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Poster Extended Abstract Form

Name of Author…Simone Casadei*
Co-Authors…Davide Faedo* Francesco Avella

Affiliation:*INNOVHUB-SSI, SSC Division
Mailing address:.Via Galileo Galilei 1, 20097 San Donato Milanese – (MI) Italy
Phone / Fax…+39 02 51604249 / +39 02 514286…. E-mail…casadei@ssc.it

Nanoparticle size distribution, soot and ammonia emissions from a NGVs fleet

1. Background

In Italy natural gas vehicles (NGVs) constitute more than 50% of the European NGVs fleet and recently a significant increase of conventional fuels costs and environmental awareness led to further favorable conditions towards the use of natural gas as automotive fuel. Natural gas is in fact considered as an alternative "bridge fuel" from the conventional fuels to the supply of vehicles using electricity generated from low or no environmental impact sources (solar, wind, hydro …). Some scenario analysis predict a significant growth in the European NGVs market, also related to the bio-methane production development [1]. Many vehicles manufacturers have recently produced new natural gas models, in order to meet not only the European demand but also the one of other NGVs most successful markets (e.g., Iran, Pakistan, Argentina, Brazil, India, China) [2].

2. Aims

In literature only few data are available on particles, PM soot fraction and ammonia (atmospheric secondary aerosol precursor) exhaust emissions by recent technology engine vehicles and these are mainly related to the comparison between NGVs and conventional fuelled vehicles [3].
In the reported project a fleet of seven bi-fuel vehicles (from EURO 2 to EURO 5) was tested and the gasoline/NG fuelling associated emissions were compared. A focus is reported about the particle emissions related to a typical European urban driving condition (cold starts and stop&go speed profile), to evaluate the potential consequences of a diffusion of a NGVs fleet in an urban area in terms of negative effects on air quality and human health.

3. Materials and test method

In Table 1 the NGVs main characteristics are reported. All vehicles engines were equipped with phased sequenced MPI gas supply systems and three-way conversion catalysts (TWC). During tests each vehicle was powered with the gasoline available at that time, taken as a reference fuel, and with the natural gas compressed in the cylinder. Since the periods of each vehicle testing were strongly timely distant, it has not been possible to employ the same gasoline and the same natural gas for all. Samples of reference gasoline of the NGVs were taken from the respective tanks for their analytical characterization: they were all compliant with the EN 228 specification [4]. Instead, the chemical composition and the main physical properties of natural gas used during each vehicle testing were obtained from the analysis reports provided by ENI Gas & Power and reported in Table 1.

Table 1 - Main vehicles characteristics and corresponding gaseous fuels composition

Vehicle Model		Fiat Marea Bipower	Fiat Doblò Bipower	Fiat Panda Natural Power	VW Touran EcoFuel	Fiat Doblò Natural Power	Fiat Multipla Natural Power	Fiat Grande Punto Natural Power
ID code		A	B	C	D	E	F	G
Emission Homologation Category		EURO 2	EURO 3	EURO 4	EURO 4	EURO 4	EURO 4	EURO 5
Tests performance period		Feb-05	Oct-07	Jul-10	Aug-10	Apr-11	Apr-12	Oct-11
Accumulated mileage (km)		97000	30500	10600	15350	15400	54058	18900
Displacement (cc)		1581	1596	1242	1984	1596	1596	1368
Max power (kW@rpm) Gasoline		76 @ 5750	76 @ 5750	44 @ 5000	-	76 @ 5750	76 @ 5750	57 @ 6000
Max power (kW@rpm) Natural Gas		68 @ 5750	68 @ 5750	38 @ 5000	80 @ 5400	68 @ 5750	68 @ 5750	51 @ 6000
Characteristics of natural gas inside the gaseous fuel tank at the test time								
GCV	KJ/Sm^3	38620	38460	38738	38305	38218	38691	38527
LCV	KJ/Sm^3	34821	34683	34961	34517	34490	34908	34739
LCV	kJ/kg	48188	47187	45680	48999	44894	46399	47641
Wobbe Index		-	49660	49018	50522	48268	49379	49944
density [kg/Sm^3]	kg/Sm^3	0.723	0.735	0.765	0.704	0.768	0.752	0.729
methane	% vol	94.2	92.77	89.08	96.83	88.42	90.6	93.64
ethane	% vol	2.980	3.390	5.240	1.520	4.520	4.490	3.130
propane	% vol	0.710	0.789	1.072	0.483	1.126	0.943	0.792
isobutane	% vol	0.100	0.120	0.151	0.078	0.196	0.165	0.076
nbutane	% vol	0.120	0.124	0.165	0.082	0.211	0.146	0.084
isopentane	% vol	0.020	0.027	0.040	0.016	0.054	0.040	0.030
npentane	% vol	0.020	0.020	0.035	0.012	0.044	0.037	0.025
hexanes	% vol	0.030	0.024	0.052	0.011	0.059	0.055	0.027
CO_2	% vol	0.260	0.836	1.713	0.159	1.424	1.153	0.777
N_2	% vol	1.530	1.867	2.413	0.799	3.912	2.342	1.402
He	% vol	0.030	0.025	0.039	0.011	0.036	0.036	0.023
H_2	% vol	-	0.000	0.000	0.000	0.000	0.000	-
O_2	% vol	-	0.000	0.000	0.000	0.000	0.000	-
CO	% vol	-	-	0.000	0.000	0.000	0.000	-

By comparing the natural gas compositional characteristics, the minimum methane content was detected in the gaseous fuel that fed the vehicle E (88.42 %vol), while the highest was found in the vehicle D fuel (96.83 %vol). Sensitive oscillations were also found for the higher homologues hydrocarbons content. Despite the different chemical composition, the seven natural gases were characterized by a poorly differentiated energy content, i.e. both upper and lower heating contents had approximately the same value.
The laboratory scheme is reported in Figure 1.

