissions, particularly from light-duty vehicles, emerging as a prominent source of GHGs. Despite extensive research on gaseous pollutants, studies on GHG emissions within the framework of carbon neutrality remain scarce. This study delves into the emission characteristics of three primary GHGs (carbon dioxide  $(CO_2)$ , methane  $(CH_4)$ , and nitrous oxide  $(N_2O)$  from various light-duty vehicles, encompassing conventional gasoline and hybrid vehicles and bi-fuel taxis. As anticipated, with advancements in emission standards and powertrains, vehicular GHG emissions have significantly decreased. However, our findings also revealed surprising trends. While engine technology upgrades reduced CO<sub>2</sub>, they unexpectedly increased CH<sub>4</sub> and N<sub>2</sub>O emissions. Additionally, hot starts, beneficial for reducing CO<sub>2</sub> and CH<sub>4</sub> emissions, caused heightened N<sub>2</sub>O emissions, which is noteworthy under operating conditions with frequent start-stop events. Intriguingly, compressed natural gas (CNG), generally perceived as cleaner, increased CH<sub>4</sub> emissions. Regarding the impact of three-way-catalyst (TWC) converters on GHG emissions, under "TWC deteriorated" conditions, N<sub>2</sub>O emissions from CNG-powered vehicles were approximately 3 times higher than those under "TWC worked" conditions, which can be attributed to the significant increase in nitrogen oxides  $(NO_x)$ . Considering the global warming potential (GWP), the "TWC deteriorated" scenario paradoxically decreased GWP values, highlighting the complex interaction between emission control technologies and their environmental impacts. This study provides crucial insights into vehicular GHG emissions, which are essential for developing effective strategies aimed at mitigating emissions and enhancing the efficiency of emission control systems.

## 1 Introduction

As the issue of global climate change becomes increasingly prominent, reducing greenhouse gas (GHG) emissions has emerged as a critical objective for environmental protection. In 2023, global energy-related carbon dioxide (CO<sub>2</sub>) emissions grew by 1.1 %, an increase of 410 million tonnes (Mt), reaching a new record high of 37.4 billion tonnes (Gt). This is in comparison to the rise of 490 Mt in 2022, which was a 1.3 % increase (IEA, 2024). Transportation is a significant source of GHG emissions, with the global transportation sector accounting for 23% of all energy-related CO<sub>2</sub> emissions (Liu et al., 2023). In 2023, the number of motor vehicles in China reached 435.6 million; among these vehicles, the number of automobiles reached 336 million, a yearon-year increase of 5.3 % (Ministry of Ecology and Environment, 2024). Due to the large number of vehicles, the road transportation sector's contribution to CO<sub>2</sub> emissions is significant. One study indicated that in 2019, China's vehicle CO<sub>2</sub> emissions were 952 Mt, with gasoline and other fuels (natural gas, alcohol fuels) comprising 47.5 % of this figure (Huang et al., 2022b). A recent study found that GHG emissions rose from 431 Mt in 2005 to 807 Mt in 2015, with an annual growth rate of 6.5 % (Li et al., 2019). Many studies have shown that the transportation sector of China will not peak before 2030 due to huge increasing transport demand (Yuan et al., 2021; Liu et al., 2018; Yin et al., 2015). Furthermore, road-based transport is the largest emission source in the transportation sector (Xue et al., 2019), accounting for 82.7 % of total CO<sub>2</sub> emissions in the entire transportation sector in 2015 (Zhang et al., 2019); thus, road-based transport has a much larger CO<sub>2</sub> emission reduction potential compared to other transport modes (Wang et al., 2017).

In addition to CO<sub>2</sub>, vehicular activities are closely associated with the greenhouse gases methane (CH<sub>4</sub>) and nitrous oxide  $(N_2O)$ . The N<sub>2</sub>O emissions are primarily produced by the combustion of nitrogenous compounds in fuel, while CH<sub>4</sub> is generated from incomplete combustion in natural gas vehicles or the thermal cracking of alkanes (such as n-alkanes) in gasoline at high temperatures. Although the emissions of N<sub>2</sub>O and CH<sub>4</sub> from vehicles are 3 to 6 orders of magnitude smaller than those of CO<sub>2</sub>, their global warming potential (GWP) is significantly higher. The GWP of N<sub>2</sub>O and CH<sub>4</sub> is 298 times and 25 times greater than that of CO<sub>2</sub>, respectively (IPCC, 2014). While the emissions of CH<sub>4</sub> from vehicles may be negligible on a global scale, in urban areas, particularly those with heavy traffic, CH<sub>4</sub> emissions can account for up to 30 % of the regional CH<sub>4</sub> emissions (Nam et al., 2004). In northern China, where natural-gas-powered vehicles are more common, cities need to consider the impact of fuel types and emission standards on N<sub>2</sub>O and CH<sub>4</sub> emissions when developing carbon reduction plans for vehicles (Da et al., 2020). However, earlier studies have paid less attention to this aspect. Additionally, the population of light-duty hybrid electric vehicles in China is currently experiencing rapid growth, but comprehensive research on their GHG emission profiles remains inadequate. Precise quantification of GHG emissions from hybrid electric vehicles (HEVs) is crucial for developing effective strategies to mitigate emissions from the light-duty-vehicle sector. Meanwhile, the three-way-catalyst (TWC) system remains the predominant aftertreatment technology for gasoline vehicles. This catalytic system facilitates the abatement of exhaust pollutants by catalyzing the oxidation of carbon monoxide (CO) and unburned total hydrocarbons (THC), alongside a reduction in nitrogen oxides  $(NO_x)$ . Notably, the NO<sub>x</sub> reduction process involves undesirable side reactions, particularly the interaction between NO and nitrogen species on the catalyst, resulting in the formation of N<sub>2</sub>O (Wallington and Wiesen, 2014). Therefore, there is a need to strengthen comprehensive research on vehicular GHG emissions to further provide data and theoretical support for controlling greenhouse gas emissions. The Chinese government has promised to reach peak CO<sub>2</sub> around 2030 and strives to achieve it as soon as possible. The government has incorporated climate change into its ecological planning and built a low-carbon system for society and the economy.

