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On-Road and Laboratory Emissions from Three Gasoline Plug-In Hybrid Vehicles—Part 1: Regulated and Unregulated Gaseous Pollutants and Greenhouse Gases

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Abstract: Road transport is a relevant source of greenhouse gas emissions. In order to meet the European decarbonisation targets, the share of electrified vehicles, including battery electric vehicles and plug-in hybrid electric vehicles (PHEVs), is rapidly growing, becoming the second most popular powertrain in the European market. PHEVs are of interest since they are expected to deliver a reduction in gaseous pollutants such as NOx as well as in greenhouse gases such as CO_2. Herein, we explored both categories of emissions for three PHEVs with gasoline direct-injection engines, meeting the latest European emission standards (Euro 6d and Euro 6d-TEMP). They were studied in laboratory and on the road, in different modalities and temperatures. All tested vehicles met the Euro 6 emission limits in the Worldwide Harmonised Light-Duty Vehicles Test Procedure (WLTP) and the real driving emissions (RDE) test procedure. Still, when their internal combustion engine ignited even for a few km, their emissions were comparable to, and in some cases higher than, the average emissions reported for a fleet of eight conventional Euro 6d-TEMP gasoline direct-injection vehicles. The tested PHEVs presented similar trends to those of conventional vehicles, such as the increase in all pollutants considered at low ambient temperature or the high CO emissions during acceleration events, concomitantly with NH_3. Moreover, depending on the boundary conditions, emissions were higher for the vehicles with a battery fully charged with respect to tests performed with the depleted battery. Furthermore, the use of an operating mode that allowed charging the vehicles' high voltage battery using the internal combustion engine had a very strong impact on the vehicles' CO_2 emissions, offsetting the benefits in terms of greenhouse gas reduction demonstrated in other conditions. The results indicate that for the sample tested, the expected reduction in pollutants emission due to the presence of a hybrid gasoline-electric traction seemed in some cases limited, also showing high variability. CO_2 emissions were also affected by the initial state of charge of the vehicles' high voltage battery as well as from the user-selectable operating mode, also in this case with high variability.

Keywords: pollutants emissions; greenhouse gas emissions; plug-in hybrid vehicles; unregulated pollutants; transport sector

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1. Introduction

With the European Green Deal [1], the European Commission (EC) has set the ambitious objective of a carbon-neutral European Union (EU) by 2050, calling for decarbonisation of all sectors. At the end of 2020, the European Council accepted the EC proposal for a Climate Law [2] to increase the intermediate greenhouse gases (GHG) emission reduction target for 2030 to 55%.

The mobility system, in general, is responsible for nearly a quarter of Europe's greenhouse gas emissions and has so far proven difficult to decarbonise, with limited success in reducing GHG emissions and in shifting toward more sustainable solutions. This is true for the road transport sector in particular, where GHG emissions have increased by 27% since 1990. In 2017, road transport alone was responsible for almost 72% of total greenhouse gas emissions from transport (including international aviation and international shipping). Of these emissions, 44% were from passenger cars, 9% from light commercial vehicles, and 19% came from heavy-duty vehicles [3,4].

In December 2020, the EC published its 'Sustainable and Smart Mobility Strategy' [5], laying out its vision to ensure that the EU transport system can achieve a green transformation. This is the last of a long series of strategic documents and binding regulations demonstrating the commitment of the EU not only to a reduction in GHG emissions from the transport sector but also to a strict control of atmospheric pollutants impacting on air quality and human health. Indeed, the EU has introduced since the early 1990s a series of progressively more stringent emission regulations to control air pollution from the transport sector, commonly known as Euro standards. Detailed reviews on the topic have been recently published for the interested reader [6,7]. These standards had the net positive effect of significantly reducing different air pollutants. Indeed, the EEA reported a reduction in the following pollutants for the period between 1990 and 2017: carbon monoxide and non-methane volatile organic compounds (both by around 87%), sulphur oxides (66%), nitrogen oxides (40%), and since 2000 particulate matter (44% for $PM_{2.5}$ and 35% for PM_{10}). Emissions from road transport have continued to decrease (except emissions of sulphur oxides) in recent years. In 2017, emissions were lower than in the previous year: emissions of nitrogen oxides decreased by 3% and those of carbon monoxide by 3.2%, those of PM_{10} and $PM_{2.5}$ decreased by 1.4% and 3.6%, respectively. Emissions of sulphur oxides increased by 2.7% in 2017, compared with 2016, but it is still less than 1% of what was emitted in 1990 [4]. More recently, in the framework of the European Green Deal, the EC has indicated the intention to further review the current air pollutant emissions standards.

In this context, the share of battery electric vehicles (BEVs) and plug-in hybrid electric vehicles (PHEVs) is rapidly growing, reaching 7.6% and 8.6% of the new registrations in the Q1–Q3 2021 period. During Q3 2021, the registration of hybrid passenger cars accounted for 20.7% of the total EU market, becoming the second most popular powertrain option after gasoline in the EU [8]. Increasing the market penetration of such vehicles is seen as an important step toward effective decarbonisation of the mobility system [3,9].

PHEVs are of interest since they are expected to deliver a reduction in both gaseous pollutants such as NOx and greenhouse gases such as CO_2, also in challenging conditions such as low temperature and/or high altitude. However, in such conditions, improvements are not consistent for all pollutants, with, on the contrary, an increase in emissions with respect to conventional internal combustion engine (ICE) vehicles reported for HC, CO, and particle number [10,11]. PHEVs can encounter operative conditions previously unseen on traditional vehicles, e.g., the necessity to trigger the ICE under high-power demand conditions without a heated after-treatment system [12]. This can lead to significant emissions [13], even higher with respect to those registered in cold start for conventional vehicles.

For this reason, in this work, we explored the environmental performance (i.e., performance for both pollutants and GHG emissions) of three modern PHEVs, all types approved according to the Euro 6d-ISC or Euro 6d-TEMP-EVAP-ISC provisions. They were tested both in the laboratory and on-road for regulated and unregulated pollutants, including low-temperature conditions. Herein, only gaseous emissions will be discussed: a detailed investigation on solid particle number (SPN) emissions will be presented in a follow-up work.

The results herein reported explore in which conditions the adoption of PHEVs can contribute to the decarbonisation and the zero pollution ambitions set out by the EU for the next decades, highlighting at the same time some areas of concerns for such vehicles, in relation to specific operative modes.

2. Methods

2.1. Vehicles and Tests

Three plug-in hybrid gasoline fuelled vehicles meeting the Euro 6d-ISC or Euro 6d-TEMP-EVAP-ISC standards were tested at the Joint Research Centre (JRC) vehicle emissions laboratories (VELA 8) of the European Commission. All vehicles were tested over the type-approval Worldwide harmonised Light-duty vehicles Test Cycle (WLTC) at 23 °C and −10 °C, two of them were also tested at 40 °C. Additionally, all of them were also tested on both a route compliant to the real driving emissions (RDE) regulation (RDE from now on) and a non-compliant (motorway from now on) one. The motorway route is constituted by a predominant section of high-speed motorway, punctuated with sequences of decelerations and accelerations to simulate overtakes. This mimics what is done in the ADAC (Allgemeiner Deutscher Automobil-Club e.V.) motorway cycle for laboratory testing described elsewhere [14] (also known as BAB 130, where BAB stands for Bundesautobahn, i.e., federal motorway). Speed profiles and altitude profiles, when relevant, together with the main features of the used cycles, can be found in Figure S1 and Table S1 in the SI. An exhaustive list of the performed tests can be found in Table S2 in the SM.

The vehicles tested were characterised by plug-in hybrid powertrains where the internal combustion engine featured a gasoline direct-injection technology. The after-treatment system of all vehicles tested included a three-way catalyst (TWC) to control gaseous pollutants and a gasoline particulate filter (GPF) to control particulate emissions. Table S3 in the SI summarises the main characteristics of the vehicles tested.