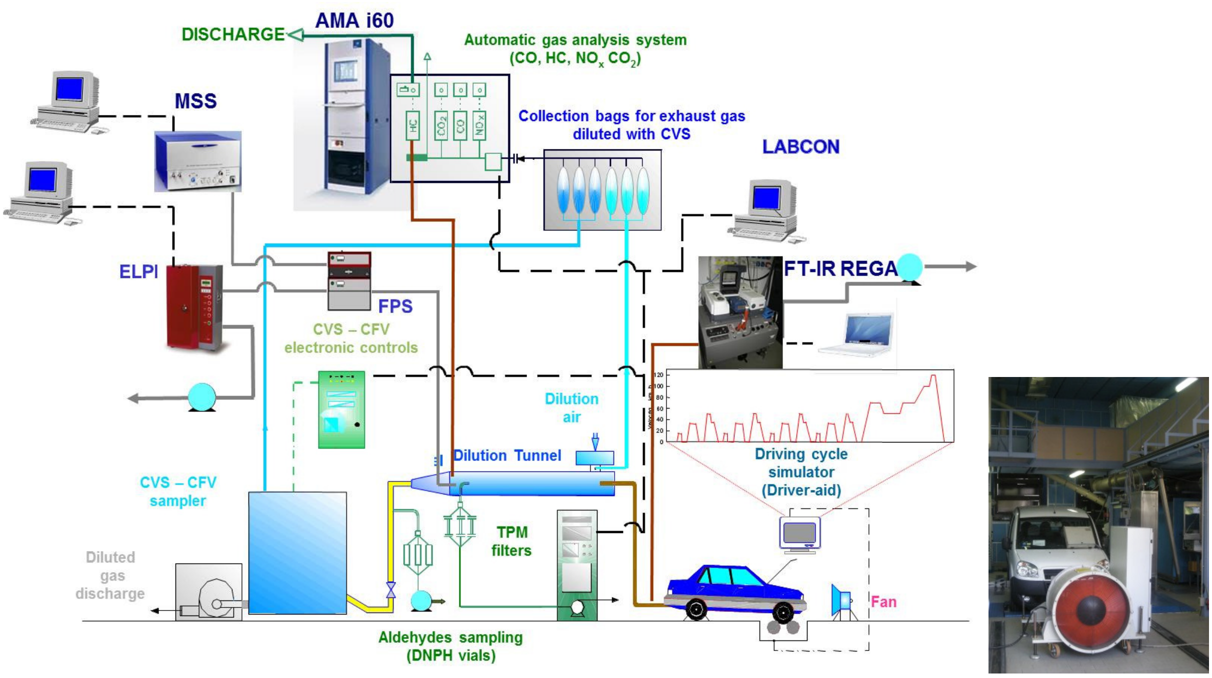

Figure 1 - Exhaust sampling and emissions analysis system

PM and its dimensional characterization, soot and ammonia emissions were determined during chassis dyno standard tests (CVS-CFV dilution tunnel system according to the UN ECE Regulation N.83) following a NEDC + CADC Urban driving cycles sequence. Regulated gaseous emissions were also determined. The test protocol considered four repetitions of cold start cycles sequences performed for each of the two fuels in order to verify the repeatability of the measurements. For each parameter (regulated and unregulated emissions) the statistical significance of the variation between the gasoline and the natural gas fuelling was determined through a 95% confidence t-Student test.

Total PM was sampled on conditioned Pallflex TX60A20 membranes; the emitted particles number (PN) and their dimensional distribution were measured by an Electrical Low Pressure Impactor (ELPI Dekati) in the 7 nm ÷ 9.6 μm aerodynamic diameter (Dp) range by using a suitable probe and a further dilution (with FPS Dekati system). A further part of the diluted exhausts was analysed in order to evaluate the particulate soot fraction (by Microsoot Sensor AVL). On-line ammonia emission was detected only for five vehicles, using a FT-IR Nicolet 6700 integrated with the REGA ThermoFisher automotive module.

4. Test results

Nearly all tested vehicles respected the Type I standard emissions homologation limits when fuelled with both gasoline and the gaseous fuel. The few data exceeding the standard limits were complying with the permitted tollerances for the in-use vehicles.

