Exhaust Gas Particle Number Measurement.
Round Robin Test Using ELPI.

E. Zervas*[1], P.Dorlhène[1], L. Forti[2], C. Perrin[2], J.C. Momique[3], R. Monier[3], H. Ing[4], B. Lopez[4]

[1]Renault, 1, Allée Cornuel - 91510 Lardy - France,

[2]Institut Français du Pétrole (IFP), 1 et 4 Avenue du Bois Préau - 92500 Rueil-Malmaison - France

[3]PSA Peugeot Citröen - 18, rue des Fauvelles - 92250 La Garenne-Colombes - France

[4]Union Technique de l'Automobile, du Motocycle et du Cycle (UTAC), Autodrome de Linas-Montlhéry - 91310 Montlhéry - France

1. INTRODUCTION

Current European regulations demand for a gravimetric measurement of particles emitted from Diesel passenger cars. The Particle Measurement Programme (PMP) of the Working Party on Pollution and Energy (GRPE) of the United Nations at Geneva, is mandated to work on the development of an improved method for the particulate matter (PM) measurement, which can include, if feasible and necessary, the measurement of the particle number of the exhaust particles. This article presents the results of the French PMP subgroup, composed by IFP, PSA Peugeot-Citroën, Renault and UTAC, of an inter-laboratory comparison of the passenger cars particle number measurement using ELPI, which was, at the moment this study has been curried out, the only commercially available instrument that can determine both particle number and size (but with a low size resolution). Three Euro3 passenger cars are used in this study. A pragmatic protocol, using the European regulatory conditions (tests on the NEDC) is used. The intra-laboratory variability, the repeatability and the reproducibility are presented in the case of the three vehicles. The CO_2, fuel consumption (FC) and regulated pollutants are also measured, and their repeatability and reproducibility are compared with these of the particle number. Other particular aspects that are not very well established today, as calibration of the analytical instruments, are not treated here.

2. EXPERIMENTAL SECTION

The passenger cars used for this study were: a Renault Mégane 1.6L gasoline PC operating under stoichiometric conditions, a Citröen 307 2.0L Diesel PC and a Citröen 307 2.0L Diesel PC equipped with DPF. All vehicles were fed with fuels with less than 10 ppm of sulphur and the same lubricant (<0.4% of S). Three tests were performed on the European Driving Cycle (cold start). Number and size distribution measurements were performed using a DEKATI ELPI, with a sampling of 10 L/min (three labs, while the lab1 used 20 L/min). An ejector type dilutor heated at 130°C with hot nitrogen or air (120°C) was used for dilution to minimise the nucleation particles. The four laboratories did not use the same set-up of the dilution tunnels: Labs 1 and 2 had the same dilution tunnel for gasoline and Diesel vehicles, Lab4 used two dilution tunnels (one for gasoline and one for Diesel vehicles), but with a common sampling point, while Lab3 had two dilution tunnels with two sampling points. The intra-laboratory variability is expressed as the RSD (relative standard deviation) of the measured values. The repeatability and reproducibility between the four laboratories are calculated following the ISO 5725 standards (with a confidence interval of 63%).

3. RESULTS AND DISCUSSION

The reproducibility RSD values of the three vehicles are 7, 11 and 58% respectively for the CO emissions, 14, 12 and 34% for the HC, 24, 6 and 9% for the NOx, 35, 15 and 84% for the PM, and less than 1.8% for the CO_2 and FC. The repeatability RSD vlues are 6, 10 and 19% respectively in the case of CO, 7, 9 and 14% in the case of HC, 23, 2 and 3% for the NOx, 31, 7 and 97% for the PM, and less than 0.8% for the CO_2 and FC. The particulate matter emissions are less repeatable than the other regulated pollutants in the case of the low emitting vehicles. The gasoline vehicle and the DPF equipped Diesel one have high RSD values due to their very low PM emissions, which are similar to the tunnel background levels.

The particle number of the tunnel background tests is around $1x10^{11}$ 1/km. These tests are not very repeatable due to the low particle number; the repeatability is also very dependent on the tests performed previously. The reproducibility RSD value is 172% while the repeatability RSD is 119%. The particle number of blank measurements is very near or even below the ELPI limits of detection (LOD) or limits of quantification (LOQ=3xLOD).

efthimios.zervas@renault.com

The mean total particle number of the gasoline vehicle is $1.3x10^{12}$ 1/km, with reproducibility and repeatability RSD values respectively of 30% and 23%. The mean total particle number of the Diesel vehicle without DPF is two orders of magnitude higher than the particle number of the gasoline vehicle: $1.3x10^{14}$ 1/km, with reproducibility and repeatability RSD values respectively of 24% and 7%. It must be noted that this vehicle is representative of the current European fleet (Euro3). The mean total particle number of the DPF equipped Diesel vehicle is lower than the two previous ones: $1.8x10^{11}$ 1/km, with reproducibility and repeatability RSD values respectively of 67 and 49%. It must be noted that the particulate emissions of this vehicle is representative of these of the future European passenger cars. The particle number of this vehicle is very close or even lower than the ELPI limits of detection or quantification, which induces high values of reproducibility. These high values of reproducibility might not be adapted for regulatory purposes.

The particle distributions of these vehicles are quite similar. Furthermore, the size distribution shows that there is no nucleation during the measurements. The particle numbers emitted by the gasoline vehicle are about one order of magnitude higher than the tunnel blanks particle numbers, up to about 300 nm. For the bigger particles, the ratio between the particles of exhaust gas and the tunnel background drops to only two. The particle number of the Diesel vehicle is 2-3.5 orders of magnitude higher than the particle number of the tunnel blank measurements, while this of the DPF equipped Diesel vehicle remains very close to the blank measurements for the entire distribution, especially in the area of the very fine particles. It is clearly shown that the particle numbers of tunnel blanks, of the DPF equipped Diesel PC and of the upper part of the gasoline PC distribution are very close or even lower to the LOD and LOQ.