2.2. Laboratory Set-Up

Laboratory tests were performed at the VELA 8 testing facility. This is a 4WD chassis dynamometer climatic test cell with controlled temperature and relative humidity. The characteristics of the climatic test cell are described elsewhere [11,15]. For all the tests conducted, all regulated gaseous emissions (CO, HC, NMHC, and NOx) were measured from the full dilution tunnel in real-time using an AMA i60 bench (AVL, Graz, Austria). Unregulated gaseous emissions (NH_3, N_2O, HCHO, CH_4, aromatics, <C_5 and C_5 hydrocarbons) were measured using a SESAM i60 FTIR spectrometer from AVL. The system was equipped with a spectrometer (Nicolet Antaris IGS Analyser—Thermo Electron Scientific Instruments LLC, Madison, WI, USA), a multi-path gas cell with a 2 m optical path with a working pressure of 860 hPa, a downstream sampling pump (6.5 L/min sampling rate) and had the acquisition frequency of 1 Hz. The instrument was connected to the exhaust tailpipe with a heated polytetrafluoroethylene sampling line at 191 °C. The exhaust flow rate was determined by subtracting the flow of dilution air introduced into the tunnel, measured with a Venturi system, to the total flow of the dilution tunnel, measured by a critical flow Venturi. Mass emissions of unregulated pollutants were derived from the exhaust gas flow rates (m^3/s) and from the measured concentration (ppmV). The emission factors (mass per distance units) presented at 23 °C over the WLTC correspond to the average of at least one test per vehicle performed at the same temperature and over the same cycle. These were tested in both charge-depleting and charge-sustaining modes, i.e., starting with the battery at different states of charge (SOC, namely 100%, 50%, 25%, and minimum). The default user-selectable mode was always used for the tests in the laboratory, with the only exception of the tests performed in "battery charging" mode, i.e., using the internal combustion engine to recharge the electric battery. This default mode can have different names depending on the vehicle (e.g., "comfort", "hybrid"), but for the cases tested is always the mode in between fully electric functioning and more dynamic/sport mode. Unless specified differently, laboratory tests were performed following the WLTP procedure and, in particular, option 3 of the regulation. This requires the execution of consecutive tests, starting with the cold vehicle with the high voltage (HV) battery at maximum SOC until the brake-off criteria are reached (as defined in EU regulation 2017/1151 [16]) (these are called charge-depleting tests, CD from now on). This is followed by a cool-down period (6 to 36 h) and, subsequently, a charge-sustaining (CS) test is performed.

2.3. On-Road Set-Up

On-road tests were also performed at the JRC, using different portable emissions measurement systems (PEMS). In particular, the measurement of the instantaneous on-road emissions of NOx, CO, PN, and CO_2 was performed using either an AVL MOVE system (AVL, Graz, Austria) or a HORIBA OBS ONE system (Horiba, Kyoto, Japan). Both PEMS used consisted of a tailpipe attachment, an exhaust flow meter (EFM), heated exhaust sampling lines, exhaust gas analysers, a data logger connected to the vehicle on-board diagnostic (OBD) port, a GPS, and a weather station for ambient temperature and humidity measurements. The AVL MOVE system measured exhaust gas concentrations of CO and CO_2 by a non-dispersive infrared sensor and NOx by a non-dispersive ultra-violet sensor. The HORIBA OBS ONE system measured exhaust gas concentrations of CO and CO_2 by a heated non-dispersive infrared sensor and NOx by a heated chemiluminescence detector. Both systems were equipped with EFMs with a Pitot tube to estimate the exhaust mass flow rate. All relevant emissions data were recorded at a frequency of 1 Hz. The gas analysers were all kept inside the vehicles and connected to the mains during the soaking period. The emission factors (mass per distance units) or the absolute emissions (in mass units) presented for the on-road tests correspond to the average of at least one test per vehicle, performed in conditions as reproducible as possible. As for the laboratory tests, vehicles were tested in both charge-depleting and charge-sustaining modes, i.e., starting with the battery at different states of charge (namely 100%, 50%, 25%, and minimum). The default user-selectable mode was always used for the tests on-road, with the only exception of the tests performed in "battery charging" mode, i.e., using the internal combustion engine to recharge the electric battery of the vehicle. The vehicles were generally soaked in a temperature-controlled area at 20 °C, with the only exception of vehicle #1 (see Table S2 in the SM for additional details) that was soaked outside at approximately 1 °C for a single test. On-road tests were generally performed according to the RDE test procedure (as defined in EU regulation 2017/1151 [16]), unless differently specified. Emission values are calculated integrating instantaneous emissions in g/s for the whole trip without any corrections for boundary conditions or CO_2 ratio.

In order to measure currently unregulated pollutants on-road, a portable FTIR instrument installed in parallel to a conventional PEMS was used for two of the tested vehicles (namely vehicle #1 and #2, see Table S3 in the SI for additional details). The instrument (PEMS-LAB from CERTAM and ADDAIR, Saint-Étienne-du-Rouvray, France), p-FTIR in the following, is equipped with: a portable FTIR, a Pegasor Particle Sensor (PPS-M from PEGASOR, Tampere, Finland) for solid particle number (PN) measurement, an EFM module, a dedicated weather station for ambient temperature, pressure and relative humidity, a GPS module and a stand-alone energy supply battery. The portable FTIR includes a shock-resistant interferometer (spectral resolution of 8 cm^{-1}, spectral range: 900–4200 cm^{-1}), a mercury cadmium telluride (MCT) thermoelectrically cooled detector, a gas cell with an internal volume of 200 cm^3 (working at 180 °C and atmospheric pressure), and a fixed optical path of 2 m. The sampling line is made of polytetrafluoroethylene (PTFE), and it is heated to 220 °C. The instrument is the same previously used for the measurement of unregulated pollutants from a heavy-duty vehicle [17].

Additional schematics of laboratory and on-road experimental setups can be found in Figure S2 in the SM.

3. Results and Discussion

In this section, we discuss the emissions for regulated pollutants (Section 3.1) and how these are impacted by the ambient temperature (Section 3.2). In Section 3.3, this is similarly performed for unregulated pollutants, especially in the laboratory. Then, some peculiarity of PHEV and their impact on emissions of pollutants and GHG are discussed, including: repeated cold-start events during urban operation in (Section 3.4) and battery-charging operating mode (in Section 3.5).

3.1. Regulated Pollutants Emissions

3.1.1. Laboratory Tests

Figure 1 shows the results of emissions tests conducted in the laboratory at 23 °C, according to the procedure prescribed in EC regulation 2017/1151 [16] for PHEV vehicles. In particular, testing was performed following option 3 of the regulation as already detailed in the "Methods" section. The figure reports the charge-depleting test in which the internal combustion engine ignites for the first time. Incidentally, this was the third test for all the vehicles (CD3 from now on). This behaviour was expected due to the similar electric range of the three vehicles tested (52–61 km), as detailed in Table S3 in the SI, and given the length of the WLTC test (23 km ca.). The CD3 test can be considered a cold start, i.e., with a coolant temperature equal to the ambient one ±3 °C, due to full electric functioning in the previous two repetitions. For comparison, in the figure, a charge-sustaining test (CS from now on), i.e., a test with a minimum initial SOC of the HV battery, also in cold conditions, performed the day after was also reported. In addition, the results of the weighted emissions calculated according to the procedure reported in the currently in-force EU regulation [16], which considers the full set of charge-depleting and -sustaining tests performed for each vehicle, were also inserted for comparison purposes. Finally, for reference, both the Euro 6 regulatory limit (red dashed line) and also the average emissions (black dotted line) for the whole fleet of pure ICE (i.e., non-PHEV) gasoline direct-injection (GDI) cars tested in the 2018–2019 period at the JRC, in the framework of the market surveillance (MaSu) activity have been reported [18]. The tested fleet was composed of 8 ICE GDI vehicles, all types approved as Euro 6d-TEMP.

