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Determination of Euro 6 LPG passenger car emission factors through laboratory and on-road tests: Effect on nation-wide emissions assessment for Italy

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ABSTRACT

Five Euro 6 LPG (liquefied petroleum gas) bi-fuel passenger cars well representative of the Italian circulating fleet were tested in laboratory (in repeatable conditions on a chassis dynamometer) and on road (in real traffic conditions using a portable emissions measurement system). The regulated and unregulated exhaust emissions and the energy consumptions measured with gasoline and LPG fuelling were compared. All the regulated emissions were compliant with the Euro 6 limits over the type-approval driving cycles and over the RDE (Real Driving Emissions) tests except for the particle number emission measured from the only direct injection engine tested car with gasoline fuelling, which exceeded the emission limit by 10.9% in laboratory and by 17.2% on road. Switching from gasoline to LPG fuelling, systematic carbon dioxide emission reductions were detected. Based on sub-cycles data, distance-specific exhaust emission factors and energy consumption factors were calculated for LPG fuelling and subsequently compared with those proposed by the EMEP/EEA (European Monitoring and Evaluation Programme/European Environment Agency) guidebook, which are integrated in the COPERT (COmputer Programme to calculate Emissions from Road Transport) model. The comparison showed a reasonable agreement for carbon monoxide and energy consumption for the medium segment, whereas more marked differences were found for the other compared emissions, with almost systematically lower values for the experimental factors. The comparison was then extended to the results of the national emissions calculation from Euro 6 LPG passenger cars in Italy, obtained using the EMEP/EEA factors and the experimental factors. These latter factors led to country-level lower emission estimates for all species, with the largest difference (around 90% less) for PM emission and smaller but still relevant (5%–23% range) differences for NOx, carbon monoxide, and total hydrocarbons. A considerable reduction (15.6%) was also found for the small segment with the new energy consumption factors, whereas a less marked reduction (1.8%) was found for the medium segment. The findings highlight the importance of up-to-date factors in order to obtain much more realistic and accurate estimations of exhaust emission and energy consumption within national inventories.

1. Introduction

Air pollution is a global problem: a wide range of adverse effects of air pollutants on health and environment are well documented (Manisalidis et al., 2020; WHO Regional Office for Europe, 2013) and effective actions to reduce air pollution and its impacts require a good understanding of its sources (EEA, 2020). Atmospheric emission inventories are essential tools to point out the contributions of the different emission sources and, consequently, to set up air quality improvement plans and efficient air pollution abatement strategies (Moussiopoulos et al., 2009).

According to the European Union emission inventory report (EEA, 2021), road transport is one of the main sources that contributes to emissions of key primary air pollutants in the European Union. It accounted for 39% of nitrogen oxides (NO_x), 26% of black carbon (BC),

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Abbreviations

COPERT	COmputer Programme to calculate Emissions from Road Transport
EC	Energy Consumption
EF	Emission Factor
EMEP/EEA	European Monitoring and Evaluation Programme/ European Environment Agency
ERMES	European Research for Mobile Emission Sources
EUDC	Extra Urban Driving Cycle
ISPRA	Institute for Environmental Protection and Research
LPG	Liquefied petroleum gas
NEDC	New European Driving Cycle
NTE	Not-To-Exceed
OEM	Original Equipment Manufacturer
PN-EEPS	Particle Number - Engine Exhaust Particle Sizer
PN-ELPI	Particle Number - Electrical Low Pressure Impactor
PN-PMP	Particle Number - Particle Measurement Programme
PEMS	Portable Emissions Measurement System
RDE	Real Driving Emission
UDC	Urban Driving Cycle
WLTC	Worldwide harmonized Light vehicles Test Cycle

20% of carbon monoxide (CO), 11% of fine particle matter (PM2.5) and 8% of non-methane volatile organic compounds (NMVOC) emitted in the 2018 in EU-28 (EEA, 2020). Due to these important contributions and considering that emission estimates should address emission reduction measures and air quality policies (e.g. establishment of restricted traffic areas, definition of emission standards), the calculation of road transport emissions needs to be reliable, detailed and accurate (Kousoulidou et al., 2013; Ntziachristos et al., 2009). However, the estimate of road transport emissions is complex due to its variable nature and due to the great diversity of road vehicle types; therefore, the use of emission models is necessary (Fontaras et al., 2014). COPERT is the European Union standard vehicle emissions model developed for official road transport emission inventory compilation in EEA member countries and represents the software implementation based on the Tier 3 methodology of the EMEP/EEA air pollutant emission inventory guidebook (Ntziachristos and Samaras, 2019). COPERT estimates vehicle fleet emissions on a country-level by combining activity data input by the user (e.g. number of vehicles divided into different emission categories/technologies, trip characteristics, circulation activity) with vehicle-specific emission factors (EFs), based on the mean speed of vehicles, included in the model (Kousoulidou et al., 2011). EFs are functional relations that depend on many parameters (e.g. vehicle characteristics, emission treatment technology, operating conditions) and that predict the quantity of pollutant emitted per distance driven, energy consumed, or amount of fuel used (Franco et al., 2013). Several studies claim that for a realistic and accurate development of EFs it is necessary to take into account both laboratory and on-road measurements (Fontaras et al., 2014; Franco et al., 2013; Valverde et al., 2019).

Although the EFs are already provided by COPERT model, in order to best represent the national emissions, it is envisaged that country-specific data (if available) will be privileged with respect to the default data adopted at international level. In particular, it is good practice to develop country-specific emission factors for the most relevant emission sectors, with the goal to improve the accuracy of emission assessment (IPCC, 2019).

In Italy the Institute for Environmental Protection and Research (ISPRA) has the overall responsibility for the national emission inventory, including all the work related to its compilation and submission to CLRTAP (Convention on Long Range Transboundary Air Pollution) and UNFCCC (United Nations Framework Convention on Climate Change) (ISPRA, 2021a). ISPRA has at its disposal an extensive and accurate database, continuously updated on the basis of validated results of studies and researches, for road transport emissions estimation, which is a key sector at national level. Therefore, ISPRA introduces Italian country-specific EFs in the COPERT model, using the EFs provided by the software only when country-specific data are not available. This latter was the case concerning the EFs of liquefied petroleum gas (LPG) bi-fuel passenger cars. Furthermore, updated EFs were not available for Euro 6 LPG passenger cars in the software, where for this vehicle category the EFs attributed to vehicles homologated with previous emission standards were used. As confirmed by the EMEP/EEA guidebook (Ntziachristos and Samaras, 2019), indeed, due to the limited amount of data for LPG vehicles, a large number of assumptions and extrapolations have been made on the basis of existing information to provide a consistent set of emission factors. In this context it should be emphasized that Italy has the second-largest circulating fleet of LPG passenger cars in the European Union after Poland and the sixth-largest in the world, with about 2.6 millions of LPG vehicles (WLPGA Liquid Gas Europe, 2020), accounting for 6.7% of the total Italian passenger cars circulating fleet in 2020, 29.5% of these with Euro 6 homologation (ACI, 2020). This explains the great importance for Italy to rely on EFs updated to the latest Euro standard for LPG vehicles, in order to improve the estimation accuracy of road transport emissions. Additionally, there are two main types of LPG-fuelled vehicles: those produced by the original equipment manufacturers (OEMs) to operate as bi-fuel vehicles, and conventional gasoline vehicles later retrofitted to operate with LPG. The latter are optimized to operate on gasoline and, after the LPG system installation, it has to be guaranteed that the new fuel continues to retain optimal engine-out conditions for the catalyst to operate efficiently (Ntziachristos and Samaras, 2019). For this reason and considering the wide diffusion of retrofitted vehicles (since 2010 about 1.15 million cars have been retrofitted with an LPG system in Italy (Assogasliquidi, 2021)), it is very important to properly account for the emissions from this type of vehicles when estimating national road transport emissions. In general, nitrogen oxides (NOx), volatile organic compounds (VOCs) and particulate matter (PM) emissions for LPG vehicles are reported to be lower than for diesel and petrol ones; carbon dioxide (CO_2), emissions seem to be 9–20% lower in LPG vehicles than in petrol ones (benefiting from the higher H/C ratio of LPG compared to petrol fuel) while diesel fuelled vehicles seem to be at least as good and up to 15% lower CO_2 emitters than LPG ones, because of their superior fuel efficiency (Papadopoulos et al., 2018). Regarding the impact of LPG on the anthropogenic global warming, a potential significant reduction can be achieved through its substitution with bio-LPG, originating during the production of HVO (Hydrotreated Vegetable Oil) fuels following hydrogenation of the glycerol molecule in the vegetable oil. A preliminary assessment of this potential is reported in (European Commission, 2020). A further fuel decarbonisation potential consists in the blending of bio-LPG with renewable DME (Dimethyl Ether), which is reported as a potential suitable blending component (Flekiewicz et al., 2017).