The RSD of particle emissions determined by filters are, for each vehicle, higher than the other three regulated pollutants. This difference is not very important in the case of the Diesel vehicle: reproducibility of 7% and repeatability of 15% against 6-12% of the other pollutants. As the other two vehicles emit very low particle mass, these differences become quite important: 31 and 35% for the PM emissions against 6-24% for the other three pollutants in the case of the gasoline vehicle; 84 and 97% against 3-58% for the DPF equipped Diesel vehicle. The ELPI reproducibility over the entire NEDC is not very different from the reproducibility of PM emissions: 35, 15 and 84% for the PM determination against 30, 24 and 67% for the ELPI, for the three vehicles respectively; the corresponding repeatability values are 31, 7 and 97% for the PM against 23, 7 and 49% for ELPI.

CONCLUSIONS

The results of this study showed that:

- The reproducibility RSD values are 7, 11 and 58% respectively for the three vehicles in the case of CO, 14, 12 and 34% in the case of HC, 24, 6 and 9% in the case of NOx and 35, 15 and 84% in the case of particles collected on filters. The reproducibility RSD values of CO_2 emissions and fuel consumption are always less than 1.8%.

- There is no effect of the ELPI sampling volume, between 10 and 20 L/min, as the results of the Lab1 is similar to these of the other labs.

- The reproducibility RSD of the tunnel background tests is quite high (up to 172%), due to low particle numbers, very close to the ELPI detection limits. On the entire NEDC, the reproducibility of total particle number determined by ELPI is 30, 24 and 67% for the gasoline, Diesel and DPF equipped Diesel vehicle respectively. These values are quite similar to those of the mass particle determination.

- The particle number emitted from the DPF equipped Diesel engine and at the upper part of the distribution of the gasoline PC engine is very close or even lower to ELPI limits of detection (LOD and limits of quantification (LOQ).

- These results show that the protocol used in this study allows the reliable measurement of exhaust particle number in the case of vehicles emitting at least two orders of magnitude more than the tunnel background. In the other cases, the measurement variability is too high, especially for regulatory purposes.

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[1]Renault, [2]IFP, [3]PSA Peugeot Citroën, [4]UTAC

Motivation and Objectives

- Exhaust PM measurement of Euro3 and Euro4 is based on a well adapted gravimetric method
- But what about future more stringent regulations?
- PMP programme of the UN at Geneva: proposition of an improved PM measurement method, which can includes, if feasible and necessary, particle number measurements
- French PMP subgroup (IFP, PSA Peugeot-Citroën, Renault, UTAC) evaluated the ELPI performances

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    - Particle size distribution
- Conclusions

Experimental Section (1)

Vehicles used

Type	Renault Mégane	Peugeot 307	Peugeot 307
Fuel	Gasoline	Diesel	Diesel
Emission limits	Euro3	Euro3	Euro3
Displacement	1.6 L	2.0 L	2.0 L
After-treatment device	TWC	DOC	DOC+DPF

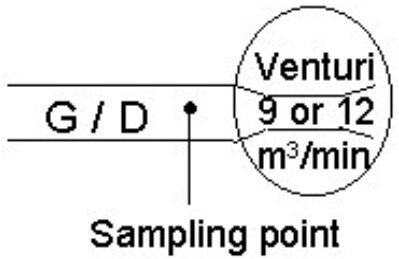
Fuel with <10ppm of S

Calculations according to the ISO 5725 standard.

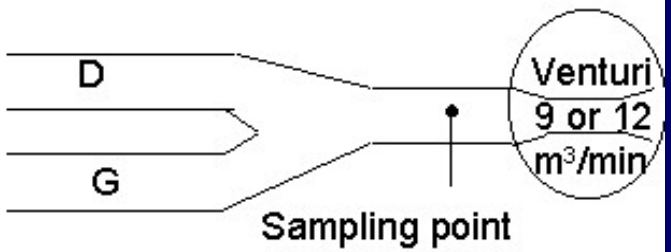
Experimental Section (2)

Sampling systems used

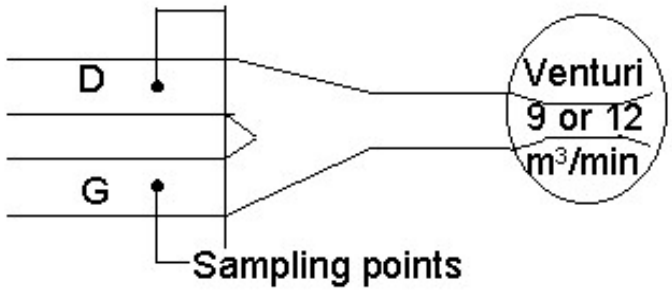
Lab1 and 2 :
Same dilution tunnel for Gasoline and Diesel
=> venturi change to obtain 9 or 12m³/min