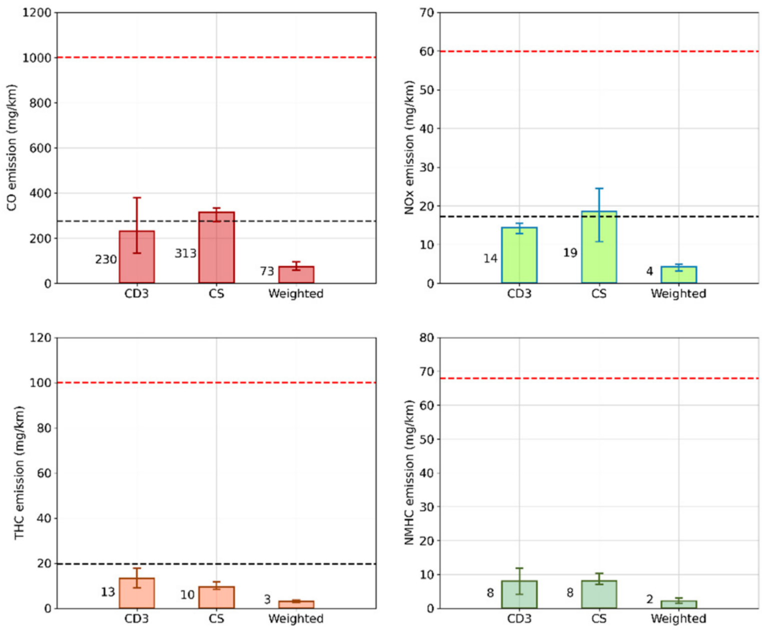

Figure 1. Regulated emissions on WLTC tests at 23 °C, averages of the three vehicles. CD3 indicates the third cycle in the charge-depleting sequence (for all vehicles, this was the cycle in which the ICE ignited for the first time). Started with an initial battery state of charge (SOC) equal to 100%; CS indicates the cold-start charge-sustaining cycle performed after the battery depleting sequence, i.e., starting with the minimum SOC. Weighted indicates emissions calculated according to the procedure reported in the currently in-force EU regulation [16]. Red dashed lines represent the Euro 6 regulatory limits, black dashed lines represent average emissions from the fleet of gasoline direct-injection vehicles tested at the JRC in the 2018–2019 period, in the framework of the MaSu activity on the same cycle [18]. All tests were performed in the default user-selectable mode of operation. Error bars on the graph represent maximum and minimum values. See the Methods section for additional details.

All the vehicles met the Euro 6 regulatory limit in the WLTP. CO emissions increased from an average of 230 mg/km to up to 313 mg/km when comparing CD3 and CS tests. In this case, it is evident the effect of dilution given by the fact that part of the CD3 is still driven in electric mode. Interestingly, in CD3 tests, some vehicles presented relatively high variability with emissions, e.g., for CO, in some cases even higher than those recorded on the CS. This was mostly related to relevant emissions during the cold-start events. Although such cold starts occurred at a later stage, with respect to those of the best-behaved vehicle (vehicle #2), as evident from Figure S3 in the SI, the additional mileage driven in electric mode was not enough to compensate for the severity of the event.

Finally, when the vehicles operated in CS mode, e.g., once their electric range was depleted, CO emissions were in line with those recorded for conventional gasoline vehicles with the same injection technology, namely 275 mg/km. CO weighted emissions were much lower and in the order of 73 mg/km.

NOx emissions were similar for both CD3 and CS tests and, respectively, 14 and 19 mg/km on average. Although CO performances varied significantly in CD tests, NOx performance was similar, indicating that it is possible to achieve suitable control in both NOx and CO emission with proper optimization. Additional details can be found in Figure S3 and Table S4 in the SI. In addition, in this case, CS operation was in line with emissions recorded for conventional GDI vehicles, namely 17 mg/km, and weighted emissions were very low and in the order of 4 mg/km.

Similar to NOx, both THC and NMHC emissions were low for all the tests, and there was no significant difference between CD3 and CS tests. Again, weighted emissions were also low and equal to 2–3 mg/km.

3.1.2. On-Road Tests

Figure 2 shows the results of emissions tests performed on-road, within the boundaries of the regulation [16], and on the same RDE compliant route. In particular, for each vehicle, charge-depleting tests (CD) with initial HV battery SOC equal to 100% (CD100), 50% (CD50), and 25% (CD25) were performed, as well as a test in charge-sustaining operation, i.e., with minimum initial SOC. For all the tests, the default user-selectable mode was used, with the notable exception of a single CD50 test for vehicle #1 (see Table S2 in the SI for additional information) in which the "full electric" mode was used. In this modality, the vehicle privileged electric operation over the use of the internal combustion engine until the SOC of the HV battery allowed for it. All the tests were performed in cold conditions, i.e., with a coolant temperature at the beginning of the test equal (±3 °C) to one of the soak area from where the test was started (ca. 20 °C). In addition to the NOx, which is regulated on-road, CO emissions were reported to allow a comparison with the regulated emissions in the laboratory.

Although well below the Euro 6 regulatory laboratory and RDE limits, on-road, the average performances of the vehicles were subpar when compared to conventional gasoline direct-injection vehicles. Indeed, both CO and NOx emissions were in line or above the average urban or total emissions of conventional gasoline direct-injection vehicles tested by the JRC in the framework of the market surveillance activity during the 2018–2019 period, namely 158 and 128 mg/km for CO and 12 and 9 mg/km for NOx [18]. This means that, at least for some vehicles in the sample tested, the expected gain in emission due to the presence of a hybrid gasoline-electric traction seemed limited, even in the shorter urban phase (36 km) and with an initial SOC equal to 100%.

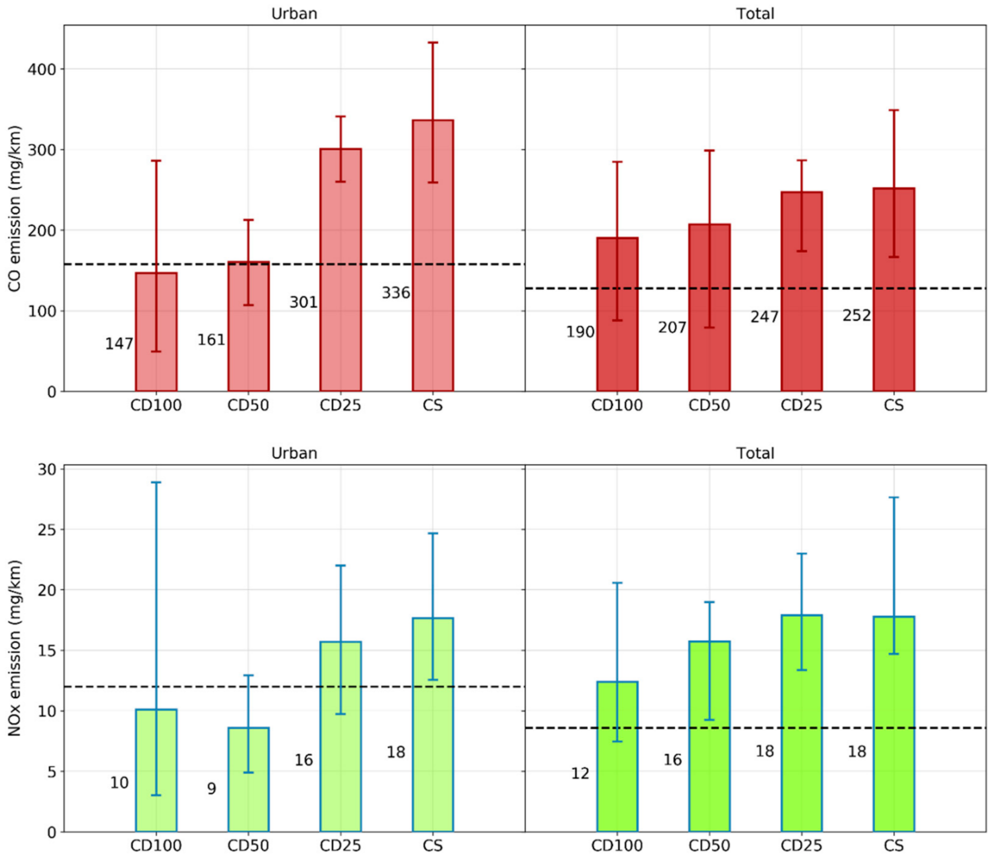

Figure 2. Regulated emissions on RDE compliant tests, averages of the three vehicles. CD100/50/25 indicates charge-depleting operation with initial battery state of charge (SOC) equal to 100/50/25%. CS indicated charge-sustaining operation, i.e., starting with the minimum SOC. Left panels report emissions for the urban phase, right panel for the complete test. Black dashed lines represent average emissions from the fleet of gasoline vehicles tested at the JRC in the 2018–2019 period in the framework of market surveillance [18]. All tests were performed in the default user-selectable mode of operation with the notable exception of a single CD50 test for vehicle #1 (fully electric mode). Error bars on the graph represent maximum and minimum values. See the Methods section and the Supplementary Materials for additional details.