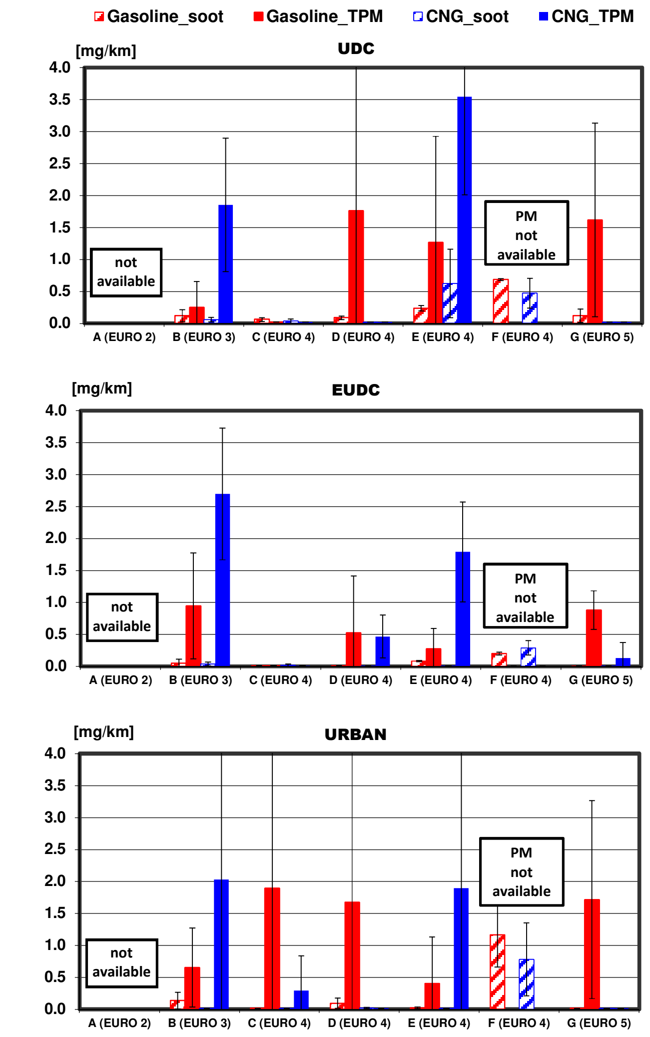

Figura 2 – TPM vs PM soot emissions for the fleet vehicles (tests average data with std. dev.)

In general for all the tested vehicles a significant reduction in the CO_2 emission and both mass and energetic fuel consumption reductions were detected by shifting the vehicle feeding from gasoline to natural gas.

Total particulate matter (TPM) emission data were not available for vehicles A (Euro 2) and F (Euro 4). For the other test vehicles the emitted quantity of TPM was very low and as a consequence sometimes not determined with the conventional gravimetric method. Because of the very small TPM sampled quantities, measurements were affected by a high variability, especially in the Urban driving cycle, when the engine and the TWC reached the thermal regime.

A decrease in the TPM emission was observed for almost all vehicles when switching from gasoline to natural gas fuelling. As observed in other experimental works [5], most of the particulate can be considered consisting of volatile substances given the very low level of soot fraction lube oil originated (Figure 2). Vehicle B (Euro 2) and vehicle E (Euro 4) emitted a greater TPM quantity when fuelled with NG. This may be due to a not optimized engine setup of the NG fuelling system of the two vehicles. The differences in the TPM emissions by replacing gasoline with NG were statistically significant only for vehicle G (Euro 5), given the high measurements variability for the other vehicles.

A significant variability was observed also for the PM soot fraction data due to the low emission levels detected, but a PM soot emission reduction was generally detected by switching from the gasoline to the NG fuelling. In the UDC warm up phase the most important soot emissions were mainly detected for almost all vehicles compared to emission levels measured in the

EUDC and Urban cycles. PM soot emission was very high during the first 20 seconds, being associated to the cold start gasoline fuelling for all tested vehicles. In fact just after about 20 seconds, when the engine and the TWC were not yet thermally conditioned, PM soot fraction emissions were lower for all the tested vehicles except vehicle F (Euro 4), whose soot emissions were significant in EUDC and Urban cycles, too. In the EUDC driving cycle the highest soot emissions were generally observed during the 100 km/h to 120 km/h acceleration phase both with gasoline and NG fuelling. In the Urban cycle a lower soot emission was observed with the NG fuelling, with the peaks detected during the most aggressive acceleration phases for all tested vehicles. Figure 3 shows the modal soot emission for vehicle D (Euro 4) as an example.

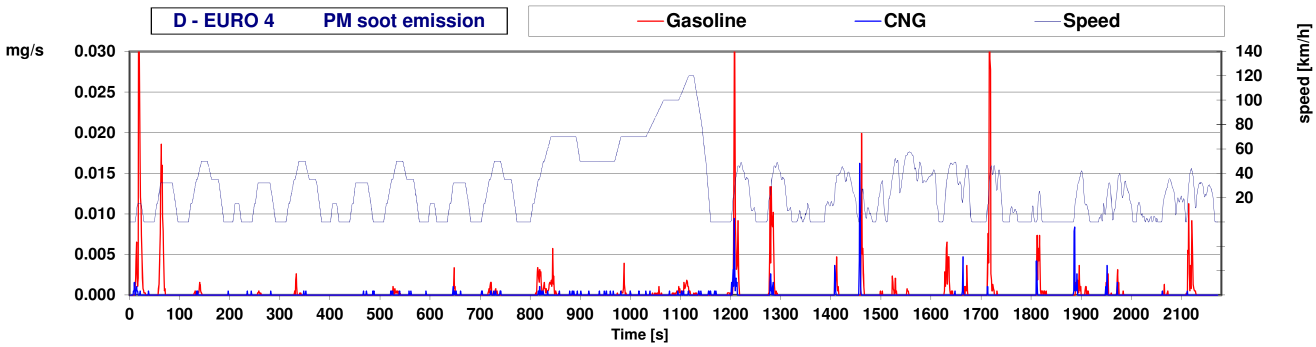

Figure 3 - Modal PM soot fraction emission - vehicle D (EURO 4)