Currently, research on vehicular GHG emissions both domestically and internationally mainly employs three methods: chassis dynamometer testing, onboard testing, and model estimation. In recent years, a significant number of scholars have explored the emission characteristics of vehicular greenhouse gases. Emission factors for GHGs for actual vehicles under different emission standards, temperature points and driving operating conditions were obtain (He et al., 2014; Zhong et al., 2023; Clairotte et al., 2020; Wang et al., 2022b). Specifically, CO<sub>2</sub> emissions for China VI (335 g km<sup>-1</sup>) diesel vehicles have been reduced compared to those for China IV (415 g km<sup>-1</sup>) and China V  $(447 \text{ g km}^{-1})$  vehicles (Li et al., 2024), and some studies have reported CO<sub>2</sub> emissions for China VI gasoline vehicles of around 200 to  $300 \,\mathrm{g \, km^{-1}}$  (Zhu et al., 2022; Wang et al., 2022b). The CH<sub>4</sub> emissions decreased from  $48 \text{ mg km}^{-1}$  for China I to 28 mg km<sup>-1</sup> for China IV light-duty gasoline vehicles, and N<sub>2</sub>O emissions were reduced from 45 (China I) to  $21 \text{ mg km}^{-1}$  (China IV) (He et al., 2014). The CH<sub>4</sub> and N2O emissions for Euro V to Euro VI vehicles were found to be  $7 \text{ mg km}^{-1}$  (Clairotte et al., 2020); the average emission factors of CO<sub>2</sub> and CH<sub>4</sub> for light-duty vehicles in Thailand were found to be  $232.25 \text{ g km}^{-1}$  and  $9.50 \text{ mg km}^{-1}$ , respectively (Sirithian et al., 2022). With advancements in powertrain technologies, hybrid electric vehicles are being progressively promoted and have demonstrated significant potential in reducing both pollutant and CO<sub>2</sub> emissions (Selleri et al., 2022). Furthermore, from a model simulation perspective, computational models such as International Vehicle Emissions (IVE), the Motor Vehicle Emission Simulator (MOVES), and the Gompertz Growth Model have been utilized to establish greenhouse gas emission inventories for different base years (Tang et al., 2018; Li et al., 2022; Zeng et al., 2016) and even to predict future greenhouse gas emissions (Zeng et al., 2016).

Overall, from the perspective of GHG emissions from domestic vehicles in China, Yin et al. (2024) explored the GHG emissions of 84 China VI light-duty gasoline vehicles, indicating that vehicular GHG emissions generally tend to increase with engine displacement and that the CO<sub>2</sub> increase rate caused by the CO2 conversion of CH4 and N2O emissions from all types of vehicles was less than 1 %; this suggests that CO<sub>2</sub> emissions from vehicle exhaust remain the primary source of greenhouse gases (Yin et al., 2024). Nevertheless, existing research reveals a critical knowledge gap in systematic comparative analysis of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O emission profiles from vehicles certified under China's most stringent emission standards. Notably, old vehicles with deteriorated aftertreatment systems have been confirmed as "super-emitters", contributing 50 %-80 % to total vehicular emissions while representing only 23 % of the fleet composition (Huo et al., 2012). However, comprehensive understanding of GHG emission characteristics from these "superemitters" remains incomplete. To address these gaps, key investigations are urgently required, including the establishment of comprehensive domestic emission profiles for vehicles complying with China's latest emission standards and their comparison with previous emission standards and the determination of GHG emission characteristics from "superemitters".

In this study, we conducted chassis dynamometer experiments on GHGs ( $CO_2$ ,  $CH_4$ , and  $N_2O$ ) from light-duty vehicles with different emission standards, engine types, fuel types, and working conditions of aftertreatment devices, aiming at improving the characteristics of GHG emission factors for light-duty vehicles and exploring the GHG emission potential of light-duty-vehicle exhaust emissions. This study provides an important data foundation for fully understanding the localized GHG emissions from light-duty vehicles in China, effectively reducing the uncertainty in greenhouse gas emission inventory calculations.

## 2 Materials and methods

#### 2.1 Tested vehicles and fuels

A total of 11 in-use light-duty vehicles, comprising 10 internal combustion engine vehicles (ICEVs) and 1 hybrid electric vehicle (HEV), from rental car companies were tested in this study. These light-duty vehicles selected cover various emission standard categories, engine techniques, powertrain technology types, and mileage accumulations, as detailed in Table 1. Among the eight light-duty gasoline vehicles (vehicles #1 to #8), representation was provided by China IV, China V, and China VI emission standards. This is mainly since China's current light-duty-vehicle fleet is predominantly composed of China IV-, China V-, and China VIcompliant vehicles. These vehicles featured two distinct engine techniques – gasoline direct injection (GDI) and port fuel injection (PFI) – and all were equipped with TWC converters. With the advancement of vehicular emission control technologies, GDI engines are progressively replacing traditional PFI engines due to their superior combustion efficiency, which is achieved by directly injecting fuel into the combustion chamber. Additionally, to assess the GHG emission differences between gasoline and hybrid electric vehicles, one non-plug-in hybrid electric light-duty vehicle (vehicle #9) adhering to the China VI emission standard was also included, utilizing both conventional gasoline engines and electric motors and potentially influencing overall GHG emissions.

To gain a deeper understanding of the effects of fuel types and TWC operating conditions on vehicular GHG emissions, two China VI compliant bi-fuel taxis (vehicles #10 to #11) were tested, one with 266 624 km (representing the "TWC deteriorated" condition) and another only 79 960 km (representing the "TWC worked" condition). Figure S1 in the Supplement demonstrates the emission characteristics of the two vehicles representing distinct TWC operational states: (1) a properly functioning under the "TWC worked" condition and (2) a deteriorated system under the "TWC deteriorated" condition. The analysis revealed that the "TWC deteriorated" vehicle emitted substantially higher levels of carbon monoxide (CO) and total hydrocarbons (THC), with emission factors elevated by over 24-fold and 97-fold, respectively, compared to the "TWC worked" vehicle. These compressed natural gas (CNG) bi-fuel taxis were originally equipped with two separate fuel delivery systems and control units from the equipment manufacturer, allowing for seamless switching between gasoline and CNG during operation, with gasoline being used during the warm-up phase (engine coolant temperature < 70 °C) before transitioning to CNG (Wang et al., 2024).

