Measurement report: Insight into Greenhouse Gas Emission Characteristics of Light-Duty Vehicles in China Driven by 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_2, they unexpectedly increased CH_4 and N_2O emissions. Additionally, hot starts, beneficial for reducing CO_2 and CH_4 emissions, caused heightened N_2O emissions, which is noteworthy under operating conditions with frequent start-stop events. Intriguingly, compressed natural gas (CNG), generally perceived as cleaner, increased CH_4 emissions. Regarding the impact of Three-Way Catalysts (TWC) on GHG emissions, under "~~TWC failed~~TWC deteriorated" conditions, N_2O emissions from CNG-powered vehicles were approximately three times higher than those under "TWC worked" conditions, attributed to the significant increase in nitrogen oxides (NOx). Considering the global warming potential (GWP), the "~~"~~TWC deteriorated"~~failure"~~ 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 (GHGs) emissions has emerged as a critical objective for environmental protection. In 2023, global energy-related carbon dioxide (CO_2) 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_2 emissions (Liu et al., 2023). In 2023, the number of motor vehicles in China had reached 435.6 million; Among them, the number of automobiles reached 336 million, a year-on-year increase of 5.3% (Ministry of Ecology and Environment, 2024). By the end of 2022, China's vehicle ownership had reached 319 million units (Ministry of Ecology and Environment, 2023). Due to the large population of vehicles, the road transportation sector’'s contribution to CO_2 emissions is significant. One research indicated that in 2019, China'’s vehicle CO_2 emissions were 952 Mtmillion tonnes, with gasoline and other fuels (natural gas, alcohol fuels) comprising 47.5% of this figure (Huang et al., 2022b)(Zhihui et al., 2022). A recent study found that GHG emissions rose from 431 million tonsMt in 2005 to 807 million tonsMt in 2015, with an annual growth rate of 6.5% (Li et al., 2019). Many researches showed 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_2 emissions in the entire transportation sector in 2015 (Zhang et al., 2019); thus, road-based transport has a much larger CO_2 emission reduction potential compared to other transport modes (Wang et al., 2017).

In addition to CO_2, vehicular activities are closely associated with the greenhouse gases methane (CH_4) and nitrous oxide (N_2O). The N_2O emissions are primarily produced by the combustion of nitrogenous compounds in fuel, while CH_4 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_2O and CH_4 from vehicles are 3 to 6 orders of magnitude smaller than those of CO_2, their Global Warming Potential (GWP) is significantly higher. According to the IPCC (2014), theThe GWP of N_2O and CH_4 is 298 times and 25 times greater than that of CO_2, respectively (IPCC, 2014). While the emissions of CH_4 from motor vehicles may be negligible on a global scale, in urban areas, particularly those with heavy traffic, CH_4 emissions can account for up to 30% of the regional CH_4 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_2O and CH_4 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 HEVs is crucial for developing effective strategies to mitigate emissions from the light-duty vehicle sectors. Meanwhile, the Three-Way Catalysts (TWC) system remains the predominant after-treatment 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 the reduction of nitrogen oxides (NOx). Notably, the NOx reduction process involves undesirable side reactions, particularly the interaction between NO and nitrogen species on the catalyst, resulting in the formation of N_2O (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 peak CO_2 around 2030 and strive to achieve it as soon as possible. The government has incorporated climate change into its ecological planning and built a low-carbon system of society and economy.

