Response to Reviewer #2:

General comments:

This manuscript presents a comprehensive investigation into the characteristics of greenhouse gas (GHG) emissions from motor vehicles under various influencing factors, including emission standards, TWC operational status, cold/hot start modes, and power-train types. The study elucidates the evolving trends of GHG emissions during technological upgrades, providing critical data support for constructing GHG emission inventories while offering scientific basis for emission control strategies.

Focusing on the current research landscape where vehicular pollutant emissions have been extensively studied but GHG emissions remain under-investigated, this work makes significant contributions to complementing existing knowledge on vehicular emission characteristics. Particularly within the context of future carbon peak and neutrality goals, the findings provide crucial insights from the mobile source perspective to support continuous improvement of air quality.

However, the following comments should be addressed before this manuscript can be accepted. The English writing of this work should also be improved. As a result, I recommend a minor revision of this manuscript.

Response: Thank you for recognizing the manuscript and all of the constructive suggestions. We have taken all these valuable suggestions into account and have made corrections in this revised manuscript. Below is our response to the specific comments, highlighted in blue, with changes to the manuscript highlighted in red.

Specific comments:

1. Introduction: In the third paragraph, please reorganize the research processes and results from the published papers. The current presentation merely lists the findings of individual studies without integrating them. Additionally, the authors have not explicitly identified the limitations in the existing research.

Response: Thank you for the valuable comment. We have reorganized the research processes and limitations of the existing research in Introduction.

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_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 both pollutant and CO_2 emissions (Selleri et al., 2022). Furthermore, from the model simulation perspective, 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 predict the future greenhouse gas emissions (Zeng et al., 2016).

Overall, from the perspective of GHG emissions from domestic vehicles in China, Yin et al. 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 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".

Additionally, we added a summary list of GHG emission factors in different studies in Table S1 in the Supplementary Information.

Table S1 Comparison of CO_2, CH_4, and N_2O emission factors in different studies.

GHG	Tested vehicles	Fuel types	Methods	Emission factors	References
CO_2	China 6	Gasoline	PEMS[1]	313.7 g/km (urban area, expressway) 304.4 g/km (suburban area, expressway)	(Zhu et al., 2022)
	China 6	Gasoline	PEMS	275.7 g/km (urban area, expressway) 273.9 g/km (suburban area, expressway)	(Zhu et al., 2022)
	China 6	Gasoline	Chassis dynamometer	≈ 300 g/km (-10 °C, cold start, TWC[2]) ≈ 289 g/km (0 °C, cold start, TWC) ≈ 230 g/km (23 °C, cold start, TWC) ≈ 259 g/km (40 °C, cold start, TWC) ≈ 257 g/km (-10 °C, cold start, TWC + GPF[3]) ≈ 259 g/km (0 °C, cold start, TWC + GPF) ≈ 210 g/km (23 °C, cold start, TWC + GPF) ≈ 238 g/km (40 °C, cold start, TWC + GPF)	(Wang et al., 2022b)
	UK fleet using 2015 new car sales car	Gasoline	PEMS	175.2 g/km (urban conditions, extra small displacement) 199.2 g/km (urban conditions, small displacement) 231.5 g/km (urban conditions, medium displacement) 340.9 g/km (urban conditions, large displacement) 140.6 g/km (motorway conditions, extra small displacement) 154.3 g/km (motorway conditions, small displacement) 174.4 g/km (motorway conditions, medium displacement) 213.0 g/km (motorway conditions, large displacement)	(O'Driscoll et al., 2018)
	China 0 ~ China 2	Diesel	IVE[4]	409.9 g/km	(Yao et al., 2011)
	China 2	Diesel	COPERT[5]	245.8 g/km	(Cai and Xie, 2010)
	China 3	Diesel	COPERT	238.2 g/km	(Cai and Xie, 2010)
	China 3	Diesel	PEMS	304 g/km	(Wu et al., 2017)
	China 4	Diesel	PEMS	310 g/km	(Wu et al., 2017)
	China 4	Diesel	COPERT	238.2 g/km	(Cai and Xie, 2010)