Total particle number (PN) and particles distributions were measured with 1 Hz frequency during the NEDC + CADC Urban cycles sequence for all vehicles. PN emission levels ranged from 10^{10}÷ 10^{12} #/km, depending from vehicle models, driving conditions and, to a lesser extend, from the type of fuel (gasoline, NG). However PN emissions were at least two orders of magnitude lower than those from diesel vehicles without DPF tested in the same conditions [5, 6].

A sensible reduction in the PN emission was observed for most vehicles in urban driving conditions, when gasoline was replaced with natural gas. Differently in extra-urban driving conditions for four vehicles an increase of the number of emitted particles was observed shifting from gasoline to natural gas fuelling. The higher PN emission peak in these driving conditions was detected in the acceleration phase up to 120 km/h. The very high PN emission was probably due to other sources than the fuel combustion, such as lubricating oil or catalyst degradation. This was also confirmed by the similar particle distribution profiles with gasoline and natural gas fuelling.

A focus on soot, nanoparticles and ultrafine particles modal emissions detected for vehicles C (Euro 4) and G (Euro 5) during the UDC warm-up phase is shown in Figure 4 (average data reported). The emissions trends were similar for the two vehicles, with both the gasoline and the NG fuelling, but a significant prevalence of these three species emissions was observed with the gasoline one. After the cold start, the emissions related to the NG feeding rapidly decrease and in the first UDC acceleration these are mainly related to the gasoline starting supply that occurs before the effective NG shifting. With the NG fuelling the aerodynamic diameters of the emitted particles were mainly below 40 nm while, with the gasoline one, 40 nm <Dp< 144 nm particles emissions were more significant, especially during the first UDC acceleration phase. A noticeable PM soot emission peak (> 140 µg/s) was detected few seconds after the cold start of the Euro 5 vehicle, gasoline fuelled.

In all test conditions the particle distribution profile was unimodal. In urban driving conditions the aerodynamic diameter of the majority of the particles emitted from all test vehicles was around 70 ÷ 140 nm, while in EUDC it was shifted towards below 30 nm, independently of the fuels.

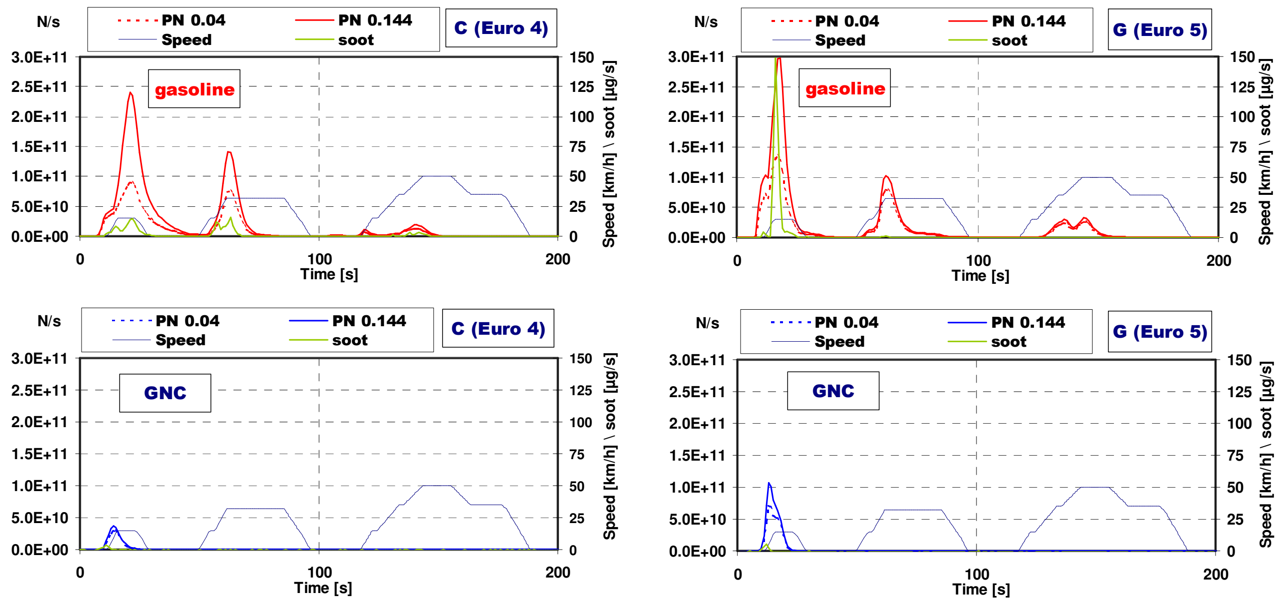

Figure 4 - Comparison between soot, nanoparticles (Dp< 40 nm) and ultrafine particles (Dp < 144 nm) emissions – UDC warm-up phase (vehicles C and G)