In this study, we employed three types of fuel: conventional gasoline, ethanol gasoline, and compressed natural gas (CNG). The conventional gasoline, exclusively utilized in light-duty gasoline vehicles and HEVs, was sourced from an automobile testing center, guaranteeing uniformity in chemical composition throughout all testing cycles. The specifications of this conventional gasoline adopted in this study have been previously detailed in our earlier work (refer to Supplement Table 2 in Zhang et al., 2024). To delve deeper into the impact of fuels on vehicular emissions, all three fuel types were applied in the case of bi-fuel taxis. Ethanol gasoline and CNG came from gasoline fueling and natural gas fueling station locations, respectively.

#### 2.2 Experimental protocol and driving cycles

We carried out vehicular emission tests utilizing a chassis dynamometer within the esteemed China Automotive Technology and Research Center (CATARC) laboratory, a typeapproval testing center accredited by the Ministry of Ecol-

Vehicle ID	Category	Model year	Emission standard	Engine technology	Displacement (mL)	Mileage (10 <sup>4</sup> km)	Max net engine power (kW)	Max authorized mass (kg)	After- treatment type
#1	ICEV <sup>a</sup>	2011	China IV	GDI <sup>b</sup>	1798	22.11	118	2000	TWC <sup>c</sup>
#2	ICEV	2011	China IV	GDI	1390	12.60	96	1930	TWC
#3	ICEV	_d	China V	GDI	1798	8.00	_d	_d	TWC
#4	ICEV	2018	China V	GDI	1798	14.66	132	2100	TWC
#5	ICEV	2022	China VI	PFI <sup>e</sup>	1490	2.90	89	1740	TWC
#6	ICEV	2022	China VI	PFI	1490	5.22	89	1740	TWC
#7	ICEV	2021	China VI	GDI	1498	6.59	83	1725	TWC
#8	ICEV	2021	China VI	GDI	1498	5.50	83	1725	TWC
#9	HEV	2022	China VI	GDI	1798	2.75	72	1845	TWC
#10	ICEV, bi-fuel taxi	2019	China VI	PFI	1591	26.66	90.2	1640	TWC
#11	ICEV, bi-fuel taxi	2021	China VI	PFI	1591	8.00	90.2	1640	TWC

 Table 1. Specifications of the light-duty vehicles in this study, including category, model year, emission standard, engine technology, displacement, mileage, max net engine power, max authorized mass, and aftertreatment types.

<sup>a</sup> Internal combustion engine vehicle. <sup>b</sup> Gasoline direct injection. <sup>c</sup> Three-way catalyst. <sup>d</sup> The unrecorded data, being a non-essential parameter, have no bearing on the subsequent analysis. <sup>e</sup> Port fuel injection.

ogy and Environment of China. The tailpipe pollutants are diluted by a constant volume sampling (CVS) method; this is followed by analysis using an array of measurement systems, as shown in Fig. 1. For GHG quantification, specific instruments were employed: a non-dispersive infrared (NDIR) analyzer for CO<sub>2</sub> and a quantum cascade laser (QCL) analyzer for N<sub>2</sub>O, ensuring precise measurements. Additionally, CH<sub>4</sub> concentrations were determined by a setup combining a non-methane cutoff (NMC) filter with a hydrogen flame ionization detector (FID) (Yin et al., 2024). The calculation of distance-based emission factors for GHGs was achieved through a meticulous process involving second-by-second concentrations, exhaust volume, pollutant density, and the actual driving distance during the driving cycles.

To comprehensively investigate the impact of diverse driving conditions on GHG emissions, we implemented three distinct types of driving cycle. For gasoline vehicles and HEVs, we adhered to the Worldwide Harmonized Light Vehicles Test Cycle (WLTC), newly introduced into the China VI emission regulations. The WLTC protocol comprises four phases: low-speed (589 s), medium-speed (433 s), highspeed (455 s), and extra-high-speed (323 s) phases. The impacts of both cold and hot startup modes on vehicular emissions were explored during the application of the WLTC protocol. Furthermore, we conducted additional driving tests on some typical vehicles using the China Light-duty Vehicle Test Cycle (CLTC), which could be more relevant to realworld driving conditions in China. This protocol, which encompasses CLTC-P (passenger) and CLTC-C (commercial), was proposed in 2019 (Wang et al., 2020; Hu et al., 2021). In addition, we conducted the emission testing based on constant-speed conditions: 30, 60, 90, and 120 km h<sup>-1</sup>. During these constant-speed tests, the driver accelerated to the target speed and maintained it for 10 min, allowing for the precise measurement of GHG emissions under stable driving conditions. Notably, for the CLTC protocol and our constant-speed tests, only the hot start mode was employed.

#### 2.3 Calculation of global warming potentials

Global warming potentials (GWPs) have been established as a widely accepted method for comparing the climate impacts of various greenhouse gas emissions over the past decades (Shine, 2009). This metric enables the conversion of CH<sub>4</sub> and N<sub>2</sub>O emissions into their CO<sub>2</sub> equivalents, thereby facilitating a standardized comparison. Furthermore, the relative growth rate of CO<sub>2</sub> after conversion can be obtained, which is calculated precisely according to Eq. (1), offering valuable insights into the long-term warming potential of these gases.

$$R_{\rm CO_2} = (\rm GWP \times M_i) / M_{\rm CO_2}, \tag{1}$$

where  $R_{CO_2}$  is the relative growth rate of CO<sub>2</sub>,  $M_i$  denotes the emissions of CH<sub>4</sub> or N<sub>2</sub>O, and  $M_{CO_2}$  denotes the emissions of CO<sub>2</sub>. The GWP of N<sub>2</sub>O and CH<sub>4</sub> is 298 and 25, respectively.