				214.1 g/km (NEDC[6])	
	China 4	Diesel	Chassis dynamometer	209.7 g/km (WLTC[7])	(Wang et al., 2022a)
	China 4	Diesel	PEMS	415.06 g/km	
	China 5	Diesel	PEMS	447.48 g/km	(Li et al., 2024)
	China 6	Diesel	PEMS	335.26 g/km	
	Euro 6	Diesel	Chassis dynamometer	239.0 g/km	(Vojtíšek-Lom et al., 2018)
				133 g/km	
				134 g/km	
	UK fleet using 2015 new car sales car	Diesel	PEMS	141.9 g/km (urban conditions, small displacement) 163.4 g/km (urban conditions, medium displacement) 205.1 g/km (urban conditions, large displacement) 137.1 g/km (motorway conditions, small displacement) 149.0 g/km (motorway conditions, medium displacement) 170.0 g/km (motorway conditions, large displacement)	(O'Driscoll et al., 2018)
	Heavy-duty vehicles in Korea	Diesel	Chassis dynamometer	320 g/km (case 1) 411 g/km (case 2) 634 g/km (case 3) 727 g/km (case 4) 877 g/km (case 5) 537 g/km (case 6)	(Seo et al., 2018)
CH_4	Euro 5	Gasoline	Chassis dynamometer	4.8 mg/km	(Clairotte et al., 2020)
	Euro 6b/c	Gasoline		3.2 mg/km	
	Euro 6d-TEMP	Gasoline		1.7 mg/km	
	Euro 5	Gasoline hybrid		0.8 mg/km	
	Euro 6b	Gasoline hybrid		4.5 mg/km	
	Euro 6b	Dual-LPG/Ga		2.3 mg/km	

			soline	
	Euro 5	Diesel		3.6 mg/km	
	Euro 6b/c	Diesel		10.3 mg/km	
	Euro 6d-TEMP	Diesel		3.6 mg/km	
	China 0	Diesel	COPERT	18 mg/km	(Wang et al., 2022a)
	China 1	Diesel	COPERT	11 mg/km	
	China 2	Diesel	COPERT	5 mg/km	
	China 0 ~ China 2	Diesel	IVE	90 mg/km	(Yao et al., 2011)
	China 2	Diesel	COPERT	7 mg/km	(Cai and Xie, 2010)
	China 3	Diesel	COPERT	2 mg/km	(Wang et al., 2022a)
	China 3	Diesel	COPERT	0 mg/km	(Cai and Xie, 2010)
	China 4	Diesel	COPERT	1 mg/km	(Wang et al., 2022a)
	China 4	Diesel	COPERT	0 mg/km	(Cai and Xie, 2010)
	China 4	Diesel	PEMS	5 mg/km	(Li et al., 2024)
	China 5	Diesel	COPERT	1 mg/km	(Wang et al., 2022a)
	China 5	Diesel	PEMS	3 mg/km	(Li et al., 2024)
	China 6	Diesel	PEMS	3 mg/km	(Li et al., 2024)
	Euro 6	Diesel	Chassis dynamometer	3 mg/km	(Vojtíšek-Lom et al., 2018)
				5 mg/km	
				7 mg/km	
N_2O	Euro 5	Gasoline	Chassis dynamometer	3.1 mg/km	(Clairotte et al., 2020)
	Euro 6b/c	Gasoline		0.9 mg/km	
	Euro 6d-TEMP	Gasoline		0.3 mg/km	
	Euro 5	Gasoline hybrid		0.3 mg/km	
	Euro 6b	Gasoline hybrid		3.2 mg/km	
	Euro 6b	Dual-LPG/Ga		0.9 mg/km	

