



EUROPEAN COMMISSION
JOINT RESEARCH CENTRE
Directorate of Energy, Transport and Climate
Sustainable Transport Unit

HD H₂ ICE emissions measurement

1. Introduction

The heavy-duty (HD) road transport sector, including: Lorries, buses and coaches, has been reported to be responsible for ~6% of EU's total CO₂ emissions, and for about 19% of the Greenhouse Gases (GHG) emissions of the road transport sector^{1,2}.

In order to fulfil the GHG reduction objectives, the road transport sector is shifting towards alternative solutions, including the use of hydrogen. However, the market deployment of this technology was hindered by the limited legal framework that would allow its approval and further commercialization. In search of evidence that could support the request of an amendment to the current emissions regulation and accommodate the type-approval of HD hydrogen in internal combustion engines (HD H₂ ICE) and vehicles operating with this technology, a validation exercise was launched by the EC. This exercise aims at evaluating if the current type-approval testing procedure laid down in the UNECE Regulation N°49 can be carried out using HD engines running on pure H₂. This procedure requires the measurement of a series of air pollutants during laboratory testing (namely: CO, NO_x, hydrocarbons (HC), particulate mass (PM), particle number (PN), CH₄ and NH₃) or on the road using portable emissions measurement systems (PEMS – only CO, NO_x, CH₄ and PN are required on the road).

When highly pure H₂ (known as Grade D) is combusted by a modern ICE, besides some air pollutants such as: NO_x and oil related particles, the resulting exhaust will contain levels of water higher than those typically obtained in Diesel engines. Under certain conditions (e.g., stoichiometric and rich fuel to air ratios) the water concentration can be >30% of the total exhaust gases, which is higher than the maximum level currently found in the technical specifications of the measurement devices used during engine/vehicle homologation (~24%). Hence, this analytical limitation could lead to measurement interferences, and erroneous estimation of the pollutant emissions.

HD H₂ ICE can also run in lean conditions that would lead to water levels that are closer to the current instrument specifications (~20%). In order to meet the Euro VI emissions standard, a lean-burned H₂ engine may need to be equipped with a typical Euro VI deNO_x after-treatment system used by lean-burned Diesel engines (DOC+SCR+ASC).³ In this case, N₂O, NH₃ and/or ammonium particles^{4,5} may also be found in the exhaust gases.

The present study investigates the feasibility to perform the HD type-approval testing procedure as described in the UNECE Regulation N°49 when using a lean-burn H₂ engine. It addresses the main constraints that the current procedure may have due to the absence of carbon and the higher level of water in the exhaust, especially on exhaust flow calculation and pollutant emissions both during laboratory testing and during PEMS testing. Moreover, the measurement of a series of pollutants that can be of interest for a future Euro emission standard (N₂O, SPN₁₀, and HCHO) were also investigated.

2. Materials and methods

A prototype lean-burn spark ignition hydrogen heavy-duty engine was tested over the regulatory cycles: WHSC, WHTCcold (hereinafter WHTCc), WHTChot (hereinafter WHTCh) and also over an In-service conformity-like test (hereinafter PEMS cycle) at 23°C.

The engine was equipped with a series production Diesel after-treatment system that included a DOC+SCR+ASC, (the DPF was removed). Emissions of NO_x (NO and NO₂), CO, CO₂, HC, CH₄, NH₃, PM, SPN₂₃, SPN₁₀, N₂O, H₂O and O₂ were measured with a series laboratory grade equipment and/or PEMS instrumentation (See Table 1). The engine was checked in two complementary experimental campaigns. The first one focused on gaseous emissions and the second focused on particulate emissions (PM and PN) and exhaust flow measurement. During the first campaign three valid tests were considered for each cycle (with the exception of the WHTCc for which only two were available), and two valid tests were considered for the second campaign.

During the first campaign all instruments sampled directly from the engine exhaust. A partial dilution system was used during the second campaign to allow following the particulate mass measurement procedure.

The measurement devices used included equipment that can be currently used during engine emissions type-approval or PEMS testing (bold text of Table 1): AVL i60, HORIBA MEXA-730 Lambda, HORIBA MEXA-2110SPCS, AVL APC 489, HORIBA FTX-ONE-CS, HORIBA MEXA-ONE-XL-NX, HORIBA MDLT-ONE, HORIBA OBS-ONE-GS (PEMS) and HORIBA OBS-ONE-PN23 (PEMS) and some that measure unregulated pollutants and hydrogen: HORIBA OBS-ONE-PN10 (PEMS) and HSENSE (Lab).

The AMA i60 was calibrated with the following span gases mixing ratios: CO (H) 3%, CO (L) 500 ppm, CO₂ 500 ppm, O₂ 8%, HC 900 ppm C₃H₈, NO_x 50 ppm, CH₄ 900 ppm. The FTIR and QCL were checked with NH₃ 50 ppm and N₂O 200 ppm. The PEMS OBS-GS was calibrated using typical PEMS mixture that includes: CO 9%, CO₂ 18%, NO 2500 ppm, CH₄ 9000 ppm and C₃H₈ 9000 ppm. As it will be shown in the Results and discussion section, it is important to highlight that these concentrations, although suitable for Diesel engines, are much higher than the typical concentrations measured for the hydrogen system under study.

Exhaust flow was obtained using two different approaches. One is determined by the combination of fuel and intake air mass flow measurement for laboratory-grade instruments. The Coriolis flowmeter (CMFS015M, Emerson) was utilized to the hydrogen mass flow measurement and obtained real-time mass flow of the fuel. The real-time intake air mass flow was measured by the ultrasonic flowmeter (FLOWSONIX, AVL). The second-by-second mass flows of the fuel and the intake air were summed to determine the exhaust mass flow ($q_{mew} = q_{mf} + q_{maw}$). The other is the Pitot-tube flowmeter for PEMS (OBS-ONE PF, HORIBA). The flowmeter was installed at the end of the tailpipe and can determine the exhaust mass flow directly. The transformation time of the analyzer was taken in account in order to determine emissions of each pollutant.