## 3 Results and discussion

#### 3.1 Scenario-based greenhouse gas emission factors

## 3.1.1 Distance-based emission factors

Figure 2 comprehensively depicts the distance-based emission factors of GHGs, illustrating the diverse impacts stemming from various scenarios, encompassing emission standard categories, engine techniques, powertrain advancements, startup modes, and driving cycles. Concerning emis-



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Figure 1. Schematic diagram for vehicular emission measurements based on the chassis dynamometer used in this study.

sion standard categories, distinct trends emerged across the three primary GHG types. Prior research has documented a pronounced reduction in CO,  $NO_x$ , and THC emissions in response to stricter emission standards (Duan et al., 2021). Similarly, in this study, CH<sub>4</sub> emission factors have undergone a substantial decline, with the China VI gasoline vehicle (during cold starts) exhibiting an emission factor of approximately one-sixth of that recorded for the China IV gasoline vehicle under similar conditions. This trend in CH<sub>4</sub> is consistent with previously reported variations in volatile organic compounds (VOCs; Qi et al., 2021; Duan et al., 2021). Turning to CO<sub>2</sub> and N<sub>2</sub>O, Fig. 2 highlights that the emission factors for these gases in China VI gasoline vehicles were notably lower than those in China IV and China V vehicles. Nevertheless, the emission factors for CO<sub>2</sub> of the China V vehicles (155 g km<sup>-1</sup> for the hot starts) were comparable to those of China IV vehicles  $(151 \text{ g km}^{-1} \text{ for the hot starts})$ , and those for  $N_2O$  of the China V vehicles (2.0 mg km<sup>-1</sup> for the hot start) were even higher than those of China IV vehicles ( $1.5 \text{ mg km}^{-1}$  for the hot starts), particularly for the hot start mode. This highlights a nuanced aspect of emission performance that needs further investigation. A comparison with previous studies shows that the CH<sub>4</sub> emissions for China V and China VI vehicles in our study were lower than those for Euro 5 and Euro 6 vehicles reported by Clairotte et al. (2020). A similar trend was observed for N2O emissions, as demonstrated by the summary of previous studies provided in Table S1 in the Supplement. Furthermore, lower CO<sub>2</sub> emissions were found in our study compared to those for gasoline vehicles reported by Zhu et al. (2022) and Wang et al. (2022b) as well as those for diesel vehicles reported by Wu et al. (2017), Cai and Xie (2010), Wang et al. (2022a), and Li et al. (2024), as shown in Table S1.

Regarding the startup modes, our analysis revealed that  $CO_2$  and  $CH_4$  emissions were slightly lower during hot starts compared to cold starts. Herein, we conducted a comparative assessment of vehicular emissions from tested vehicles equipped with GDI engines. Specifically, the  $CO_2$  emission

factors for the tested China IV vehicles were found to be  $159 \text{ g km}^{-1}$  under cold startup mode and  $151 \text{ g km}^{-1}$  under hot startup mode. Similarly, for the tested China V vehicles, the CO<sub>2</sub> emission factors were 161 and 155 g km<sup>-1</sup>, respectively, for cold and hot startup modes. The China VI vehicles also exhibited a similar trend, with CO<sub>2</sub> emission factors of 151 and 145 g km<sup>-1</sup> for cold and hot startup modes, respectively. Similarly, the CH<sub>4</sub> emissions displayed a declining trend, transitioning from cold starts to hot starts across all tested vehicle categories. For China IV vehicles, the CH4 emission factors were 7.7 mg km<sup>-1</sup> under cold starts and  $7.4 \text{ mg km}^{-1}$  under hot starts. China V vehicles showed a reduction from 2.8 to  $2.2 \text{ mg km}^{-1}$ , and China VI vehicles showed a reduction from 1.4 to  $1.0 \,\mathrm{mg \, km^{-1}}$ . Interestingly, the trend for N<sub>2</sub>O emissions was inverse to that of CO<sub>2</sub> and CH<sub>4</sub>, with higher emissions observed during hot starts compared to cold starts, particularly among China IV and China V vehicles. A systemic introduction to the detailed cause analysis will be presented in Sect. 3.1.2.

When it comes to engine techniques, a notable trend emerged in the comparison between PFI and GDI engines. Specifically, PFI engines exhibited higher CO<sub>2</sub> emission factors compared to GDI engines. Conversely, GDI engines demonstrated higher CH<sub>4</sub> and N<sub>2</sub>O emissions. In terms of testing protocols, the CLTC protocol yielded slightly lower CO<sub>2</sub> emission factors by approximately 4 % compared to the WLTC protocol. However, the CH<sub>4</sub> and N<sub>2</sub>O emission factors for CLTC were observed to be higher, with increases of around 8 % and 10 %, respectively, compared to WLTC. Additionally, a comparative analysis between the gasoline vehicles and HEV revealed insight into the advancement of powertrain technology. Despite the selected HEV having a higher overall mass and displacement, they demonstrated a significant reduction in GHG emissions. Specifically, CO<sub>2</sub> emissions were reduced by 10 % to 14 %, CH<sub>4</sub> emissions by 11 % to 37 %, and N<sub>2</sub>O emissions by 23 % to 43 %.



Figure 2. Emission factors of GHGs (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) for various scenarios involving different emission standard categories, engine techniques, powertrain technology types, startup modes, and driving cycles.

# 3.1.2 Interpreting technological upgrading through the perspective of generation mechanisms

The different trends of GHG emissions across various scenarios might be attributed to the underlying mechanism of their generation.  $CO_2$  can originate from the complete combustion of fuel in the tailpipe or from the oxidation of THC by the catalyst in TWC converters, highlighting a strong correlation between  $CO_2$  emissions, fuel consumption, and the completeness of combustion.  $CH_4$ , on the other hand, can be formed through the partial oxidation of gasoline fuels or the thermal cracking of organic compounds in fuels. Regarding N<sub>2</sub>O, it may arise from the reaction between ammonia (NH<sub>3</sub>) and NO<sub>x</sub> or from the decomposition of ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) generated in catalytic converters, suggesting a greater dependence on NO<sub>x</sub> and NH<sub>3</sub> concentrations (Yin et al., 2024; Brinklow et al., 2023).

The fuel consumption for China VI vehicles  $(5.0 \text{ L} (100 \text{ km})^{-1} \text{ for the hot start})$  was lower than that of China IV  $(6.4 \text{ L} (100 \text{ km})^{-1} \text{ for the hot start})$  and China V  $(6.6 \text{ L} (100 \text{ km})^{-1} \text{ for the hot start})$  vehicles. The lower fuel consumption for China VI vehicles caused the lower CO<sub>2</sub> and CH<sub>4</sub> emissions. Furthermore, N<sub>2</sub>O emissions are primarily derived from reactions in the aftertreatment systems. Similar aftertreatment technology routes for China IV and China V vehicles may account for their comparable N<sub>2</sub>O emissions.