The aim of this study is the development of country-specific EFs for Euro 6 passenger cars based on data from both laboratory and on-road emission tests performed on a pool of five Euro 6 LPG bi-fuel passenger cars representative of the different technologies of the circulating Italian fleet. The testing protocol was developed for the execution of several driving cycles performed with both gasoline and LPG fuelling. The emission data were compared, analysed and, starting from sub-cycle emissions detected using LPG fuelling, distance-specific (mass per unit km) hot emission and energy consumption (EC) factors (measured when the engine and the exhaust after-treatment system run at their nominal operating temperature) were determined and subsequently compared with the corresponding EFs provided by COPERT model.

Finally, the impact of these new, country-specific factors on the Italian national emissions calculation for Euro 6 LPG cars was evaluated through COPERT model simulations carried out with the default hot emission and EC factors and the experimental factors provided by this work.

2. Material and methods

The testing campaign was performed, depending on the availability of vehicles, from July 2017 to March 2018 by the Automotive Emission Laboratory of the Emissions Laboratory of Innovhub-SSI, located in San Donato Milanese (Italy).

2.1. Vehicles and fuels

Five Euro 6 LPG bi-fuel passenger cars with small (i.e. less than 1400 cm^3) and medium (i.e. between 1400 cm^3 and 2000 cm^3) engine displacement were selected for the testing campaign to represent almost totally (about the 99.95%) the Italian circulating fleet of Euro 6 LPG bi-fuel passenger cars (ACI, 2020). In order to take into account the different engine technologies, each tested car was from a different car manufacturer, three cars were equipped with retrofit LPG powertrain and two with original equipment manufacturer (OEM) LPG powertrain. The two types of LPG systems can be considered equivalent in terms of equipment. The main characteristics of the tested cars are summarized in Table 1.

All the cars had 5-gear manual transmission, 4-cylinder engine, front-wheel drive and were equipped with three-way catalyst (TWC). In order to obtain a better repeatability of emission tests, the start and stop system, whether present in the vehicle, was disabled before starting each test. The cars were tested with both petrol and LPG fuelling according to the protocol described in the following chapter. Commercial gasoline and LPG, compliant respectively with the European technical standard (CEN, 2017) and (CEN, 2012), were used to fuel the tested cars. Car engines always started-up in gasoline mode even if LPG mode was selected and, after 1–2 min, automatically switched to LPG.

2.2. Test protocol and equipment

2.2.1. Laboratory tests

Laboratory tests were carried out driving the cars on a chassis dynamometer in a climatically controlled test cell (temperature: 23 ± 3 °C; relative humidity: 50 ± 5%) in order to assure repeatable and reproducible testing conditions. The driving cycles performed in laboratory were the NEDC, the ERMES and the WLTC one:

Table 1
Characteristics of the tested cars.

	Car 1	Car 2	Car 3	Car 4	Car 5
Homologation Emission Standard	Euro 6b	Euro 6b	Euro 6b	Euro 6b	Euro 6c
Type approval driving cycle	NEDC	NEDC	NEDC	NEDC	WLTC
Mileage at test start [km]	6780	1966	6650	2000	1984
Engine displacement [cm^3]	1248	1598	1197	1590	1598
COPERT segment	Small	Medium	Small	Medium	Medium
Test mass [kg]	1130	1355	1165	1470	1370
Max power [kW]	63 @ 6000 rpm	115 @ 6000 rpm	66 @ 4800 rpm	86 @ 6000 rpm	75 @ 5500 rpm
Max torque [Nm]	120 @ 4000 rpm	155 @ 3900 rpm	160 @ 1400 rpm	154 @ 4000 rpm	156 @ 4000 rpm
Injection type	Port Fuel Injection – PFI	Port Fuel Injection – PFI	Direct injection – DI	Port Fuel Injection – PFI	Port Fuel Injection – PFI
LPG powertrain	Retrofit	Retrofit	Retrofit	OEM	OEM

- NEDC (New European Driving Cycle), whose first version was introduced since the Euro 1 standard, as performed in this research (UN/ECE, 2012) was the European Type I type-approval driving cycle for light-duty vehicles from Euro 3 to Euro 6b homologation emission standard. NEDC covers a distance of 11 km, has a duration of 1180 s, and is divided into two phases: UDC (average speed of 18.7 km/h) followed by EUDC (average speed of 62.6 km/h) which represent, respectively, an urban driving condition and an extra urban driving condition. NEDC was carried out with cold engine start and preceded by a soaking phase, in which the car remained on the test bench, at the ambient conditions of the cell, for at least 6 h.
- ERMES cycle is a research cycle developed by the ERMES group (European Research Group on Mobile Emission Sources) suitable to drive vehicles in laboratory emission tests in similar or even more demanding conditions compared to on-road tests. ERMES cycle covers a distance of 24.2 km, has a duration of 1320 s, imposes a fixed gear strategy, and includes five full load accelerations that allow to cover a wide vehicle engine emission map (Matzer and Rexeis, 2016). This cycle was carried out with hot engine start immediately after the NEDC cycle.
- WLTC (Worldwide harmonized Light vehicles Test Cycle) (European Commission, 2017a) is the cycle that in 2017 replaced the NEDC as European Type I type-approval driving cycle for light-duty vehicles according to Euro 6c homologation emission standard. WLTC is a more realistic driving cycle than NEDC, which was widely criticized for not being representative of real-life driving conditions because of its stylized driving path (Mock et al., 2013). WLTC covers a distance of 23.3 km, has a duration of 1800 s, and is divided into four phases characterized by increasing average speeds, intended to represent different driving conditions: Low (average speed of 18.9 km/h, representative of urban driving condition), Medium and High (average speeds of 39.5 km/h and 56.7 km/h, both representative of extra urban driving condition) and Extra-high (average speed of 92.0 km/h, representative of motorway driving condition). As for NEDC, WLTC was carried out with cold engine start and preceded by a soaking phase, in which the car remained on the test bench at the ambient conditions of the cell for at least 6 h.

For each passenger car and for each fuelling type, the sequence NEDC cycle followed by ERMES cycle was performed until three tests with good repeatability were achieved (coefficient of variation of CO_2 emission for both cycles close to 2%), while WLTC cycle was performed only once.

Measurements performed in laboratory tests regarded both regulated and unregulated emissions: the former included nitrogen oxides (NOx), total hydrocarbons (THC), non-methane hydrocarbons (NMHC), carbon monoxide (CO), particulate matter mass (PM) and solid particle number (PN-PMP compliant (UN/ECE, 2015)); the latter included carbon dioxide (CO_2), methane (CH_4), ammonia (NH_3), nitrogen dioxide (NO_2), nitrous oxide (N_2O), formaldehyde, soot and particle number (PN) in different size ranges. NOx was measured through a chemiluminescence detector, THC, NMHC and CH_4 through a double channel flame ionization detector, CO and CO_2 through a non-dispersive infrared detector, PM through filters and a 10^{-7} g precision scales, PN through a PMP compliant system with 23 nm particle aerodynamic diameter cut-off. As to unregulated emissions, NH_3, NO_2, formaldehyde and N_2O were measured through a Fourier transform infrared spectroscope, soot through a micro soot sensor, PN through an electrical low pressure impactor (ELPI) for particles in the 0.007 ÷ 10 μm aerodynamic diameter range and through an engine exhaust particle sizer (EEPS) for particles in the 5.6 ÷ 560 nm aerodynamic diameter range. The experimental devices layout is fully described elsewhere (Puricelli et al., 2021). All the gas analysers have been purged and calibrated, through zero and span checks, with certified gas cylinders. Before testing each car, the chassis dynamometer was calibrated by setting the breaking equivalent inertia of the car, whose coefficients are reported in Table S1 of the supplementary material. For Car 1 and 4 the coastdown curves were provided by the car manufacturer, whereas for the remaining cars the procedure developed by the ERMES group (Matzer and Rexeis, 2016) was applied. For each car the same coastdown coefficients were used for all the tests.

Exhaust emissions were measured and calculated as prescribed by (European Commission, 2017a). Gasoline ($FC_{gasoline}$) and LPG (FC_{LPG}) fuel consumptions were calculated through the carbon balance equations reported in (European Commission, 2008) and showed in Equation (1) for gasoline and Equation (2) for LPG:

$$FC_{gasoline} = 0.118 * [(0.848 * HC) + (0.429 * CO) + (0.273 * CO_2)] / \rho_{fuel} \quad \text{Equation1}$$

$$FC_{LPG} = 0.1212 * [(0.825 * HC) + (0.429 * CO) + (0.273 * CO_2)] / 0.538 \quad \text{Equation2}$$

where HC, CO, and CO_2 are the measured species emissions [g/km], ρ_{fuel} is the gasoline's density [kg/l] and the value 0.538 kg/l is the reference LPG's density. Actually, we used Equations (1) and (2) with the densities calculated after the chemical analysis of the fuels in the tanks of the different vehicles, according to the (UNI EN ISO 12185:1999) method for gasoline and (UNI EN ISO 8973:2001) for LPG.