Table 1. List of measurement devices used during the test campaigns. Bold indicate the regulated pollutants and instruments currently used during engine emissions type-approval.

Instrument	NO_x	NO	NO₂	CO	CO₂	HC	CH₄	NH₃	PM	SPN₂₃	SPN₁₀	N₂O	HCHO	H₂	H₂O	O₂
AVL i60 (Lab)	x			x	x	x	x									x
HORIBA MEXA-2110SPCS (Lab)										x	x					
AVL APC 489 (Lab)											x					
HORIBA FTX-ONE-CS (Lab)	x	x	x	x	x			x				x	x		x	
HORIBA MDLT-ONE (Lab)									x							
HORIBA MEXA-730 Lambda (Lab)																x
HORIBA OBS-ONE-GS (PEMS)	x	x	x	x	x	x										
HORIBA OBS-ONE-PN₂₃ (PEMS)										x						
HORIBA MEXA-ONE-XL-NX (Lab)	x	x	x					x				x				
HSENSE (Lab)														x		
HORIBA OBS-ONE-PN₁₀ (PEMS)											x					

3. Results and discussion

3.1. Water Content

UNECE Regulation N°49 does not prescribe the measurement of water from the engines' exhaust. Without a specific principle requested by the regulation, an FTIR – instrument, widely used in the field for this purpose, was used. Hence, the concentration of water in the exhaust was measured using the HORIBA FTX-ONE-CS (hereinafter FTIR).

During the four tests cycles, WHSC, WHTCc, WHTCh and PEMS cycle, maximum water concentration measured was 20-21%. The average concentration ranged from 10% for the WHTCc, WHTCh, 11% for the PEMS cycle and 13% for the WHSC. The repeatability of the measurements was very good with a standard deviation <0.01 for the WHTCh and PEMS cycle, <0.1 for the WHSC and <0.3 for the WHTCc.

The exhaust water content for this lean-burn HD H₂ engine was comparable to what is typically measured on stoichiometric HD CNG engines, ~18%. These concentrations are near, but below, the maximum value declared for state-of-the-art PEMS instrumentation, ~24%. Thus, it was expected that water concentration should not be an obstacle to apply the current test procedure for the engine under study.

3.2. Exhaust flow measurement

UNECE Regulation N°49 describes different approaches to estimate engine/vehicle exhaust flow, including the carbon balance methodology. However, the carbon balance method can only be applied in case carbon based fuels are used. With hydrogen as a fuel, the exhaust flow can be calculated for instance: i) as the difference between the constant flow volume and the dilution air volume; ii) using air and fuel flow (see Methods section); iii) using air or fuel flow and lambda; iv) or using an exhaust flow meter (EFM - typically used during PEMS testing).

Figure 1 and 2 illustrate the real-time data and the correlation between the exhaust sample flow obtained as the difference between the constant diluted exhaust and the dilution air flows and the exhaust flow calculated using intake-air and fuel flow as described in the Method section (sample flow raw exhaust flow) for the tests performed over the WHSC and WHTC. These are two approaches commonly used in laboratory testing. On the other hand, Figure 3 and 4 illustrate the real-time data and the correlation between the exhaust flow measured with the EFM (PEMS EFM) and the exhaust flow calculated using intake-air and fuel flow measurement (Exhaust mass flow) for the tests performed over the PEMS cycle and the WHSC. As previously indicated the EFM is the approach used during on-road testing.

The correlation for the two laboratory approaches (sample flow and raw exhaust flow) was very good, with an $R^2 > 0.99$ and a relative difference $\sim 0.1\%$ during both tests. The good alignment can also be appreciated thanks to the real-time data. When comparing the results obtained with the EFM and the laboratory over the PEMS cycle at standard conditions at 20°C , 101.325 kPa , which are the working conditions of the EFM PEMS, the relative difference decreases to $\sim 2\%$. Data scatter decreased when comparing the results obtained during the WHSC ($R^2 > 0.99$).

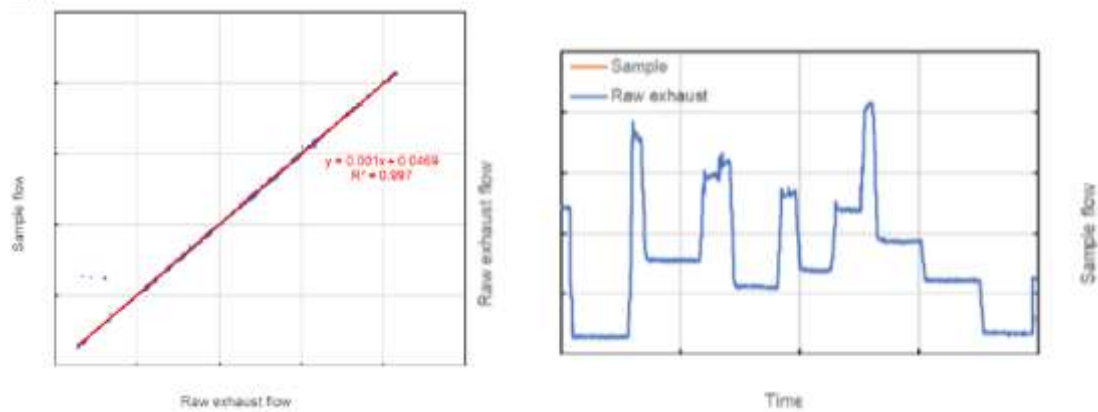


Figure 1. (Right panel) Real-time data and (left panel) correlation between the exhaust sample flow obtained as the difference between the constant diluted exhaust and the dilution air flows and the exhaust flow calculated using intake-air and fuel flow for the tests performed over the WHSC.