Urban CO emissions were 147, 161, 301, and 336 mg/km for initial SOC equal to 100%, 50%, 25%, and minimum, respectively. They reach up to 190, 207, 247, and 252 mg/km when considering total emissions instead. The situation is qualitatively similar for NOx, both in the case of urban or total emissions. In particular, urban emissions were, respectively, 10, 9, 16, and 18 mg/km, increasing up to 12, 16, 18, and 18 mg/km in case total emissions were considered. Interestingly, also NOx emissions were always well above and almost twice those recorded for conventional direct-injection gasoline vehicles, in particular in charge-sustaining tests. Nevertheless, the absolute levels were low.

Generally speaking, the tests showed fair reproducibility, considering the intrinsic variability involved in testing on-road, with the only notable exception of NOx emissions for the CD100 test. This is due to a single vehicle (vehicle #1) with highly variable performances and up to approximately four times higher with respect to the others. Additional details can be found in Figure S4 in the SM.

3.2. Effect of Ambient Temperature

3.2.1. Laboratory Tests

Figure 3 shows the results of emissions tests conducted in the laboratory at 23, −10, and 40 °C, on the WLTC and for different initial SOC. In particular, in this specific case, CD indicates the first cycle in which the internal combustion engine ignited during a charge-depleting sequence similar to the one foreseen in EC regulation 2017/1151 [16] for PHEVs. For tests at 23 °C, the first ignition occurred during the third cycle, as already discussed in section "Regulated pollutants emissions" above. At −10 °C, for all the vehicles, the engine ignited during the first charge-depleting cycle despite a fully charged HV battery (SOC 100%). Note that at 40 °C, only vehicles #2 and #3 were tested and solely in charge-sustaining conditions. Emissions at 23 °C were the same as those reported in Figure 1 above and were included just to ease the comparison.

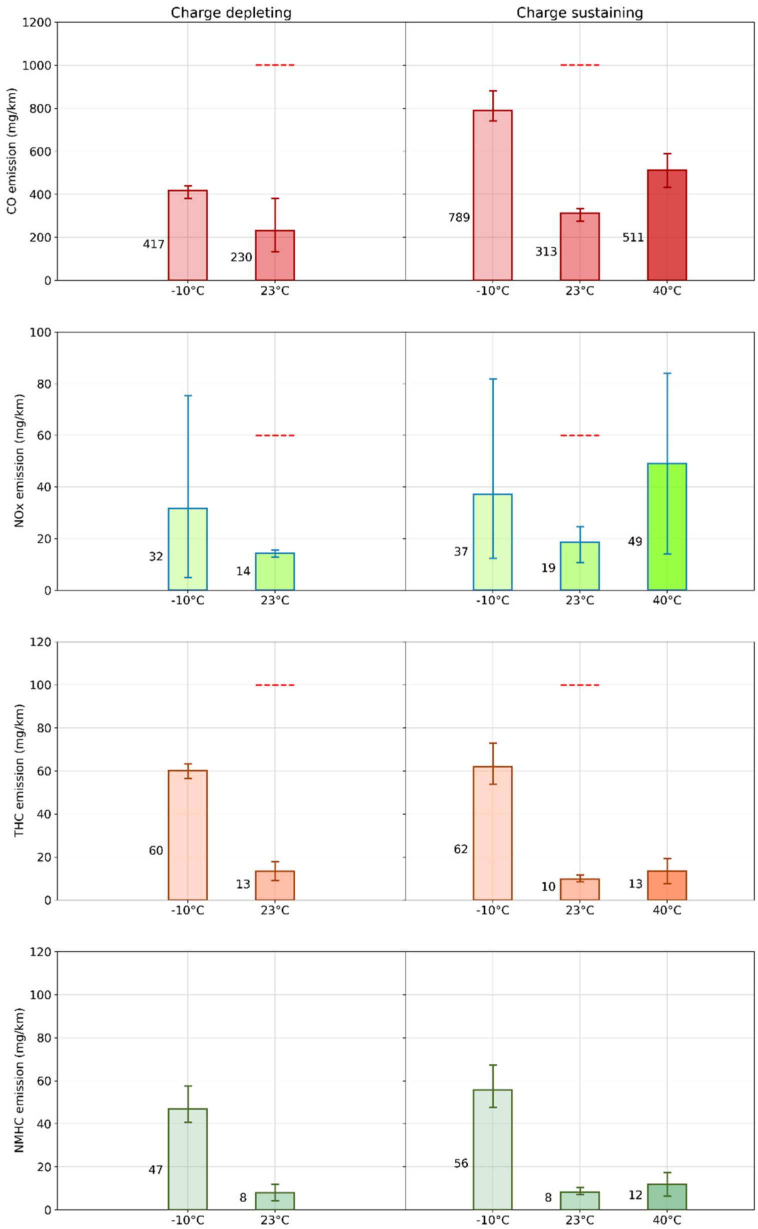

Figure 3. Emissions on WLTC tests as a function of temperature. Emissions at 23 °C are the same as those reported in Figure 1 above and have been included just to ease the comparison. CD indicates the cycle in the charge-depleting sequence, started with an initial battery state of charge (SOC) equal to 100%, in which the internal combustion engine ignited for the first time (3rd at 23 °C, 1st at −10% °C); CS indicates the cold-start charge-sustaining cycle performed after the battery depleting sequence, i.e., starting with the minimum SOC. All tests were performed in the default user-selectable mode of operation. Error bars on the graph represent maximum and minimum values. See the Methods section for additional details.

For CD tests, emissions of all pollutants considered were significantly higher at low temperature, with increments ranging from two times up to almost six times depending on the pollutant considered. CO emissions at −10 °C were 417 and 789 mg/km for CD and CS test, respectively, approximately twice as much as those recorded at 23 °C. The behaviour of NOx at low temperature was not very reproducible among the different vehicles, with a significant scatter with respect to the average values of 32 and 37 mg/km for CD and CS tests, respectively. Indeed, the worst-performing vehicle emitted 75 and 82 mg/km in the same tests while, on the contrary, for the best performing one, emissions were substantially lower, namely 5 and 12 mg/km. In general, vehicle #2 showed the highest emissions for both CO and NOx, while vehicle #3 was slightly better performing with respect to vehicle #1 in CD tests due to better management of the electric range. This can be seen in more detail in Table S4 and Figure S5 in the SM. Reproducibility was much better in the cases of THC and NMHC, with average low-temperature emissions of up to approximately 60 and 50 mg/km, respectively, in both CD and CS tests. Notably, also at low temperatures, no large differences existed for such pollutants between CD and CS operations, as already explained in the "Regulated pollutants emissions" section above.

This worsening of the performances at low temperature is in line with literature data [11,15,19] and mostly due to a slower heat up of the catalytic converter, with the increased importance of the cold-start events, as also evident from Figure S5 in the SM. Moreover, it is worth noticing that at such low temperatures, even the modern PHEVs tested did not operate in electric mode during this critical cold-start phase, despite a fully charged HV battery.

At high temperatures, both CO and NOx emissions increased. NOx emissions were even higher with respect to those recorded at low temperatures. This effect is not new [15,20] and was possibly more related to issues in engine calibration rather than to the performances of catalytic converters. In particular, for CS tests, CO emissions were 511 mg/km with an increase in approximately 60% with respect to 23 °C. NOx emissions increased up to 49 mg/km, more than twice those recorded at 23 °C, although again with high variability among vehicles.

3.2.2. On-Road Tests

Figure 4 shows the effect of the overnight soaking temperature on emissions tests performed on-road, within the boundaries of RDE regulation [16], and on the same route. This was possible only for one vehicle that was tested during winter (i.e., vehicle #1, see Table S3 in the SM for additional information). In particular, a CD100 test, i.e., with initial SOC of the main vehicle battery equal to 100%, was repeated twice: (i) performing the overnight soak outside, at an average temperature in the order of 1 °C; (ii) using the conventional soak area at controlled temperature (namely 20 °C). Both tests were performed in "full electric" mode of operation, which should privilege the electric traction over the conventional one. The average ambient temperature in the urban phase during the soak out test was approximately 1 °C, while during the soak in test was 12 °C.

