



Article

# Emissions from a Modern Euro 6d Diesel Plug-In Hybrid

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Abstract: Plug-in hybrid electric vehicles (PHEVs) are promoted as an alternative to conventional vehicles to meet European decarbonisation and air quality targets. However, several studies have shown that gasoline PHEVs present similar criteria and particulate emissions as their conventional gasoline counterparts. In the present work, we investigate the environmental performance of a modern plug-in hybrid Diesel-fuelled vehicle meeting the Euro 6d standard under a large variety of driving patterns, ambient temperatures, and battery states of charge (SOC). Emissions of regulated pollutants, currently unregulated pollutants, and CO<sub>2</sub> were measured in the laboratory and following various on-road routes. The vehicle, whose electric range was 82 km, presented emissions below the Euro 6 regulatory limits in all the different driving cycles performed at 23  $^{\circ}\text{C}$  and all the on-road tests at the different battery SOC. The emissions were lower than the average of the conventional Diesel vehicles tested at JRC in 2020-2021 for all the SOC tested, the exception being solid particle number emissions >23 nm (SPN<sub>23</sub>) emissions that were comparable at all SOC. Moreover, the emissions obtained with the high voltage battery fully charged during on-road tests were comparable to those obtained with the battery at the minimum SOC for the entire test (ca. 91 km) as well as for the urban section (ca. 36 km). Overall, NOx and SPN23 emissions increased at lower temperatures, showing that at very low temperatures, there is no benefit in terms of particulate emissions from the electric range. Finally, it is shown that the emissions of N2O, the only unregulated pollutant presenting relevant emissions for this vehicle, and which are of catalytic nature, were proportional to the utilisation of the internal combustion engine. The scope of the manuscript is thus to deepen the knowledge on the emission performances of Diesel PHEVs through the systematic testing of a modern representative of this class of vehicles in a wide range of driving and environmental conditions.

**Keywords:** regulated pollutants; unregulated pollutants; greenhouse gas emissions; on-road vehicle testing; laboratory vehicle testing



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## 1. Introduction

The road transport sector has significantly reduced the emissions of criteria pollutants in recent years [1,2] with reductions of 88% for CO, 60% for NOx, 89% for NMVOCs, 99% for SOx, and 34% for PM $_{10}$  in the period 1990–2017. Still, road transport remains an important source of greenhouse gas (GHG) emissions and air pollutants in Europe [3]. Following the introduction of progressively more stringent pollutant emissions standards as well as the introduction of CO $_{2}$  fleet reduction targets for passenger vehicles, electrified vehicles, including plug-in hybrid (PHEVs) and pure electric vehicles, have rapidly increased their market share. In fact, in 2021 hybrid passenger cars became the second most popular powertrain in the European market, right after conventional gasoline-fuelled vehicles [4]. Due to the possibility to be driven fully electric, PHEVs are often considered to have lower pollutant and GHG emissions than their conventional counterparts. However, in a series of recent studies, we have shown that gasoline PHEVs present comparable criteria, particulate, and unregulated pollutant emissions to their conventional gasoline counterparts [5–7].

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These relatively high emissions, considering that these vehicles could run at least one third of the studied distance on full electric mode, resulted from triggering the ICE under high-power demand conditions without a heated after-treatment system or engine [8]. It was also shown that for gasoline PHEVs, the CO<sub>2</sub> emissions can vary largely depending on the PHEV user selectable modes used [5,9].

Most of the latest PHEV models available in the European market use a gasoline directinjection (GDI) engine. Thus, it does not come as a surprise that virtually all the studies that investigate the environmental performance (i.e., performance for both pollutants and GHG emissions) of modern PHEVs have focused on the gasoline version of this powertrain [5–9]. The studies of Diesel hybrid vehicles are limited and with older technologies with almost non-existent on-road or low temperature application [10–13]. Taking a step to fill this information gap, and to close a series of three studies dedicated to PHEVs, in this study we investigate the emissions of regulated pollutants (NOX, SPN<sub>23</sub>, HC + NOx, CO), currently unregulated pollutants (SPN<sub>10</sub>, NH<sub>3</sub>, N<sub>2</sub>O, HCHO, CH<sub>4</sub>), and CO<sub>2</sub> of a modern plug-in hybrid Diesel-fuelled vehicle meeting the Euro 6d-ISC-FCM standard under a large variety of real-world driving patterns and ambient temperature, as well as the potential impact of using a PHEV with different battery state of charge (SOC). The study also includes laboratory and on-road regulatory cycles and procedures currently in force in Europe.

Together with the other two recently published papers [5,14], the results presented describe a series of conditions under which modern PHEVs could contribute to the decarbonisation and the zero pollution ambitions set out by EU for this decade. At the same time, it also highlights areas of concern, mainly linked to the operation modes, that need to be improved for such vehicles.

#### 2. Methods

The emissions of a modern plug-in hybrid Diesel vehicle meeting the Euro 6d-ISC-FCM standard were investigated at the Joint Research Centre (JRC) vehicle emissions laboratories (VELA 8) of the European Commission, in a large variety of conditions. Specifically, it was tested according to the type-approval Worldwide harmonised Light-duty vehicles Test Procedure (WLTP), a duty cycle simulating congested urban traffic (Traffic for London, TfL cycle), and a dynamic motorway driving cycle (BAB 130 cycle, where BAB stands for Bundesautobahn, i.e., German federal motorway). In addition, a laboratory cycle replicating an on-road Real Driving Emissions (RDE) route ("labRDE" from now on) was also performed. All the tests with the exception of the WLTC were performed at four different temperature levels, namely:  $23 \,^{\circ}$ C,  $-10 \,^{\circ}$ C,  $5 \,^{\circ}$ C, and  $40 \,^{\circ}$ C. The vehicle was tested also on-road on a route compliant to the RDE regulation [15] ("RDE" from now on), a non-RDE compliant motorway route ("Motorway" from now on), and a non-compliant RDE route including repeated stops and idling, simulating congested urban driving ("City" from now on). The Motorway route includes section of high-speed motorway, punctuated with sequences of decelerations and accelerations to simulate overtakes. This mimics what is done in the BAB 130 motorway cycle during laboratory testing (developed by the Allgemeiner Deutscher Automobil-Club e.V., ADAC and also described elsewhere [16]). Speed 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 Supplementary Materials. An exhaustive list of the performed tests can be found in Table S2 in the Supplementary Materials.

The vehicle tested was characterised by plug-in hybrid powertrain where the internal combustion engine featured a Diesel engine. The after-treatment system included a Diesel oxidation catalyst (DOC), a selective catalytic reduction (SCR) catalyst, and an ammonia slip catalyst (ASC) to control gaseous pollutants and a Diesel particulate filter (DPF) to control particulate emissions. Table S3 in the Supplementary Materials summarises the main characteristics of the vehicle tested.