	soline			
Euro 5	Diesel		6.2 mg/km	
Euro 6b/c	Diesel		13.5 mg/km	
Euro 6d-TEMP	Diesel		15.2 mg/km	
China 0 ~ China 2	Diesel	IVE	197 mg/km	(Yao et al., 2011)
China 0	Diesel	COPERT	0 mg/km	(Wang et al., 2022a)
China 1	Diesel	COPERT	2 mg/km	
China 2	Diesel	COPERT	5 mg/km	
China 2	Diesel	COPERT	167 mg/km	(Cai and Xie, 2010)
China 3	Diesel	COPERT	8 mg/km	(Wang et al., 2022a)
China 3	Diesel	COPERT	200 mg/km	(Cai and Xie, 2010)
China 4	Diesel	COPERT	8 mg/km	(Wang et al., 2022a)
China 4	Diesel	COPERT	200 mg/km	(Cai and Xie, 2010)
China 4	Diesel	PEMS	2 mg/km	(Li et al., 2024)
China 4	Diesel	COPERT	8 mg/km	(Wang et al., 2022a)
China 5	Diesel	PEMS	4 mg/km	(Li et al., 2024)
China 6	Diesel	PEMS	3 mg/km	(Li et al., 2024)
Euro 6	Diesel	Chassis dynamometer	13 mg/km	(Vojtíšek-Lom et al., 2018)
			5 mg/km	
			7 mg/km	

Notes:

[1] Portable Emission Measurement System

[2] Three-Way Catalyst

[3] Gasoline Particulate Filter

[4] International Vehicle Emissions model

[5] Computer Programme to Calculate Emissions from Road Transport

[6] New European Driving Cycle

[7] Worldwide Harmonized Light Vehicles Test Cycle

2. Introduction: In the background section, succinctly summarize why it is important to test hybrid vehicle exhaust greenhouse gas emissions. Describe the unique characteristics of hybrid vehicle exhaust emissions and discuss the effects of post-treatment technologies on exhaust emissions in vehicles.

   Response: Thank you for the helpful suggestions, and we added the detailed information on HEVs and post-treatment in the *Introduction*:

   Additionally, the population of light-duty hybrid electric vehicles in China is currently experiencing rapid growth. However, 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 post-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).

3. Line 100-105: How was the determination made that the TWC failed? Does a mileage of approximately 27 km necessarily indicate failure? Are the authors referring to TWC degradation here?

   Response: We sincerely appreciate your constructive feedback. Regarding this issue, our consideration was indeed insufficiently comprehensive. As you pointed out, high mileage does not necessarily indicate TWC failure, but rather reflects performance degradation, despite of their substantially elevated CO and THC emissions (see Figure S1 in the Supplementary Information). As suggested, we have revised "TWC failed" to "TWC deteriorated".

   Since the "TWC failed" appears frequently in the manuscript, we have uniformly revised it to "TWC degradation" throughout the manuscript and will not list each instance here. Furthermore, we added some information on Figure S1 in Section 2.1:

   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 degraded 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.
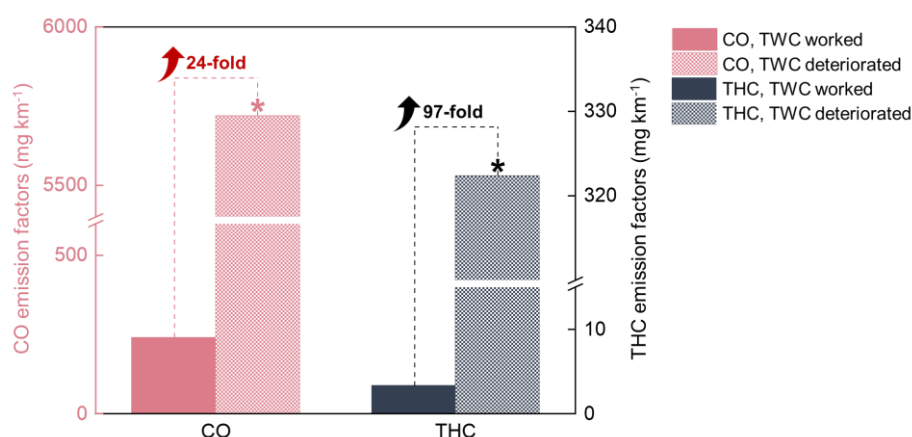
Figure S1. CO and THC emission factors from the fueled taxi fueled by conventional gasoline under the "TWC worked" and "TWC deteriorated" conditions under the WLTC protocol. The symbol * represents that the pollutant concentrations exceeded the upper limit of the measurement instrument.