The overall emissions were, respectively, 18 and 9 mg/km for NOx and 242 and 287 mg/km for CO when comparing the inside and the outside soak, respectively. Urban emissions were instead 17 and 5 mg/km for NOx and 211 and 298 mg/km for CO in the above-mentioned cases. Similar to what occurred in the laboratory but also in much milder conditions, the vehicle was not able to start in electric mode at a temperature of 1 °C when soaked outside, despite the fully charged battery and at variance with what occurred at warmer temperatures.

NOx cold-start emissions are comparable in the two cases, as shown by both emissions profiles and cumulative emissions at 0 and 2000 s approximately. The situation is very different for CO in which the cold start in the case of outside soak, occurring at test start, is significantly more important (7.18 vs. 1.42 g in the first 150 s after the first ignition of the ICE).

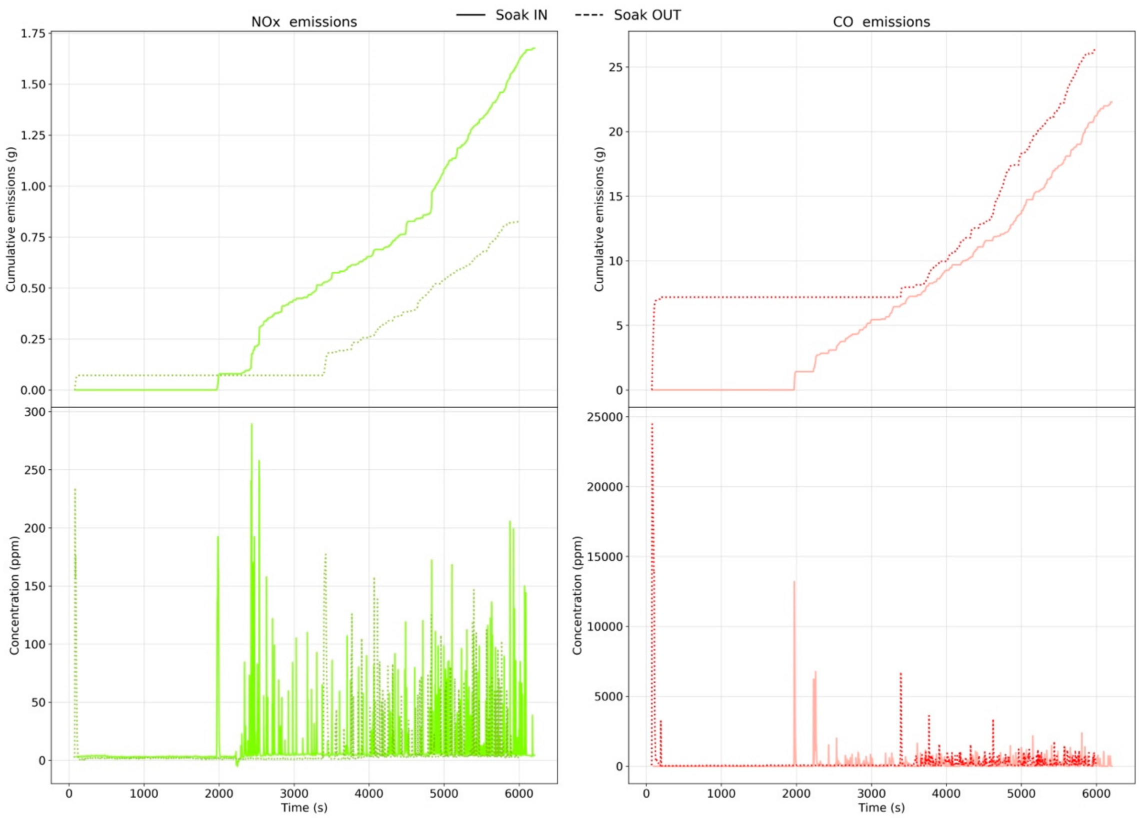

Figure 4. NOx and CO emissions on RDE compliant tests. Both tests were performed with an initial state of charge (SOC) equal to 100% and in "full electric" mode of operation. Solid lines refer to a test performed after soaking the vehicle in a closed area at 20 °C, dashed lines refer to a test performed after a soaking performed outside at 1 °C ca. See the Methods section for additional details.

Interestingly, also, the time of operation in electric mode after this first cold start is higher with respect to what occurs in the test with the soak performed inside. This might be related to different factors: (i) initial ignition of the ICE might have preserved part of the electric range for a subsequent part of the cycle; (ii) not reproducible driving (e.g., due to traffic, dynamicity); (iii) not reproducible ambient conditions (e.g., ambient temperature).

3.3. Emissions of Unregulated Pollutants

3.3.1. Laboratory Tests

Figure 5 shows the results of tests conducted in the laboratory at 23, −10, and 40 °C on the WLTC and for different initial SOC. Emissions of NH_3, N_2O, HCHO, CH_4, aromatics, small hydrocarbons (<C_5), and C_5 hydrocarbons were measured. However, being low (<2 mg/km) in all the different conditions tested, N_2O, alcohols, and HCHO will not be discussed further. As reported above in the "Effect of ambient temperature" section, CD indicates the first cycle in which the internal combustion engine ignited during the charge-depleting sequence. At −10 °C, for all the vehicles, the engine ignited during the first charge-depleting cycle despite a fully charged HV battery (SOC 100%). On the contrary, for tests at 23 °C, first ignition occurred during the third cycle, as already discussed in section "Regulated pollutants emissions" above. Note that for the three vehicles, no charge-depleting tests were performed in the laboratory at 40 °C, and only vehicles #2 and #3 were tested in charge sustaining at this temperature.