Given the interest in the environmental and health effects due to the use of natural gas, which are supposed to be minor than those associated to the use of gasoline and diesel fuel in transport in a urban context (where a pollution or a congestion charge is active, i.e. the Milan Area C [7]), a focus on nanoparticles and ultrafine particles emissions is reported for the UDC warm-up phase (Figure 5) and for the CADC Urban cycle (Figure 6). These operating conditions are representative, respectively, of the most important emission phase (the engine cold start) and of an effective real urban driving in the European context.
During the engine warm-up PN decreased for four vehicles (up to an order of magnitude for vehicle C), when they were powered by natural gas rather than gasoline and the observed decrease affected all the twelve size classes detectable by ELPI; it was comparable for the other vehicles. The emission peak was around 70 nm for almost all tested vehicles; only for vehicle A (Euro 2) a stronger emission was found for the particulate fraction detectable in the ELPI last stage (Dp < 20 nm), regardless of the used fuel. Only for vehicle E (Euro 4), when natural gas fuelled, it was detected a greater emission in all particles sizes. The examination of the cumulative Dp distribution curves indicated that 95% of the particles emitted from the majority of tested vehicles had an aerodynamic diameter less than or equal to 140 nm. For only vehicles A (Euro 2) and G (Euro 5), 95% of the emitted particles had an average size below 70 nm.
In the cold start phase (first UDC subcycle) the number of emitted nanoparticles and ultrafine particles in the exhaust gas was at least double for all vehicles fed with NG and gasoline except for the vehicle F (Euro 4) in comparison with the hot phases of the subsequent driving cycles. Moreover most of the ultrafine particles Dp was lower than 40 nm when vehicles were fuelled with both gasoline and natural gas. However the nano and ultrafine particles emission level was visibly lower when five of the seven vehicles were fuelled with the gaseous fuel (Figure 5).
Under CADC Urban driving conditions the size distribution curves were similar to the UDC ones, i.e. unimodal with an emission peak in the 70-140 nm range for almost all vehicles fed with both fuels. The emission level was higher in all size classes with the gasoline supply than with natural gas for vehicle C (Euro 4) and E (Euro 4), while no significant difference was found for all the others.
Even in the CADC Urban cycle 95% of the emitted particles by all vehicles, regardless of the fuel, had an aerodynamic diameter lower than 70 nm. Except for the vehicle E (Euro 4), whose nanoparticles emitted fraction was greater when it was fuelled with natural gas, all other vehicles presented overlapping cumulative emission profiles with both fuels and the Dp distribution profiles were practically similar one to the other.

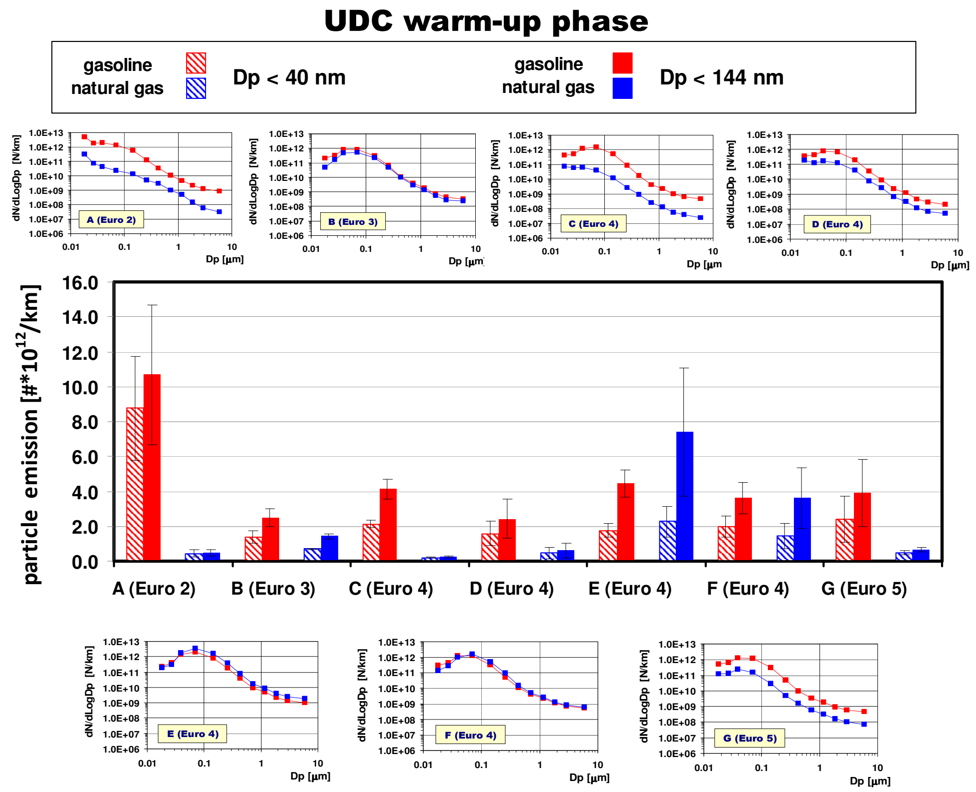

Figure 5 - Nanoparticles and ultrafine particles (Dp< 144 nm) NGVs fleet emissions with each vehicle particle distribution comparing gasoline and NG fuelling - UDC warm up phase