Currently, research on vehicular GHG emissions both domestically and internationally mainly employs three methods: chassis dynamometer testing, on-board 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 ~~He et al. and Zhong et al. conducted tests on a~~actual vehicles under different emission standards, temperature points and driving operating conditions ~~to~~ were obtain ~~emission factors for CH_4 and N_2O~~ (He et al., 2014;Zhong et al., 2023;Clairotte et al., 2020;Wang et al., 2022b). Specifically, CO_2 emissions for China VI (335 g/km) diesel vehicles have been reduced compared to China IV (415 g/km) and China V (447 g/km) vehicles (Li et al., 2024), and some studies reported CO_2 emissions for China VI gasoline vehicles were around 200 g/km to 300 g/km (Zhu et al., 2022;Wang et al., 2022b). The CH_4 emissions decreased from 48 mg/km for China I to 28 mg/km for China IV light-duty gasoline vehicles, and N_2O emissions were reduced from 45 mg/km (China I) to 21 mg/km (China IV) (He et al., 2014). The CH_4 and N_2O emissions for Euro V to Euro VI vehicles were found to be 7 mg/km (Clairotte et al., 2020), the average emission factors of CO_2 and CH_4 for light-duty vehicles in Thailand, which were 232.25 g/km and 9.50 mg/km, respectively (Sirithian et al., 2022). With advancements in powertrain technologies, hybrid electric vehicles are being progressively promoted and have demonstrated significant potential in reducing~~Selleri et al. used the WLTC and the Real Drive Emission (RDE) test procedures to test three types of plug-in hybrid electric vehicles, finding that the emissions of~~ both pollutants and ~~carbon dioxide~~CO_2 emissions~~were significantly reduced in hybrid vehicles~~ (Selleri et al., 2022). Furthermore, from the model simulation perspective, ~~Tang et al., Li et al., and Zeng et al. used~~ computational models such as International Vehicle Emissions (IVE), Motor Vehicle Emission Simulator (MOVES), and Gompertz Growth Model have utilized to establish greenhouse gas emission inventories for different base years (Tang et al., 2018;Li et al., 2022;Zeng et al., 2016), even to ~~. Zeng et al. also~~ predict~~ed~~ the future greenhouse gas emissions (Zeng et al., 2016). ~~Wang et al. used the World Light Vehicle Test Cycle (WLTC) protocol to test the CO_2 emission characteristics of vehicles at different temperature points (Wang et al., 2022b). International researchers have also conducted numerous studies: Clairotte et al. monitored the average emission factors of CH_4 and N_2O for light-duty vehicles from Euro 5b to Euro 6d, which were found to be 7 mg/km (Clairotte et al., 2020); Sirithian et al. measured the average emission coefficients of CO_2 and CH_4 for light-duty vehicles in Thailand, which were 232.25 g/km and 9.50 mg/km, respectively (Sirithian et al., 2022); Selleri et al. used the WLTC and the Real Drive Emission (RDE) test procedures to test three types of plug-in hybrid electric vehicles, finding that the emissions of pollutants and carbon dioxide were significantly reduced in hybrid vehicles (Selleri et al., 2022).~~

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Overall, from the perspective of GHG emissions from ~~The level of GHG emissions from~~ domestic vehicles in China, ~~during driving is still in need of further research.~~ Yin et al. ~~adopted the WLTC to study~~explored the GHG emissions of 84 China VI light-duty gasoline vehicles, ~~including six hybrid vehicles (Yin et al., 2024). The results~~ indicating~~ed~~ that vehicular ~~g~~ GHG emissions generally tend to increase with engine displacement~~. Vehicles with larger displacements often have more complex engine designs, which adds to the uncertainty of greenhouse gas emissions.~~ and ~~The~~ the CO_2 increase rate caused by the CO_2 conversion of CH_4 and N_2O emissions from all types of vehicles was less than 1%, suggesting that CO_2 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_2, CH_4, and N_2O emission profiles from vehicles certified under China's most stringent emission standards. Notably, old vehicles with deteriorated after-treatment 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 "super-emitters" remains incomplete. To address these gaps, the key investigations are urgently required, one is the comprehensive domestic emission profiles for vehicles complying with China's latest emission standards and their comparison with previous emission standards, another is the GHG emission characteristics from "super-emitters".

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 different working conditions of ~~post~~after-treatment ~~-treatment~~ devices, aiming at improving the characteristics of GHG emission factors for light-duty vehicles and explore 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 of greenhouse gas emission inventory calculations.

2 Materials and methods

2.1 Tested vehicles and fuels

Eleven in-use light-duty vehicles, including ten Internal Combustion Engine Vehicles (ICEVs) and one 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, and mileage accumulations, as detailed in Table 1. Among the eight light-duty gasoline vehicles (vehicles #1 to #8), representation was provided from 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 VI compliant vehicles. These vehicles featured two distinct engine techniques: Gasoline Direct Injection (GDI) and Port Fuel Injection (PFI), all equipped with ~~Three-Way Catalysts (~~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~~ to assess the GHG emission differences between gasoline and hybrid electric vehicles, one non-plug-in

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hybrid electric light-duty vehicle (vehicle #9) adhering to China VI emission standard was also included, utilizing both conventional gasoline engines and electric motors, potentially influencing overall GHG emissions.

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 after-treatment types.

Vehicle ID	Category	Model year	Emission standard	Engine technology	Displacement (mL)	Mileage (10^4 km)	Max. net engine power (kW)	Max. authorized mass (kg)	After-treatment
#1	ICEV[a]	2011	China IV	GDI[b]	1798	22.11	118	2000	TWC[c]
#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[~~d~~e]	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

[a] Internal Combustion Engine Vehicle. [b] Gasoline Direct Injection. [c] Three-Way Catalysts. [d] The unrecorded data, being a non-essential parameter, have no bearing on the subsequent analysis. [~~d~~e] Port Fuel Injection.

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 failed~~TWC deteriorated" condition) and another only 79,960 km (representing the "TWC worked" condition). Figure S1 in the Supplementary Information 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 gasoline~~-/~~compress 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 ℃) before transitioning to CNG (Wang et al., 2024).