Based on the fuel consumptions, LPG energy consumptions (EC) were subsequently calculated using the calorific values (ranging from 45.730 MJ/kg to 46.344 MJ/kg) measured according to the (DIN 51612:1980–06) technical standard, whereas, not being available the calorific values of gasoline in the tank of the tested cars, the calorific value of gasoline sold in Italy (i.e. 42.817 MJ/kg) was used (ISPRA, 2021b).

2.2.2. RDE tests

RDE tests were carried out driving the test cars on road in and around the city of Milan. For all the tests the same predetermined route, shown in Fig. 1, was driven and a similar planned driving style was maintained in order to get emission results as much as possible comparable among the tests. The RDE tests, performed in compliance with mandatory conditions imposed by RDE regulation (European Commission, 2016a, 2016b, 2017b), covered a distance of about 73 km and were composed by three phases: Urban (i.e. with speed less than 60 km/h), Rural (i.e. with speed between 60 km/h and 90 km/h) and Motorway (i.e. with speed greater than 90 km/h). For each car and for each fuelling type RDE tests were carried out twice. In section S2 of the supplementary material detailed trip characteristics as well as altitude and speed profiles are reported.

Measurements in RDE tests were performed through a Portable Emissions Measurement System (PEMS) module that allowed to detect both gaseous and non-gaseous emissions: NOx through an ultraviolet detector, THC and CH_4 through a double channel flame ionization detector, CO and CO_2 through a non-dispersive infrared detector, and PN through a volatile particle remover followed by a diffusion charger detector. Before and after each test, all the gas analysers have been purged and calibrated, through zero and span checks, with certified gas cylinders. All PEMS data were processed and final distance-specific emission results were obtained by AVL Concerto software using the Rel_9_B162_HF1_v2 work environment.

3. Results and discussion

3.1. Gasoline and LPG fuelling comparison

For any car and fuelling the emission results were calculated as average values of three tests for NEDC and ERMES cycles, as a single value for WLTC, and as average values of two tests for the RDE (Table S3 in the supplementary material). The statistical significance of the differences between the emissions with LPG and gasoline fuelling was investigated by applying the Welch's *t*-test with 95% confidence level for each vehicle over NEDC and ERMES cycles only. Table 2 summarises the percentage differences between LPG and gasoline fuelling with the

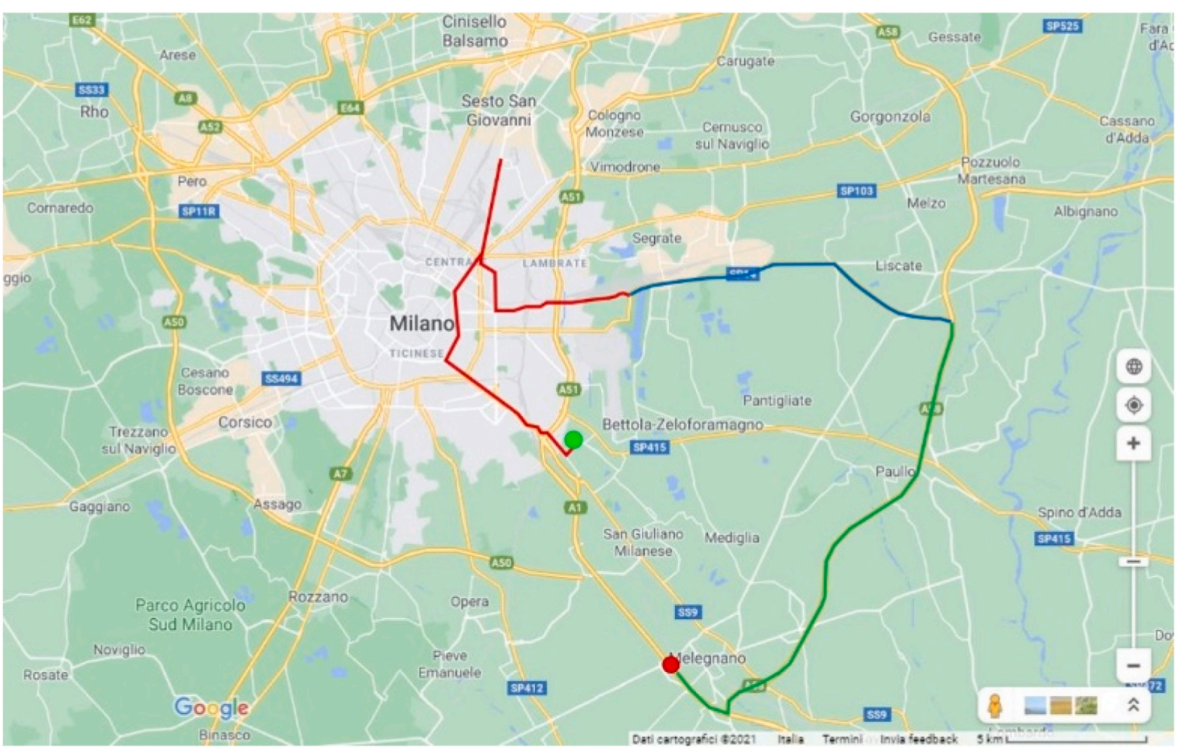

Fig. 1. Route of the RDE tests traced on a Google Maps image. Red line: urban phase; blue line: rural phase; green line: motorway phase; green and red circles are the starting and ending point of the route, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Percentage differences found switching from gasoline to LPG feeding.

Emission change LPG vs gasoline fuelling [%]													
		Car 1	Car 2	Car 3	Car 4	Car 5			Car 1	Car 2	Car 3	Car 4	Car 5
Regulated emissions													
CO	NEDC	+239.2	−18.8	+126.2	+197.5	+81.0	THC	NEDC	+54.7	+11.3	+171.5	+87.0	+15.2
	ERMES	−34.2	−41.4	+334.2	−58.6	+32.9		ERMES	−5.9	+1.9	+183.4	–	+22.2
	WLTC	+285.1	−33.6	+48.0	+101.4	+7.6		WLTC	+96.4	−27.0	+80.0	+10.8	+5.6
NMHC	NEDC	+48.2	+11.1	+177.1	+85.2	+8.5	NO_x	NEDC	−36.2	+132.8	−7.3	−36.0	−64.6
	ERMES	−0.3	+8.8	+185.4	−27.2	+6.6		ERMES	+59.9	+281.6	−23.9	+285.5	−7.6
	WLTC	+90.4	−27.9	+63.6	+2.0	+5.3		WLTC	−31.1	+85.9	−15.7	+12.5	−24.7
PN_PMP Compliant	NEDC	+1.1	−46.5	−36.5	+49.4	−50.5	PM	NEDC	−10.6	+14.1	+324.8	−98.9	+53.1
	ERMES	−36.2	−61.0	−85.6	−89.8	+40.7		ERMES	−28.0	+67.4	−4.8	−90.6	–
	WLTC	−18.3	−37.2	−31.9	−62.7	−49.5		WLTC	+27.9	−4.4	+93.0	−91.3	−49.4
Unregulated emissions and energy consumption													
CO_2	NEDC	−9.1	−8.7	−5.7	−9.7	−12.3	CH_4	NEDC	+240.0	+16.8	+137.5	+126.3	+68.8
	ERMES	−9.1	−8.0	−6.7	−4.5	−11.6		ERMES	−29.1	−19.3	+166.3	−61.8	+57.7
	WLTC	−9.9	−7.9	−5.8	−5.9	−9.4		WLTC	+217.5	−18.2	+142.4	+106.2	−2.3
PN-ELPI	NEDC	+11.4	−52.3	−59.7	+63.0	−37.7	PN-EEPS	NEDC	+5.6	−49.7	−39.7	+64.1	−39.1
	ERMES	−30.2	+10.1	−85.6	−54.1	−56.6		ERMES	−44.7	+14.2	+28.1	−72.2	−6.5
	WLTC	−9.8	−39.9	−28.9	−63.4	−43.4		WLTC	−14.8	−27.8	−30.5	−42.6	−19.2
Soot	NEDC	+21.5	−54.9	−29.4	+106.8	−40.5	Energy Consumption (EC)	NEDC	+0.9	+3.2	+3.9	+3.0	−2.3
	ERMES	−3.5	−26.3	−84.7	−46.8	+34.1		ERMES	−0.1	+3.8	+2.7	+7.8	−1.6
	WLTC	+22.9	−5.6	−33.0	−57.0	−63.5		WLTC	−0.3	+4.0	+3.8	+7.2	+0.8
NH_3	NEDC	+619.1	+2.9	+82.1	+183.4	+701.1	NO_2	NEDC	+6.6	−1.2	+25.1	+10.8	+9.9
	ERMES	+42.9	−17.8	+83.2	+6.0	+74.0		ERMES	+26.0	+5.1	+24.0	−22.0	−0.9
	WLTC	+112.0	−11.6	+51.0	+137.3	+95.3		WLTC	+14.2	−0.1	+23.9	+19.4	+10.2
Formaldehyde	NEDC	−39.8	+56.3	+93.4	+437.9	+294.6	N_2O	NEDC	+49.3	−6.0	+15.0	−24.0	−11.6
	ERMES	–	–	–	–	+6988.2		ERMES	+74.2	−5.8	+35.8	−1.8	−14.5
	WLTC	+10.5	−69.4	+26.4	–	−27.9		WLTC	+28.4	+44.2	−14.3	−3.1	+41.9

statistically significant differences highlighted in yellow. Figs. 2–4 summarise the emission results (mean value and 95% confidence interval), also indicating the cars for which statistically significant differences were observed. Additionally, for the regulated emissions (Fig. 2, cars 1–4 over the NEDC and car 5 over the WLTC) the red lines show the Euro 6 emission standards according to the type-approval homologation of each testing car. Because the on-road driving is characterized by unpredictable external conditions, the RDE tests are intrinsically not repeatable; therefore, no statistical analysis was developed. For this reason, any comparisons between gasoline and LPG emissions over RDE tests shall be considered as qualitative. Also, although RDE test was not mandatory for Euro 6b and Euro 6c vehicles in the homologation phase, PN and NO_x emission values obtained from RDE tests have been compared to the Euro 6d-temp not-to-exceed (NTE) emission limits (Fig. 5) (European Commission, 2017a).