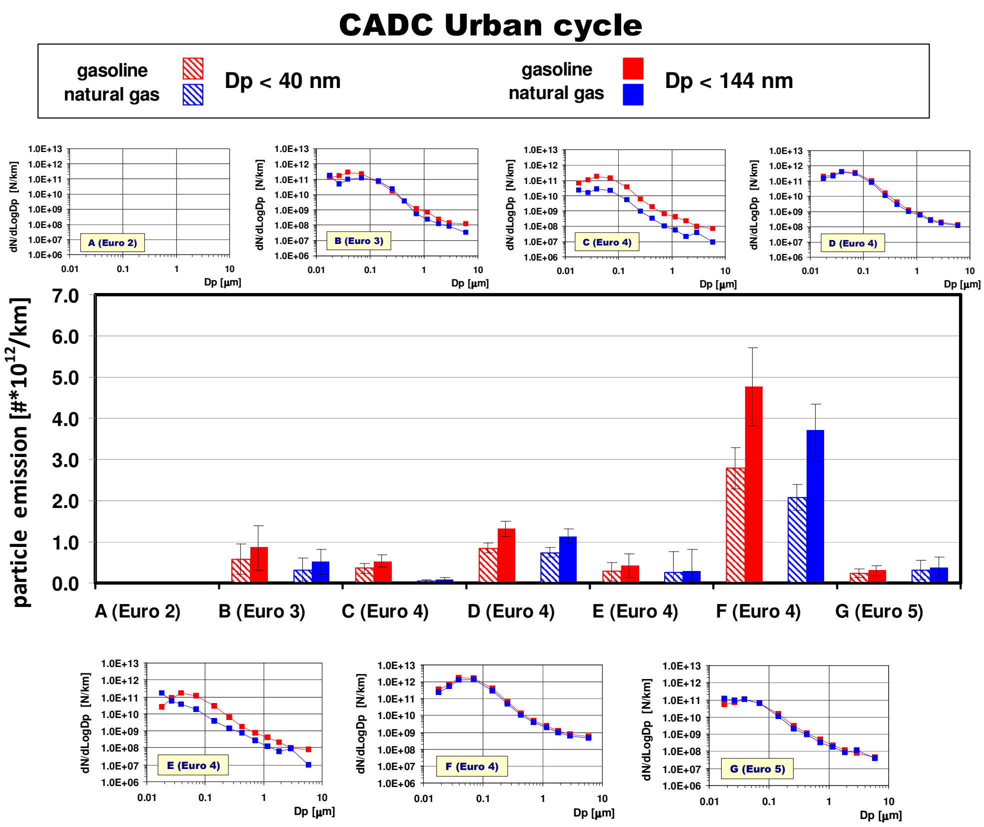

Figure 6 - Nanoparticles and ultrafine particles (Dp< 144 nm) NGVs fleet emissions with each vehicle particle distribution comparing gasoline and NG fuelling - CADC Urban cycle

Under real urban driving conditions, with thermally stabilized engines, for almost all test vehicles (except vehicle F) the ultrafine particles emitted with both fuels were mostly composed of nanoparticles. Moreover a slight decrease of nano and ultrafine particles was detected by switching from gasoline to natural gas (Figure 6).

Very few informations are available in literature regarding gasoline/NG passenger cars ammonia (NH_3) emissions [8,9] suggesting this chemical species generates in catalytic devices during speed transients (acceleration) engine operating conditions, requiring a temporary enrichment of the air/fuel mixture. Other possible causes are related to the effects of aging of this parameter control system (lambda probe) and of the catalyst itself.
In all driving conditions the detected NH_3 emission level was quite variable between the tested vehicles.
UDC NH_3 emissions for all vehicles, except vehicle C, were significantly lower when they were fed with natural gas instead of gasoline, these reductions ranging between 40 and 95%. Conversely, EUDC and CADC Urban emissions increased when vehicles were fed with natural gas compared to gasoline feeding, except vehicle D whose NH_3 emission was significantly lower when fed with the gaseous fuel.
Due to the strong variability of the NH_3 measured values, only in few cases the variations were found statistically significant, suggesting that more tests are necessary.