The startup modes have a significant impact on the engine combustion state and the effectiveness of the aftertreatment systems. Generally, the cold start mode tends to enhance hydrocarbon emissions, partly due to the incomplete combustion during the warm-up phase (Drozd et al., 2016) and partly because the temperature does not reach the light-off temperature of the exhaust catalyst (Saxer et al., 2006), leading to enhanced CH<sub>4</sub> emissions during the cold start mode. In terms of CO<sub>2</sub>, lower emissions might be primarily attributed to the lower fuel consumption during the hot startup mode. Regarding N<sub>2</sub>O emissions, the higher N<sub>2</sub>O emissions observed for China IV and China V vehicles during hot startups compared to cold startups can be explained by the increased  $NO_x$  emissions during the hot start mode. Specifically, for the tested GDI vehicles, China IV vehicles exhibited  $NO_x$  emission factors of 24.2 and 32.7 mg km<sup>-1</sup> during cold and hot startup modes, respectively, while China V vehicles displayed factors of 24.9 and 27.3 mg km<sup>-1</sup> and China VI vehicles exhibited factors of 6.5 and 8.2 mg km<sup>-1</sup> for the same conditions. This disparity underscores the influence of the startup mode on  $NO_x$  and consequently on  $N_2O$  emissions.

Furthermore, GDI engines exhibited lower CO<sub>2</sub> emissions compared to PFI engines. This is attributed to the fact that GDI engines inject fuel directly into the combustion cylinder, enabling more precise control over injection time, fuel volume, and oil-gas mixing and hence a higher brake thermal efficiency and power output (Awad et al., 2020). As a result, the improved engine efficiency for GDI engines can reduce fuel consumption, ultimately leading to decreased emissions of CO2 and CH4. However, it is possible that due to the differences in vehicle configurations, the advantages of GDI engines may not fully offset the increase in CH<sub>4</sub> emissions, causing higher CH<sub>4</sub> emissions from vehicles equipped with GDI engines. Regarding the strong correlation between N<sub>2</sub>O and  $NO_x$ , herein, we compared the  $NO_x$  emissions of different engine technologies. Compared to PFI engines, GDI engines have a higher air : fuel ratio, which can have two opposing effects on the generation mechanism of  $NO_x$ , which originates from the oxidation of N2 in the air within the combustion chamber (Huang et al., 2016). On the one hand, a high air : fuel ratio provides more air to oxidize N2, but on the other hand, it can reduce the mixture temperature, constraining  $NO_x$  production. For the China VI vehicles in our study, the NO<sub>x</sub> emissions for GDI engines (6.5 and 8.2 mg km<sup>-1</sup> in cold and hot startup modes, respectively) were significantly higher than those for PFI engines (4.6 and  $3.8 \text{ mg km}^{-1}$  in cold and hot startup modes, respectively). Additionally, NO can selectively form N2O at lower temperatures (Brinklow et al., 2023). Consequently, compared to PFI engines, the higher  $NO_x$  emissions and the lower temperature for GDI engines contributed to enhanced N<sub>2</sub>O emissions.

When considering the impact of driving protocols on GHG emissions from vehicles, it is notable that the CLTC protocol, compared to the WLTC protocol, represents a low average speed, a high idle-speed ratio, and more frequent acceleration and deceleration characteristics (Liu et al., 2020). Additionally, the idling conditions during the CLTC protocol exceed 20 %, which is significantly higher than the 12.7 % in the WLTC protocol, ultimately possibly leading to lower fuel consumption. Specifically, the fuel consumption of China VI vehicles during hot starts for WLTC and CLTC protocols was 5.0 and  $4.8 L (100 \text{ km})^{-1}$ , respectively. Moreover, previous studies have reported that HEVs facilitate reduced emissions of gaseous and particulate pollutants (Zhang et al., 2024; Huang et al., 2022a). In this study, we utilized non-plug-in hybrids, which operate on the principle that the engine and generator complement each other to maintain optimal engine performance for minimizing electricity consumption. This type of vehicle cannot be charged externally, and the battery is charged by the internal combustion engine through electricity generation. Consequently, HEVs achieve lower fuel consumption (4.7 and  $4.3 \text{ L} (100 \text{ km})^{-1}$  in cold and hot startup modes, respectively) compared to gasoline vehicles with the same engine technology and emission standard (5.2 and  $5.0 L (100 \text{ km})^{-1}$  in cold and hot startup modes, respectively). Additionally, HEVs exhibit higher combustion efficiency as the engine can maintain optimal operating conditions during operation. Both of these factors combine to result in lower GHG emissions for HEVs compared to gasoline vehicles.

Overall, the China VI emission regulation marks a significant milestone in the control of GHG emissions by introducing a specific emission limit of  $20 \text{ mg km}^{-1}$  for N<sub>2</sub>O. Notably, the N<sub>2</sub>O emissions recorded from all tested vehicles in this study, including even the older China IV vehicles, fell well below this limit, indicating a positive trend toward reduced emissions. However, the current emission standards in China have yet to establish emission limit values for CO<sub>2</sub> and CH<sub>4</sub>. This lack of stringent regulations for these GHGs underscores the need for a more comprehensive and systematic approach to greenhouse gas control.

# 3.2 Influence of fuel types and three-way-catalyst operation conditions on GHG emissions

In this study, we investigated two bi-fuel taxis exhibiting substantial disparities in mileage. By comparing the CO and THC emissions between these two taxis (241 and 5719 mg km<sup>-1</sup> for CO under "TWC worked" and "TWC deteriorated" conditions and 3 and 329 mg km<sup>-1</sup> for THC under "TWC worked" and "TWC deteriorated" conditions, respectively), it was found that the taxi with a mileage of 266 624 km was equipped with a deteriorated TWC, whereas the taxi with a significantly lower mileage of 79 960 km was equipped with a worked TWC. Herein, Fig. 3 illustrates the GHG emission factors associated with various fuel types and TWC operating conditions for our tested bi-fuel taxis.