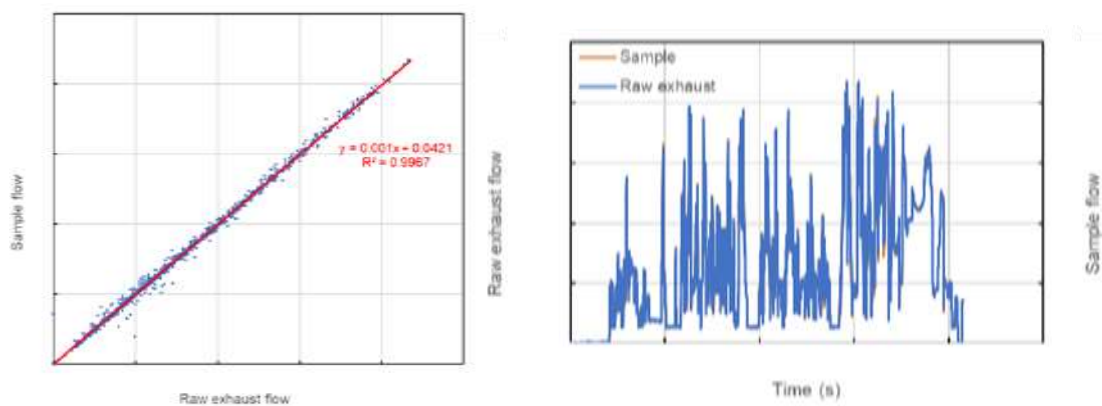


Figure 2. (Right panel) Real-time data and (left panel) correlation between the exhaust sample flow obtained as the difference between the constant diluted exhaust and the dilution air flows and the exhaust flow calculated using intake-air and fuel flow for the tests performed over the WHTC.

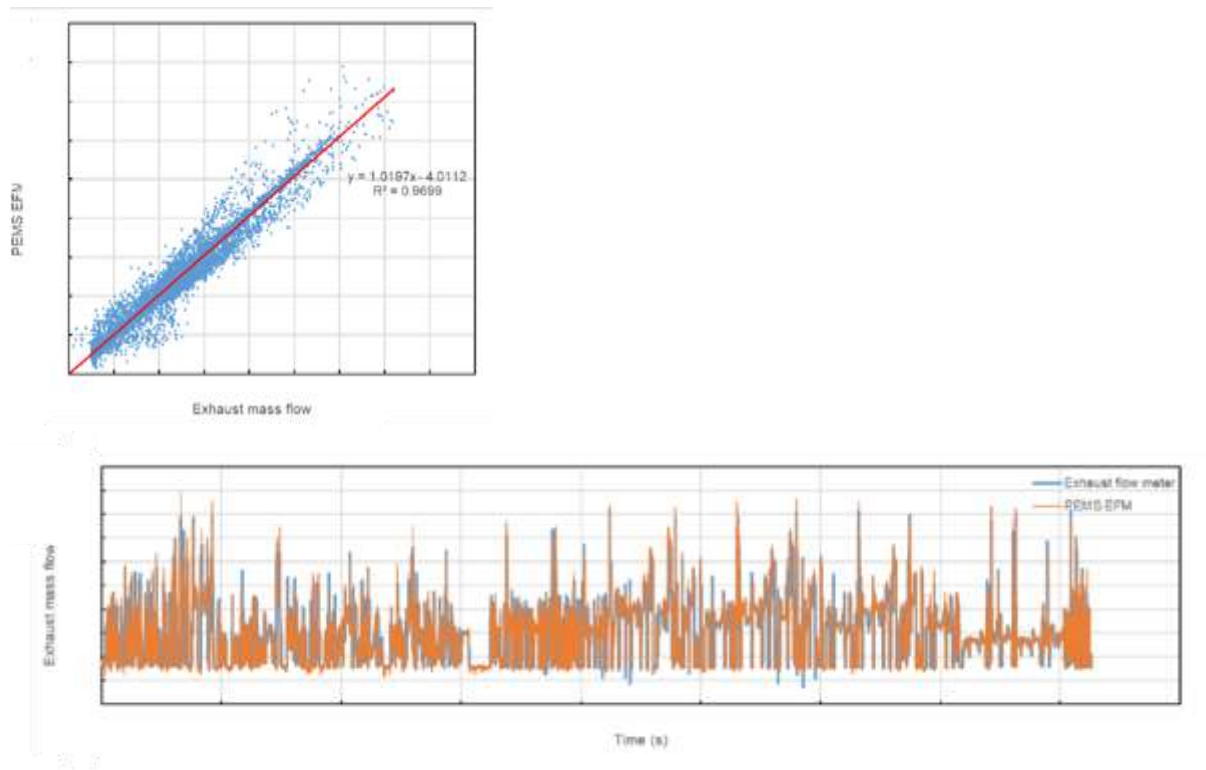


Figure 3. (Bottom panel) Real-time data and (top panel) correlation between the exhaust sample flow measured with the PEMS' EFM and the exhaust flow calculated using intake-air and fuel flow for the tests performed over the PEMS cycle.

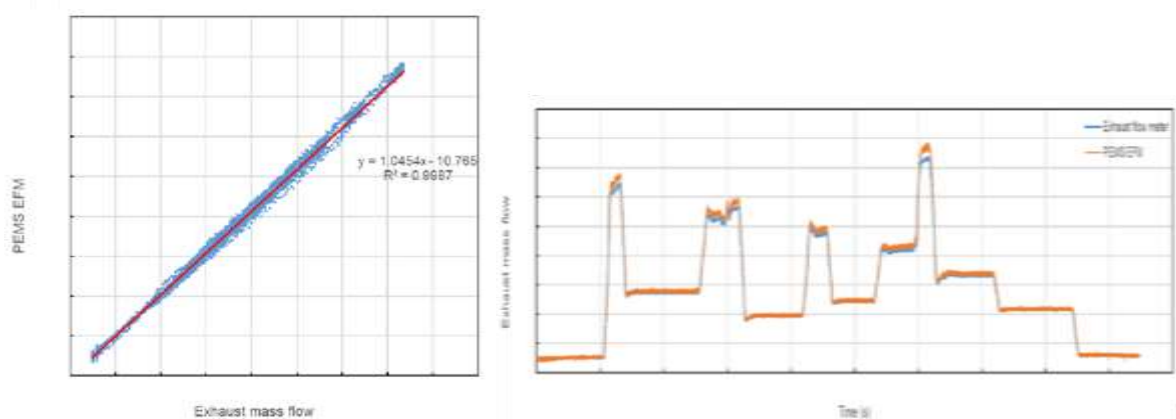


Figure 4. (Right panel) Real-time data and (left panel) correlation between the exhaust sample flow measured with the PEMS' EFM and the exhaust flow calculated using intake-air and fuel flow for the tests performed over the WHSC.