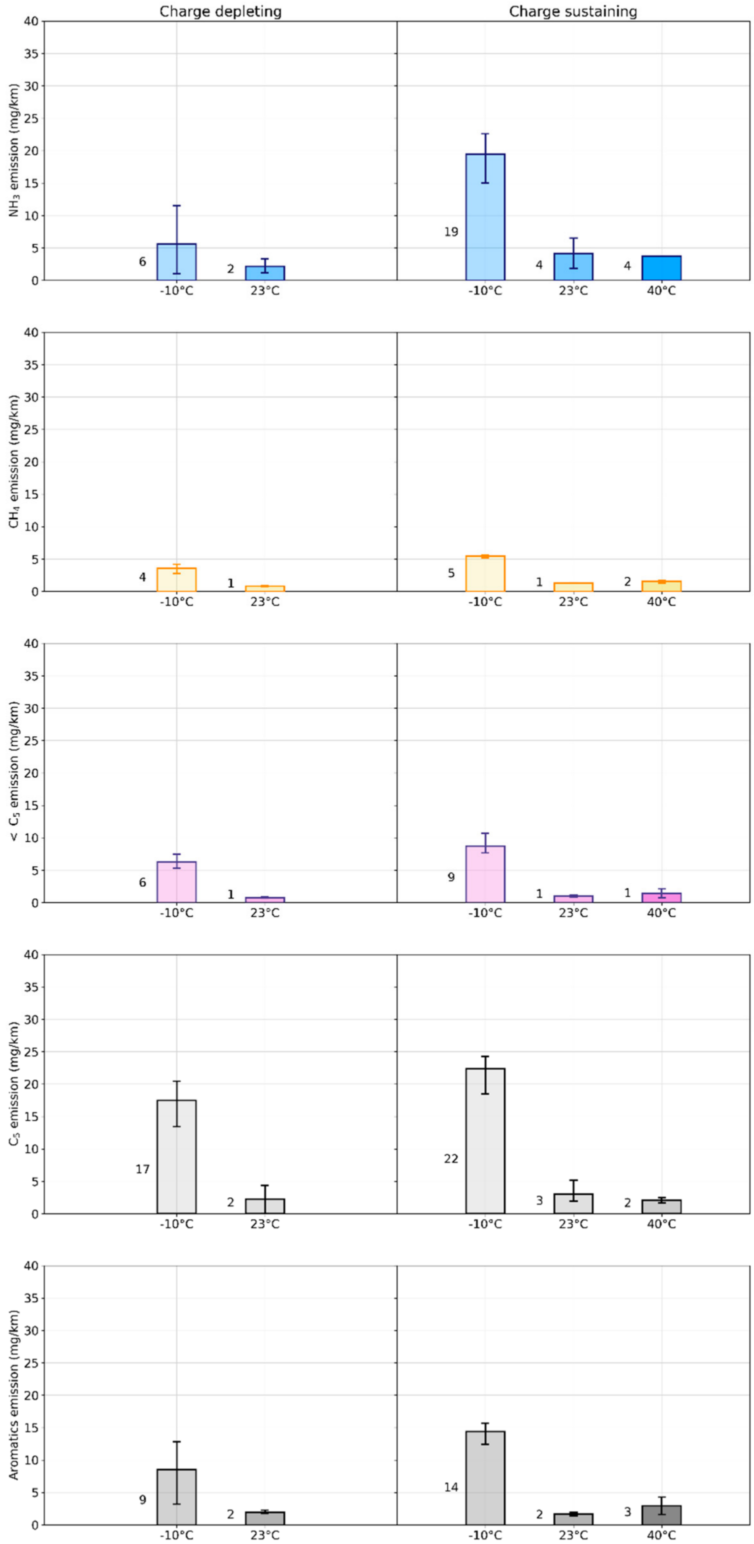

Figure 5. Emissions of unregulated pollutants (NH_3, CH_4, <C_5, C_5, and aromatics hydrocarbons) in WLTC tests as a function of temperature. CD indicates the cycle in the charge-depleting sequence, started with an initial battery state of charge (SOC) equal to 100%, in which the internal combustion engine ignited for the first time (3rd cyle at 23 °C, 1st cycle at −10 °C); CS indicates the cold-start charge-sustaining cycle performed after the battery depleting sequence. All tests were performed in the default user-selectable mode of operation. Error bars on the graph represent maximum and minimum values. See the Methods section for additional details.

Inspecting Figure 5 below, some general trends can be highlighted. Similar to what occurred for regulated pollutants, also, in this case, all emissions increased at low temperature both in CD and CS tests, with the only notable exception of NH_3 in CD tests that remained, however, at very low levels. Unregulated pollutants were very low or negligible at both 23 °C and 40 °C. For example, NH_3 average emissions were 4 mg/km, and C_5 hydrocarbons reached 3 mg/km both in CS tests at 23 °C [20,21].

The situation was significantly different at low temperatures. Hydrocarbons emissions were in the range of 10–20 mg/km, with C_5 hydrocarbons being the more abundant compounds (17 and 22 mg/km in CD and CS tests, respectively). NH_3 emissions also increased significantly: reaching, respectively, 19 and 6 mg/km in CS and CD tests. The results in Figure 5 below were in line with those reported in a recent study available in the literature [14]. Higher emissions at low temperatures (e.g., −7 °C) have been correlated to rich combustion, in particular during cold-start phases, to improve combustion performances and/or to overcome the poor mixing of the inlet charge as a result of the cylinder walls being cold [22,23]. CH_4 remained low at all the tested temperatures, with maximum emissions of 5 mg/km during CS tests at −10 °C.

3.3.2. On-Road Tests

Figure 6 shows the results of an emissions test performed on-road, on a route with a prevalent motorway section, driven dynamically and thus not compliant to the prescriptions of the RDE regulation [16]. Additional details on the road characteristics can be found in Table S1 in the SI. Although this test was routinely performed on all the tested vehicles, the portable FTIR instrument (p-FTIR) for the measurement of unregulated pollutants was only available for the motorway test on vehicle #2 (see Table S2 in the SM for additional information). Although the instrument is able to measure numerous different compounds, as detailed in the "Methods" section, the analysis in the following will focus on NH_3 emissions since it is the only additional pollutant, among those recorded, to be released in significant amounts for this vehicle. The test was performed in CS mode, i.e., with the initial SOC of the main vehicle battery equal to the minimum, selecting the default operation mode among the different ones selectable by the user. In addition, the test was performed in the afternoon and after a 3 h soak from the morning test; thus, the vehicle was not completely cold at test start. In the following, the specific case related to vehicle #2 will be discussed.

From the inspection of Figure 6, it is evident that the vehicle presented extremely relevant CO emissions, summing up to more than 200 g, or, in distance-specific units, more than 1000 mg/km (considering that the overall test length is approximately 186 km). Such emissions occurred coherently to acceleration events, as evident from the speed profile reported in gray. Notice that such accelerations had the purpose of simulating "normal" overtaking during motorway driving. Interestingly, considering an approximate global warming potential (GWP) of CO between 1 and 3 [24], this will lead to an additional CO_2 equivalent production of 1–3 g/km ca.

This behavior is very likely due to an auxiliary emissions strategy (AES), approved during the EU type-approval process and which allows using fuel enrichment to reduce the temperature of exhaust gas entering the catalyst when a threshold temperature is reached. For a more detailed discussion on AES and strategies for their identification, the interested reader could refer to the latest JRC MaSu report available in the literature [18].

Concomitantly with CO emissions, NH_3 emissions were also consistently recorded up to approximately 12 mg/km. This effect is not new and is again due to side reactions promoted on the three-way catalyst when burning rich mixtures in gasoline engines. Correlation between acceleration events, NH_3, and CO emissions in gasoline engines had been extensively studied in a recent work available in the literature for the interested reader [14]. It is also worth mentioning, as also detailed in Figure S6 in the SM, that all the vehicles performed in a qualitatively similar way, i.e., showing significant CO emissions concomitantly with acceleration events.

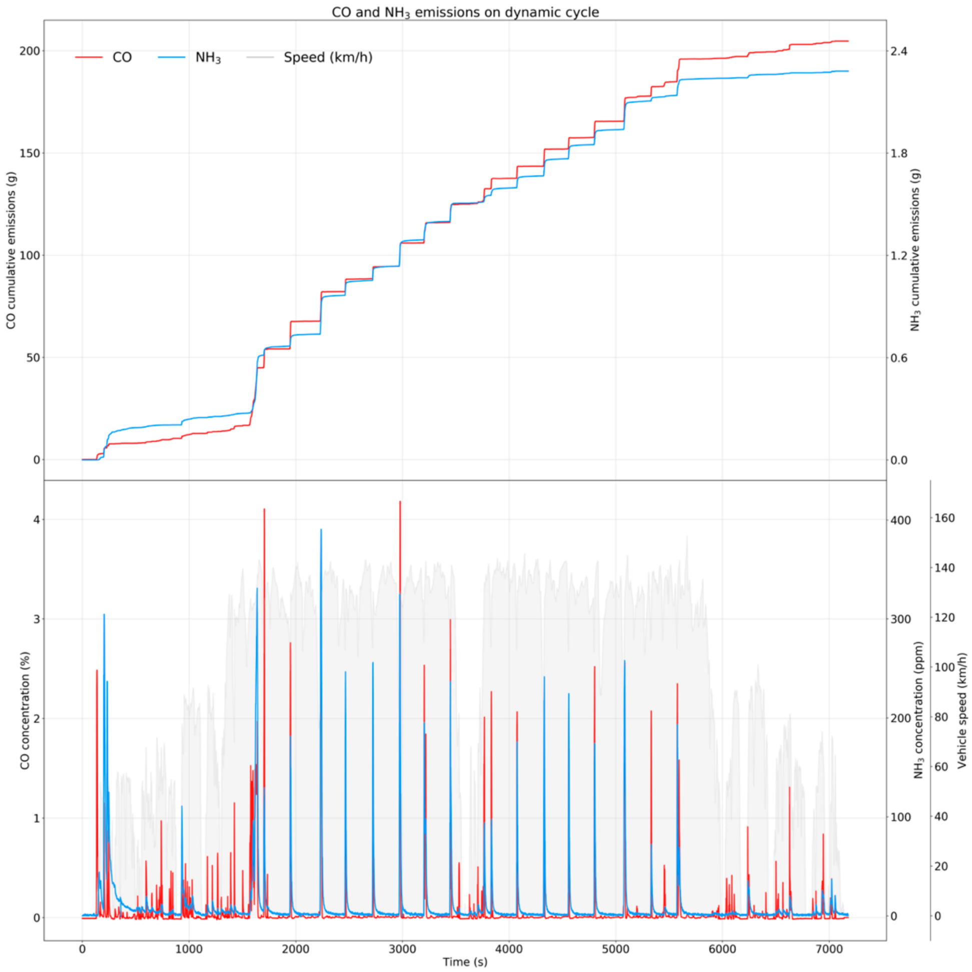

Figure 6. CO and NH_3 emissions on a dynamic motorway test on-road. The test was performed with an initial state of charge (SOC) equal to the minimum and the default user-selectable mode. Upper panel reports cumulated emissions of CO (red line) and NH_3 (blue line). Lower panel reports instantaneous tailpipe concentrations of CO (red line, measured with conventional PEMS) and NH_3 (blue line, measured with p-FTIR) as well as the speed profile of the cycle (gray). See the Methods section for additional details.