In ~~our~~ this study, we employed three types of fuels: conventional gasoline, ethanol gasoline, and compressed natural gas (CNG). The conventional gasoline, exclusively utilized in light-duty gasoline vehicles and HEVs, was sourced from the

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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 Supplementary 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 the 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 type-approval testing center qualified by the Ministry of Ecology and Environment of China. The tailpipe pollutants are diluted by a Constant Volume Sampling (CVS) method, followed by analysis using an array of measurement systems, as shown in Figure 1. For GHG quantification, specific instruments were employed: the Non-Dispersive InfraRed (NDIR) analyzer for CO_2 and the Quantum Cascade Laser (QCL) analyzer for N_2O, ensuring precise measurements. Additionally, CH_4 concentrations were determined by a setup combining a Non-Methane Cut-off (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.

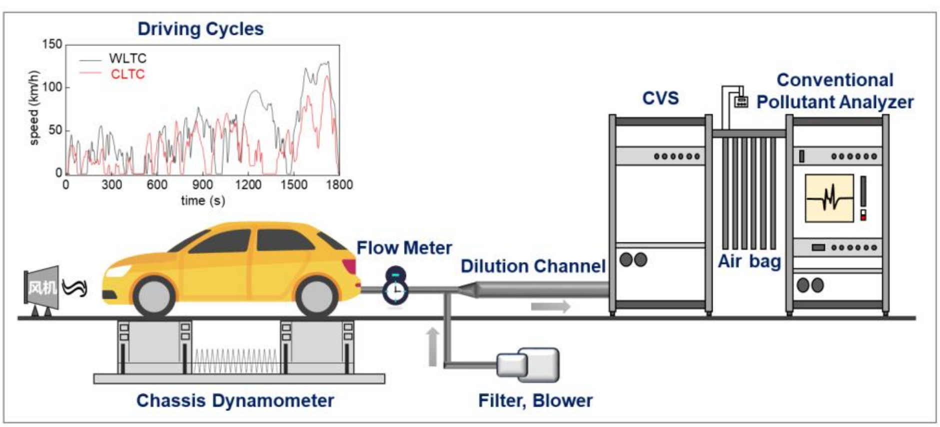

Figure 1: Schematic diagram for vehicular emission measurements based on chassis dynamometer in this study.

To comprehensively investigate the diverse driving conditions on GHG emissions, we implemented three distinct types of driving cycles. For gasoline vehicles and HEVs, we adhered to the Worldwide Harmonized Light Vehicles Test Cycle (WLTC) newly introduced in the China VI emission regulation. The WLTC protocol comprises four phases: low-speed (589

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seconds), medium-speed (433 seconds), high-speed (455 seconds), and extra-high-speed (323 seconds) phases. The impacts of both cold and hot startup modes on vehicular emissions were explored during 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 real-world driving conditions in China. This protocol, which encompasses the 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 km/h, 60 km/h, 90 km/h, and 120 km/h. During these constant speed tests, the driver accelerated to the target speed and maintained it for 10 minutes, 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 (GWP) 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_4 and N_2O emissions into their CO_2 equivalents, thereby facilitating a standardized comparison. Furthermore, the relative growth rate of CO_2 after conversion can be obtained, which is precisely calculated according to Eq. (1), offering valuable insights into the long-term warming potential of these gases.

$$R_{CO_2}=(\mathrm{GWP} \times M_i)/M_{CO_2} \quad (1)$$

where R_{CO_2} is the relative growth rate of CO_2, M_i denotes the emissions of CH_4 or N_2O, and M_{CO_2} denotes the emissions of CO_2. The GWP of N_2O and CH_4 is 298 and 25, respectively.

3 Results and Discussion

3.1 Scenario-Based Greenhouse Gas ~~(GHG)~~ Emission Factors

3.1.1 Distance-based emission factors

Figure 2~~1~~ 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 emission standard categories, distinct trends emerged across the three primary GHG types. Prior research has documented a pronounced reduction in ~~carbon monoxide (~~CO~~)~~, ~~nitrogen oxides (~~NOx~~)~~, and ~~total hydrocarbons (~~THC~~)~~ emissions in response to stricter emission standards (Duan et al., 2021). Similarly, in this study, CH_4 emission factors have undergone a substantial decline, with the China VI gasoline vehicle (during cold start) exhibiting an emission factor ~~that was~~of approximately one-sixth of that recorded for the China IV gasoline vehicle under similar conditions. This trend in CH_4 is consistent with previously reported variations in VOCs (Qi et al., 2021;Duan et al., 2021). ~~Clairotte et al. (2020)~~Turning to CO_2 and N_2O, Figure 2~~1~~ highlights that the emission factors for these gases in China VI

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gasoline vehicles were notably lower ~~compared to~~than those in China IV and China V vehicles. Nevertheless, the emission factors for CO_2 of the China V vehicles (155 g/km for the hot start) were comparable to those of China IV vehicles (151 g/km for the hot start) and those for N_2O of the China V vehicles (2.0 mg/km for the hot start) even were higher than those of China IV vehicles (1.5 mg/km for the hot start), particularly for hot start mode. It highlights a nuanced aspect of emission performance that needs further investigation. A comparison with previous studies shows that the CH_4 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)~~,~~ ~~and a~~. A similar trend was observed for N_2O emissions, as demonstrated by the summary of previous studies provided in Table S1 in the Supplementary Information. Furthermore, lower CO_2 emissions was found in our study than 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) in Table S1 in the Supplementary Information.