Under "TWC worked" conditions, CO<sub>2</sub> emission factors of conventional and ethanol gasoline were observed to be virtually identical, with both fuels exhibiting comparable CH<sub>4</sub> emission factors as well. As a gaseous fuel, CNG demonstrated a noteworthy reduction in CO<sub>2</sub> emissions by approximately 27 % compared to the two liquid fuels. However, this reduction was accompanied by a marked increase in CH<sub>4</sub> emissions, with CNG emitting 10.8 times more CH<sub>4</sub> than conventional gasoline and 6.5 times more than ethanol gasoline. This trend of reduced CO<sub>2</sub> emissions and elevated CH<sub>4</sub> with CNG usage has been consistently reported in previous studies (Lv et al., 2023; Rašić et al., 2017; Bielaczyc et al., 2014). The lower CO<sub>2</sub> emissions from CNG can be attributed to its lower C / H ratio. Conversely, the elevated CH<sub>4</sub> emissions from CNG are primarily due to the direct emission of unburned CN4, the primary component of CNG, from the tailpipe. Turning to N<sub>2</sub>O emissions, CO and NO serve as the principal precursors, while H<sub>2</sub> and THC also play important roles as precursor species (Brinklow et al., 2023; Nevalainen et al., 2018). The three fuel types exhibited comparable N2O emission factors, ranging from 0.3 to  $0.4 \text{ mg km}^{-1}$ . Specifically, ethanol gasoline exhibited slightly higher N2O emissions than conventional gasoline, which could be attributed to the combined effects of increased precursor concentration and reduced combustion temperatures stemming from ethanol's relatively low combustion flame temperature and calorific value compared to conventional gasoline (Qu et al., 2020). Between the two liquid fuels,  $NO_x$  emission factors were comparable, with conventional gasoline at  $11 \text{ mg km}^{-1}$  and ethanol gasoline at 10 mg km<sup>-1</sup>. Nevertheless, ethanol gasoline would emit much higher CO and THC concentrations: the CO and THC emission factors for conventional gasoline were  $240 \,\mathrm{g \, km^{-1}}$  and  $3 \,\mathrm{mg \, km^{-1}}$ , whereas those for ethanol gasoline were 407 g km<sup>-1</sup> and 7 mg km<sup>-1</sup>, respectively. Nevertheless, previous studies have revealed that ethanol gasoline would increase the emissions of non-methane organic gases (NMOGs), acetaldehydes, 1,3-butadiene, and benzene and would not lead to statistically significant changes in  $NO_x$ ,  $CO_2$ ,  $CH_4$ ,  $N_2O$  or formaldehyde emissions (Graham et al., 2008). When using gasoline with higher ethanol concentrations (e.g., E85), distinct emission characteristics can be observed. The  $NO_x$ , 1,3-butadiene, and benzene are significantly reduced, while the emissions of formaldehyde and acetaldehyde increase statistically significantly. In contrast, there are no statistically significant changes in the emissions of CO, CO<sub>2</sub>, or NMOGs (Graham et al., 2008). When comparing ethanol gasoline to CNG, CNG demonstrated lower CO emission factors (158 g km<sup>-1</sup> for CNG versus 407 g km<sup>-1</sup> for ethanol gasoline) but higher NO<sub>x</sub> and THC emission factors (15 mg km<sup>-1</sup> for NO<sub>x</sub> and 9 mg km<sup>-1</sup> for THC versus 10 and 7 mg km<sup>-1</sup>, respectively, for ethanol gasoline). Notably, CNG has a higher combustion temperature (Lv et al., 2023; Chen et al., 2019), which may be detrimental to conversion of NO to  $N_2O$  (Brinklow et al., 2023). Consequently, in the  $N_2O$  generation process, the potential advantage of higher precursor concentrations in CNG may be counteracted by the unfavorable effects of high temperatures, resulting in comparable  $N_2O$  emission factors for CNG and ethanol gasoline.

TWC converters are instrumental in mitigating vehicular emissions. Under "TWC deteriorated" conditions, notable enhancement in CH<sub>4</sub> emissions was recorded for the three fuel types, which was similar to the good removal ability of TWC for THC as reported in previous studies (Woo Jeong et al., 2024). Specifically, the CH<sub>4</sub> emission factors for conventional gasoline under "TWC deteriorated" conditions were around 130 times higher, while those for ethanol gasoline were approximately 110 times higher and those for CNG were around 9 times higher, compared to the "TWC worked" conditions. Intriguingly, CO2 emissions from both conventional and ethanol gasoline exhibited lower emission factors under "TWC deteriorated" conditions compared to "TWC worked" conditions. On the one hand, this may be related to the unexpectedly lower fuel consumption for "TWC deteriorated" conditions  $(4.8 L (100 \text{ km})^{-1} \text{ for conventional gaso-}$ line and  $4.9 L (100 \text{ km})^{-1}$  for ethanol gasoline) compared to "TWC worked" conditions  $(5.8 L (100 \text{ km})^{-1} \text{ for conven-}$ tional gasoline and  $5.8 L (100 \text{ km})^{-1}$  for ethanol gasoline). On the other hand, from the perspective of considering the aftertreatment system, this could also be explained by the underlying operating mechanism of TWC converters. When the high-temperature exhaust passes through TWC, the catalyst within it enhances the reactivity of CO, HC, and  $NO_x$ , catalyzing oxidation and reduction reactions. This process converts CO into CO<sub>2</sub>, HC into H<sub>2</sub>O and CO<sub>2</sub>, and NO<sub>x</sub> into N<sub>2</sub> and O<sub>2</sub> (De Abreu Goes et al., 2021). Consequently, under "TWC deteriorated" conditions, these conversions are hindered, leading to reduced CO<sub>2</sub> generation and correspondingly lower emission factors.