Lab4 :
2 dilution tunnels with
commun sampling point

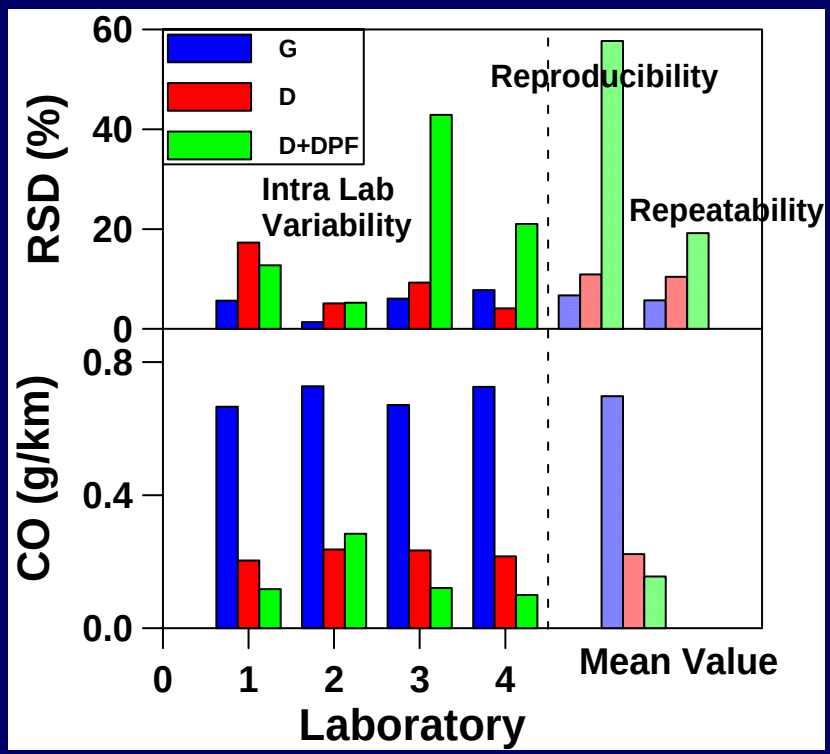


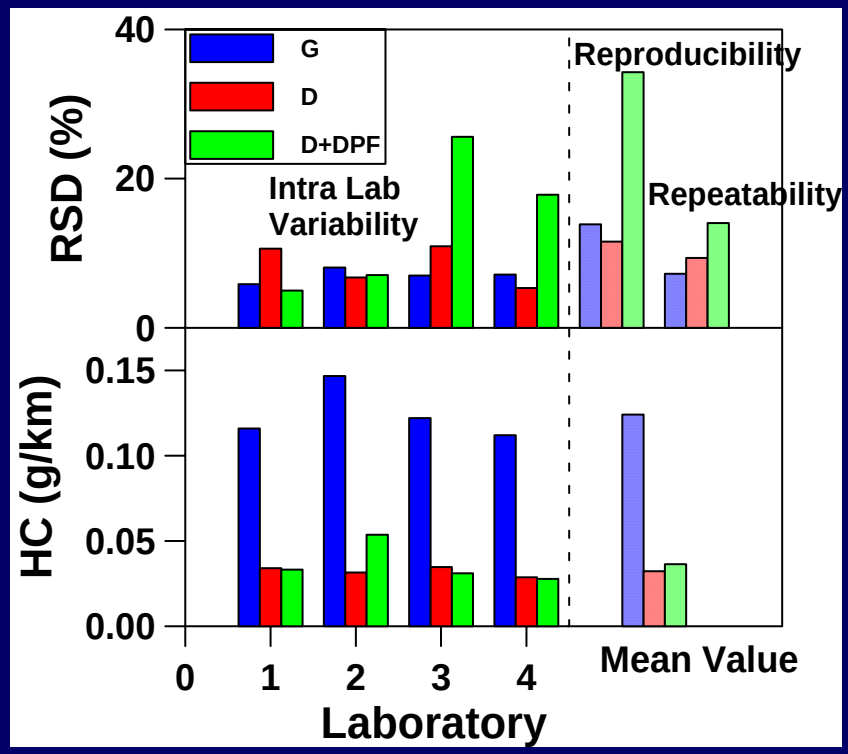
Lab3 :
2 dilution tunnels
with two different
sampling points



Results (1): *Emissions of regulated pollutants (CO, HC)*

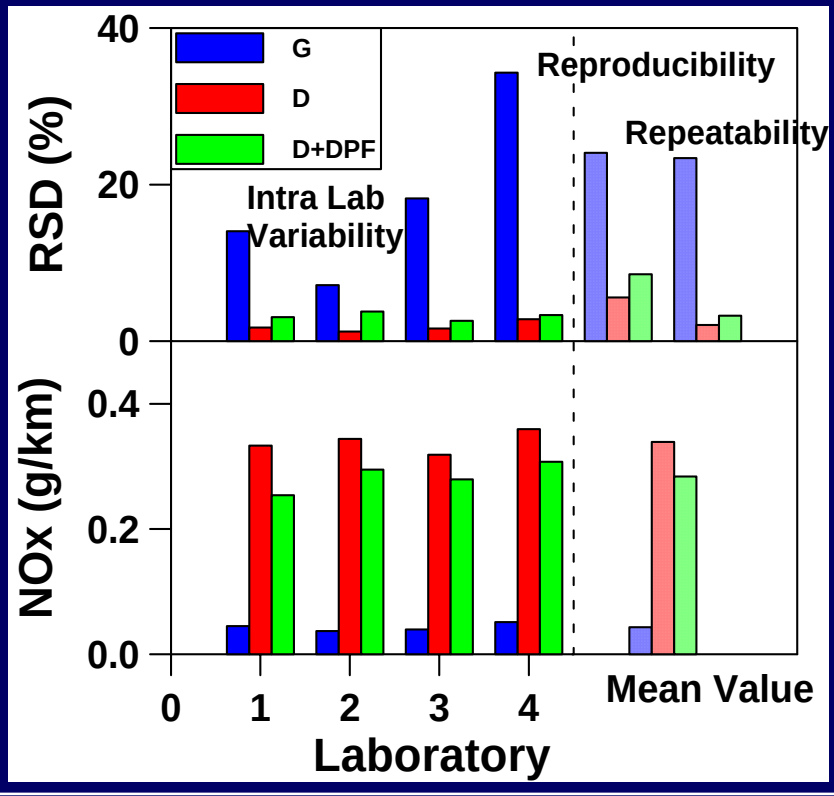
- Good agreement between labs
- Relatively low RSD reproducibility and repeatability values, except D+DPF vehicle