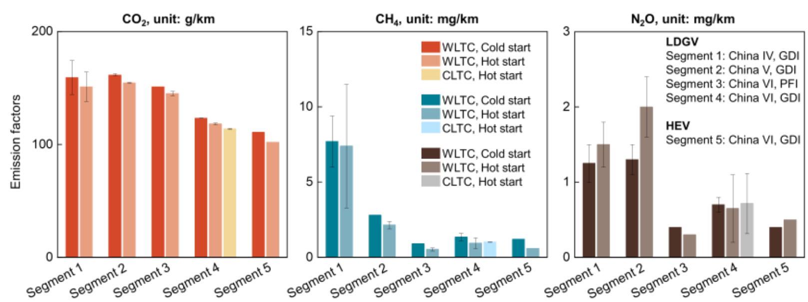

Figure 2~~1~~: Emission factors of GHGs (CO_2, CH_4, and N_2O) for various scenarios involving different emission standard categories, engine techniques, powertrain technology, startup modes, and driving cycles.

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 g/km under cold startup mode and 151 g/km under hot startup mode. Similarly, for the tested China V vehicles, the CO_2 emission factors were 161 g/km and 155 g/km, respectively, for cold and hot startup modes. The China VI vehicles also exhibited a similar trend, with CO_2 emission factors of ~~15~~151~~9~~ g/km and ~~151~~ 145 g/km for cold and hot startup modes, respectively. Similarly, the CH_4 emissions displayed a declining trend, transitioning from cold starts to hot starts across all tested vehicle categories. For China IV vehicles, the CH_4 emission factors were 7.7 mg/km under cold starts and 7.4 mg/km under hot starts. China V vehicles showed a reduction from 2.8 mg/km to 2.2 mg/km, and China VI vehicles from 1.4~~35~~ mg/km to ~~0.95~~1.0 mg/km. Interestingly, the trend for N_2O emissions was inverse to that of CO_2 and CH_4, 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 Section 3.1.2.

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When it comes to engine techniques, a notable trend emerged in the comparison between PFI and GDI engines. Specifically, PFI engines exhibited higher CO_2 emission factors compared to GDI engines. Conversely, GDI engines demonstrated higher CH_4 and N_2O emissions. In terms of testing protocols, the CLTC protocol yielded slightly lower CO_2 emission factors by approximately 4% compared to the WLTC protocol. However, the CH_4 and N_2O 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_2 emissions were reduced by 10% to 14%, CH_4 emissions by 11% to 37%, and N_2O emissions by 23% to 43%.

3.1.2 Interpreting technological upgrading through the ~~lens~~perspective of generation mechanisms

The different trends of GHG emissions across various scenarios might be attributed to the underlying mechanism of ~~GHG~~ 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_2O, it may arise from the reaction between ammonia (NH_3) and NOx or from the decomposition of ammonium nitrate (NH_4NO_3) generated in catalytic converters, suggesting a greater dependence on NOx and NH_3 concentrations (Yin et al., 2024;Brinklow et al., 2023).

The fuel consumption for China VI vehicles (5.0 L/100km for the hot start) was lower than that of China IV (6.4 L/100km for the hot start) and China V (6.6 L/100km for the hot start) vehicles. The lower fuel consumption for China VI vehicles caused the lower CO_2 and CH_4 emissions. Furthermore, N_2O emissions are primarily derived from reactions in the aftertreatment systems. Similar after-treatment technology routes for China IV and China V vehicles may account for their comparable N_2O 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_4 emissions during the cold start mode. In terms of CO_2, lower emissions might be primarily attributed to the lower fuel consumption during the hot startup mode. Regarding N_2O emissions, the higher N_2O emissions observed for China IV and China V vehicles during hot startup compared to cold startup can be explained by the increased NOx emissions during the hot start mode. Specifically, for the tested GDI vehicles, China IV vehicles exhibited NOx emission factors of 24.2 mg/km and 32.7 mg/km during cold and hot startup modes respectively, while China V vehicles displayed factors of 24.9 mg/km and 27.3 mg/km and China VI vehicles exhibited factors of 6.5 mg/km and 8.2 mg/km for the same conditions. This disparity underscores the influence of the startup mode on NOx and consequently N_2O emissions.