Laboratory tests were performed at the VELA 8 testing facility. This laboratory is a 4WD chassis dynamometer climatic test cell with controlled temperature and relative humidity. For all the tests conducted, all regulated gaseous emissions (CO, NOx, and

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HC) were measured from the full dilution tunnel using an AMA i60 bench (AVL, Graz, Austria). Unregulated gaseous emissions (NH $_3$ , N $_2$ O, HCHO, CH $_4$ ) were measured using a SESAM i60 FTIR spectrometer from AVL. Solid particle number (SPN $_{23}$ ) emissions from approximately 23 nm were measured with an advanced particle counter (APC 489) (AVL, Graz, Austria) connected to the full dilution tunnel. The system included an evaporation tube at 350  $^{\circ}$ C to remove volatiles. A 3010 condensation particle counter (CPC) (TSI, Shoreview, MN, USA) connected to the APC measured particles from approximately 10 nm (SPN $_{10}$ ). For the calculation of SPN $_{10}$ , a correction factor equal to 1.2 for additional losses in the 10 to 23 nm size range was used [17].

During the on-road tests, the instantaneous emissions of NOx, CO, SPN<sub>23</sub>, and CO<sub>2</sub> were measured using an AVL MOVE (AVL, Graz, Austria) Portable Emissions Measurement Systems (PEMS). On-road, unregulated pollutants were measured with a portable FTIR instrument (instrument (PEMS-LAB from CERTAM and ADDAIR, Saint-Étienne-du-Rouvray, France) used in parallel to the PEMS. The instrument was validated in a previous study [18].

The emission factors (mass per distance units) or the absolute emissions (in mass units) presented for the on-road tests are averaged if more than one test is available. Tests are performed in conditions as reproducible as possible, except when indicated differently. As for the laboratory tests, vehicles were tested 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, except for a few tests performed in fully electric mode clearly identified in the following. The vehicle was soaked in a temperature-controlled area at 20 °C. Unless differently specified, on-road tests were performed according to the RDE test procedure (as defined in EU regulation 2017/1151 [15]). Emission values were calculated integrating instantaneous emissions in g/s for the whole trip without any corrections for boundary conditions or CO<sub>2</sub> ratio.

WLTP was performed following option 3 of the EU regulation 2017/1151 [15]. Hence, the vehicle was tested in two different operation modes: charge depleting mode (CD) and charge sustaining (CS). CD testing is composed of a number of WLTC tests carried out with a fully charged battery until break-off criterion (relative electric energy change (REEC) < 0.04) is reached. At this point, the vehicle is at its minimum SOC. CS test is a test performed following the procedures used for the conventional vehicles at cold start conditions. Weighted emissions are calculated weighting the CD and CS operations using regulatory predefined Utility Factors (UF). These UF represent the ratio of the distance covered in CD mode to the total distance covered between two subsequent charges.

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

### 3. Results and Discussion

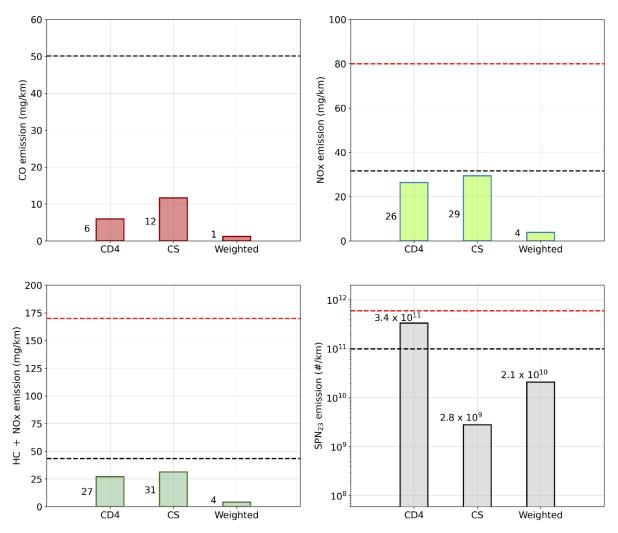
3.1. Emissions over Regulated Laboratory Cycles and On-Road Routes

# 3.1.1. Regulated Pollutants

Figure 1 shows the emissions of CO, NOx, HC + NOx, and SPN<sub>23</sub> recorded during emissions tests conducted in the laboratory at 23 °C and performed within the provisions of the WLTP included in EC regulation 2017/1151 [15] for PHEV vehicles. More specifically, testing was done following option 3 of the regulation as already detailed in "Methods" section. The figure reports the charge depleting test in which the internal combustion engine ignited for the first time which, for this specific vehicle, corresponds to the 4th test (CD4 from now on, engine ignited 11 km after test start). This behaviour was expected due to the electric range of the vehicle (82 km), as detailed in Table S3 in the Supplementary Materials, and given the length of the WLTC (23 km ca.). The CD4 test has been considered as cold, i.e., with a coolant temperature equal to 23  $\pm$  3 °C, due to fully electric functioning in the previous three cycles of the WLTP. For comparison, in Figure 1, a charge sustaining test, i.e., a test with a minimum initial SOC of the HV battery, also in cold conditions, performed the day after was also reported, together with the weighted emissions calculated

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in accordance with EU regulation [15] (see Section 2 Methods above). These emissions accounted for the whole sequence of charge depleting and sustaining tests performed. Finally, for reference, both the Euro 6 regulatory limit (red dashed line, 500 mg/km in case of CO, not reported) and the average emissions (black dashed line) for the whole fleet of pure ICE (i.e., not hybrid or PHEV) Diesel cars tested in the 2020–2021 period at the JRC in the framework of the Market Surveillance (MaSu) activity have been reported [19]. The tested fleet was composed of five vehicles, three type-approved as Euro 6d-TEMP and two type-approved Euro 6d.



**Figure 1.** Regulated emissions on WLTP at 23 °C. CD4 indicates the 4th cycle in the charge depleting sequence (i.e., the cycle in which the ICE ignited for the first time), started with a battery initial 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. Red dashed lines represent the Euro 6 regulatory limits (500 mg/km for CO not reported), black dashed lines represent average emissions from the fleet of Diesel vehicles tested at the JRC in the 2020–2021 period, in the framework of the Market Surveillance (MaSu) activity on the same cycle. All tests were performed in the default user selectable mode of operation. See Methods section for additional details.

The vehicle met the Euro 6 regulatory limits in the WLTP. CO emissions were generally low, even with respect to those of the reference fleet of conventional Diesel vehicles tested in MaSu, namely 50~mg/km. They varied between 6~mg/km and 12~mg/km in CD4 and CS, respectively. CO weighted emissions were almost negligible and in the order of 1~mg/km. Low CO emissions are expected for conventional Diesel cars and in line with experimental evidence in the specialised literature [19].