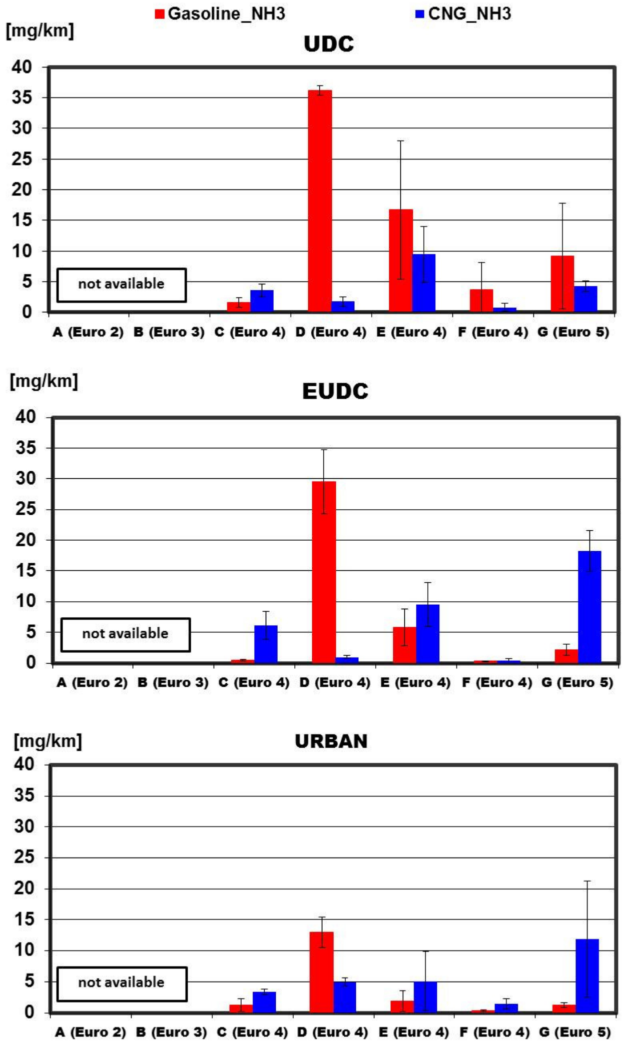

Figure 7 – Ammonia emissions for the fleet vehicles (tests average data with std. dev.)

5. Conclusions

A fleet of seven bi-fuel vehicles (from EURO 2 to EURO 5) was tested and the gasoline/NG fuelling particle distribution, soot and ammonia associated emissions were compared driving NEDC + CADC Urban driving cycles.
A PM soot emission reduction was generally detected by switching from the gasoline to the natural gas fuelling: the main peaks were detected during the cold start gasoline fuelling for all tested vehicles, no significant differences between the two fuels were detected in the EUDC cycle while in the Urban cycle a lower soot emission was observed with the natural gas fuelling.
When gasoline was replaced with natural gas a sensible reduction in the PN emission was observed for most vehicles in urban driving conditions while in extra-urban driving conditions for four vehicles an increase was noticed. A focus on particle emissions related to urban driving conditions pointed out a particle emission reduction during the engine warm-up phase shifting from gasoline to natural gas feeding in all the twelve size classes detectable by ELPI and for almost all the vehicles (with the emission peak around 70 nm); also with thermally stabilized engines and in real driving conditions, for almost all test vehicles, a slight decrease of nano and ultrafine particles was detected by switching from gasoline to natural gas.
For ammonia more tests were found to be necessary for a better understanding, but anyway the UDC NH_3 emissions for all vehicles, except one, were found to be significantly lower when they were fed with natural gas (reductions ranging between 40 and 95%).

With a suitable engine set-up in urban driving conditions the natural gas feeding, compared to the gasoline one, was shown to have lower emissions in terms of nano and ultrafine particles, soot and ammonia.

Acknowledgements
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NANOPARTICLE SIZE DISTRIBUTION, SOOT AND AMMONIA EMISSIONS FROM A NGVS FLEET

Simone Casadei, Davide Faedo, Francesco Avella

Innovhub – Stazioni Sperimentali per l'Industria, Divisione SSC Viale A. De Gasperi 3 – 20097 San Donato Milanese, Milano – Italy

Contact: casadei@ssc.it

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Test vehicles and natural gases characteristics

Vehicle Model		Fiat Marea Bipower	Fiat Doblò Bipower	Fiat Panda Natural Power	VW Touran EcoFuel	Fiat Doblò Natural Power	Fiat Multipla Natural Power	Fiat Grande Punto Natural Power
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ethane	% vol	2.980	3.390	5.240	1.520	4.520	4.490	3.130
> C2 HCs	% vol	1.000	1.104	1.515	0.682	1.690	1.386	1.034
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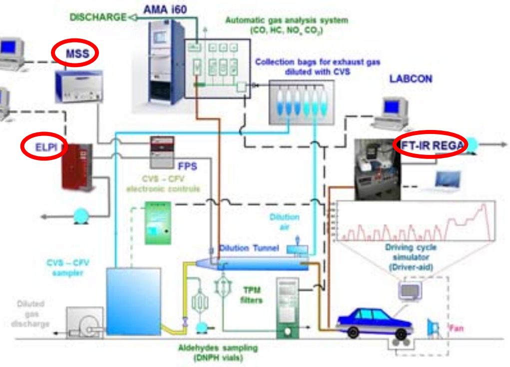
H_2	% vol	-	0.000	0.000	0.000	0.000	0.000	-
O_2	% vol	-	0.000	0.000	0.000	0.000	0.000	-
CO	% vol	-	-	0.000	0.000	0.000	0.000	-

MATERIALS AND METHODS

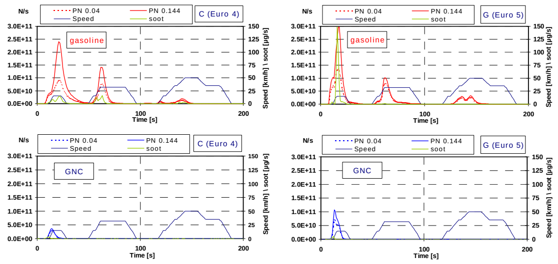
- 7 bi-fuel vehicles (from Euro 2 to Euro 5) all with MPI and TWC systems
- Test gasoline samples were all EN 228 specification compliant [3]
- NEDC + CADC Urban chassys dyno driving sequence
- 95% t-Student test for statistical significance of gasoline/NG emission variations (4 tests each vehicle/fuel)