3.3. Filter and dilution temperatures

UNECE Regulation N°49 lays down detailed prescriptions regarding the temperature ranges of the dilution air and filter. Hydrogen combustion can result in higher exhaust temperatures than those obtained with conventional fuels. This, in turn, could lead to dilution air temperatures above the prescribed ranges.

Figures 5 and 6 illustrate the filter and dilution air temperatures during the WHSC and WHTC. The temperature ranges were well within regulatory limits (42-52 °C for the filter temperature and 20-30 °C for the dilution air temperature).

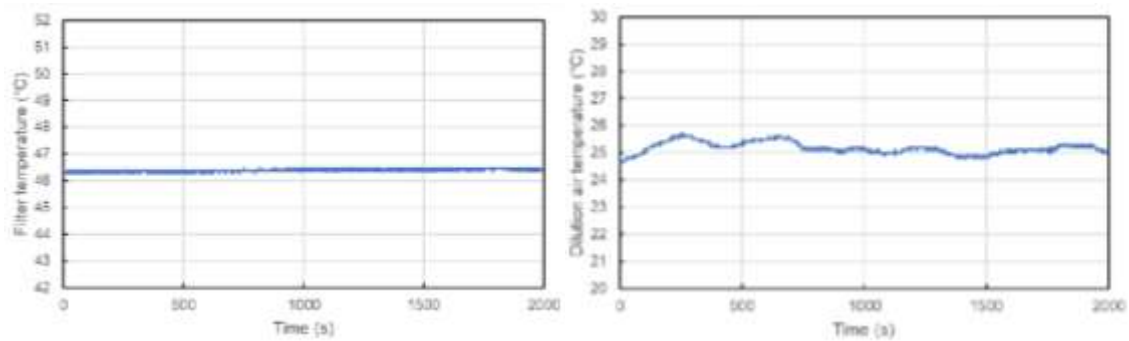


Figure 5. Filter and dilution air temperatures during the WHSC.

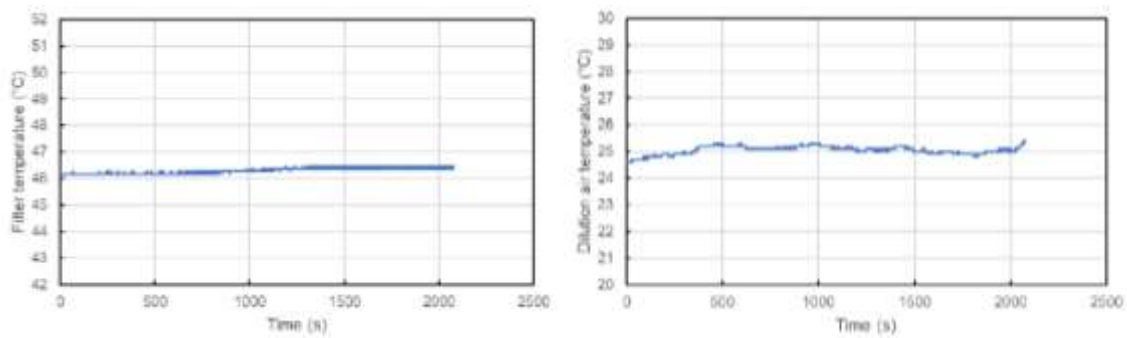


Figure 6. Filter and dilution air temperatures during the WHTC.

3.4. Gaseous emissions measurement

Table 2 summarizes the average concentrations of the different gaseous pollutants measured during the testing campaigns. The concentrations of all regulated gases were low (NO_x), very low (NH₃ and CO₂) or even close to the detection limit of the measurement instruments (HC, CH₄ and CO). In the following the emissions of each compound will be presented and discussed.

Table 2. Average concentrations of the different gaseous pollutants measured during the testing campaigns.

		NO _x	CO	THC	CH ₄	NH ₃	CO ₂	N ₂ O	H ₂
Average concentration	ppm	1	2	<1	<1	<1	560	2	33

The concentration of NO_x emissions measured during the different test cycles with the laboratory reference instrument, the PEMS (CLD) as well as with the QCL and FTIR were compared. All instruments showed very similar NO_x emission profiles during the transient (see Figure 7) and steady state tests. Average NO_x concentration measured was 1 ppm. The correlation between the reference laboratory and the PEMS analysers resulting from all the tests performed, summarized in Figure 8, was good, with a R² of 0.92 and the angular coefficient of the regression line equal to 0.85 and an absolute average deviation of 0.7 ppm. The R² of the individual laboratory tests recorded at 10 Hz (i.e., WHTC and WHSC) was

always >0.93 , while R^2 during the PEMS-test (recorded at 1 Hz) was 0.83. A very good correlation was also found for the other laboratory-grade instruments, QCL and FTIR, where R^2 was >0.96 and the angular coefficient and y-intercept of the regression line equal to $0.98 - 0.28$ and $1.05 - 4.61$, respectively.

The inter-test repeatability was very good, with R^2 ranging from 0.90 for the WHTCc tests to 0.99 for WHTCh and PEMS-test. The absolute average deviation was below 1 ppm for all test pairs, with the exception of the WHTCc for which it was 1.3 ppm.