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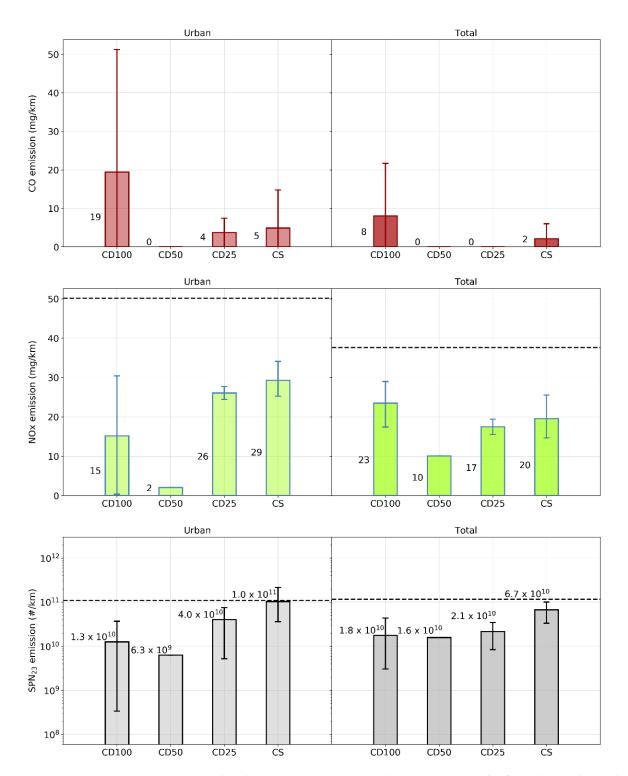
NOx emissions showed similar values between CD4 and CS tests, namely 26 mg/km and 29 mg/km, despite the different share of electrically driven kilometres in the two cases. Indeed, in CD4, the internal combustion engine ignited during the 3rd phase of the WLTC, after more than 11 km of full electric functioning. This indicates that, in this cycle, the cold start event is much more severe, and the additional mileage driven in electric mode is barely enough to compensate for it. Additional details are shown in Figure S3 in the Supplementary Materials. Interestingly, both CD4 and CS emissions are in line with those recorded for conventional Diesel vehicles, i.e., 31 mg/km. NOx weighted emissions were low and equal to 4 mg/km.

HC emissions were essentially negligible. Indeed, the main contributor to HC + NOx emissions was NOx and this was also always the case for tests performed in different conditions and on different duty cycles. For this reason, HC + NOx will not be discussed further in the following.

SPN<sub>23</sub> emissions in both CD4 and CS tests were below the applicable limit of  $6 \times 10^{11}$  #/km. In the CS cycle, the SPN<sub>23</sub> emissions were very low (2.8  $\times$  10<sup>9</sup> #/km) compared to the regulation limit and to the average SPN<sub>23</sub> emissions of the Diesel fleet used as reference and already presented. During the CD4 test, the thermal engine switched on only for 410 s out of 1800 s. Nonetheless, the vehicle emitted  $3.4 \times 10^{11}$  #/km, in line with the average of the Diesel vehicles tested at JRC in 2020–2021 [19]. These SPN<sub>23</sub> emissions were linked to the ignition of the vehicle. The thermal engine ignition occurred at a speed of ~97 km/h and the engine speed sharply increased to 2400 rpm. During the first 40 s after the ICE ignition, the vehicle emitted more than 80% of the total SPN<sub>23</sub> emissions showing the strong effect of the engine cold start in these demanding conditions. The appearance of such high concentrations during cold start of DPF-equipped vehicles has been attributed to higher engine out emissions, blow out of particles, and leaks through the DPF canister and mat. Recently, another assumption was reported for gasoline vehicles: these are heavy molecular species that are in the gaseous phase at the temperature of the DPF and pass unfiltered but became "solid" (non-volatile) at atmospheric conditions or at the 350 °C of the SPN measurement system [20]. This could be the case also for Diesel vehicles with the DPF installed close to the engine.

Summarising, this car showed a significant electric range resulting in very low weighted emissions in the WLTP. However, especially for NOx and  $SPN_{23}$ , when the engine ignited even for few km in the WLTC, this mostly overcomes the benefit of the electric driving due to the severity of the cold start event, resulting in similar or even higher emissions as compared to a CS test.

Figure 2 shows the emissions of CO, NOx, and SPN<sub>23</sub> recorded during on-road tests performed within the provisions of the RDE test procedure [15]. All the measurements were performed on the same RDE compliant route in conditions as reproducible as possible. In particular, 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: in this specific case, this was the one in between a fully electric functioning and the more dynamic, sport driving. For all tests, the coolant temperature at the beginning of the test was matching the one of the soak area (ca. 20  $\pm$  3  $^{\circ}$ C), namely they were performed in cold conditions. In addition to the NOx and SPN23 which are regulated on-road, CO emissions were reported to allow a comparison with the regulated emissions in the laboratory. Moreover, the average emissions (black dashed line) for the whole fleet of pure ICE (i.e., non-hybrid/PHEV) Diesel cars tested in the 2020–2021 period at the JRC in the framework of the Market Surveillance (MaSu) activity have also been reported [19]. The tested fleet was composed of nine vehicles, six type approved as Euro 6d-TEMP and three type approved Euro 6d.



**Figure 2.** Regulated emissions on RDE compliant tests. CD100/50/25 indicate charge depleting operation with battery initial 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 panels for the complete test. Black dashed lines represent average emissions from the fleet of Diesel vehicles tested at the JRC in the 2020–2021 period in the framework of Market Surveillance and are not reported for CO. All tests were performed in the default user selectable mode of operation. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

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In general, on-road emissions were below the Euro 6 RDE limits. CO emissions were 10 times lower than the laboratory CO limit. Urban CO emissions were 19 mg/km, negligible, 4 mg/km and 5 mg/km for initial SOC equal to 100%, 50%, 25% and minimum, respectively. They decrease to 8 mg/km, negligible values and 2 mg/km when considering total emissions instead.

Similarly, NOx emissions were lower than the laboratory NOx limit and the set of reference vehicles, i.e., 50 mg/km and 37 mg/km for the urban and total emissions, respectively. For NOx, urban emissions were respectively 15 mg/km, 2 mg/km, 26 mg/km, and 29 mg/km while 23 mg/km, 10 mg/km, 17 mg/km, and 20 mg/km in case total emissions were considered. NOx emissions were quite comparable for the RDE tests because the emissions are controlled by the SCR in a similar way independently of the battery SOC, being cold start the main contributing emissions.

Urban SPN<sub>23</sub> were  $\leq 1 \times 10^{11}$  #/km at all battery SOC levels. For battery levels 25–100%, the SPN<sub>23</sub> emissions were lower than the average Diesel fleet and one order of magnitude lower than the applicable limit. When the vehicle operated in charge sustaining mode, the SPN<sub>23</sub> emissions were near the average Diesel vehicles levels; but in all other cases lower. Note that for average Diesel vehicles levels, trips with DPF regeneration were also included as according to RDE4 regulation [15] if they comply with the limit these trips are considered valid. The tested vehicle did not regenerate during the entire campaign (approximately 580 km). A study that summarised distances between regeneration events reported ranges from <200 km to >800 km with a decreasing tendency for modern vehicles [21]. The contribution of regeneration can be significant to overall emissions and should not be neglected [20].

Interestingly, the electrical range of the vehicle was 82 km and the total length of the RDE route was 91 km ca. (see Table S1 for additional details) but, in the default user selectable mode used, the internal combustion engine ignited already in the urban phase, also in the case of a fully charged HV battery (SOC 100%). In addition, while in general the tests showed fair reproducibility, also accounting for the intrinsic variability involved in testing on-road, as already seen for gasoline PHEV [5], CD100 urban emissions showed a high variability, ranging from a fully electrical functioning with no tailpipe emissions, to a more conventional behaviour in line with standard Diesel vehicles. This underlines the importance of an effective management of Diesel and electric traction: avoiding high power or repeated engine ignition has a significant benefit in terms of environmental performance.