3.4. Emissions of Pollutants during Repeated Cold-Start Events

In general, PHEV combines both electric and conventional internal combustion engines, which can both contribute and cooperate to the displacement of the vehicle. This has as the net result that such vehicles have a much broader range of operative conditions with respect to conventional ones. In particular, in this section, some aspects related to the cold-start parts of the RDE tests conducted on-road will be explored. Normally, for conventional vehicles, it is possible to identify as cold start the initial period of time following engine ignition in which neither the engine nor the after-treatment system has reached their optimal operative temperature, after which emissions of noxious compounds should be properly controlled. This, however, is more complex in PHEV where the internal combustion engine could ignite, for example, at the beginning of the cycle for very few seconds and then reignite several times for very short periods before starting to operate continuously. In the following, at variance with what is currently indicated in the regulation, we have considered for the analysis a period, from now on called “cold operation”, of 300 s of functioning of the internal combustion engine, not necessarily consecutive. A fixed time interval was, in this case, preferred with respect to criteria based on coolant temperature in order to have perfectly comparable intervals across the different vehicles. In any case, most of these emissions occurred in the urban zone, being the maximum distance covered in the interval considered, also during charge-depleting tests, well aligned with the length of the urban segment (37 km on average with respect to the 35.5 km span of the urban segment).

Figure 7 below shows the NOx and CO cumulative emissions in mass units on tests performed on-road and within the boundaries of RDE regulation [16]. In particular, both CD100 tests, i.e., with the HV battery fully charged (SOC 100%) and CS tests, i.e., with the SOC at minimum. During cold operation, both CO and NOx emissions in CD100 were higher with respect to those recorded for the CS test, namely 4.164 and 0.324 g against 3.417 and 0.236 g, respectively. Interestingly, the variability was also high, with the best performing vehicles actually improving the cold operation performance, despite the electric functioning and the discontinuous operation of the internal combustion engine with repeated cold-start events. However, the worst-performing vehicles emitted more than twice the amount of NOx during cold operation with an initial SOC equal to 100% with respect to the case in which the battery was depleted. This behaviour, already reported in the literature [12], is of concern and require attention: PHEVs can, in some cases, have worse overall performances in their initial phases of operation with respect to conventional gasoline ones, in which a rapid heat up of the after-treatment system prevent the occurrence of repeated cold-start events. The only difference compared to conventional ICEs is that the "cold" start emissions are spread over a distance of 35 km instead of 1–2 km [15]. Additional details, including the emission profiles for the different cases explored, can be found in Figure S7 in the SM.

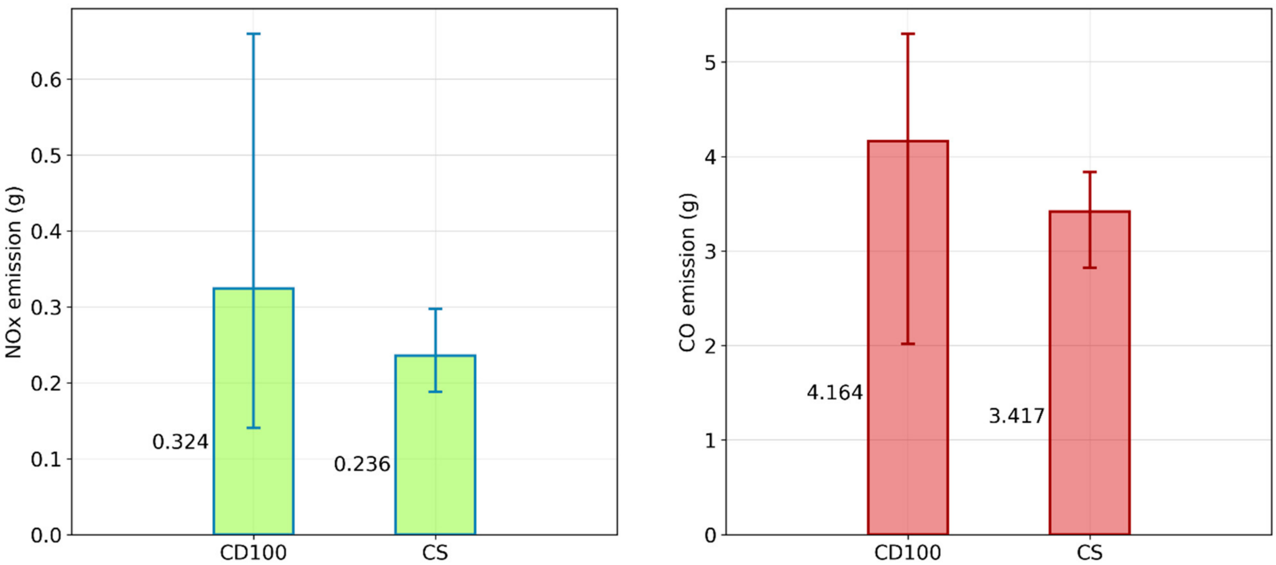

Figure 7. CO and NOx emissions on RDE compliant tests during the first cumulative 300 s of internal combustion engine operation, not necessarily consecutive. CD100 indicates charge-depleting operation with initial battery state of charge (SOC) equal to 100%; CS indicated charge-sustaining operation, i.e., starting with the minimum SOC. All tests were performed in the default user-selectable mode of operation. Error bars on the graph represent maximum and minimum values. See the Methods section for additional details.

3.5. Emissions of CO_2 as a Function of the Mode of Operation

Figure 8 shows the comparison of CO_2 emissions both in the laboratory (left panel) and on-road (right panel) for different operation modes of the PHEV. The on-road tests were performed according to the RDE regulation. This is not the situation for the laboratory, where the regulation prescribes the use of the default mode for this purpose [16]. In general, as already described, these vehicles could operate in a wide range of different modalities that can be either selected by the user or imposed by the SOC of the main battery. Specifically, charge-depleting operation indicates that the vehicle moves while drawing energy from the battery, the ICE, or both at the same time, while charge sustaining refers to a mode in which the SOC of the battery is minimal, and the vehicle moves mainly by using energy coming from the internal combustion engine, although some of this excess energy could be stored in the battery (e.g., after breaking) and thus some electrical traction could still occur. On top of this, the final user can (this was the case for all the three vehicles tested) select some mode of operation that tends to promote electric traction over conventional one (e.g., "electric mode"), promoting a use of the battery or a balanced mode (e.g., "comfort mode" or "eco mode") which is usually the default one. In addition to such modes, for the tested vehicles, a "battery charging mode" was present. In this modality, the vehicle used the internal combustion engine of the car to recharge the battery during normal driving. Such modality can be, for example, used to increase the electric range of the PHEV before entering the zero-emissions zone of a city and can even be used with the car parked in idling. It is important to note that the use of such a modality had a limited effect on the emission of regulated pollutants, which is the reason why they will not be discussed in the following.