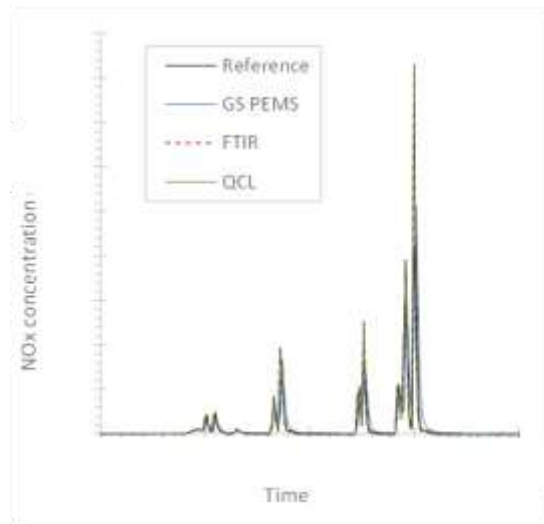


Figure 7. Example of NO_x emission profiles over a PEMS-test.

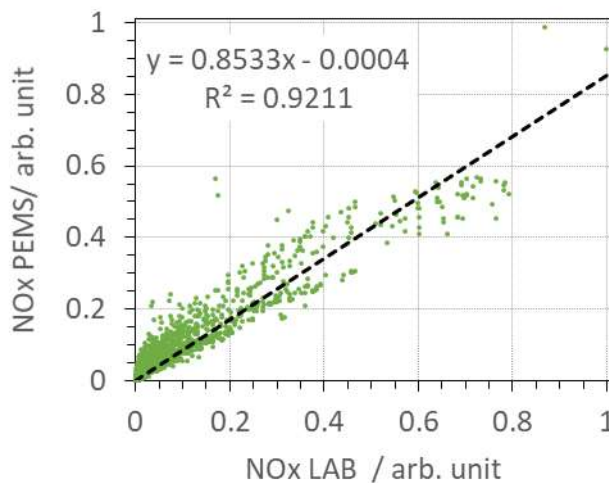


Figure 8. Correlation between NO_x concentration measured in the laboratory and by the PEMS analyser over all the cycles tested.

The concentrations of CO and THC (also labelled as HC) measured with the laboratory reference and the PEMS during all tests were very low, reaching the detection limit of the analysers. High purity of the fuel (Grade D) and good performance of engine and after-treatment system lead to this expected result. Although the measurements of THC at such low concentration appear to suffer from cross-interferences with O₂ (see Figure 9; R^2 between THC and O₂ up to 0.8). Hence, the levels of emissions measured were within the accuracy tolerances currently required for such PEMS measurements. Given the emission profile measured for the CO with the PEMS (see Figure 10) and the good correlation with H₂O emissions for FTIR and PEMS (R^2 up to 0.9), it can be considered that at these relatively low

concentrations FTIR and PEMS instruments could be affected by H₂O. It should be noted that while FTIR are factory calibrated for a wide range of concentrations, the span gases used for the laboratory and PEMS tests were substantially high, if compared to the concentrations of the emissions measured. Finally, the measurement of CH₄ presented the same characteristics described for THC (Figure 11).

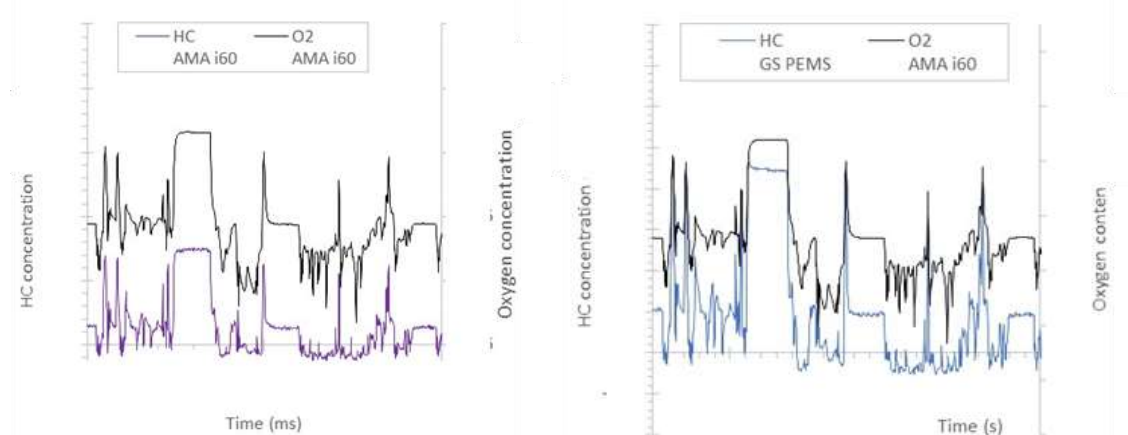


Figure 9. Example of THC emission profile measured by laboratory grade instrument (left panel) and PEMS analyser (right panel) along with O₂ measured by the laboratory instrument.

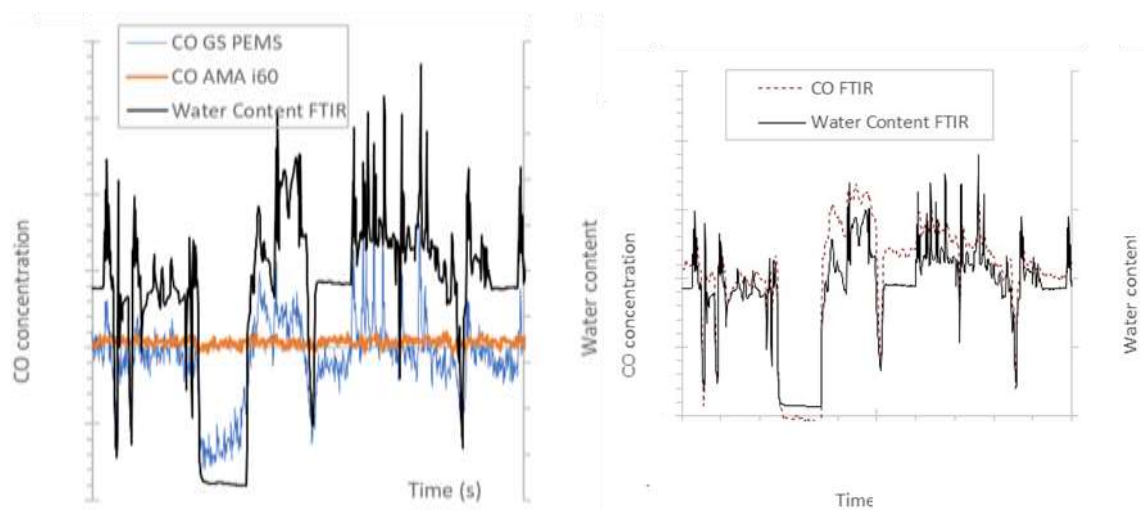


Figure 10. Example of CO emission profile measured with the reference laboratory analyser (orange) PEMS (blue) analysers (left panel) and FTIR laboratory (red) (right panel)