Regarding N<sub>2</sub>O, its generation pathway is related to the primary precursors, notably CO, NO<sub>x</sub>, and THC, with lower temperatures facilitating N<sub>2</sub>O formation (Brinklow et al., 2023). The formation of  $N_2O$  over TWC converters is the result of a complex interplay of factors, including the active metal composition, converter aging, exhaust temperature, air: fuel ratio, and feed gas composition (Nevalainen et al., 2018; Mejía-Centeno et al., 2007). For both conventional and ethanol gasoline, despite the substantial increases in CO and THC emissions observed under "TWC deteriorated" conditions compared to "TWC worked" conditions, N<sub>2</sub>O emission factors did not correspondingly become elevated. This phenomenon could be attributed to the significant reduction in NO<sub>x</sub> emissions under "TWC deteriorated" conditions, which were only one-seventh of those recorded under "TWC worked" conditions. This suggests that  $NO_x$  may be a pivotal precursor in determining N2O production other than CO and THC. Similar to prior research highlighting that catalyst aging would change the structure of oxygen storage material and thereby cause an enhancement of N<sub>2</sub>O formation (Nevalainen et al., 2018), our findings reveal a notable increase in N<sub>2</sub>O emissions for CNG under "TWC deteriorated" conditions, reaching approximately 3 times that of "TWC worked" conditions. This elevation is likely attributable to heightened precursor concentrations, particularly NO<sub>x</sub>. Our study underscores the variability in the impact of TWC deterioration on N<sub>2</sub>O emissions across different fuel types corresponding to different combustion states. This highlights the need for future research to delve deeper into identifying the primary driver of N<sub>2</sub>O generation and elucidating the underlying mechanism governing N<sub>2</sub>O generation under diverse fuel usage conditions.

Overall, the difference in GHG emissions between conventional gasoline and ethanol gasoline is not significant. The application of CNG can effectively reduce  $CO_2$  emissions but significantly increase  $CH_4$  emissions. In the context of CNG usage,  $CO_2$  was not sensitive to TWC aging, while the combined effect of CNG usage and TWC aging led to a substantial increase in  $CH_4$  emissions. Additionally, N<sub>2</sub>O emissions are minimally affected by the substitution of clean fuels such as CNG but are greatly influenced by TWC deterioration.

#### 3.3 GHG emissions from hybrid electric vehicles

Previous studies have reported that HEVs facilitate reduced emissions of gaseous and particulate pollutants (Zhang et al., 2024; Huang et al., 2022a). In this study, we focused on non-plug-in hybrids, which operate on the principle that the engine and generator complement each other to maintain optimal engine performance for minimizing electricity consumption. This type of vehicle cannot be charged externally, and the battery is charged by the internal combustion engine through electricity generation. Consequently, HEVs exhibited lower fuel consumption, achieving 4.7 and  $4.3 \text{ L} (100 \text{ km})^{-1}$  in cold and hot startup modes, respectively, compared to gasoline vehicles, which consumed 5.2 and  $5.0 \,\mathrm{L} \,(100 \,\mathrm{km})^{-1}$  under the same conditions. Furthermore, HEVs showed higher combustion efficiency as the engine can maintain optimal operating conditions during operation. These factors, when combined, lead to a substantial reduction in GHG emissions for HEVs compared to gasoline vehicles. Specifically, CO<sub>2</sub> emission factors were 110.8 and  $102.0 \,\mathrm{g \, km^{-1}}$  for cold and hot startup modes, respectively, while  $CH_4$  emission factors stood at 1.2 and  $0.6 \text{ mg km}^{-1}$ and N<sub>2</sub>O emission factors at 0.4 and  $0.5 \text{ mg km}^{-1}$  for the same conditions. A comparative analysis between gasoline vehicles and HEVs underscores the advancements in powertrain technology. Notably, despite the HEV in our study having a higher overall mass and higher displacement, it demonstrated remarkable reductions in GHG emissions. In particular, CO<sub>2</sub> emissions were reduced by 10 % to 14 %, CH<sub>4</sub> emis-



Figure 3. Emission factors of GHGs (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) for different fuel types and TWC operating conditions of our tested bi-fuel taxis.

sions by 11 % to 37 %, and  $N_2O$  emissions by 23 % to 43 %, highlighting the environmental benefits of HEV technology.

HEVs, when compared to conventional gasoline vehicles, utilize electric motors for propulsion during startup and lowspeed driving conditions (approximately  $20 \text{ km h}^{-1}$ ). As depicted in Fig. 4, HEVs exhibited a delayed and significantly reduced CO<sub>2</sub> emission profile during these phases. This reduction is attributed to the electric motor's efficiency and the absence of fuel combustion. However, when the vehicle accelerates beyond a certain threshold, the internal combustion engine (ICE) is activated to provide additional power. At this juncture, there was a marked increase in CO<sub>2</sub> emissions, reflecting the higher fuel consumption required to initiate the engine's operation. This transient spike underscores the instantaneous fuel demand and combustion process associated with engine startup in HEVs.

As presented in Fig. 5, an examination of the various speed phases within the WLTC revealed that HEVs demonstrated notably lower CO<sub>2</sub> emissions during low-speed segments, especially under hot start conditions. This reduction in CO<sub>2</sub> emissions can be attributed to the efficient operation of the electric motor at lower speeds. Conversely, the emission factors for CH<sub>4</sub> and N<sub>2</sub>O were observed to be relatively high in these low-speed segments, indicating a tradeoff in emission profiles. When compared to conventional gasoline vehicles, HEVs generally exhibited lower emissions across a range of pollutants. The exception to this trend was the CH<sub>4</sub> emissions during cold starts at low speeds, where HEVs display elevated levels. This anomaly suggests that while HEVs are more efficient overall, certain conditions may lead to increased emissions of specific pollutants. For example, the analysis of CO emissions from hybrid vehicles reveals significant advantages when compared to conventional fuel-powered vehicles. Specifically, during cold starts, hybrid vehicles exhibit a CO emission factor of  $70.9 \,\mathrm{mg}\,\mathrm{km}^{-1}$ , whereas their conventional counterparts register 95.7 mg km<sup>-1</sup>, reflecting a 26.8 % reduction in CO emissions under these conditions. Similarly, during hot starts, hybrid vehicles achieve an impressive CO emission factor of 43.3 mg km<sup>-1</sup>, compared to the 71.9 mg km<sup>-1</sup> emitted by conventional vehicles, demonstrating a 40.5 % decrease. This indicates that hybrid vehicles have more advantages in terms of combustion efficiency comparison.