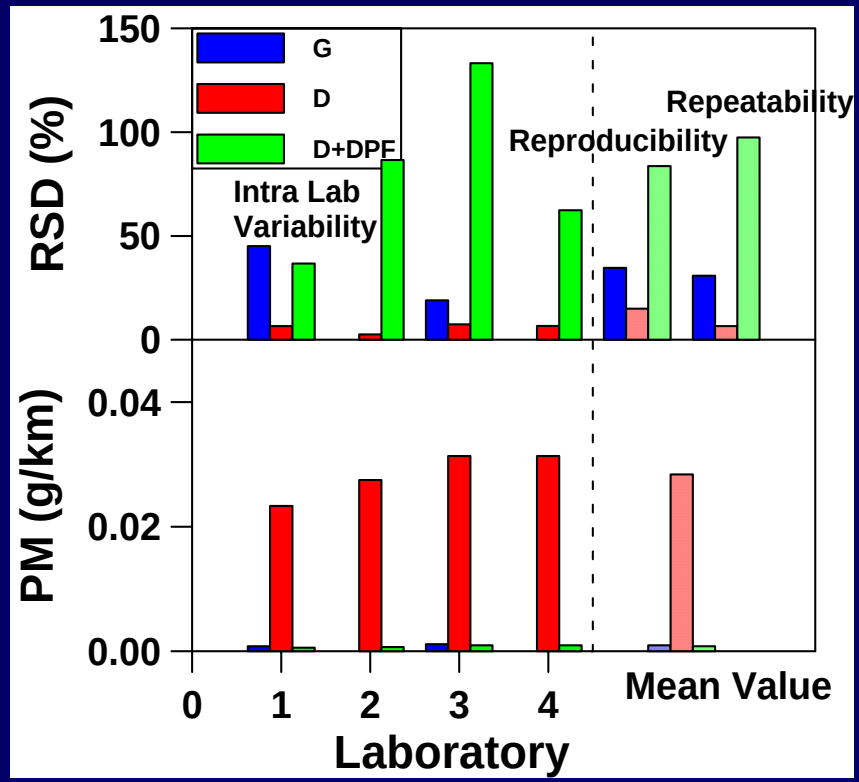




Results (2): *Emissions of regulated pollutants (NOx, PM)*

- Good agreement between labs
- Relatively low RSD reproducibility and repeatability values, except PM of the Gasoline and D+DPF vehicles



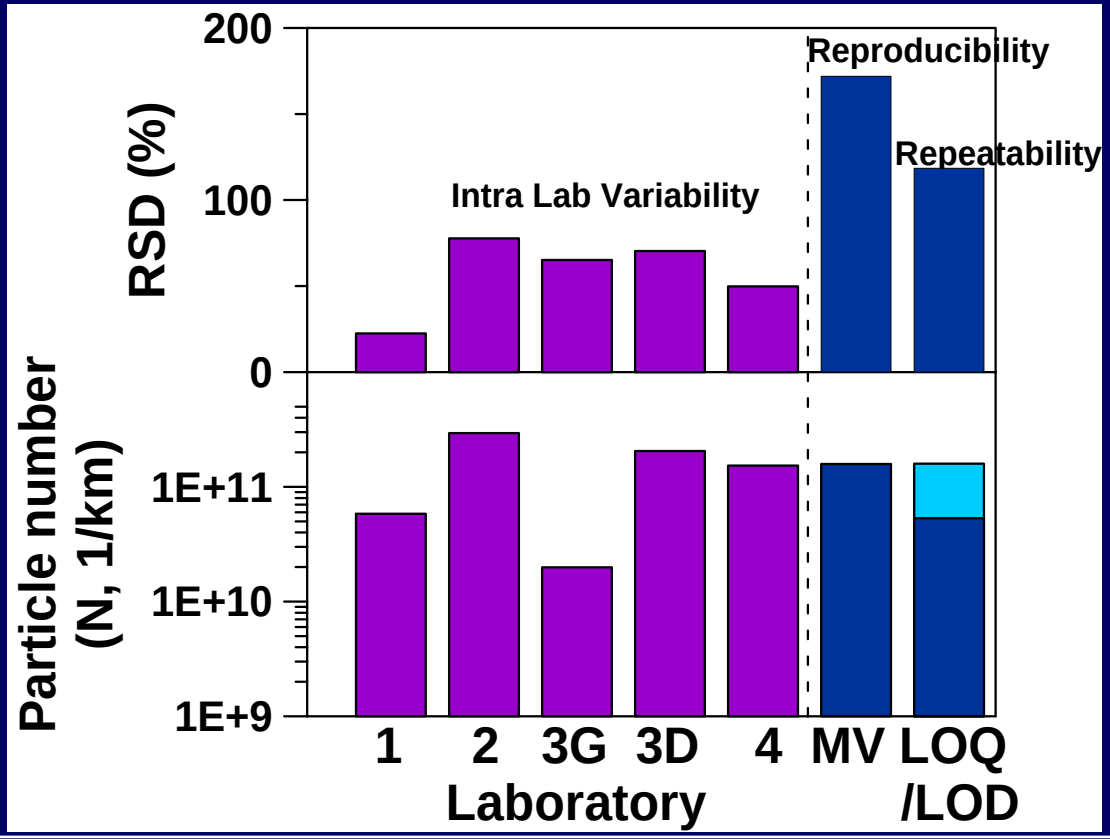


Results (3): *Particle number of the tunnel background tests*

- Good agreement between labs

- Very low values of particle numbers (mean=1.4x10^{11} 1/km), very close or lower of LOD/LOQ (5.3x10^{10}, 1.6x10^{11}, 1/km)