## 3.1.2. Unregulated Pollutants

Figure 3 shows the emissions of  $N_2O$ , currently unregulated, recorded during tests conducted in the laboratory at 23 °C and performed according to the WLTP [15] for PHEV vehicles. As already discussed, CD4 indicates the 4th charge depleting test in the WLTP sequence, i.e., the first cycle in which the engine ignited. Emissions were 5 mg/km and 9 mg/km in CD4 and CS tests, respectively. This was mostly due to the different utilisation of the internal combustion engine in the two cycles (approximately 12 km vs. 23 km). Indeed, at variance with other pollutants such as NOx, there is no significant release of  $N_2O$  during cold start which is mainly produced by unselective temperature-activated processes occurring in the after-treatment system. This is also in line with what was observed in a previous experimental campaign [22] and also reported in the specialised literature [23]. The  $N_2O$  emissions were slightly lower than the average that has been reported for SCR-equipped Euro 6 vehicles tested over the WLTC (~13 mg/km) [24].

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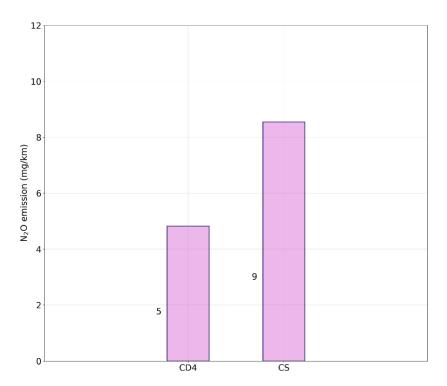
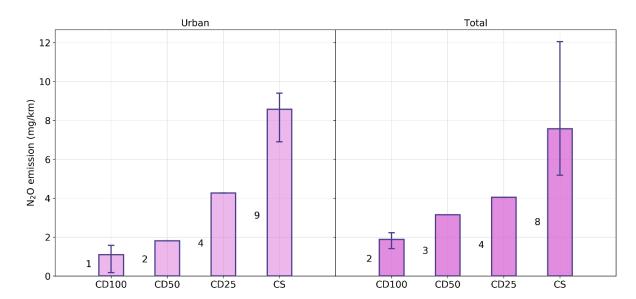


Figure 3. Emissions of  $N_2O$  in WLTC tests at 23 °C. CD4 indicates the 4th cycle in the charge depleting sequence (i.e., the cycle in which the ICE ignited for the first time), started with a battery initial state of charge (SOC) equal to 100%; CS indicates the cold start charge sustaining cycle performed after the battery depleting sequence, i.e., starting with the minimum SOC. See Methods section for additional details.

For this specific vehicle, no other relevant emissions of currently unregulated compounds, among those measured (NH $_3$ , HCHO, CH $_4$ ; see Methods for details), were recorded. This suggests good engine calibration (given the low emissions of HCHO and CH $_4$ ), and/or optimal after-treatment, DOC (in the case of HCHO and CH $_4$ ), and SCR + ASC (in the case of NH $_3$ ), design/operation/control. For this reason, in the following the discussion will focus on N $_2$ O only.

Figure 4 shows the emissions of N2O, currently unregulated, recorded during on-road tests performed within the provisions of the RDE test procedure [15]. Also in this case and similarly to what presented for laboratory tests, N<sub>2</sub>O was the only compound emitted by this particular vehicle in relevant amounts among those measured (see Methods for details), in the specific testing conditions explored. Results indicated that N<sub>2</sub>O emissions were again proportional to the utilisation of the ICE, increasing for the urban part of the trip from 1 mg/km to 2 mg/km, 4 mg/km, and up to 9 mg/km as a function of the initial SOC of the HV battery (100%, 50%, 25% and minimum, respectively). A quantitatively similar situation was recorded when the overall trip was considered, with emissions ranging from 2 mg/km up to 8 mg/km depending on the initial SOC. The three CS tests analysed include hot tests and one cold test, see Table S4 for more details. However, the high variability recorded is related to higher emissions in a single hot test. In this case and at variance with other pollutants (such as NOx), the moment in which cold start occurs had a minimum impact on the overall emissions. This is in line with the fact that N<sub>2</sub>O is normally a by-product of the modern Diesel after treatment systems (DOC/SCR/ASC) and not a main product of combustion, as already discussed. Additionally, the trend highlighted indicates that N<sub>2</sub>O once is formed is essentially not controlled by current catalytic devices [2] or, in other words and at variance with what happens for NOx, there is not a threshold temperature above which its abatement starts to occur.

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**Figure 4.** Emissions of  $N_2O$  in RDE tests. CD100/50/25 indicate charge depleting operation with battery initial 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 panels for the complete test. All tests were performed in the default user selectable mode of operation. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

## 3.2. Effect of Ambient Temperature on Emissions

Figure 5 shows the results of emissions tests conducted in the laboratory at  $-10\,^{\circ}$ C,  $5\,^{\circ}$ C,  $23\,^{\circ}$ C, and  $40\,^{\circ}$ C, on the WLTC and for different initial SOC. In particular, at  $-10\,^{\circ}$ C and  $23\,^{\circ}$ C, the full WLTP was performed (i.e., sequence of charge depleting and sustaining tests) while at  $5\,^{\circ}$ C and  $40\,^{\circ}$ C, only WLTC in charge sustaining tests were performed. Specifically, for this figure, charge depleting 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 [15] for PHEVs. This was the 4th cycle at  $23\,^{\circ}$ C and, despite a fully charged HV battery (SOC 100%), the 1st one at  $-10\,^{\circ}$ C. Emissions at  $23\,^{\circ}$ C were the same of those reported in Figure 1 above and were included just to ease the comparison.

For both CD and CS tests, emissions of all gaseous pollutants considered were significantly higher at lower temperatures, with increments up to more than 13 times depending on the pollutant considered with respect to 23  $^{\circ}$ C, with the notable exception of N<sub>2</sub>O. This is in line with previous literature studies [22] and with the fact that N<sub>2</sub>O in Diesel engines mostly originates from unselective catalytic reactions in the after-treatment system, as already discussed. These are mainly occurring in specific temperature windows (e.g., not during cold start) and thus are affected by the temperature at which the after-treatment system (ATS) is operating rather than the outside ambient temperature.

CO emissions at -10 °C were 22 mg/km and 44 mg/km for CS and CD test, respectively, hence, 2 to 7 times those recorded at 23 °C. Despite this increment, they were well below the Euro 6 limit, namely 500 mg/km. In the CS test at 5 °C and 40 °C, CO emissions were negligible.