Furthermore, GDI engines exhibited lower CO_2 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 CO_2 and CH_4. 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_4 emissions, causing higher CH_4 emissions from vehicles equipped with GDI engines. Regarding the strong correlation between N_2O and NOx, herein, we compared the NOx 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 NOx, which originates from the oxidation of N_2 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 N_2, but on the other hand, it can reduce the mixture temperature, constraining NOx production. For the China VI vehicles in our study, the NOx emissions for GDI engines (6.5 mg/km and 8.2 mg/km in cold and hot startup modes, respectively) were significantly higher than those for PFI engines (4.6 mg/km and 3.8 mg/km in cold and hot startup modes, respectively). Additionally, NO can selectively form N_2O at lower temperatures (Brinklow et al., 2023). Consequently, compared to PFI engines, the higher NOx emissions and the lower temperature for GDI engines contributed to enhanced N_2O 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 start for WLTC and CLTC protocols was 5.0 L/100km and 4.8 L/100km, 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 L/100km and 4.3 L/100km in cold and hot startup modes, respectively) compared to gasoline vehicles with the same engine technology and emission standard (5.2 L/100km and 5.0 L/100km 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 mg/km for N_2O. Notably, the N_2O 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_2 and CH_4. 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 mg/km and 5719 mg/km for CO under "TWC worked" and "~~TWC failed~~TWC deteriorated" conditions, 3 mg/km and 329 mg/km for THC under "TWC worked" and "~~TWC failed~~TWC deteriorated" conditions, respectively), it can be found that the taxi with 266,624 km of mileage was equipped with a deteriorated ~~failed~~ TWC, whereas the taxi with a significantly lower mileage of 79,960 km was equipped with a worked TWC. ~~Furthermore~~Herein, Figure 3~~2~~ illustrates the GHG emission factors associated with various fuel types and TWC operating conditions for our tested bi-fuel taxis.

Under "TWC worked" conditions, ~~the~~ CO_2 emission factors of conventional and ethanol gasoline were observed to be virtually identical, with both fuels exhibiting comparable CH_4 emission factors as well. As a gaseous fuel, CNG demonstrated a noteworthy reduction in CO_2 emissions by approximately 27% compared to the two liquid fuels. However, this reduction was accompanied by a marked increase in CH_4 emissions, with CNG emitting 10.8 times more CH_4 than conventional gasoline and 6.5 times more than ethanol gasoline. This trend of reduced CO_2 emissions and elevated CH_4 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_2 emissions from CNG can be attributed to its lower C/H ratio. Conversely, the elevated CH_4 emissions from CNG are primarily due to the direct emission of unburned CN_4, the primary component of CNG, from the tailpipe. Turning to N_2O emissions, CO and NO serve as the principal precursors, while H_2 and THC also play important roles as precursor species (Brinklow et al., 2023;Nevalainen et al., 2018). The~~In terms of N_2O emissions, the~~ three fuel types exhibited comparable N_2O emission factors, ranging from 0.3 mg/km to 0.4 mg/km. Specifically, ethanol gasoline exhibited slightly higher N_2O 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). Among the two liquid fuels, NOx emission factors were comparable, with conventional gasoline at 11 mg/km and ethanol gasoline at 10 mg/km. Nevertheless, ethanol gasoline would emit much higher CO and THC concentrations, the CO and THC emission factors for conventional gasoline were 240 g/km and 3 mg/km, whereas those for ethanol gasoline were 407 g/km and 7 mg/km, respectively. Nevertheless, previous studies have revealed that ethanol gasoline would increase the emissions of Non-Methane Organic Gases (NMOG), acetaldehydes, 1,3-butadiene, and benzene; and no statistically significant changes in NOx, 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 NOx, 1,3-butadiene, and benzene are significantly reduced, while the emissions of formaldehyde and acetaldehyde increase statistically significantly. In contrast, there are no statistically changes in the emissions of CO, CO_2, or NMOG (Graham et al., 2008). When comparing ethanol gasoline to CNG, CNG demonstrated

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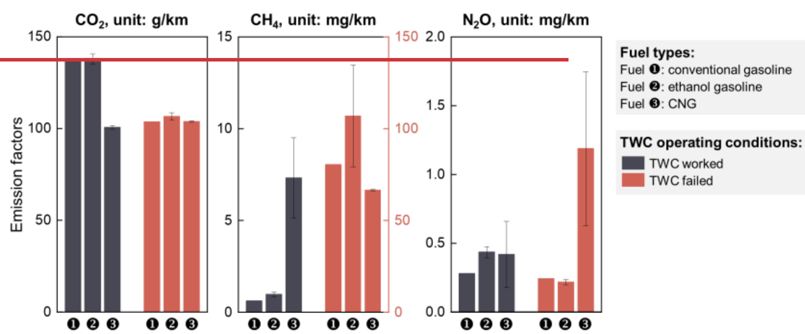
lower CO emission factors (158 g/km for CNG versus 407 g/km for ethanol gasoline) but higher NOx and THC emission factors (15 mg/km for NOx and 9 mg/km for THC versus 10 mg/km and 7 mg/km respectively for ethanol gasoline. Notably, CNG has a higher combustion temperature (Lv et al., 2023;Chen et al., 2019), which may be detrimental to ~~the~~ conversion of NO to N_2O (Brinklow et al., 2023). Consequently, in the N_2O generation process, ~~The~~ 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.