- Very high values of RSD reproducibility and repeatability, due to very low numbers

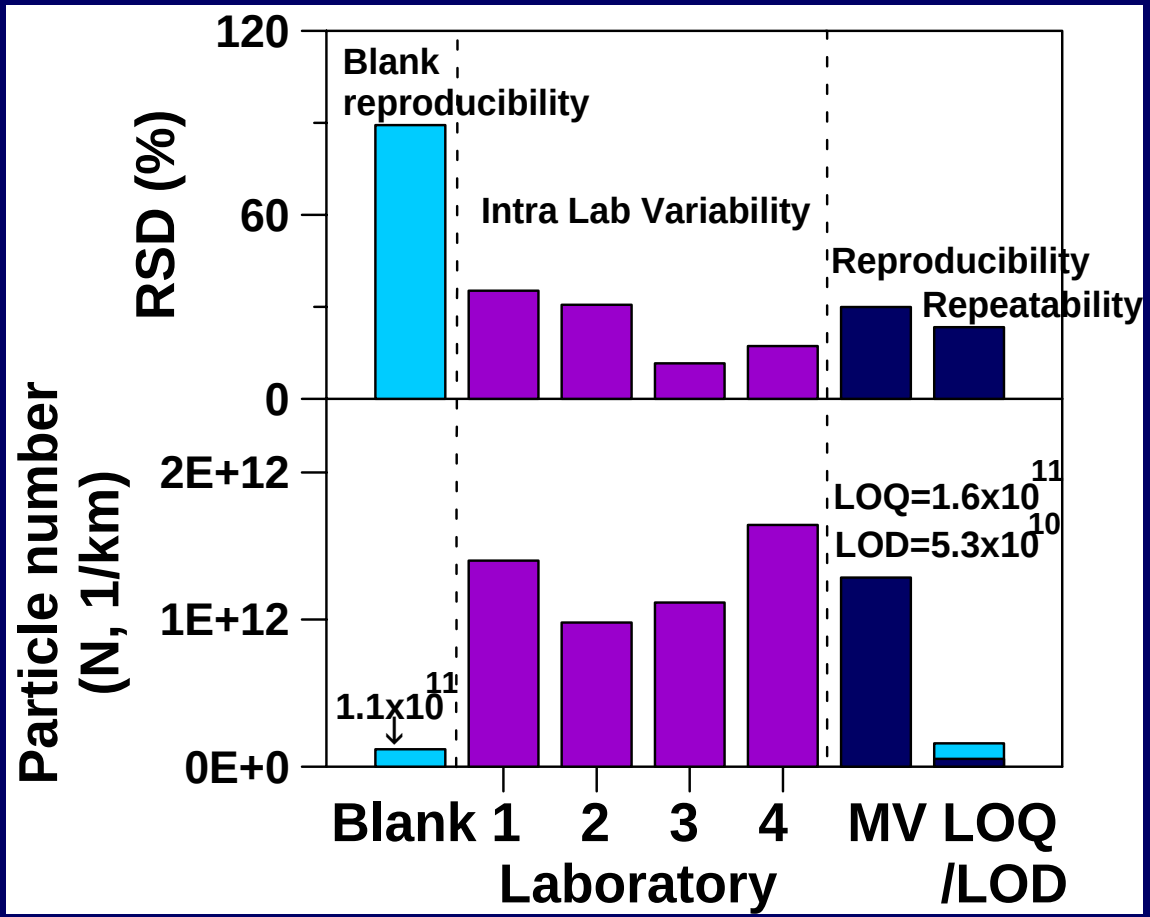


Results (4): *Particle number on the NEDC. Gasoline Vehicle*

- Good agreement between labs

- Very low values of particle numbers

- Very high values of RSD reproducibility and repeatability, due to very low numbers