3.1.1. Laboratory tests

As shown in Fig. 2, all regulated gaseous emissions (CO, THC, NMHC and NO_x) over the type-approval driving cycle were below the Euro 6 limits (European Commission, 2012). Particulate emissions (in Fig. 2 reported both as PN-PMP compliant and PM), which are only regulated for direct injection engines in positive ignition vehicles (i.e. only Vehicle 3 in this experimental campaign), were compliant with both Euro 6 limits (European Commission, 2012) over the NEDC type-approval driving cycle (PM, PN-PMP compliant) for LPG fuelling only; regarding the gasoline fuelling, although PM was complied with the limit, a 10.9% exceedance of the PN-PMP compliant limit (6.65E+11 #/km vs. 6.0E+11 #/km) was observed for PN.

As shown in Table 2, over the NEDC LPG fuelling resulted in some statistically significant increased emissions of CO, THC and NMHC and in some statistically significant reductions of NO_x and PN-PMP compliant emissions. Over ERMES cycle the statistically significant increases of THC and NMHC with LPG fuelling are confirmed only for Car 3 whereas statistically significant reductions of PN-PMP compliant emissions are confirmed for most cars. Compared to the NEDC ones, the NOx and CO emission behaviours over ERMES cycle are changed: one NOx statistically significant increase and two CO statistically significant reductions came up for LPG fuelling. Very low PM emissions were measured for all the tested cars and for both the fuels (0.005–0.426 mg/km range) and no statistically significant variations were detected. It should be noted that the weight of PM filters was often close to the 10^{-7} g scales detection limit, therefore measurements in this order of magnitude may be less accurate. Very low emissions of THC (2.25 mg/km with gasoline, 2.91 mg/km with LPG as average emission among all vehicles) and NMHC (1.83 mg/km with gasoline, 2.13 mg/km with LPG) were detected over the hot-start ERMES cycle, clearly showing a high hydrocarbons abatement effectiveness by the three-way catalyst at high temperatures. Compared to the NEDC cycle, these emissions were significantly and remarkably lower with both fuelling types: 91.0% of THC and −93.6% of NMHC with gasoline and −93.6% of THC and −94.7% of NMHC with LPG.

The emission measured over the WLTC basically mirrored the emission levels observed over the NEDC for all the tested cars. Nevertheless, clearly higher PN-PMP compliant emissions were observed for both fuels (on average + 89.0% for gasoline, +62.0% for LPG) except for car 4 with LPG fuelling, for which a lower emission was detected over WLTC (−54.0%).

The experimental results did not show any systematically different behavior for the emission of OEM and retrofitted testing cars for all the regulated pollutants. These results may emphasize the accuracy of the retrofitting processes, which were carefully carried out on testing cars 1, 2 and 3. Conversely, despite very limited and not updated for recent technologies, literature studies report that retrofit LPG vehicles have higher NOx and PM emissions compared to OEM ones. For instance, by analysing the emission levels of Euro 4 bi-fuel LPG cars with different LPG powertrain systems, (Vonk et al., 2010) observed that retrofitted vehicles emitted, on average, more than twice NO_x and more than 2.5 times PM compared to the gasoline fuelling, whereas the OEM LPG-fuelled cars emitted NO_x and PM at the same level as for gasoline fuelling. This behaviour was only partially confirmed by our study. Indeed, among the retrofitted vehicles only Car 2 emitted on average more than about 2.5 times NO_x than its gasoline fuelling, whereas the other retrofitted cars (1 and 3) emitted on average less NO_x when fuelled with LPG. Regarding PM, only GDI Car 3 emitted, when run on LPG,

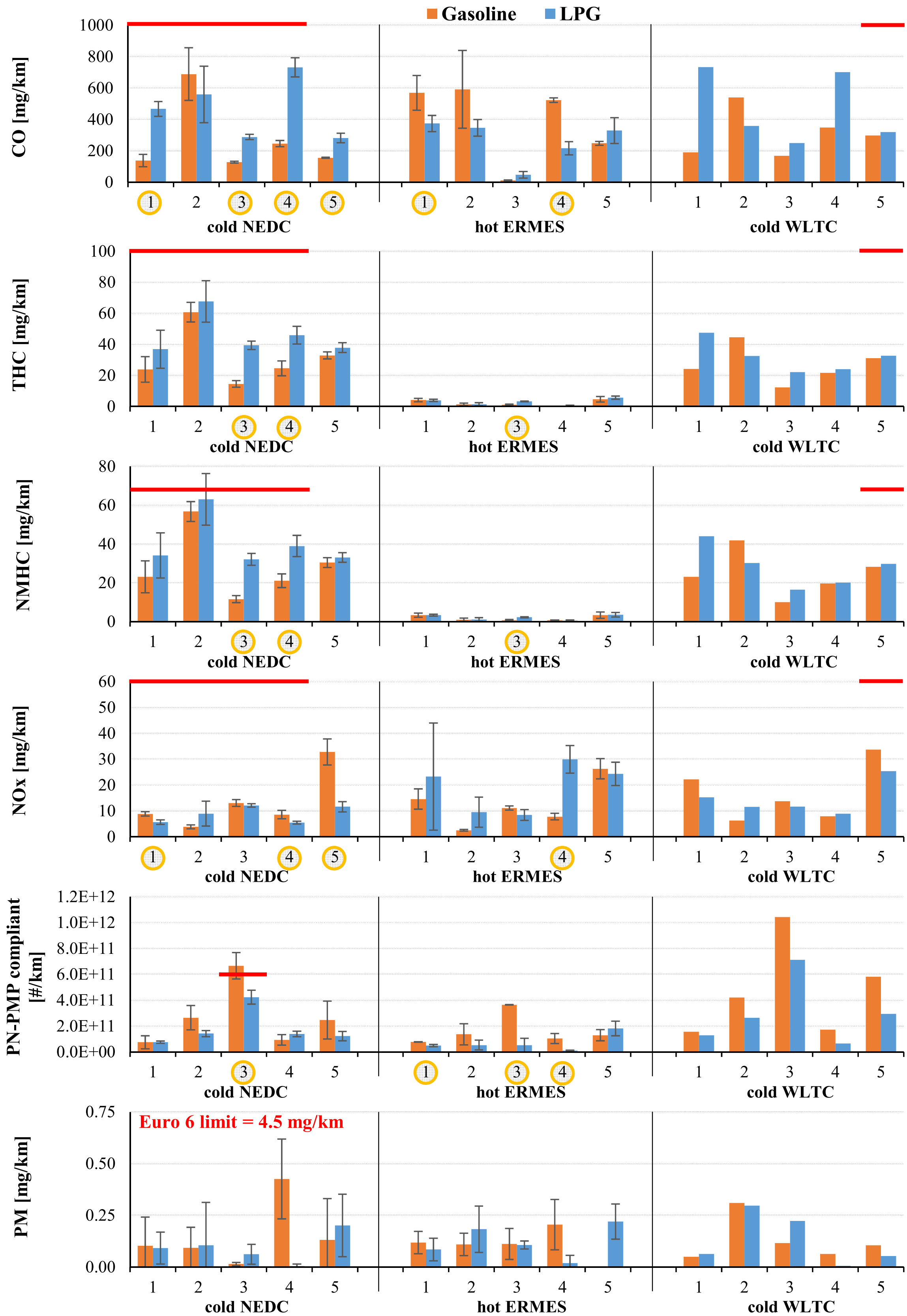

Fig. 2. Comparison of average emissions of regulated pollutants detected over NEDC, ERMES and WLTC (1 test) cycles for the testing cars fed with gasoline and LPG.