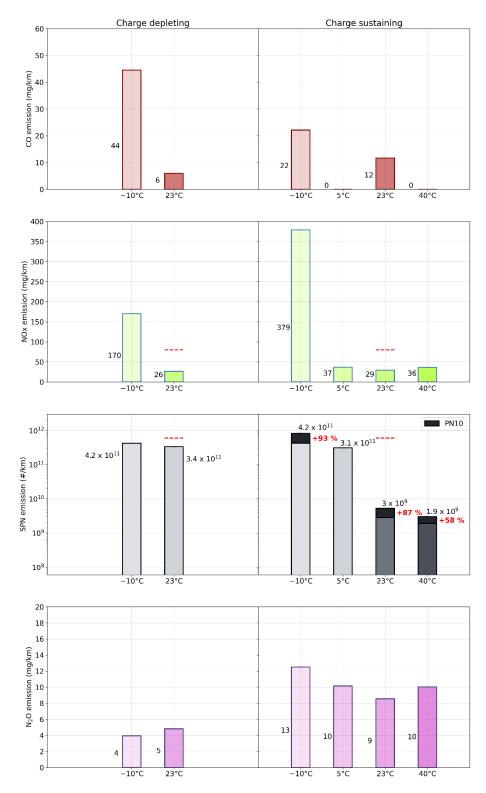


Figure 5. Emissions of regulated pollutants and  $N_2O$  on WLTC tests as a function of temperature. Emissions at 23 °C are the same of those reported in Figures 1 and 3 above and have been included just to ease the comparison. CD indicates the cycle in the charge depleting sequence, started with a battery initial state of charge (SOC) equal to 100%, in which the internal combustion engine ignited for the first time (4th 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. See Methods section for additional details.

NOx emissions at -10 °C for this particular vehicle were respectively more than 6 and more than 13 times higher for CD and CS with respect to those at 23 °C. It is worth noticing that, in the worst case, NOx emissions exceeded 379 mg/km. Similar results were also obtained in previous studies [6]. In the CS test at 5 °C, the overall NOx emitted were 37 mg/km, with an increase of approximately 30% with respect to the reference conditions. The behaviour of NOx at low temperature showed the typical trend already extensively discussed and reported in the specialised literature [5,7,21,25], with a sharp increase at sub-zero temperatures. This worsening of the performances at low temperature is in line with literature data [5,7,21,25] and mostly due to a slower heat up of the catalytic converter, with an increased importance of the cold start events, as also shown in Figure S5 in the Supplementary Materials. In this period of time, urban emissions are essentially uncontrolled. Moreover, it is worth noticing that at such low temperatures, even this modern PHEV did not operate in electric mode during this critical cold start phase, despite a fully charged HV battery.

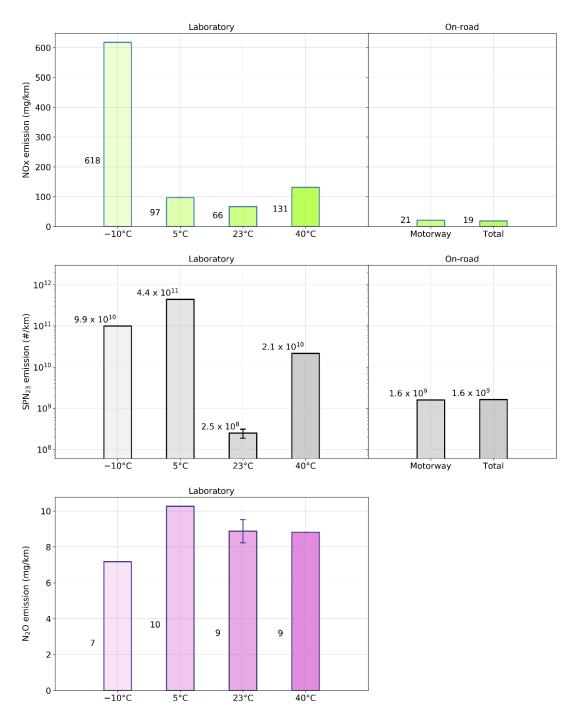
As already noted,  $N_2O$  emissions are barely affected by the ambient temperature during testing. Indeed, besides a slight increase in the CS test at  $-10\,^{\circ}$ C, where emissions reached 13 mg/km, in all other conditions, including higher temperatures,  $N_2O$  was in the order of 10 mg/km for CS tests and 5 mg/km in CD ones.

At high temperatures, NOx emissions increased while CO emissions remained negligible, at variance with what is normally observed in gasoline engines [5]. NOx emissions were 36 mg/km, with an increase of approximately 30% and incidentally similar to those recorded at  $5\,^{\circ}$ C. This effect is not new [21,26] and was possibly more related to issues in engine calibration rather than to the performances of catalytic converters.

SPN<sub>23</sub> emissions in the charge depleting tests were slightly higher at -10 °C compared to 23  $^{\circ}$ C. However, at -10  $^{\circ}$ C even if the battery was fully charged the thermal engine ignited at the beginning of the test. At the CS tests, SPN<sub>23</sub> increased at lower temperature in agreement with previous studies with pure ICEs that observed increased SPN emissions at low ambient temperature [17]. The variation was very high; at 40 °C, SPN<sub>23</sub> was  $1.9 \times 10^9$  #/km while at -10 °C  $3.2 \times 10^{11}$  #/km. Interestingly, the CS SPN<sub>23</sub> emissions at -10 °C were at the same level with the CD test performed at the same temperature even if at the latter the battery was fully charged, showing that at this low temperature, there is no benefit in terms of particulate emissions from the state of charge. During the CS cycles at -10 °C, 23 °C, and 40 °C, SPN<sub>10</sub> was also measured and presented in Figure 5. The sub-23 nm contribution, defined as  $(SPN_{10}-SPN_{23})/SPN_{23}$ , is indicated. Sub-23 nm contribution to SPN was 93%, 87%, and 58% at -10 °C, 23 °C, and 40 °C, respectively. High percentages for DPF-equipped vehicles are often reported for low emission levels  $(<10^{10} \text{ #/km})$  and are attributed to the cold start. For high concentration levels, we are not aware of studies reporting percentages higher than 20%, as these levels are typically soot particles measured after regeneration events [27]. Here it was demonstrated that a cold start at unfavourable engine conditions can result in high sub-23 nm fraction even at emission levels  $>10^{11}$  #/km.

# 3.3. Emissions Performance during Highway Driving

Figure 6 shows the results of emissions tests conducted in the laboratory on the BAB cycle at  $-10\,^{\circ}\text{C}$ ,  $5\,^{\circ}\text{C}$ ,  $23\,^{\circ}\text{C}$ , and  $40\,^{\circ}\text{C}$  and, on-road, on the Motorway route for comparison. All the tests were started in hot conditions (i.e., with the vehicle not in thermal equilibrium with ambient) and with a depleted HV battery. Some battery recharging occurred during breaking at high speed during motorway driving. Only NOx, SPN<sub>23</sub>, and N<sub>2</sub>O (only for the lab tests) were reported and discussed in the following, the emissions of other compounds measured (see Methods for details) being very low, as shown in Table S4 in the Supplementary Materials.