Exhaust sampling and emissions analysis system



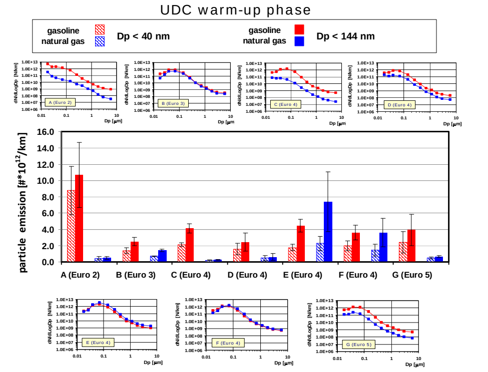
RESULTS: NANOPARTICLES (NP), ULTRAFINE PARTICLES (UFP), SOOT EMISSIONS

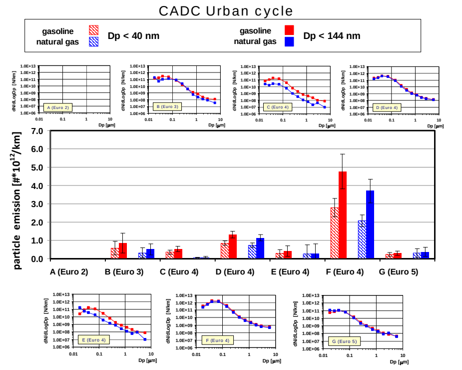
NP (Dp < 40 nm), UFP (Dp < 144 nm) and soot emissions in UDC warm-up phase - vehicles C (Euro 4) and G (Euro 5)



- In the first seconds after engine starting, particle emissions were always related to gasoline feeding
- Significant prevalence of NP, UFP and soot with gasoline
- NG fed vehicles emitted particles with Dp mainly < 40 nm
- Lower particle emissions in 40 ÷ 144 nm Dp range with NG
- A noticeable soot peak after the Euro 5 vehicle cold start, when fuelled with gasoline

NP and UFP NGVs fleet emissions and dimensional distribution: NG vs. gasoline



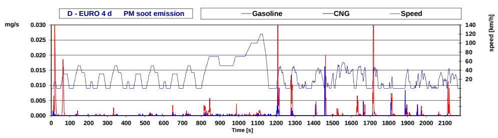


- PN emissions decreased for four vehicles NG fuelled in the Dp full range, measurable by ELPI
- PN emission peak at Dp ~ 70 nm for almost all vehicles
- Most of the UFP had Dp < 40 nm with both gasoline and NG feeding
- NP and UFP emission level was visibly lower when five vehicles were fuelled with NG

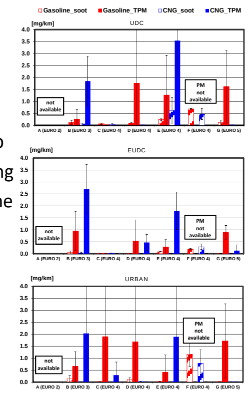
- Lower PN emission with NG for vehicles C and E, no significant difference for all the others
- PN emission peak ranged in 70 ÷ 140 nm Dp for almost all vehicles
- Slight decrease of NP/UFP emissions with NG feeding
- Most of the UFP had Dp < 40 nm with both gasoline and NG feeding

RESULTS: SOOT

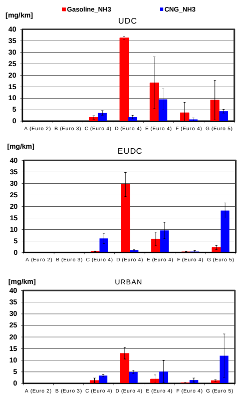
- Significant variability of total particulate matter (TPM) and soot emissions (low levels)
- TPM (mostly semi-volatile substances [4]) decreased with NG except for vehicles B and E → not optimized NG set-up
- Soot emission reduction generally detected with NG feeding
- Very high soot emission in the warm-up phase with gasoline
- Focus on vehicle D: highest soot emissions in warm-up phase and in Urban cycle, gasoline fed



Modal soot fraction emission - vehicle D (Euro 4)



TPM vs soot average emissions for the fleet vehicles



Ammonia average emission for the fleet vehicles

RESULTS: AMMONIA

Very few informations found in literature [5; 6] suggesting NH_3 generates in catalytic devices during speed transients (acceleration) engine operating conditions, that require a temporary enrichment of the air/fuel mixture. Other possible causes are related to the effects of aging of the lambda probe (air/fuel controller) and of the catalyst itself.

- In all driving conditions NH_3 emission level was quite variable between the tested vehicles
- NH_3 emission in UDC was significantly lower with NG for all vehicles except C: reduction range 40% ÷ 95%.
- NH_3 emission in EUDC and CADC Urban cycles increased with NG feeding, except vehicle D
- Due to the strong variability of measures (no statistical significance), more tests were found to be necessary

CONCLUSION

With a suitable engine set-up in urban driving conditions the natural gas feeding, compared to the gasoline one, was shown to have lower emissions in terms of nano- and ultrafine particles, soot and ammonia.

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