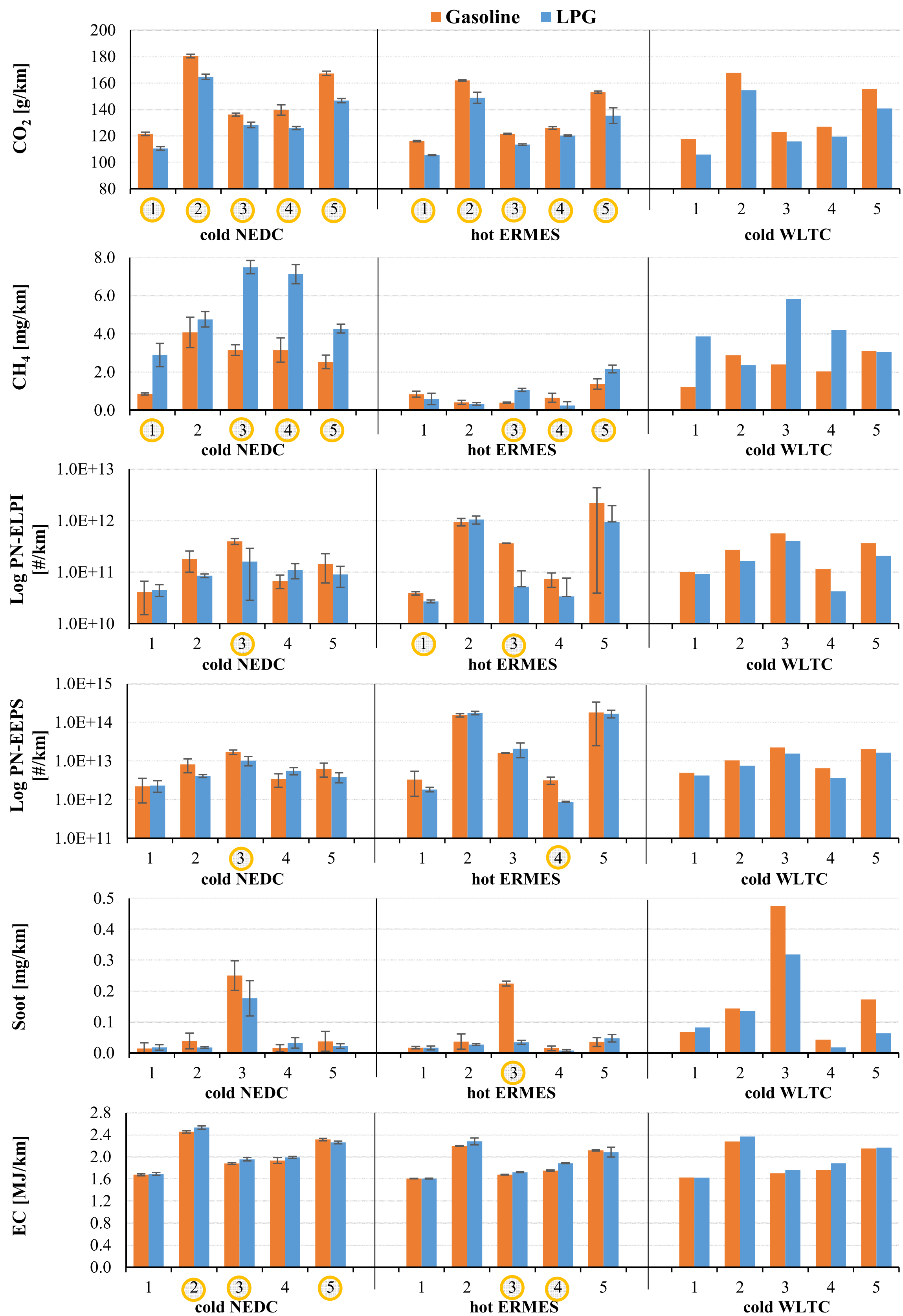

Fig. 3. Comparison of average CO_2, CH_4, PN-ELPI, PN-EEPS, Soot emissions and EC values detected over NEDC, ERMES and WLTC (1 test) cycles from the testing cars fed with gasoline and LPG.

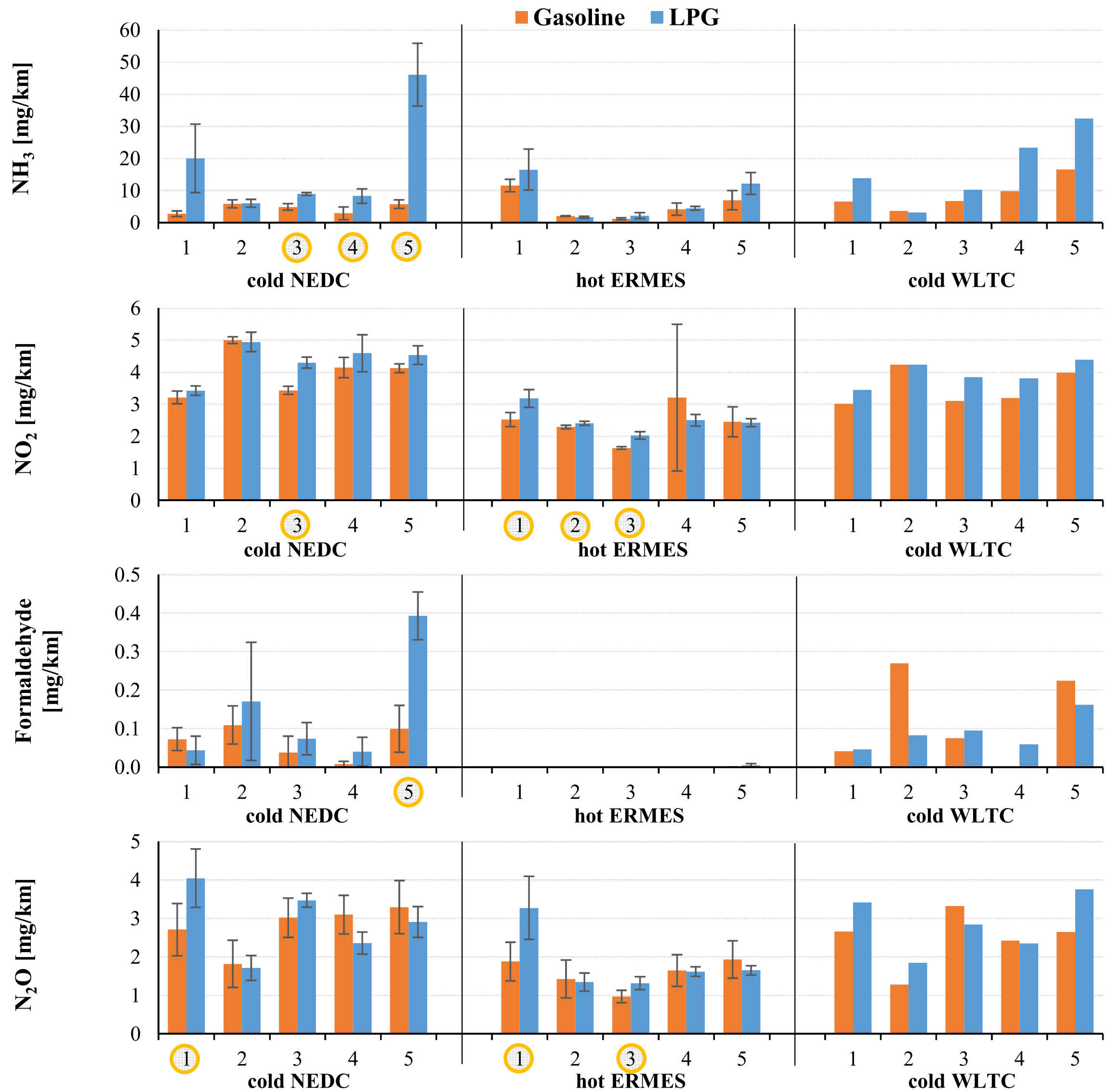

Fig. 4. Average emissions of unregulated gaseous pollutants detected over NEDC, ERMES and WLTC (1 test) cycles for the testing cars fed with gasoline and LPG.

more than twice PM as its gasoline counterpart, whereas Car 1 and Car 2 showed similar PM emissions with both fuelling. Instead, comparing NO_x and PM average emission levels from OEM vehicles (Car 4 and Car 5) fuelled with gasoline and LPG, no similar values were detected and no similar and common behaviours were observed either: Car 4 showed higher NO_x and lower PM average emissions switching from gasoline to LPG whereas the opposite happened with Car 5. Moreover, the limited number of cars did not allow to draw conclusions about the difference between OEM and retrofit systems emissions based on the fuel injection type (port fuel vs direct injection).