**Figure 6.** Emissions of regulated pollutants and  $N_2O$  on: (left panels) BAB tests as a function of temperature; (right panels) Motorway route (when available). All the tests were performed with the minimum SOC and with the default user selectable mode of operation. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

NOx emissions were 618 mg/km, 97 mg/km, 66 mg/km, and 131 mg/km at  $-10\,^{\circ}$ C,  $5\,^{\circ}$ C, 23 °C, and 40 °C, respectively. Emissions at  $-10\,^{\circ}$ C were more than 9 times higher with respect to those recorded at 23 °C, despite the tests being performed in hot conditions (coolant and oil temperature were respectively 71 and 86 °C at test start). Nevertheless, almost 90% of such emissions occurred during the first 300 s, indicating that despite a conditioned vehicle, the engine start phase was still important. It was however not possible to clarify if this induction period was related to the fact that, despite a hot vehicle

temperature on the SCR system still being insufficient or due to a type-approved emission strategy that was present on the car. These data aligned well with the trend reported in the literature for NOx emission both at low and high temperatures from pure ICE Diesel vehicles [5,7,21,25,26]. Generally, the significant NOx increase at sub-zero temperatures is associated with the increased duration of the cold start phase. At higher temperature, NOx increase is often associated to a deterioration of engine performances, as detailed in [21,26]. In particular, the laboratory emissions at  $-10\,^{\circ}\text{C}$  in the first 300 s were 14.46 g (93% of the total). Additional details and modal emission profiles were reported in Figure S6 in the Supplementary Materials.

 $N_2O$  emissions were fairly constant at the different temperatures and in the range of 7–10 mg/km. This is related to what was extensively discussed above and in line with the  $N_2O$  production mechanism in the ATS of Diesel vehicles.

For comparison on-road emissions on a Motorway route designed to mimic the laboratory cycle were also reported. When considering this comparison, however, two main aspects need to be considered: (1) the Motorway route has an initial and final urban and rural part accounting for approximately 32.9 km and characterised by a low speed/load driving; (2) the frequency of acceleration-deceleration events in the high speed part of the Motorway route is lower with respect to the laboratory cycle, with an event every 8.6 km against 2.5 km, respectively. Those differences were mainly due to physical constraints (traffic on the highway, location of the facility where the test starts) which could not be modified. Differences between the two routes can be better appreciated in Figure S1 in the Supplementary Materials. For NOx, the distance specific emissions in the motorway section were very similar to those for the total trip (19 mg/km in the Figure 6) and equal to 21 mg/km. This means that given the ratio of approximately 3 between the number of acceleration-deceleration events per km between the BAB and the Motorway route, NOx emission in the two cases are reasonably aligned. Additional details, together with the emission profiles for NOx in the two cases explored, can be found in Figure S7 in the Supplementary Materials.

SPN<sub>23</sub> emissions on BAB cycle were well below  $10^{11}$  #/km at 23 and 40 °C in the laboratory and in the Motorway route on-road tests. Low SPN emissions on the motorway (on-road tests) were also observed in [28]. Instead, at lower ambient temperature, SPN<sub>23</sub> emissions increased to around  $10^{11}$  #/km (9.9 ×  $10^{10}$  #/km at -10 °C) or even higher (4.4 ×  $10^{11}$  #/km at 5 °C). Even if the test was performed at hot engine conditions, SPN<sub>23</sub> emissions in the laboratory originated mainly from the first 3 sharp accelerations that occur at the driving cycle the first ~300 s.

# 3.4. Emissions Performance during Urban Driving

Figure 7 shows the results of emissions tests conducted in the laboratory on the TfL cycle at  $-10\,^{\circ}\text{C}$ ,  $5\,^{\circ}\text{C}$ ,  $23\,^{\circ}\text{C}$ , and  $40\,^{\circ}\text{C}$  and, for comparison, on-road, on the City route. All the laboratory tests were started in hot conditions (i.e., with the vehicle not in thermal equilibrium with ambient) and with a depleted HV battery. Only NOx, SPN<sub>23</sub>, and N<sub>2</sub>O (only for the lab tests) were reported and discussed in the following, being the emissions of other compounds measured (see Methods for details) very low, as shown in Table S4 in the Supplementary Materials.

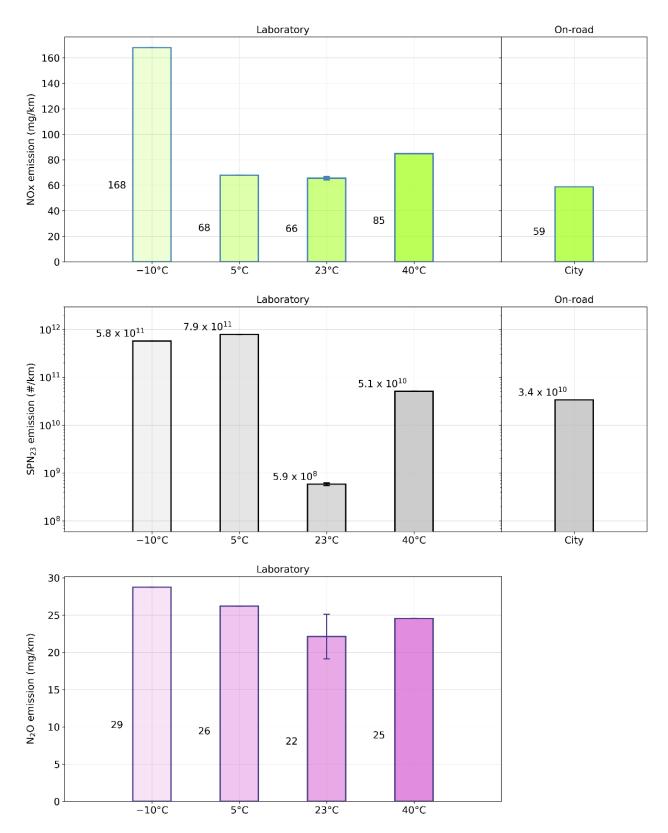


Figure 7. Emissions of regulated pollutants and  $N_2O$  on: (left panels) TfL tests as a function of temperature; (right panels) City route (when available). All the tests were performed with the minimum SOC and with the default user selectable mode of operation. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

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NOx emissions were 168 mg/km, 68 mg/km, 66 mg/km, and 85 mg/km at  $-10\,^{\circ}$ C,  $5\,^{\circ}$ C,  $23\,^{\circ}$ C, and  $40\,^{\circ}$ C, respectively. Emissions at  $-10\,^{\circ}$ C were more than 2 times higher with respect to those recorded at  $23\,^{\circ}$ C, despite the test being performed in hot conditions (coolant and oil temperature were respectively  $48\,^{\circ}$ C and  $77\,^{\circ}$ C at test start). Qualitatively, the trend was similar to the one previously discussed for motorway driving with a significant NOx increase at sub-zero temperatures and a milder increase at  $40\,^{\circ}$ C, with similar motivations. In general, emissions during the cold start phase were approximately 40–61% of the total ones, depending on the temperature (higher fractions at  $-10\,^{\circ}$ C and  $40\,^{\circ}$ C).

 $N_2O$  emissions were fairly constant at the different temperatures as already seen previously. However, in this case, they were more than doubled with respect to motorway driving and in the range of 22–29 mg/km.