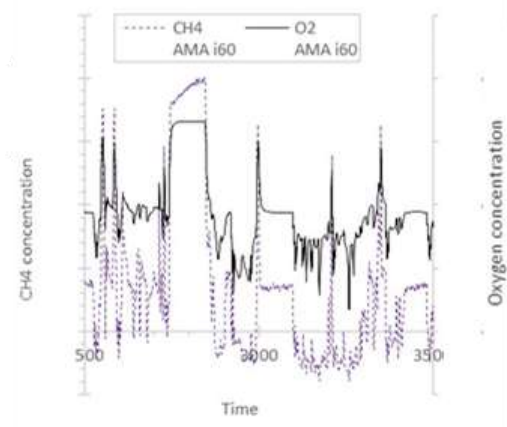


Figure 11. Example of CH₄ and O₂ profiles measured by the laboratory instrument.

The concentrations of NH₃ measured with the laboratory reference (FTIR) and the QCL-IR during all tests were very low. Several studies have reported of NH₃ emissions resulting from the slip of this pollutant when an SCR is used as deNO_x system in lean-burn engines. Figure 12 summarizes the correlation between the FTIR and the QCL-IR analysers resulting from all the tests performed. The correlation was good with an R² of 0.85 and angular coefficient and y-intercept of the regression line equal to 0.73 + 0.02 and an absolute average deviation of 0.2 ppm. Good correlation between these instruments has also been reported previously for Diesel and CNG engines.^{6,7} Both instruments also showed comparable dynamics during the transient cycles (see Figure 13).

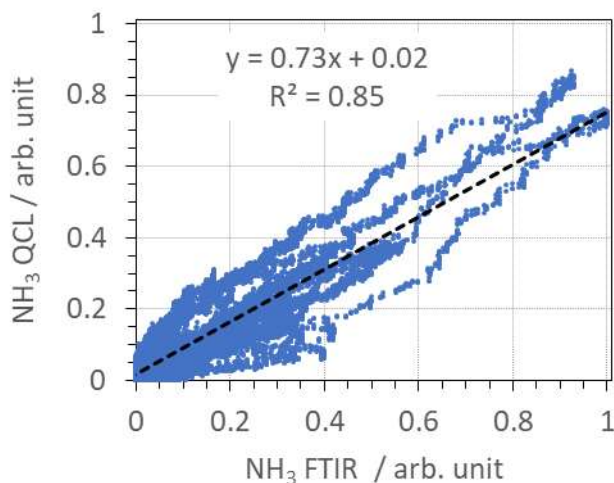


Figure 12. Correlation between NH₃ concentration measured in the FTIR and by the QCL-IR over all the cycles tested.

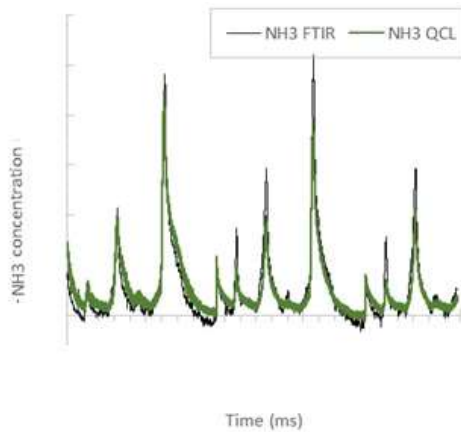


Figure 13. Example of NH₃ emission profile measured by the FTIR and the QCL-IR.

CO₂ tailpipe emissions from a pure hydrogen engine running on Grade D hydrogen can arise from: The ambient air used during combustion, lubricant burning and/or hydrolyzation of the Diesel exhaust fluid (reagent) needed in the SCR to reduce NO_x emissions. The CO₂ emissions measured during the tests performed were very low compared to engines running on carbon-based fuels with average concentrations around 550 ppm. It should be considered that the CO₂ background concentration of the ambient air during testing was around 450 ppm. The reference CO₂ from AMA i60 is measured in dry exhaust gas. Dry to wet correction was not applied, hence CO₂ emissions could be overestimated. As shown in Figure 14 the PEMS analyser followed a similar emission profile as that of the laboratory reference, their correlation was poor ($R^2 \approx 0.55$). At least two elements should be considered for this comparison: i) It should be also noted that although the reference CO₂ analyser (AMA i60) was calibrated to an appropriate span gas, the measurement cuvette was standard (up to 18% CO₂), resulting in a low accuracy; ii) the constant variations recorded with the PEMS (see Figure 14) that could be linked to the fact that the PEMS was calibrated for a much higher range (8%) (as also highlighted for CO and THC) that could lead to this less accurate measurement when testing an engine running on pure H₂, indicating, as one could have expected, a simple 'fit for all' solution cannot be used for the span gases and that dedicated span gases are needed when measuring CO₂ with the PEMS analysers. Indeed, the Regulation requires the span gases to be in the range of 80% of the maximum expected concentration of the component. The inter-test relative repeatability at these low CO₂ emissions ranged from 0.3% to 6.7%, including a 1% deviation during the PEMS-test. The maximum absolute difference (WHTCc) was lower than 1g/kWh.