As shown in Fig. 3 and according to the literature (Heidt et al., 2013; Huss and Weingerl, 2020; Ristovski et al., 2005), due to the higher hydrogen/carbon ratio of LPG compared to gasoline (H/C about 2.52 for LPG, 1.86 for gasoline (Ntziachristos and Samaras, 2019)), LPG fuelling resulted in lower CO_2 emissions, with statistically significant variations for all the tested cars over all the cycles. Compared with gasoline fuelling, the average decrease was −9.1% over NEDC, −8.0% over ERMES, and −7.8% over WLTC. The highest CO_2 emission values were detected over the NEDC cycle for both fuellings: 149 g/km and 135 g/km as average values for gasoline and LPG fuelling, respectively. The energy consumptions (EC) showed very similar results for all the testing vehicles and testing cycles (mostly within the −0.3% - –4.0% range), with two statistically significant slight increases both over NEDC and ERMES and only one statistically significant slight decrease over NEDC with LPG fuelling respect to gasoline. The overall average differences found with LPG fuelling are +1.8%, +2.5% and +3.1% over NEDC, ERMES and WLTC, respectively. However, it must be taken into account that the EC for gasoline were calculated using an average gasoline calorific value for all the testing cars and this may have affected the comparison's accuracy.

Very low CH_4 emissions were measured with both fuels over the driving cycles and particularly over ERMES, with average emissions among all vehicles equal to 0.738 mg/km using gasoline and 0.884 mg/km using LPG. Nevertheless, statistically significant increased CH_4 emissions were observed with LPG fuelling for 4 cars over NEDC and for 2 cars over ERMES cycle. The only statistically significant reduction observed in CH_4 emission was for Car 4 over ERMES cycle.

The unregulated PN emission results confirmed the behaviours detected by the PN-PMP compliant system: considering LPG fuelling, statistically significant reductions were detected both for PN-ELPI and for PN-EEPS. Compared with gasoline fuelling results, it was observed an average decrease of both PN-ELPI (−37.1%) and PN-EEPS (−27.0%) emission values with LPG fuelling over WLTC for all the tested cars. The PN-EEPS values were at least an order of magnitude higher than the PN-

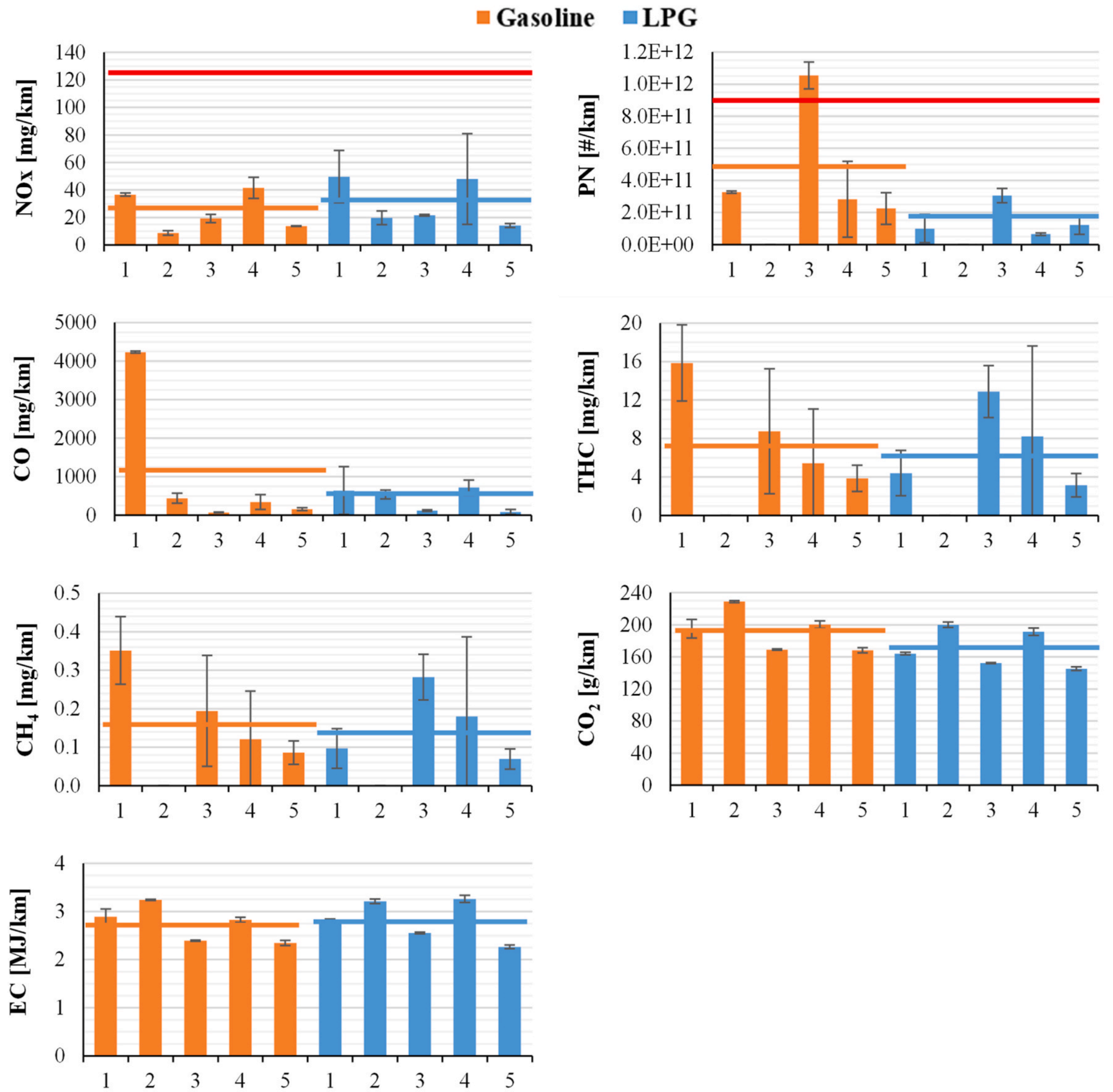

Fig. 5. Average emission and energy consumption values detected over RDE tests. Red lines represent the Euro 6d-temp NTE limits whereas the orange and blue lines represent the average values with gasoline and LPG fuelling respectively. Particle number emission was not measured during tests on Car 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

PMP compliant ones, due to two main reasons. The first one is the different measuring range of the analysers, which allows the EEPS to detect smaller particles (therefore, taking into account the particle number, it would include a bigger part of the exhaust particles) compared to the PMP-compliant system. The second one is the different sampling technique: the PMP system includes an engine exhaust thermodiluter that removes the volatile and semi-volatile particles from the sample whereas, as the EEPS detected a non-treated sample, total particles within its measuring range were measured.

Soot emission values were in general very low, with the highest emission on each cycle detected for Car 3 operating on gasoline: 0.25 mg/km over NEDC, 0.22 mg/km over ERMES and 0.48 mg/km over WLTC. Comparing the two testing fuelling, the only statistically significant variation on soot emission was the decrease detected for Car 3 operating on LPG over ERMES cycle.

As shown in Fig. 4, the gaseous unregulated emissions were mostly very low and only statistically significant increases with LPG fuelling were detected: NH_3 and Formaldehyde over NEDC, NO_2 and N_2O both over NEDC and over ERMES cycle. However, differences between gasoline and LPG fuelling were on average very low and, although a few statistical differences came up regarding the measured unregulated pollutants, they wouldn't seem sufficient to raise any general concern for LPG fuelling.

3.1.2. RDE tests

As shown in Fig. 5, all the tested cars would have met the Euro 6d-temp NTE limits (calculated according to (European Commission, 2016a) with conformity factors equal to 2.1 for NOx and 1.5 for PN), except for Car 3 with gasoline fuelling that exceeded by 17.2% the 9.0E+11 #/km PN emission limit. Switching from gasoline to LPG, PN, CO_2 and energy consumption behaviours were comparable with those observed in laboratory tests: a significant reduction (up to 70%) of PN emission (average value of 4.7E+11 #/km with gasoline and 1.5E+11 #/km with LPG), a decrease of CO_2 emission (about −11%, with average value of 192.3 g/km with gasoline and 170.6 g/km with LPG), and very similar energy consumptions, with average value equal to 2.7 MJ/km with gasoline and 2.8 MJ/km with LPG, were observed.

Concerning CO emissions, the comparison between the two fuels is strongly affected by the gasoline-powered Car 1 data: indeed, both RDE tests showed significantly higher emissions (around 4200 mg/km) than for the other four vehicles (on average 251 mg/km) likely due to the high load required in the high speed phase, which led to a greater effort of the less powerful car among those tested. Indeed, high CO emissions were detected with Car 1 during accelerations in the motorway phase for both tests performed with gasoline fuelling, highlighting a critical condition for this vehicle that was also confirmed by CO measurements during the strong accelerations of the ERMES cycle. Neglecting Car 1, the comparison showed an average increase of CO emissions with LPG fuelling for Car 2, Car 3 and Car 4 (on average + 68%) and an average decrease (−45%) for Car 5. Average NO_x emissions from LPG-powered testing cars were generally slightly higher than the gasoline ones (30.6 mg/km with LPG vs. 24 mg/km with gasoline). Concerning THC and CH_4 emissions, the cars showed different behaviours switching from gasoline to LPG: for both pollutants reductions were observed for Car 1 and at a smaller extent for Car 5; on the contrary, increased emissions were observed for Car 3 and 4. THC and CH_4 emission measured for Car 2 were instead systematically below the PEMS detection limit.