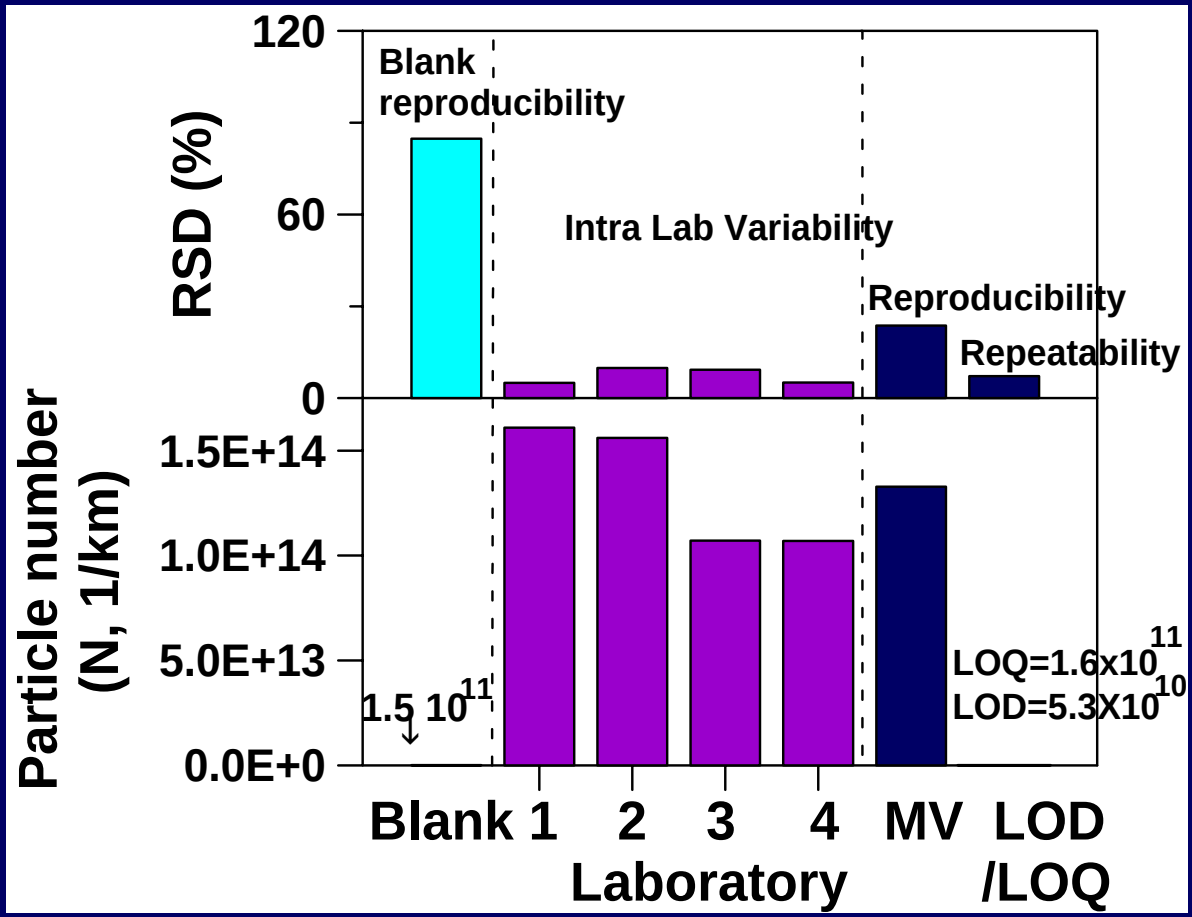


Results (5): *Particle number on the NEDC*. *Diesel Vehicle*

- Good agreement between labs

- Quite high values of particle numbers

- Due to high numbers, low values of RSD reproducibility and repeatability

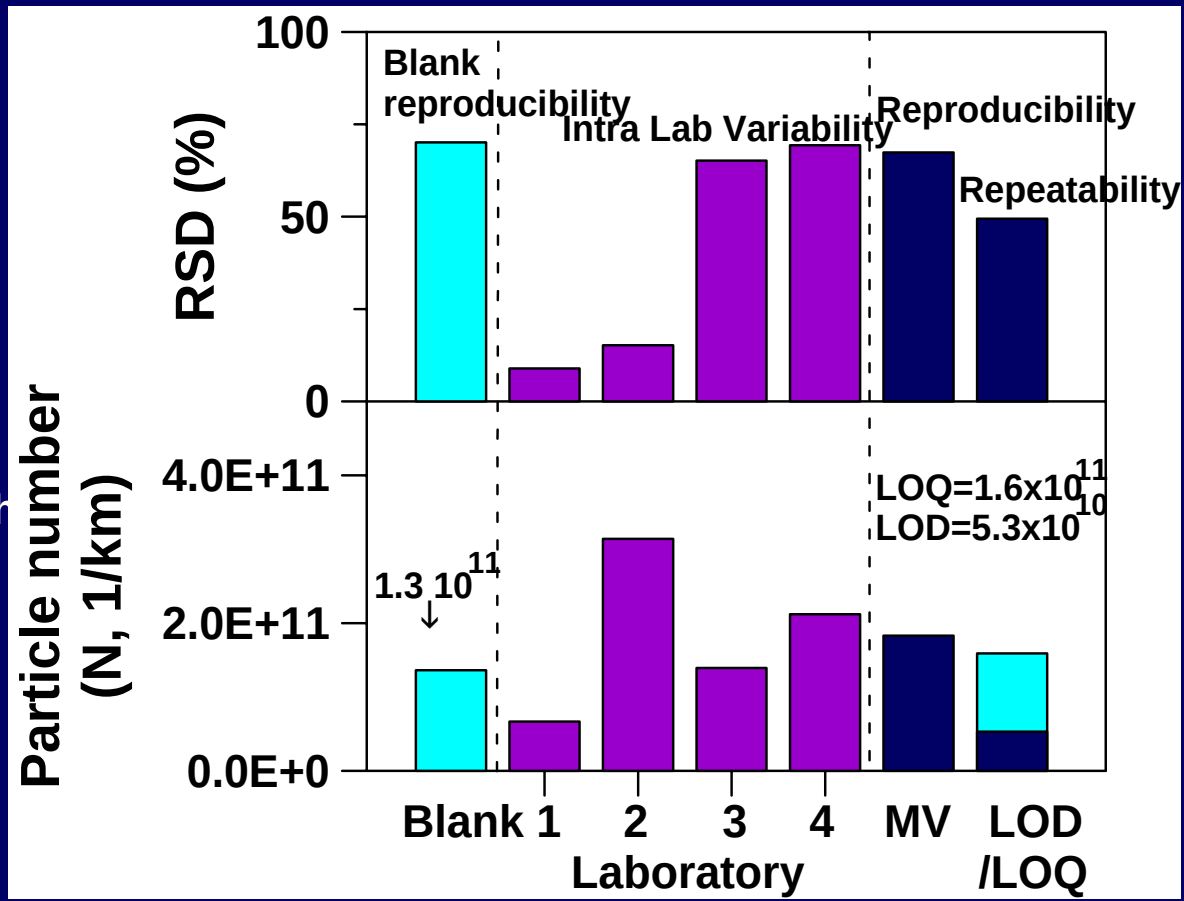


Results (6): *Particle number on the NEDC*. *Diesel DPF Vehicle*

- Good agreement between labs

- Low values of particle numbers, very close to or lower to LOD/LOQ