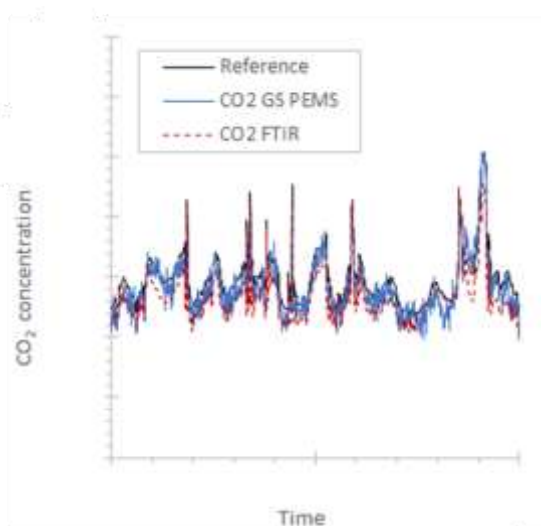


Figure 14. Example of CO₂ emission profile measured by the laboratory grade instrument, the PEMS analyser, and the FTIR.

3.5. Particulates emissions measurement

As illustrated in Figure 15. PM measurements were highly repeatable for most tests where the difference was < 0.01 mg/kWh, and with a slightly larger deviation, albeit small, in the case of the WHTCc.

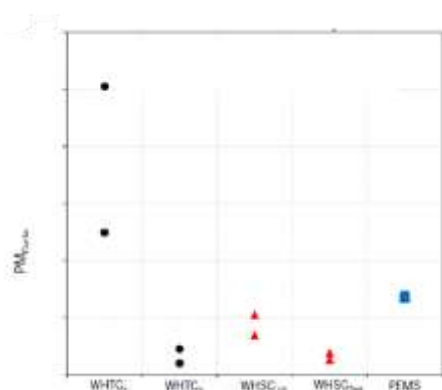


Figure 15. PM measured during the WHTCc, WHTCh, WHSC and PEMS cycle.

Figure 16 plots SPN₂₃ concentrations with the reference instrument SPCS and the PEMS OBS. The slopes were 1.98 ± 0.31 , while the slopes in most cases were >0.92. A few exceptions were tests with a spike not measured by the reference instrument, which however did not affect the slope. This spike is probably due to instrument noise and not a result of condensation, because it appeared only once and was not seen by the 10 nm PEMS. The specific instrument had issues with the analog reading of the signal and could be related to this data transmission problem. The slope indicates that the PEMS was measuring almost two times the values compared to the reference instrument. However, as it will be shown with the 10 nm instruments, similar slopes were found for the 10 nm PEMS and the second reference instrument, indicating that the reference instrument had calibration issues.

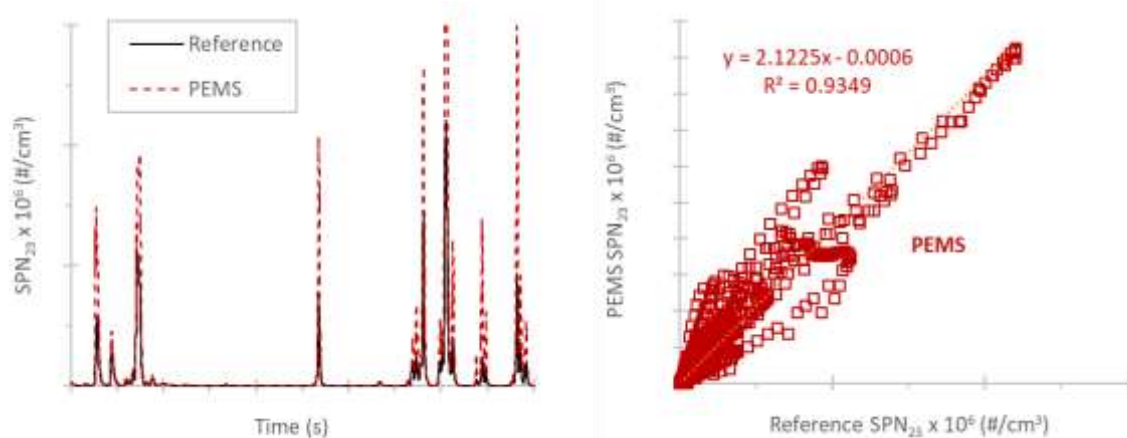


Figure 16. (Left panel) SPN₂₃ concentrations over a part of cWHTC. (Right panel) correlation of PEMS vs reference for the same cycle.

3.6. Euro VI unregulated emissions measurement

N₂O emissions for transport are of high interest, as they have continuously increased following the progressive emission standards and the implementation of different after-treatment technologies used to meet their NO_x and particulates requirements (for more information see Selleri et al. 2021 and 2022 and references therein). N₂O can be formed on the DOC, the SCR and/or the ASC following different pathways. The concentrations of N₂O measured with the laboratory reference (FTIR) and the QCL-IR during all tests performed were overall in good agreement with an R² of 0.95 and angular coefficient and y-intercept of the regression line equal to 0.87 (see Figure 17) and an absolute average deviation of 0.3 ppm. Good alignment was also observed for the emission profiles as the example in Figure 18 illustrates.

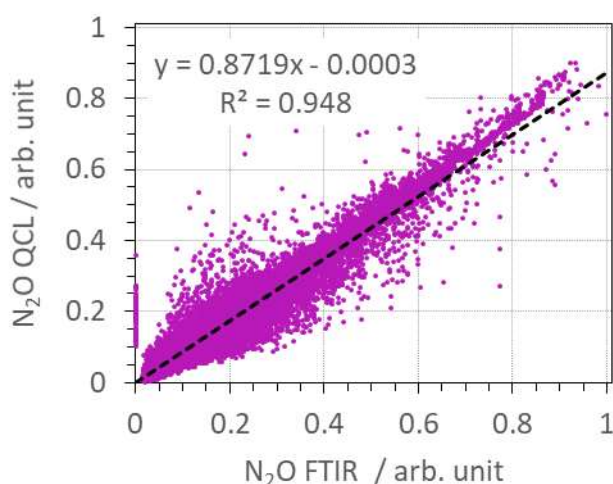


Figure 17. Correlation between N₂O concentration measured in the FTIR and by the QCL-IR over all the cycles tested.