For comparison, on-road emissions on a City route similar to the laboratory cycle were also reported. The two tests have a similar length (9 km) and both present repeated stops to simulated congested traffic. The TfL has an average speed of 14 km/h with a maximum one of 53 km/h. These values were, respectively, 14 km/h and 52 km/h for the City route. This suggests that in principle the two tests should be easily comparable considering however that the ambient temperature in the on-road test was approximately 11 °C and, at variance with laboratory tests, it was started in cold conditions. The speed profile of the two routes can be better appreciated in Figure S1 in the Supplementary Materials. For NOx, the distance specific emissions in the two tests were very similar and equal to 66–68 mg/km in the laboratory at 23 °C/5 °C and 59 mg/km on the road. In both cases, most of the emissions occurred during the cold start. In particular, the laboratory emissions in the first 300 s were 0.305 g (52% of the total), while on the City route, these figures were 0.327 g (62%) respectively.

Laboratory SPN<sub>23</sub> emissions in an urban driving cycle at 23 °C were  $5.9 \times 10^8$  #/km. At 40 °C, SPN<sub>23</sub> emissions still remained one order of magnitude lower than the limit  $(5.1 \times 10^{10}$  #/km). On-road SPN<sub>23</sub> emissions during urban driving were also much lower than the respective limit  $(3.4 \times 10^{10}$  #/km). Instead, at lower temperature in the laboratory, SPN<sub>23</sub> emissions were near  $(5.8 \times 10^{11}$  #/km at -10 °C) or even exceeded  $(7.9 \times 10^{11}$  #/km at 5 °C) the SPN<sub>23</sub> regulatory limit. Note, however, that since the route is significantly shorter than what is required in the RDE, the emissions cannot be directly compared to the limit for this test. Interestingly, at low temperatures, the thermal engine of the vehicle switched off after ~400/450 s for around 300/400 s. Nonetheless, at low temperature, the emissions were high and in one case even higher than  $6 \times 10^{11}$  #/km. At the low temperature tests (5 °C and -10 °C), the SPN<sub>23</sub> emissions during the second thermal engine ignition were higher than the first considering a period of 100 s showing the importance of thermal engine ignition at high engine speeds. Previous studies reported higher SPN<sub>23</sub> of Diesel vehicles at urban driving [20,27,29].

### 3.5. Emissions Performance during Laboratory Simulated RDE Driving

Figure 8 shows the results of emissions tests conducted in the laboratory on a simulated RDE cycle at  $-10\,^{\circ}\text{C}$ ,  $5\,^{\circ}\text{C}$ ,  $23\,^{\circ}\text{C}$ , and  $40\,^{\circ}\text{C}$  and, on-road, on the RDE route for comparison. All the laboratory tests started in cold conditions (i.e., with the vehicle in thermal equilibrium with ambient) and with a depleted HV battery. Only CO, NOx, SPN<sub>23</sub>, and N<sub>2</sub>O (only for the lab tests) were reported and discussed in the following, being the emissions of other compounds measured (see Methods for details) very low.

CO emissions were low. They were 69 mg/km, 9 mg/km, 10 mg/km, and practically zero at -10 °C, 5 °C, 23 °C, and 40 °C, respectively. The higher emissions at -10 °C with respect to those recorded at 23 °C are in line with the literature [5,7,21,25] and mostly due to the longer period required at cold start to reach operative temperature in the ATS.

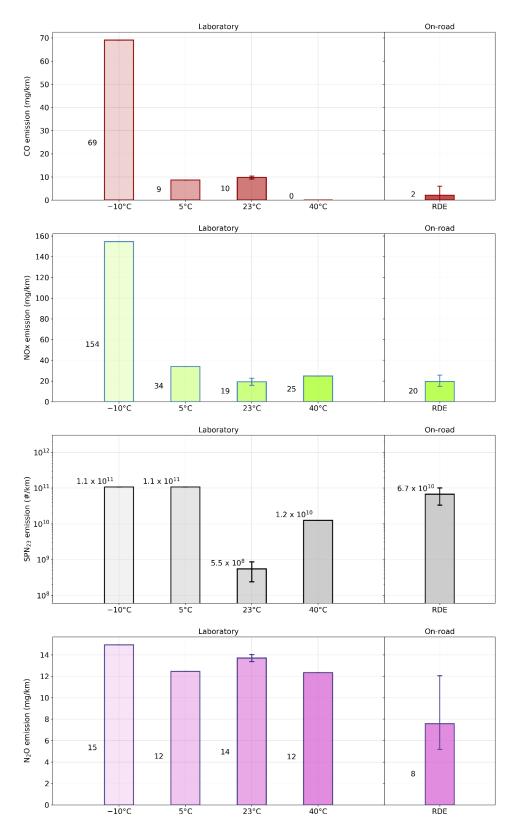


Figure 8. Emissions of regulated pollutants and  $N_2O$  on: (left panels) labRDE tests as a function of temperature; (right panels) RDE route (when available). All the tests were performed with the minimum SOC and with the default user selectable mode of operation. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

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The situation was qualitatively similar for NOx, except for the increase already observed above and also in the literature of emissions at high temperature (i.e.,  $40 \,^{\circ}$ C). NOx emissions were 154 mg/km, 34 mg/km, 19 mg/km, and 25 mg/km at  $-10 \,^{\circ}$ C,  $5 \,^{\circ}$ C,  $23 \,^{\circ}$ C, and  $40 \,^{\circ}$ C, respectively. Emissions at  $-10 \,^{\circ}$ C were more than 7 times higher with respect to those recorded at 23  $\,^{\circ}$ C, which is in line with the scientific literature and with the trends previously reported [5,7,21,25].

 $N_2O$  emissions were fairly constant at the different temperatures as previously seen. However, in this case, they laid between what was recorded during predominantly motorway or urban cycles and in the range of 12–15 mg/km.

For comparison, average emissions from four RDE road driving tests similar to the laboratory cycle were also reported. Indeed, the laboratory test was created on the basis of the data recorded during the on-road test and was then shortened to keep the total distance below 30 km (and the total time duration below approximately 30 min). This was done trying to keep similar shares of urban, rural, and motorway driving to the original cycle, as also evident from the Table S1 in the Supplementary Materials. Specifically, the urban, rural, and motorway shares were 29%, 36%, and 35% for the laboratory test and 39%, 32%, and 29% on-road. All RDE road driving tests started with a depleted HV battery, two of them started in cold condition and two in hot condition. On average, the ambient temperature on road tests was 13 °C. The speed profile of the two routes can be better appreciated in Figure S1 in the Supplementary Materials. For NOx, the distance specific emissions in the two tests were very similar and equal to 19 mg/km in the laboratory at 23 °C and 20 mg/km on the road. In both cases, most of the emissions occurred during the cold start. In particular, in the laboratory emissions in the first 300 s were 0.349 g (64% of the total) while on the City route, these figures were on average 0.627 g (36%).

The variation of SPN<sub>23</sub> emissions at different temperature was very high (from  $10^8$  #/km to  $10^{11}$  #/km). The trend was similar to the gaseous emissions; i.e., higher SPN<sub>23</sub> emissions at lower temperatures. Specifically, at 5 °C and -10 °C, they were  $1.1 \times 10^{11}$  #/km, at 23 °C < $10^9$  #/km, and at 40 °C  $1.2 \times 10^{10}$  #/km. The urban RDE value at CS testing mode was  $6.7 \times 10^{10}$  #/km. The contribution of the cold start was different at 5 °C and -10 °C. At 5 °C, the vehicle emitted ~38% of the total emissions during the first 300 s, while at -10 °C 18%, showing that at this low temperature high SPN is emitted even after the first seconds of the trip.