- Due to low numbers, quite high values of RSD reproducibility and repeatability



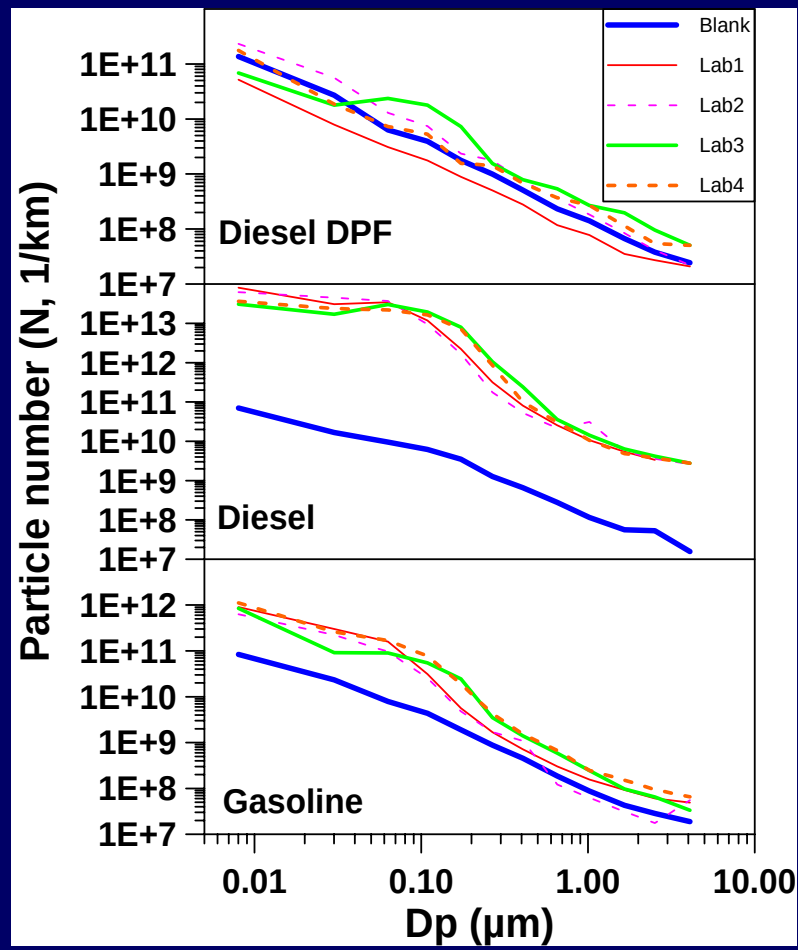
Results (7): *Particle size distribution*

-Good agreement between labs

- Gasoline: 1 order of magnitude higher than tunnel blanks (close to blank at higher Dp)

- Diesel: quite higher than blanks

- Diesel+DPF: equivalent to blanks



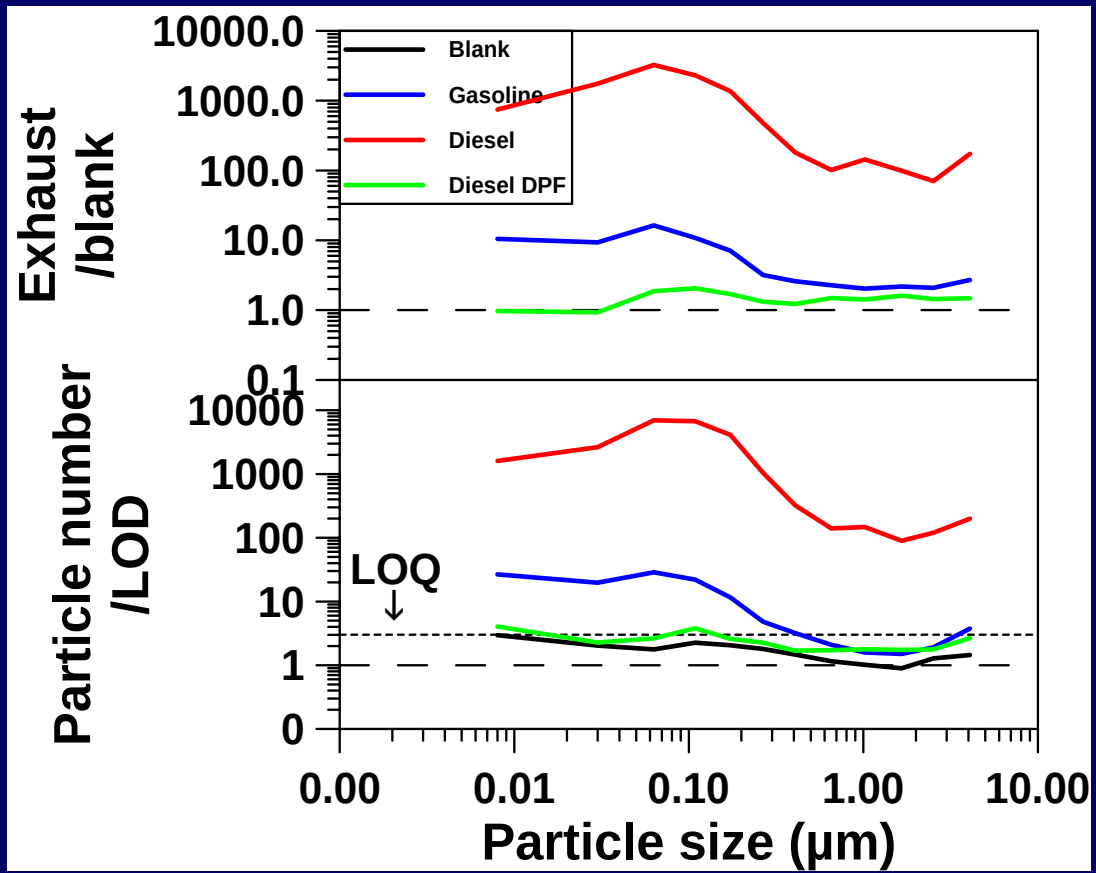
Results (8): *Comparison with tunnel background tests and ELPI limits*