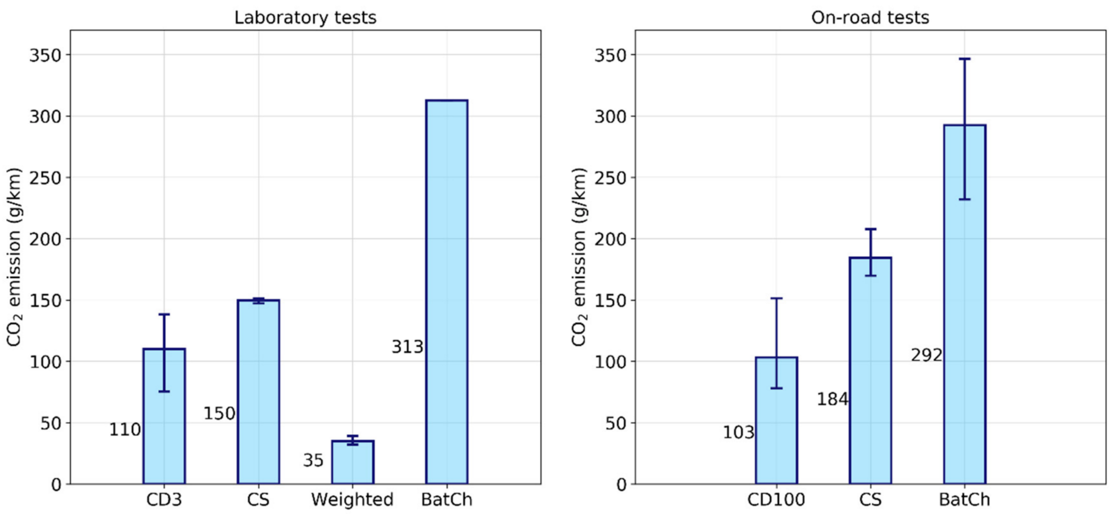

Figure 8. CO_2 emissions on laboratory (**left panel**) and on-road (**right panel**) tests performed according to the WLTP and RDE test procedures, respectively. CD100 and CS indicate tests with an initial state of charge (SOC) equal to 100% or the minimum, respectively. BatCh (battery charging) indicates the user-selectable mode of operation in which the internal combustion engine is used to charge the vehicle battery. Error bars on the graph represent maximum and minimum values. See the Methods section for additional details.

As expected, it is immediately evident how CO_2 emissions increased when moving from charge-depleting tests to charge-sustaining ones, both in the laboratory and on-road. For example, on the WLTC, CO_2 emissions during CD3 were 110 g/km and increased to 150 g/km during the CS test. However, the CO_2 calculated, taking into account also the cycles performed in full electric mode and weighted with the proper utility factors, following the provisions in [16], is 35 g/km. On-road, a CD100 test performed in the default mode, resulted in CO_2 emissions equal to 103 g/km, while a CS test, in the same default mode, up to 184 g/km. This is due to the limited amount of electric traction available in CS mode or, in other words, to the fact that in charge-depleting tests, the internal combustion engine operated for a more limited amount of time. Nevertheless, CO_2 almost doubled when the same CS test was performed in "battery charging" mode, both in the laboratory and on-road, with CO_2 emissions, respectively, equal to 313 g/km and 292 g/km. Importantly, battery-charging tests in the laboratory were performed only for vehicle #3. Vehicle #3 was generally well behaved with respect to others (both on-road and in laboratory), displaying, however, the worst battery-charging CO_2 emissions among the vehicles tested.

It is not the scope of this manuscript to perform the detailed calculations that would have been required to perform a comprehensive energy balance for the tested PHEV as a function of the operative conditions (e.g., accounting for CO_2 intensity of the generated electricity, losses over the network, CO_2 emissions related to fuel extraction, production, and distribution, just to name a few). However, it still important to notice how relevant is the impact of user-selectable modes on the CO_2 emissions of such vehicles.

4. Conclusions

Three plug-in hybrid gasoline fuelled vehicles meeting the latest vehicle emissions standards, Euro 6d or Euro 6d-TEMP, were tested at the vehicle emissions laboratories (VELA 8) of the Joint Research Centre (JRC) of the European Commission. All vehicles were tested over the type-approval Worldwide harmonised Light-duty vehicles Test Cycle (WLTC) at 23 °C, and −10 °C, two of them were also tested at 40 °C. In VELA 8 regulated gaseous emissions (CO, HC, NMHC, and NOx) and a series of unregulated gaseous emissions (NH_3, N_2O, HCHO, CH_4, aromatics, $<C_5$ and C_5 hydrocarbons) were measured. Additionally, the on-road emissions of NOx, CO, and CO_2 were measured with a PEMS. Unregulated pollutants were measured with a portable FTIR.

The results showed that while all tested vehicles met the Euro 6 emission limits in both the WLTP and RDE test procedures, their emissions were comparable to, and in some cases higher than, those reported for conventional gasoline vehicles of the same Euro standard and engine technology (direct injection), during on-road testing. On the contrary, in the laboratory, the weighted emissions for which the regulation requires to take into account a specific series of charge-depleting and charge-sustaining tests were well below those of conventional cars. Moreover, under certain operations, the presence of a hybrid gasoline-electric traction did not improve the vehicles' performance in terms of pollutant emissions even when using their full electric capabilities. Although the results were generally reproducible, in some conditions, variability occurred, mostly due to the different performance of the tested PHEV. The tested PHEVs presented higher emissions of all pollutants considered at cold temperature, including currently unregulated ones. The increase was registered not only in the laboratory but also on the road, where the vehicles were tested after being parked overnight outdoors in the winter period. In these cold conditions, the tested PHEVs were not able to start in electric mode, even with a fully charged HV battery. This resulted in significant emissions in cold start. The PHEVs presented high CO emissions during acceleration events. This behaviour was probably due to fuel enrichment. It was shown that these enrichment events also resulted in elevated NH_3 emissions during real-world operation of PHEVs. Other unregulated pollutants remained low during laboratory and on-road driving.

CO_2 emissions recorded during on-road tests showed significant variability depending on the PHEV operating modality. Emissions in charge-depleting operation were effectively lower with respect to those in charge-sustaining operation. However, certain user-selectable modes drastically increased CO_2 emissions, offsetting the gain obtained during PHEV electric operation. The additional contribution to global warming due to other substances that can be considered both greenhouse gases and air pollutants (e.g., N_2O or CH_4) was limited for the specific vehicles and tests performed. Summarising, for the sample tested, the expected reduction in emission of pollutants and CO_2 due to the presence of a hybrid gasoline-electric traction was highly variable and strongly dependent on the way of use of the vehicles. In particular, for CO_2, the modality selected by the end user had an influence. Especially for pollutants emission, the ignition of the internal combustion engine, even for a few km, compensated any benefit.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/en15072401/s1, Figure S1: Speed profiles for laboratory cycles. Examples of speed and altitude profiles for on-road routes, Figure S2: Experimental set-up used during the tests performed at VELA 8 laboratory (top) and on-road (bottom), Figure S3: Modal emissions on CD3 WLTC tests. CD3 indicates the third cycle in the charge depleting sequence (for all vehicles this was the cycle in which the ICE ignited for the first time), started with a battery initial State of Charge (SOC) equal to 100%. All tests were performed in the default user selectable mode. Figure S4: Modal NOx emissions on the urban part of RDE compliant tests for vehicle #1. Tests were performed with a battery initial State of Charge (SOC) equal to 100%. All tests were performed in the default user selectable mode. Figure S5: Modal emissions on CD WLTC tests as a function of temperature. CD indicates the cycle in the charge depleting sequence, started with a battery initial State of Charge (SOC) equal to 100%, in which the internal combustion engine ignited for the first time (different depending on the test temperature but the same for all vehicles); CS indicates the charge sustaining cycle performed after the battery depleting sequence, i.e., starting with the minimum SOC. All tests were performed in the default user selectable mode. Figure S6: CO on a dynamic motorway test on-road for the vehicle #1 (top figure) and vehicle #3 (bottom figure). The tests were performed with a depleted HV battery (minimum SOC) equal to the minimum. Upper panels in the figure report cumulated emissions of CO (red line). Lower panels reports instantaneous tailpipe concentrations of of CO (red line, measured with conventional PEMS) as well as the speed profile of the cycle (grey). Figure S7: CO and NOx modal emissions on RDE compliant CD100 test during the first cumulative 300 s of internal combustion engine operation, not necessarily consecutive. CD100 indicate charge depleting operation with battery initial State of Charge (SOC) equal to 100. All tests were performed in "hybrid" mode of operation. Table S1: RDE route details for selected examples. Table S2: List of performed experiments. Table S3: Tested vehicles. Table S4: Data summary.

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