4. Introduction: Based on the latest 2023 data, provide an overview of the passenger vehicle ownership growth trends in China.

   Response: Thanks for your valuable suggestions. Based on the latest reported data, we have performed corresponding updates:

   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).

5. Section 2: To provide a more intuitive representation of the experimental process, it is recommended that the authors include a schematic diagram of the experiment.

   Response: Thanks for your helpful suggestions. We have added the schematic diagram in the revised manuscript in Section 2.2.
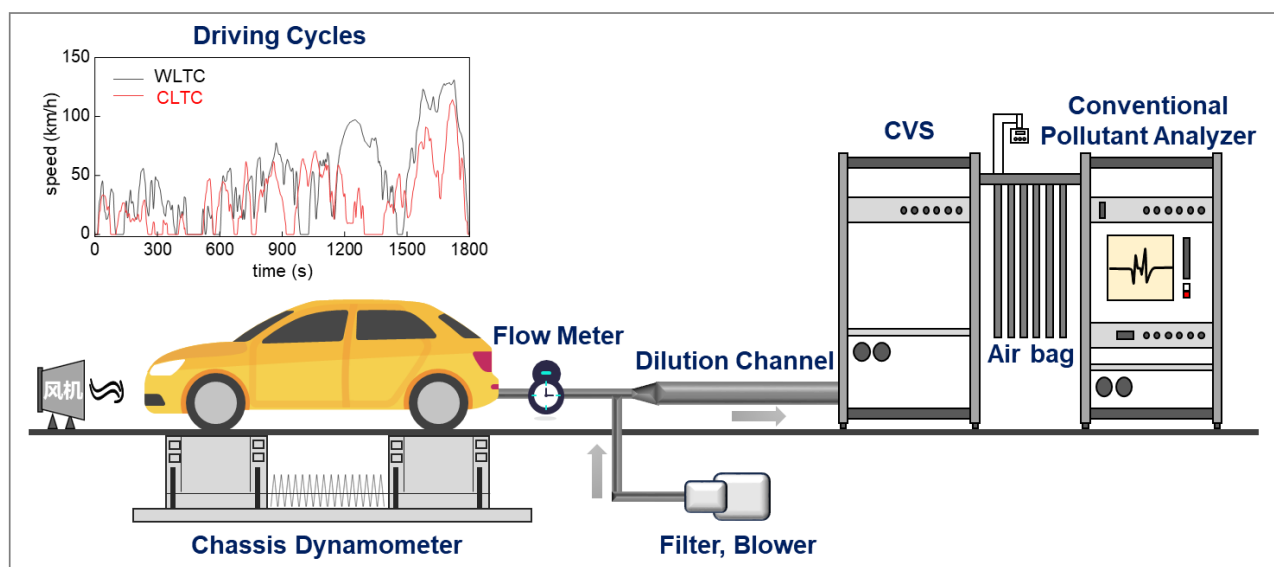
Figure 1: Schematic diagram for vehicular emission measurements based on chassis dynamometer in this study.

6. Section 3.2: What is the ethanol content in Chinese gasoline? How does the ethanol content affect greenhouse gas emissions from vehicle exhaust?

    Response: Thanks for your helpful suggestions. The ethanol content in Chinese gasoline is usually 10%, which is E10 ethanol gasoline. We have added more detailed information on ethanol gasoline in Section 3.2:

    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).

7. Line 315-320: The authors only mention that hybrid vehicles exhibit higher combustion efficiency but provide no experimental data to support this claim. If relevant data is available, please conduct a quantitative comparison between the combustion efficiency of hybrid vehicles and conventional fuel-powered vehicles.

    Response: Thanks for your valuable comments. We further the description in Section 3.3:

    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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