## 3.6. Emissions of CO<sub>2</sub> as a Function of the Mode of Operation

Figure 9 shows  $CO_2$  emissions in both WLTP and RDE test procedures performed according to the provisions of the regulation [15]. The car tested had an electric range of 82 km which, differently from what sometimes happens with pollutants, allowed a significant reduction in  $CO_2$  tailpipe emissions when operating in charge depleting as compared to charge sustaining mode. Indeed, in the laboratory, the emissions during the CD4 and CS were 192 g/km and 308 g/km, respectively, with weighted emissions as low as 29 g/km (due to the 3 CD tests performed in full electric mode and thus with no  $CO_2$  emissions at the tailpipe), and 48 g/km if the total  $CO_2$  emissions measured during the CD sequence (4.47 g) are divided by the distance covered (ca. 93 km).

Still an important benefit is seen on-road when comparing two RDE tests (ca. 92 km long) starting with a completely depleted or fully charged HV battery. In this case, the reduction in CO<sub>2</sub> emitted at the tailpipe is more than 4 times (from 207 g/km to 43 g/km).

While  $CH_4$  contribution to  $CO_2$  equivalent emissions was negligible, the  $N_2O$  contribution, considering a global warming potential (GWP) for  $N_2O$  of 298 [30], ranged from 0.6 g/km during the CD100 mode to 2.7 g/km during the CS operation.

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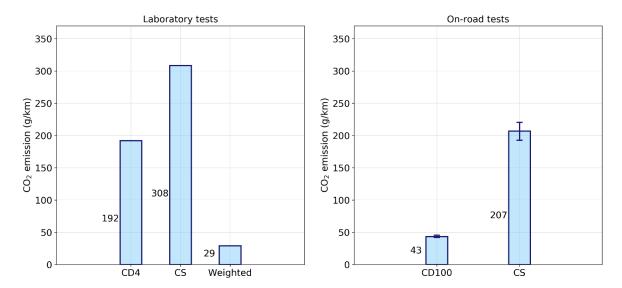


Figure 9. CO<sub>2</sub> emissions on laboratory and on-road tests performed according to the WLTP and RDE test procedures, respectively. For laboratory tests, CD4 indicates the 4th cycle in the charge depleting sequence (i.e., the cycle in which the ICE ignited for the first time). For on-road tests, CD100 indicates tests with an initial state of charge (SOC) equal to 100%. In both cases, CS indicates tests with an initial state of charge (SOC) equal to the minimum. Error bars on the graph represent maximum and minimum values for conditions having more than one test. See Methods section for additional details.

## 4. Conclusions

A modern plug-in hybrid Diesel vehicle meeting the Euro 6d-ISC-FCM standard was investigated under a large variety of laboratory and real world conditions. The car had an electric range of 82 km. Differently from what sometimes happens with pollutants, this consistently allowed a significant reduction in  $CO_2$  tailpipe emissions when operating in charge depleting (29–43 g/km) as compared to charge sustaining mode (207–308 g/km).

The vehicle presented emissions below the Euro 6 regulatory limits in the WLTP and all the on-road tests (RDE compliant and non-compliant) at the different battery SOC. The SPN<sub>23</sub> emissions were lower than the Euro 6 limit (6  $\times$  10  $^{11}$  #/km) for all the tests performed, with some cases being close to the limit and one case, the test over the TfL at 5 °C, that was slightly higher (7.9  $\times$  10  $^{11}$  #/km). Moreover, at 23 °C, the vehicle showed emissions below the Euro 6 limit over all the cycles tested.

The WLTP weighted emissions (that account for the electric range of the vehicle and apply the regulatory utility factors) were very low, and lower than the average of the conventional Diesel vehicles tested at JRC in 2020–2021. The emissions during charge sustaining operation were however comparable to those of the conventional Diesel vehicles. Moreover, the emissions obtained with the vehicle's HV battery fully charged during onroad tests were comparable to those obtained with the battery at the minimum SOC for the entire test (ca. 91 km) as well as for the urban section of the test (ca. 36 km). Urban emissions of the charge depleting test from a fully charged battery showed high variability, ranging from a fully electrical functioning, with no tailpipe emissions, to a more conventional behaviour in line with standard Diesel vehicles. All this underlines the importance of an effective management of Diesel and electric traction: avoiding high power or repeated engine ignition have a significant benefit in terms of environmental performance.

During on-road RDE tests the emissions were lower than the average of the conventional Diesel vehicles tested at JRC in 2020–2021 for all the SOC tested (100%, 50%, 25% and minimum or charge sustaining), with the exception of the  $SPN_{23}$  emissions in charge sustaining mode, which were comparable at all SOC.

Laboratory WLTP weighted CO and NOx emissions were substantially lower than those obtained during the on-road RDE tests performed with the fully charged battery

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(1 and 4 mg/km for CO and NOx weighted compared to 8 and 23 mg/km for RDE tests). SPN<sub>23</sub> were comparable under these conditions ( $\sim$ 2 × 10<sup>10</sup> #/km). On the other hand, during charge sustaining operation, emissions of CO (12 mg/km) and NOx (29 mg/km) emissions were higher than RDE tests (2 and 20 mg/km for CO and NOx, respectively) and SPN<sub>23</sub> were substantially lower (WLTP 3 × 10<sup>9</sup> #/km; RDE  $\sim$ 7 × 10<sup>10</sup> #/km).

NOx emissions were high under the WLTC at  $-10~^{\circ}\text{C}$  (379 mg/km) and even higher on the BAB 130 at the same temperature, where after just 300 s, NOx emissions were 14.46 g. In general, emissions of criteria pollutants increased as temperatures decreased. It was also shown that at  $-10~^{\circ}\text{C}$ , there is no benefit in terms of particulate emissions from the state of SOC, as the charge sustaining and charge depleting SPN<sub>23</sub> emissions were at the same level and that at 23  $^{\circ}\text{C}$ , the charge depleting SPN<sub>23</sub> emissions are higher than at charge sustaining.

 $N_2O$  was the only unregulated pollutant measured presenting relevant emissions for this vehicle both in the laboratory and on the road. The  $N_2O$  emissions were proportional to the utilisation of the ICE for all the tested conditions and contributed up to 2.7 g/km of  $CO_2$  equivalents.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/atmos13081175/s1, Table S1—On-road route details for selected examples: RDE-compliant route (RDE) , mainly highway driving (Motorway), or mainly city driving (City). Table S2—List of performed experiments. Table S3—Tested vehicles. Table S4—Data summary. Figure S1—Profiles of speed and slope (when relevant) 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—Emissions of NOx during CD4 of the WLTP. Figure S4—Emissions of NOx during two RDE test with initial SOC equals to 100. Figure S5—Emissions of NOx during CD (top) and CS (bottom) of the WLTP at -10 °C. Figure S6—Emissions of NOx during BAB test at -10 °C. Figure S7—Emissions of NOx during highway tests (Motorway route (bottom) and BAB (top)).

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