- **Diesel+DPF: very close to the tunnel background tests**

- **Gasoline: very close to the tunnel background tests at high Dp**

- **Diesel+DPF, blanks: very close to the LOD ,lower than LOQ**

- **Gasoline at high Dp: lower than LOQ**

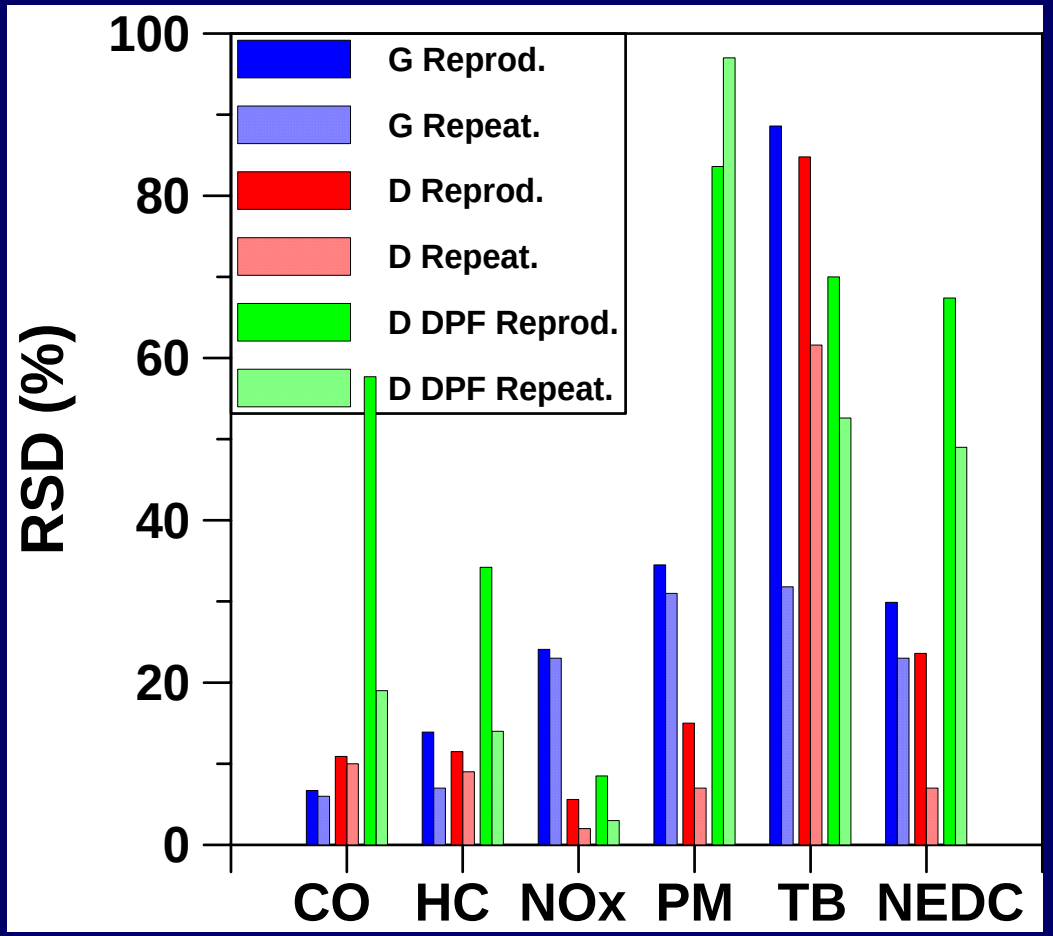


Results (10): *Comparison between RP and ELPI*

- RSD of PM: D=low, G=quite high, D+DPF=high

- TB: higher than the other measurements, due to low values

- PM number on the NEDC: comparative to PM measurements



Conclusions

- The reproducibility RSD of the tunnel background tests is quite high (up to 172%), due to low particle numbers, very close to the ELPI detection limits.
- On the entire NEDC, the reproducibility of total particle number determined by ELPI is 30, 24 and 67% for the gasoline, Diesel and DPF equipped Diesel vehicle respectively. These values are quite similar to those of the mass particle determination.
- The particle number emitted from the DPF equipped Diesel engine and at the upper part of the distribution of the gasoline PC engine is very close or even lower to ELPI LOD and LOQ.
- These results show that the protocol used in this study allows the reliable measurement of exhaust particle number in the case of vehicles emitting at least two orders of magnitude more than the tunnel background. In the other cases, the measurement variability is too high, especially for regulatory purposes. It must be noticed that other aspects of this methodology, as calibration of analytical instruments, not very well established today, are not taken into account here.