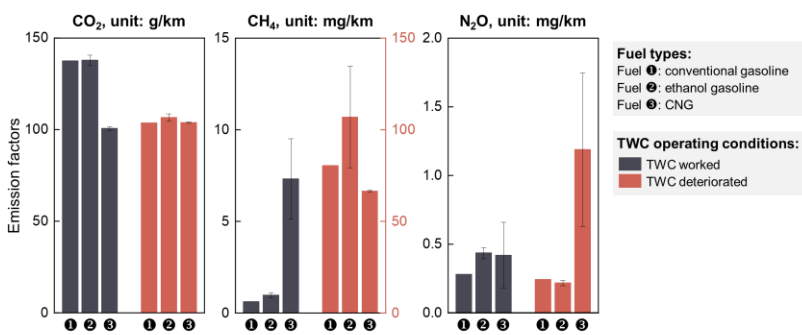


Figure 3~~2~~: Emission factors of GHGs (CO_2, CH_4, and N_2O) for different fuel types and TWC operating conditions of our tested bi-fuel taxis.

TWC converters are instrumental in mitigating vehicular emissions. Under "~~TWC failed~~TWC deteriorated" conditions, notable enhancement in CH_4 emissions ~~were~~ 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_4 emission factors for conventional gasoline under "~~TWC failed~~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

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"TWC worked" conditions. Intriguingly, CO_2 emissions from both conventional and ethanol gasoline exhibited lower emission factors under "~~TWC failed~~TWC deteriorated" conditions compared to "TWC worked" conditions. On the one hand, this may be related to the unexpectedly lower fuel consumption for "~~TWC failed~~TWC deteriorated" conditions (4.8 L/100km for conventional gasoline and 4.9 L/100km for ethanol gasoline) compared to "TWC worked" conditions (5.8 L/100km for conventional gasoline and 5.8 L/100km for ethanol gasoline). On the other hand, from the prospective of the after-treatment 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 NOx, catalyzing oxidation and reduction reactions. This process converts CO into CO_2, HC into H_2O and CO_2, and NOx into N_2 and O_2 (De Abreu Goes et al., 2021). Consequently, under "~~TWC failed~~TWC deteriorated" conditions, these conversions are hindered, leading to reduced CO_2 generation and correspondingly lower emission factors.

Regarding N_2O, its generation pathway is related to the primary precursors, notably CO, NOx, and THC, with lower temperatures facilitating N_2O formation (Brinklow et al., 2023). The formation of N_2O over TWC converters is 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 failed~~TWC deteriorated" conditions compared to "TWC worked" conditions, N_2O emission factors did not correspondingly elevate. This phenomenon could be attributed to the significant reduction in NOx emissions under "~~TWC failed~~TWC deteriorated" conditions, which were only one-seventh of those recorded under "TWC worked" conditions. This suggests that NOx may be a ~~more~~ pivotal precursor in determining N_2O production other than CO and THC. Similar to prior research highlighting that catalyst aging would change the structure of oxygen storage material thereby causing an enhancement of N_2O formation (Nevalainen et al., 2018), our findings reveal a notable increase in N_2O emissions for CNG under "~~TWC failed~~TWC deteriorated" conditions, reaching approximately three times that of "TWC worked" conditions. This elevation is likely attributable to heightened precursor concentrations, particularly NOx. Our study underscores the variability in the impact of TWC deterioration ~~failure~~ on N_2O 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_2O generation and elucidating the underlying mechanism governing N_2O 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_2O emissions are minimally affected by the substitution of clean fuels such as CNG but are greatly influenced by TWC deterioration~~failure~~.

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 L/100km and 4.3 L/100km in cold and hot startup modes, respectively, compared to gasoline vehicles, which consumed 5.2 L/100km and 5.0 L/100km 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_2 emission factors were 110.8 g/km and 102.0 g/km for cold and hot startup modes, respectively, while CH_4 emission factors stood at 1.2 mg/km and 0.6 mg/km, and N_2O emission factors at 0.4 mg/km and 0.5 mg/km 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 displacement, ~~they~~ it demonstrated remarkable reductions in GHG emissions. In particular, CO_2 emissions were reduced by 10% to 14%, CH_4 emissions by 11% to 37%, and N_2O emissions by 23% to 43%, highlighting the environmental benefits of HEV technology.