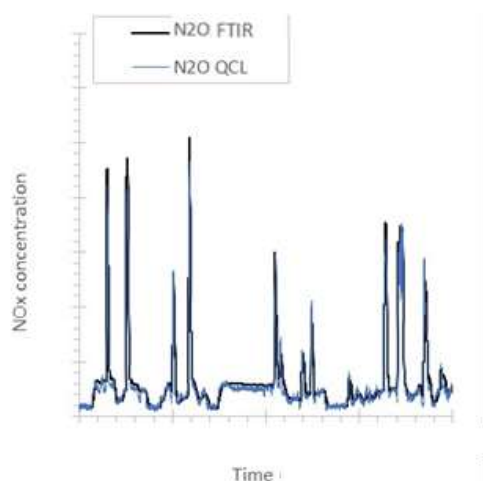


Figure 18. Example of N₂O emission profile measured by the FTIR and the QCL-IR.

The HCHO emissions were unsurprisingly very low at the level of the FTIR limit of detection (~1 ppm) with the maximum concentrations measured being 2 ppm. While other H₂ grades can contain HCHO, the Grade D used in this study is of high level of purity of H₂. H₂ emissions

were on average ~30 ppm. The repeatability of the emissions was ~1% for the WHTCh, PEMS and WHSC tests and 14% for the WHTCc.

Figure 19 plots SPN_{10} concentrations with the reference instrument SPCS, the reference APC and the PEMS OBS. The PEMS instrument's slopes were 1.81 ± 0.56 , with the lower values for the high emitting cycles. The R^2 of the slopes in most cases were >0.94 , except for the PEMS cycles that were around 0.8. For comparison, the second reference versus the APC reference had slope of 1.66 ± 0.12 and slopes >0.92 in all cases. The results indicate that the APC reference might have had calibration issues. The behaviour of the PEMS is within the expected performance.

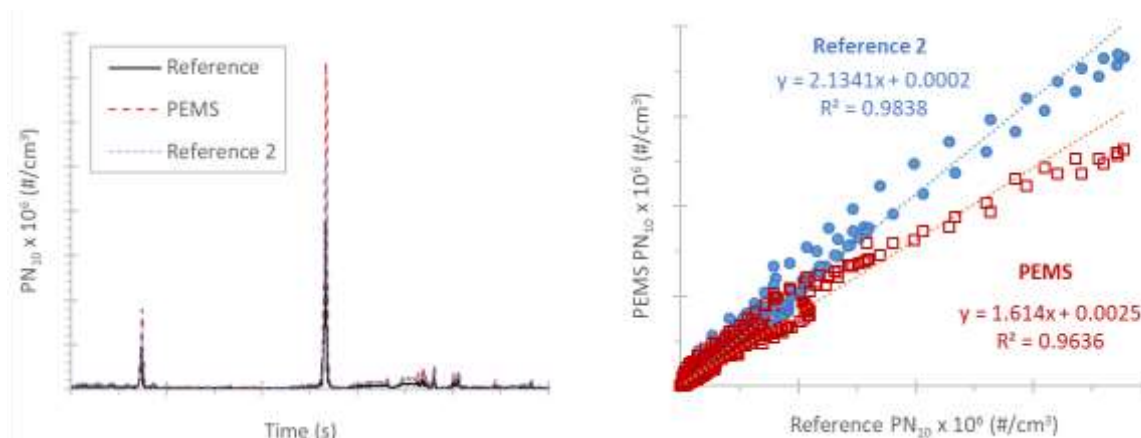


Figure 19. (Left panel) SPN_{10} concentrations over a part of cWHTC. (Right panel) correlation of PEMS and second reference vs reference for the same cycle.

4. Conclusions

A prototype lean-burn spark ignition hydrogen heavy-duty engine was tested over the regulatory cycles: WHSC, WHTCcold, WHTChot and also over an In-service conformity-like test at 23°C. The engine was equipped with a series production Diesel after-treatment system that included a DOC+SCR+ASC, (the DPF was removed). Emissions of NO_x (NO and NO₂), CO, CO₂, HC, CH₄, NH₃, PM, SPN_{23} , SPN_{10} , N₂O, H₂O and O₂ were measured with a series laboratory grade or PEMS instrumentation.

Overall, the concentrations of all regulated gases were low (NO_x), very low (NH₃ and CO₂) or even close to the detection limit the instruments (HC, CH₄ and CO). The results suggest that criteria pollutant emissions, CO₂ and particulates can reliably be measured using the analysers currently used for the type-approval of heavy-duty engines in the laboratory.

Given the low levels reached by CO, CH₄ and HC, interferences could be noticed for these pollutants. However, these interferences were at the level of the tolerances currently allowed by the UNR 49.

The tests were highly repeatable in most cases. Not surprisingly the WHTCcold tests presented the highest deviations among all tests.

The same pollutants were measured using a wet-based PEMS that included 23 nm and 10 nm CPC for the measurement of SPN and good correlations were observed. The PN PEMS was measuring on average almost twice the reference instrument, but this difference was on the same level with a second reference, indicating that the reference instrument needed recalibration.

When emissions of gaseous pollutants were non-negligible, good correlations between concentrations measured by laboratory-grade instruments and PEMS were also observed. The correlation between PEMS and laboratory was poor for CO₂ emissions. This could in part

be attributed to the low levels measured (average concentration was 550 ppm) and the fact that the span gas used on the PEMS had an unsuitable mixing ratio for this application (8%), indicating the importance of using dedicated concentrations as requested by the regulation for criteria pollutants.

Good correlations were also registered between FTIR and QCL-IR for NH₃ and N₂O measurements.

5. References

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