#### 3.4 Global warming potentials of light-duty vehicles

As shown in Fig. 6, vehicle exhaust emissions, particularly the emission of  $CO_2$ , remain the primary contributor to global warming potential, accounting for over 99% of the total of the three GHGs. Even though CH<sub>4</sub> and N<sub>2</sub>O have global warming potentials 25 and 298 times higher, respectively, than that of  $CO_2$ , the sheer volume of  $CO_2$  emissions from vehicle tailpipes still dominates. Furthermore, for conventional gasoline-powered vehicles, the GWP values tended to decrease as emission standards became more stringent. However, for China VI vehicles, those equipped with PFI engines exhibited higher GWP compared to those with GDI engines, with GDI vehicles showing a 17.6 % reduction compared to PFI vehicles. This difference highlights the varying efficiency and emission profiles between the two types of engine technology. GDI technology allows for more precise fuel delivery and better combustion efficiency, which translates into lower CO<sub>2</sub> emissions. Furthermore, the impact of hybrid engine technology on GWP values is particularly noteworthy. HEVs combine an internal combustion engine with an electric motor, which enhances fuel efficiency and reduces emissions. The data indicate that HEVs achieve a further 12.0 % reduction in GWP compared to conventional GDI vehicles. Therefore, vehicles with GDI engines and hybrid technology exhibit lower GWP values.

For bi-fuel taxis, under "TWC worked" conditions, the fuel consumption is higher when using ethanol gasoline and conventional gasoline compared to using CNG. At the same time, due to the catalytic effect of TWC, CO and HC can be converted into  $CO_2$ , resulting in higher GWP values. This indicates that bi-fuel vehicles can benefit from alternative fuels, especially the use of CNG, which can effectively reduce the emissions of vehicular GHGs. Unexpectedly, under "TWC



Figure 4. Time series of CO<sub>2</sub> and vehicle speed in the WLTC protocol with cold starts (a) and hot starts (b) of the HEV and gasoline vehicle.



Figure 5. Emission factors of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O under different speed phases under the WLTC protocol for the China VI HEV and China VI gasoline vehicle.

deteriorated" conditions, lower GWP values were observed compared to those under "TWC worked" conditions, with a decrease of approximately 27.8 %. This contradictory finding indicates that although TWC is effective in reducing pollutants such as CO, HC, and  $NO_x$ , its level of control over greenhouse gas emissions may still be worth further discussion. Overall, the deterioration of TWC may lead to a reduction in GWP, but this is also accompanied by the risk of increased emissions of other pollutants, indicating a complex interaction between emission control technologies and overall environmental impacts.

#### 4 Conclusions

Vehicular emissions constitute a pivotal contributor to atmospheric pollutants and greenhouse gases in the troposphere. While extensive research has been conducted on the emission characteristics of gaseous pollutants emanating from vehicle exhausts, there remains a notable scarcity of studies focusing on greenhouse gas emissions, particularly in the context of achieving carbon neutrality. To address this gap, our study conducted a comprehensive investigation into the emission characteristics of three important GHG (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O) emanating from a diverse fleet of 11 light-duty vehicles, encompassing conventional gasoline cars, hybrid electric vehicles, and bi-fuel taxis. The main findings of our research are outlined below.

Compared to China IV and China V vehicles, China VI vehicles significantly reduce greenhouse gas emissions, which might be related to the low  $CO_2$  and  $CH_4$  emissions of China VI vehicles being associated with their low fuel consump-



Figure 6. Global warming potentials of private car and taxi exhaust emissions.

tion, while the low  $N_2O$  emissions are associated with their aftertreatment technologies.

Compared to PFI engines, GDI engines can reduce  $CO_2$  emissions but have adverse effects on  $CH_4$  and  $N_2O$  emissions. The different responses of these three greenhouse gases to engine technologies may stem from variations in combustion efficiency, the air:fuel ratio, and combustion temperature between the two engine technologies.

In contrast to hot startup mode, cold starts promote  $CO_2$ and  $CH_4$  emissions, while N<sub>2</sub>O emissions exhibit an opposing trend, which is mainly related to the higher NO<sub>x</sub> concentrations during hot starts.

Under the "TWC worked" conditions, the GHG emissions from conventional gasoline and ethanol gasoline are roughly equivalent. However, CNG, as a clean fuel, can significantly reduce  $CO_2$  emissions, whereas it promotes  $CH_4$  emissions since  $CH_4$  is its primary component.

Unexpectedly, the CO<sub>2</sub> emissions from the two liquid fuels under the "TWC deteriorated" conditions exhibit lower emission factors than those under the "TWC worked" conditions. In the case of N<sub>2</sub>O emissions, the TWC deterioration does not lead to higher N<sub>2</sub>O emissions when using conventional gasoline or ethanol gasoline, mainly because of the significant reduction in NO<sub>x</sub> under the "TWC deteriorated" conditions despite the significant increase in CO and THC. However, under CNG usage, the emission factor of N<sub>2</sub>O under "TWC deteriorated" conditions is approximately 3 times that under "TWC worked" conditions. This significant increase is primarily associated with a substantial increase in NO<sub>x</sub>.

In terms of greenhouse gas emission reduction, hybrid vehicles demonstrate a significant advantage over conventional gasoline vehicles primarily due to the usage of electric motors. Specifically, hybrid vehicles exhibit notably lower  $CO_2$  emissions in low-speed phases, while emissions of  $CH_4$  and  $N_2O$  tend to be relatively high at these speeds.

The global warming potential from vehicle exhaust is primarily attributed to CO<sub>2</sub> emissions. HEVs demonstrate a more advantageous environmental impact compared to conventional gasoline vehicles. Surprisingly, however, under the "TWC deteriorated" conditions, a lower GWP value has been observed, indicating a complex interaction between emission control technologies and environmental impact.

This study aims to provide insights into the emission characteristics of GHGs from vehicles, which are crucial for developing targeted strategies to reduce GHG emissions and enhance the overall efficiency and environmental performance of vehicle emission control systems. However, a limitation of this study is that the vehicle fleet remains relatively small. In future research, we should investigate motor vehicle GHG emissions more systematically while simultaneously measuring auxiliary parameters such as catalyst temperature to facilitate a mechanistic understanding of GHG formation. This will aid in managing and controlling the GHG emissions from vehicles in the future.

**Code and data availability.** The data used in this publication are available at https://doi.org/10.5281/zenodo.15253351 (Yang et al., 2025), and they can be accessed by request to the corresponding authors.

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