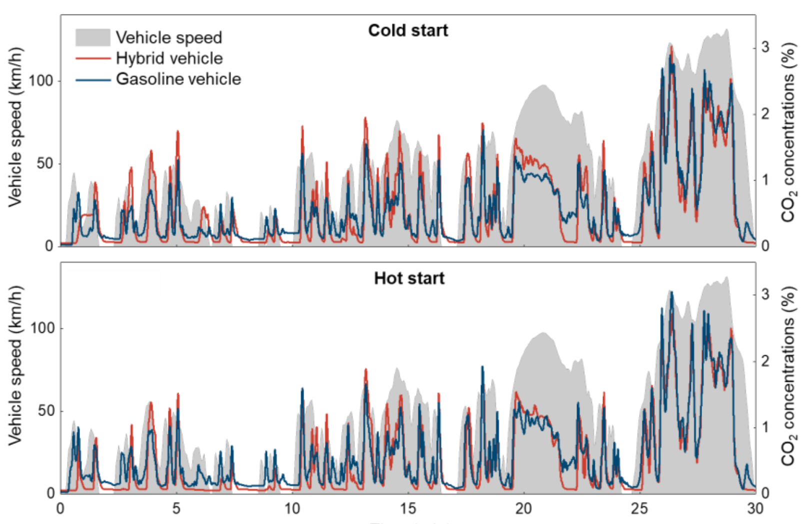

Figure 4~~3~~: Timeseries of CO_2 and vehicle speed in the WLTC protocol with ~~hot~~ cold start (upper panel) and ~~cold~~ hot start (bottom panel) of the HEV and gasoline vehicle.

HEVs, when compared to conventional gasoline vehicles, utilize electric motors for propulsion during startup and low-speed driving conditions (approximately 20 km/h). As depicted in Figure 4~~3~~, HEVs exhibited a delayed and significantly reduced CO_2 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_2 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 Figure 5~~4~~, an examination of the various speed phases within the WLTC revealed that HEVs demonstrated notably lower CO_2 emissions during low-speed segments, especially under hot start conditions. This reduction in CO_2 emissions can be attributed to the efficient operation of the electric motor at lower speeds. Conversely, the emission factors for CH_4 and N_2O were observed to be relatively higher in these low-speed segments, indicating a trade-off 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_4 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 against conventional fuel-powered vehicles. Specifically, during cold starts, hybrid vehicles exhibit a CO emission factor of 70.9 mg/km, whereas their conventional counterparts register 95.7 mg/km, 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, compared to the 71.9 mg/km emitted by conventional vehicles, demonstrating an 40.5% decrease. This indicates that hybrid vehicles have more advantages in terms of combustion efficiency comparison.

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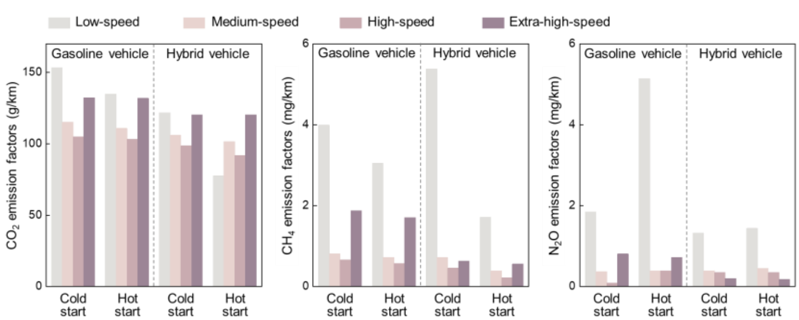
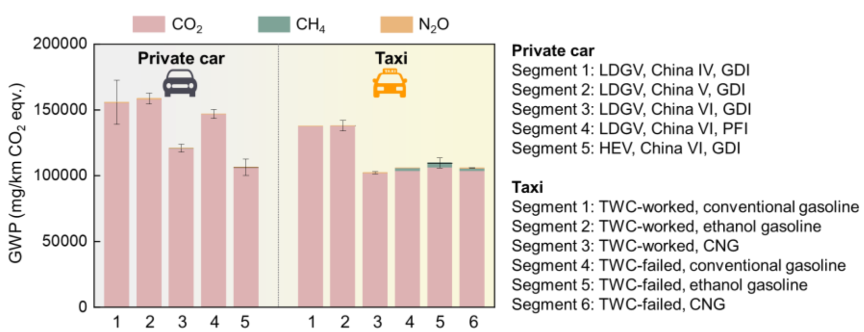


Figure 5~~4~~: Emission factors of CO_2, CH_4, and N_2O under different speed phases under the WLTC protocol for the China VI HEV and China VI gasoline vehicle.

3.4 Global warming potentials of light-duty vehicles

As shown in Figure 65, vehicle exhaust emissions, particularly the emission of CO_2, remain the primary contributor to the global warming potential, accounting for over 99% of the total three GHGs. Even though CH_4 and N_2O have global warming potentials 28 25 and 265 298 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 typestechnology. GDI technology allows for more precise fuel delivery and better combustion efficiency, which translates to lower CO_2 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 indicates 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.