3.2. LPG emission factors development

In order to obtain an indication of the accuracy of the COPERT model estimations regarding the Euro 6 LPG passenger cars, model predictions were compared with experimental results. In particular hot emission and energy consumption factors, which are integrated in the COPERT model, were extracted from the EMEP/EEA inventory guidebook (considering its September 2020 update (EMEP/EEA, 2020)) and compared with the sub-cycle distance-specific hot emission factors (Fig. 6) and energy consumption values detected using LPG fuelling (Fig. 7). Hot emission factors correspond to vehicle emissions when the engine and the exhaust after-treatment systems have reached their nominal operating temperature. For this reason, following the cold start definition introduced in Regulation (2017)/1151 for RDE test, the emissions detected during the first 300 s of each cold-start test were excluded from the calculation of the hot emission and consumption factors. Given that NEDC is not representative of real-life driving conditions, the results of this cycle were excluded from the calculation and the same was done for the full load accelerations of the ERMES cycle, as they are useful to better investigate the vehicle engine emission map but not suitable for finding EFs representative of an average driving. Each average ERMES, WLTC and RDE sub-cycle factor was plotted in correspondence of the average sub-cycle speed (yellow circles in Figs. 6–7) and overall average factors were calculated for the different driving conditions: urban, rural, and highway, which have been highlighted in Figs. 6–7 with the red, blue and green areas, respectively. Emission data of Urban (ERMES), Low (WLTC) and Urban (RDE) phases were averaged to obtain the average emission and consumption factors for the urban driving, those of Extra Urban (ERMES), Middle (WLTC), High (WLTC), and Rural (RDE) phases for the rural driving, and those of Motorway (ERMES), Extra-High (WLTC) and Motorway (RDE) phases for the highway driving. The experimental results of each sub-cycle were classified by a certain driving condition based on the average speed of that sub-cycle: 0–35 km/h, 36–85 km/h and 86–120 km/h for urban, rural and highway, respectively. For what concerns PM emissions, for which data were available for the whole driving cycle, the measured values were processed in order to assess the emissions attributable to each single phase of the driving cycle. The PM sub-cycle emission, as showed in Equation (3), was proportionally estimated based on the fraction of the PN-PMP compliant emission of each single phase with respect to the whole cycle:

$$PM_{i,k,LPG} = PM_{j,k,LPG} * (PN\text{-}PMP_{i,k,LPG} / PN\text{-}PMP_{j,k,LPG}) \qquad \text{Equation3}$$

where $PM_{i,k,LPG}$ indicates the estimate of PM emission over sub-cycle i (Urban, Extra Urban, Motorway, Low, Middle, High, Extra-high) of cycle j (ERMES, WLTC), $PM_{j,k,LPG}$ the PM emission detected over cycle j, $PN_{i,k,LPG}$ the PN-PMP compliant emission detected over sub-cycle i of cycle j, and $PN_{j,k,LPG}$ the PN-PMP compliant emission detected over cycle j for test car k. This is a very rough estimate, based on the assumption of both constant particle size distribution and PM composition (i.e. constant contribution of volatile, semi-volatile and solid fractions) in the different phases of the cycle, and thus likely affected by significant uncertainty: thus, the resulting sub-cycle PM emission values have to be considered strictly indicative and just useful for the comparison with COPERT model results.

The pollutants considered for the comparison were those that the COPERT model offers the possibility to enter user values for. Data from Car 1 and Car 3, which belong to the <1400 cm^3 displacement category, were used for the small segment whereas data from Car 2, Car 4, and Car 5, which belong to the 1400–2000 cm^3 category, were used for the medium segment.

As shown in Figs. 6–7, in the COPERT model the same EFs and EC factors are assigned to both the small and the medium segment of Euro 6 LPG passenger cars, probably due to the limited amount of available data for LPG vehicles. COPERT's hot factors were found to be in reasonable agreement only with the average experimental EFs regarding CO and with the average experimental EC factors for the medium displacement category. Conversely, more marked differences were found in the other cases, with the COPERT factors almost systematically overestimating the test results. CO average experimental EFs of the small segment resulted on average about 40% lower the COPERT's ones, despite the average emission values detected with Car 1 in the rural and motorway phases of the RDE tests were equal to 0.61 g/km and 1.35 g/km, respectively. For NO_x, the average experimental EFs of the small segment resulted by about 70% lower both in the urban and rural driving condition, but resulted about 2 times higher in the highway condition, mainly because of a single outlying result obtained in the high load Motorway phases of RDE test with Car 1. For the medium segment they were lower than the COPERT's ones in all the driving conditions, with an average reduction by about 40%. Similarly, lower EFs were found for THC, respectively by about 79% and 59% in the rural and highway driving conditions for the small category, and by about 74%, 95% and 73% in the urban, rural and highway driving conditions for the medium category. However, the average experimental EF for the small segment in the urban phase resulted 30% higher than the COPERT value, as a consequence of a single outlying data. The outcomes for CH_4 confirmed the behaviours found for THC: lower experimental EFs by about 46% and 43% in the rural and highway driving conditions for the small category and on average by about 48% considering the different driving conditions for the medium category. Also, for CH_4 one outlying data led to a 37% higher average experimental EF in the urban phase for the small category. For PM, the average experimental EFs resulted always significantly lower than the COPERT's ones, with reductions from 89% to 97% for the small segment and from 91% to 96% for the medium segment. Estimated PM EFs, showed in Fig. 6, were obtained including a rough estimate, as largely described previously. However, since these sub-cycle estimated values turned out to be in the same order of magnitude with measured PM emissions (with regard to total cycles, shown in Fig. 2), they can be considered as plausible and reliable ones, all pointing out that COPERT's PM EFs are largely affected by overestimation. The EC factors of the small segment resulted on average 19% lower than the COPERT's predictions, with a more marked 25% difference observed for the rural phase. In the red boxes of Fig. 6 the average experimental EFs obtained for the small segment neglecting the outliers are shown. In all cases the experimental EFs were lower than the COPERT's ones, with reductions of 90% and 95% respectively for THC and CH_4 urban EFs and equal to 22% for NOx highway EF.

3.3. Effect of new country-specific LPG EFs on Italian emission estimation

With the aim of highlighting the impact of these updated Euro 6 LPG country-specific emission and consumption factors on the Italian national emissions calculation for this vehicle categories, ISPRA carried out an ad hoc comparative simulation for the year 2019 through the

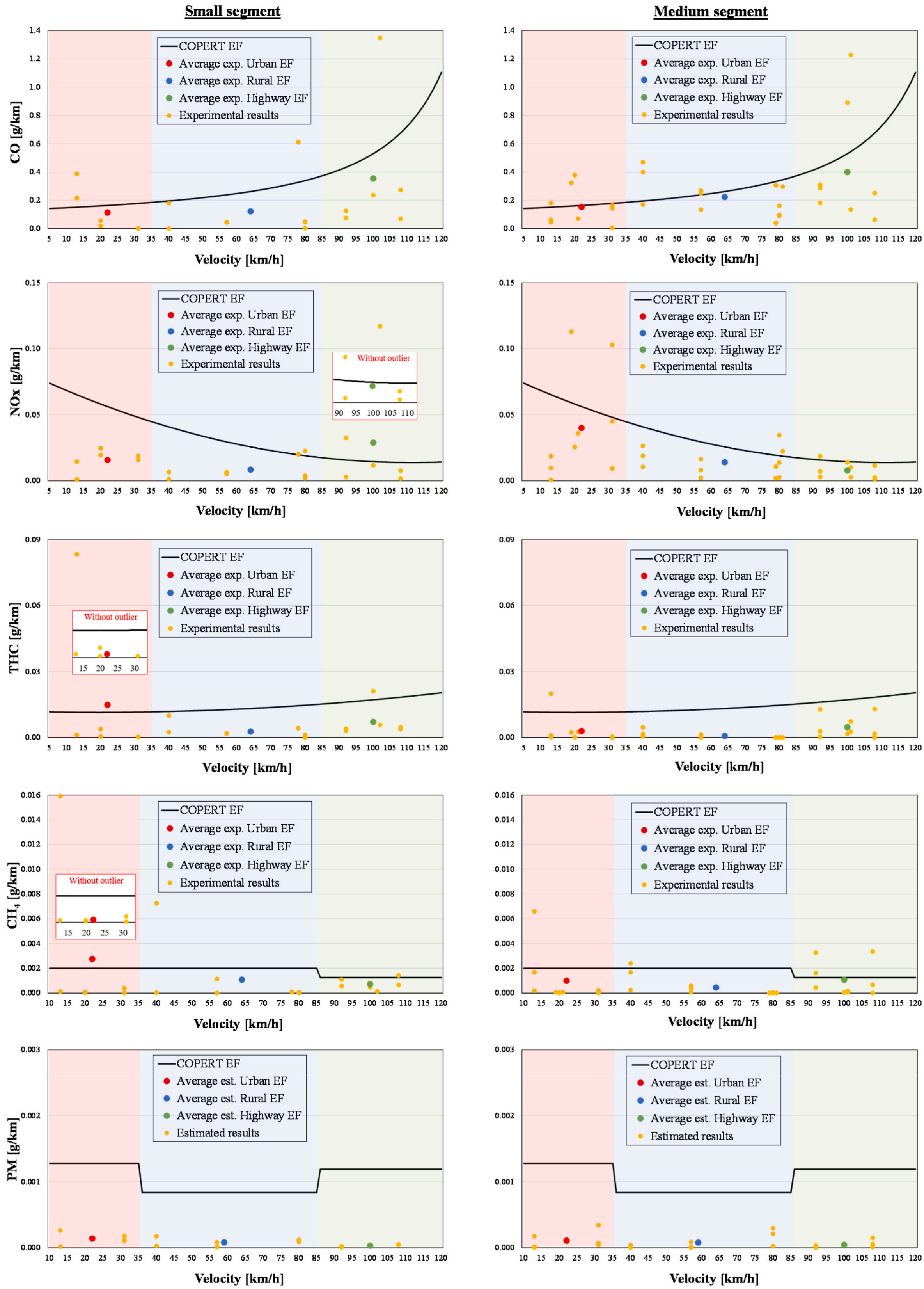

Fig. 6. Comparison between experimental hot emission factors and COPERT model predictions for Euro 6 LPG passenger cars. The red, blue and green circles show respectively the urban, rural and highway experimental average EFs. PM values are estimated from experimental results as described by equation (3). In the red boxes the average experimental EFs without the outlier data. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

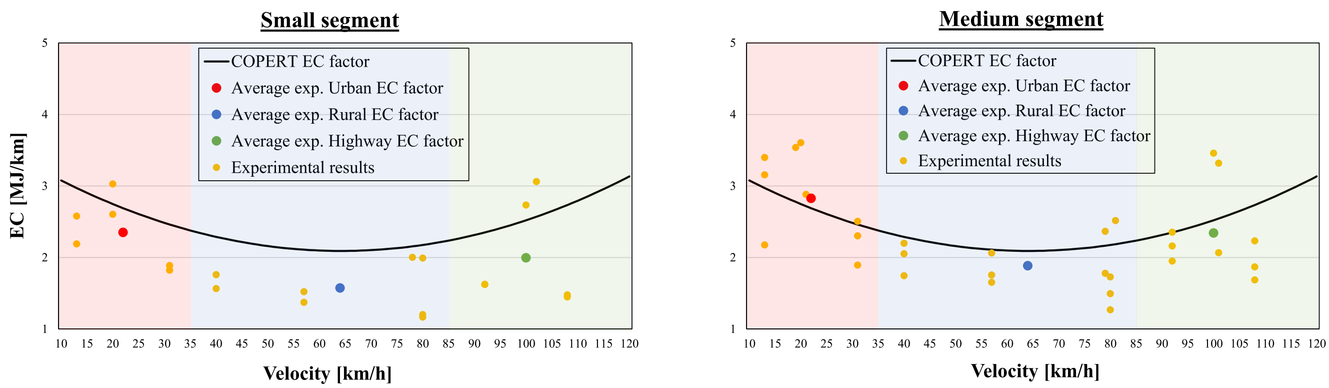

Fig. 7. Comparison between experimental hot EC factors and COPERT model predictions for Euro 6 LPG passenger cars. The red, blue and green circles show respectively the urban, rural and highway experimental average EC factors. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

5.4.36 version of the COPERT model. Keeping all the input parameters used for the estimation of Italian road transport national emissions unchanged (e.g. environmental data, trip characteristics, fuel specifications, vehicles stock, activity and circulation data), two parallel simulations were carried out: one using the default hot emission and EC factors of the COPERT model, the other using the experimental average hot factors provided by this work. In Fig. 8 the simulation results are compared separately for the small and the medium segment. In general, the experimental country-specific factors led to lower emission estimates for all species, with the highest differences in PM emission values: −91.8% and −92.5% respectively for the small and the medium segment. Smaller but still relevant differences were obtained for NOx for both segments (−22.9% for the small one and −16.8% for the medium one), for CO for the small segment (−18.1%), and for THC for the medium segment (−9.4%). Lower emission differences were instead obtained in the other cases, with very similar values especially for CH_4 (−0.2% and −1.1% for small and medium segment, respectively). Regarding the estimates of energy consumptions, a considerable −15.6% reduction was found for the small segment with the experimental hot EC factors, whereas a less marked reduction (−1.8%) was found for the medium segment.

4. Conclusions

This paper reports the results of a testing campaign performed on a pool of five Euro 6 (B and C) LPG bi-fuel passenger cars with different technical characteristics selected to well represent the Italian LPG vehicle fleet. The cars were run on both gasoline and LPG fuelling, tested over several driving cycles (NEDC, ERMES, WLTC) and on road (RDE), and their exhaust emissions as well as energy consumption were measured. Regulated emissions were basically found to meet the Euro 6 standards over each specific type-approval driving cycle except for the direct injection engine car, which exceeded the PN emission limit when running on gasoline. Switching from gasoline to LPG both the laboratory and the on-road tests showed systematic reduction in CO_2 emissions. In addition, LPG tests showed reduced particulate emissions compared to the gasoline ones on average, which has been observed both in laboratory and on road. Country-specific LPG hot emission and energy consumption factors were calculated from the experimental results and compared with those extracted from the EMEP/EEA inventory guidebook: the comparison showed a good match for CO and EC factors for the medium displacement category and slightly or widely marked gaps in the other cases. Indeed, CO and EC experimental factors for the small segment and NOx, THC, CH_4 and PM experimental EFs for both the displacement categories were almost systematically lower than the COPERT's ones, with the greatest differences (up to 97%) in the case of PM. These deviations are attributable to the lack of updated Euro 6 LPG vehicle data in the EMEP/EEA database: in fact, the EFs of this vehicle category in the database were assumed to be the same as those attributed to vehicles homologated with previous emission standards. Country-level emissions of Euro 6 LPG passenger cars estimated for Italy by means of the EFs developed in this work turned out to be actually much lower than those estimated through the default EFs of the COPERT model, with emission reductions up to 92.5% for PM and 22.9% for NOx. Therefore, the development of up-to-date Euro 6 LPG EFs based on experimental tests is fundamental to obtain much more realistic and accurate emission estimations from such vehicle category that is widely common in Italy. All the experimental data were also shared with ERMES group in order to support the update of the EFs databases of emission models. Considering the role of these tools in road transport emission inventory compilation and, consequently, in air pollution abatement measures, additional and up-to-date experimental data are

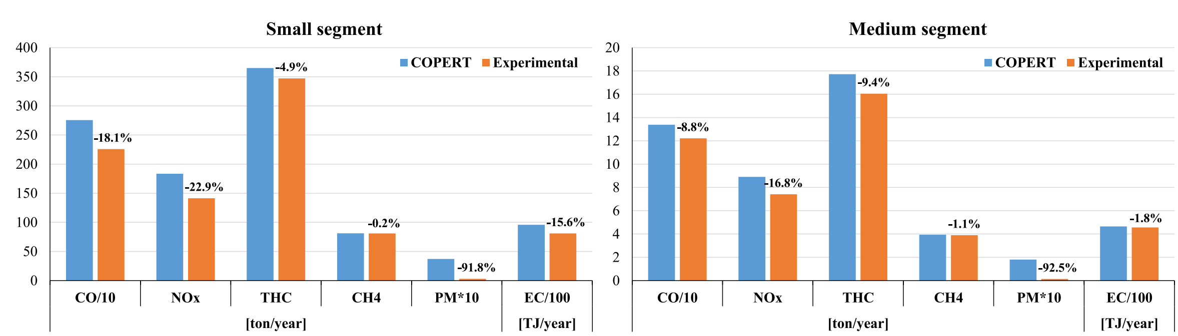

Fig. 8. Italian national emissions and energy consumptions for Euro 6 LPG passenger cars calculated using the COPERT model predefined factors and the experimental factors.

needed in particular for vehicle or homologation classes for which there could still be lacks of recent factors in the emission model databases.

CRediT authorship contribution statement

Tommaso Bellin: Conceptualization, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Simone Casadei:** Conceptualization, Formal analysis, Investigation, Supervision, Project administration, Writing – review & editing. **Tommaso Rossi:** Conceptualization, Formal analysis, Investigation, Writing – review & editing. **Antonella Bernetti:** Conceptualization, Formal analysis, Writing – review & editing. **Riccardo De Lauretis:** Conceptualization, Writing – review & editing. **Giovanni Lonati:** Conceptualization, Writing – review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

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