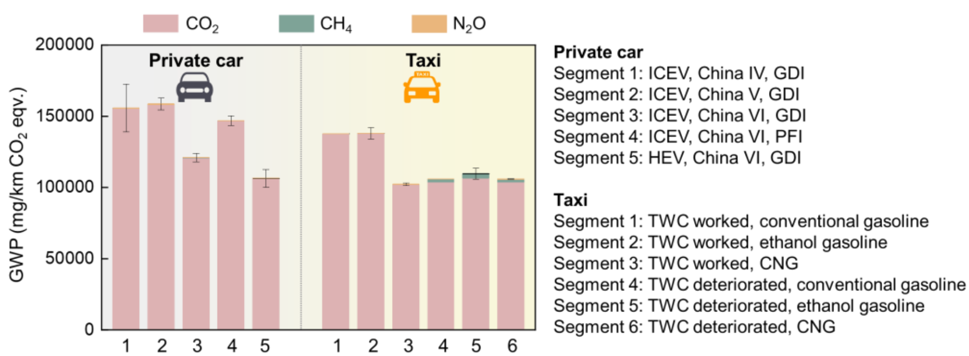


Figure ~~6~~5: Global warming potentials of private car and taxi exhaust ~~emission~~emissions.

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For ~~dual fuel~~bi-fuel taxis, under ~~normal operating conditions of TWC~~"TWC worked" conditions, the fuel consumption is higher when using ethanol ~~blended~~ gasoline and ~~regular~~ 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 ~~although dual fuel~~bi-fuel vehicles can benefit from alternative fuels, especially the use of CNG ~~fuel~~, it can effectively reduce the emissions of ~~motor vehicle~~vehicular ~~exhaust pollutants~~GHGs. ~~On the contrary~~Unexpectedly, under "TWC deteriorated" conditions,~~in the case of TWC failure,~~ lower GWP values were observed than those under "TWC worked" conditions, with a decrease of ~~. Compared to vehicles operating normally with TWC using gasoline, the GWP of vehicles with TWC failure decreased by~~ approximately 27.8%. This contradictory finding indicates that although TWC is effective in reducing pollutants such as CO, HC, and NOx, its level of control over greenhouse gas emissions ~~is not the same~~may still be worth further discussion. ~~Therefore~~Overall, the deterioration ~~failure~~ 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_2, CH_4, and N_2O) emanating from a diverse fleet of eleven light-duty vehicles, encompassing conventional gasoline ~~private~~ cars, hybrid electric ~~private~~ vehicles, and bi-fuel taxis. The main findings of our research are outlined below.

(1) 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 are related to their low fuel consumption, while the low N_2O emissions are associated with their after-treatment technologies.

(2) ~~c~~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, air-fuel ratio, and combustion temperature between the two engine technologies.

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(3) In contrast to hot startup mode, cold starts promote CO_2 and CH_4 emissions, while N_2O emissions exhibit an opposite trend, which is mainly related to the higher NOx concentrations during hot starts.

(4) 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 promotes CH_4 emissions since CH_4 is its primary component.

(5) Unexpectedly, the CO_2 emissions from the two liquid fuels under the "~~TWC failed~~TWC deteriorated" conditions exhibit lower emission factors than under the "TWC worked" conditions. In the case of N_2O emissions, the TWC deterioration ~~failure~~ does not lead to higher N_2O emissions when using conventional gasoline or ethanol gasoline, mainly because the significant reduction in NOx under the "~~TWC failed~~TWC deteriorated" conditions despite the significant increase in CO and THC. However, under CNG usage, the emission factor of N_2O under "TWC deteriorated"~~TWC failure~~ conditions is approximately three times that under "TWC worked"~~normal TWC~~ conditions, which is primarily associated with a substantial increase in NOx.

(6) 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 at low-speed phases, while emissions of CH_4 and N_2O tend to be relatively higher at these speeds.

(7) The global warming potential ~~stemming~~ from vehicle exhaust is primarily attributed to CO_2 emissions. HEVs demonstrate a more advantageous environmental impact compared to conventional gasoline vehicles. Surprisingly, however, under the "~~TWC failed~~TWC deteriorated" conditions, a lower GWP value has been observed, indicating a complex interaction between emission control technologies and the environmental impact.

~~The purpose of this study is~~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 ~~the~~ managing~~ement~~ and controlling ~~of~~ the GHG emissions from vehicles in the future.

Code and data availability. The data used in this publication are available on https://doi.org/10.5281/zenodo.15253351 https://doi.org/10.6084/m9.figshare.27303969 (Yang et al., 2024)(Yang et al., 2025), and they can be accessed by request to the corresponding authors.

Author contributions. XPY and SDL LSD performed the formal analysis, funding acquisition, and writing – original draft. JK and ZHH performed the formal analysis. YW, DLY, ZJ, and ZGY discussed the results and commented on the article. YJW, SDL, HY, and YD conceived and led the studies.

Competing interests. The authors declare that they have no conflict of interest.

Financial support. This study has been supported by the National Natural Science Foundation of China (Nos. 42205111, 42207114).

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