The effect of driving conditions and aftertreatment preconditioning on the formation of nucleation mode particles from diesel vehicles

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Diesel exhaust particles mainly appear in the nucleation and accumulation mode. While it is not possible to establish a precise size cutpoint for these two modes due to their dynamic nature, nucleation mode (NM) particles are generally considered those particles (mainly below <50 nm) which form by the condensation of exhaust semivolatile material in the exhaust line and after the dilution of the exhaust in the atmosphere. The accumulation mode particles are in principle the solid soot particles formed in the fuel-rich region within the combustion chamber. In the past, a large research effort has been invested all over the world to characterize NM particles, and to develop techniques to establish a reproducible measurement method. Due to their volatility, the concentration and size of these particles depends on sampling conditions. Having these techniques available today, the question arises, which engine, fuel and aftertreatment characteristics are responsible for the enhancement or suppression of the NM formation. The sulfur (primarily sulfate) content in the exhaust is considered the NM particle precursor. The concentration of sulfur in the exhaust though does not only depend on the rather straightforward conversion of the fuel sulfur on sulfate during the combustion or in oxidation aftertreatment devices but on storage and release mechanisms which take place in aftertreatment devices and the exhaust line. This paper studies the effect of storage and release mechanisms on the formation of NM particles by chasing a Euro 3 light duty diesel vehicle in the road and later repeating the same driving pattern on the chassis dynamometer. By driving the vehicle over repetitive acceleration – deceleration patterns and speed ramp (speed-up, speed-down) tests, it was found that the thermal pattern of the aftertreatment system affects the extent of NM formation, with speed-up tests leading to release of semi-volatile material previously stored over lower temperature conditions, hence promoting NM formation, and vice versa for speed-down tests. It was also found that the NM stabilization required more than 20 min when changing speed modes. However, this behaviour was not so intense for transient tests (cycles), due to the averaging effect of the frequently changing operation modes on the conditioning of the aftertreatment devices. Based on these tests, a proposal for vehicle and aftertreatment conditioning is proposed to reduce the variability of exhaust particle concentration during type-approval or other testing. This actually consists of having preconditioned the vehicle over the complete engine/exhaust temperature map included in the desired test, before conducting the actual test. This avoids or, at least, significantly reduces the probability for excessive storage or release of semi-volatile material which may then lead to an undesirable increase of the measurement variability.

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Introduction

Europe is guided to legislate only non-volatile particle number for emissions certificate after Euro V. This is based on the arguments that nucleation mode (NM) i) is considered a sampling artifact and ii) that it is unstable. In this paper we try to discuss and decide if these arguments are justified measuring the non-volatile and total particle number emissions of a Euro III vehicle at different speeds and preconditioning.

Experimental methods

The vehicle used was a European passenger car, equipped with oxidation catalyst and compliant with Euro III emission standards. The diesel fuel used fulfilled the EN590 regulations and had a sulphur content of 280 ppm. The lube oil was BP Vanellus C4 multi SAE 20W-50.

Particle emissions in the laboratory were measured with a partial flow sampling system, which was developed in the European Particulates project and has been lately applied to measure a large number of vehicles and engines. Constant sampling conditions were used in order to avoid the variation of NM from sampling conditions (Figure 1).

The tests were speed ramp tests in order to check the effect of the prehistory of the measurements on particle number emissions. After a 4 min warm up of the vehicle at 120 km/h, the speed was increased stepwise by 10 km/h from 80 km/h to 120 km/h (5th gear all measurements); the steps were then repeated backwards. Each step lasted 4 minutes, except the phase at 120 km/h, which lasted 8 minutes. The SMPS scans were initiated at the 1st and 3rd minute of each step (at 120 km/h also in the 5th and 7th minute), while CPC and ELPI (downstream of a thermodenuder) were continuously recording. The speed ramp test lasted 45 min.

Results: Nucleation mode

Concerning NM particles (Figure 2), at 80 and 90 km/h there was no NM, at 100 km/h it started to appear and at 120 km/h it maximized. Reducing the speed, NM decreased. At 90 km/h it disappeared.

Results: Nucleation mode volatility

The non volatile (solid) particle number concentration was measured from an ELPI downstream of a thermodenuder and the total particle number concentration from a CPC. Figure 3 shows these concentrations during the speed ramp test. In the same figure the vehicle speed and the exhaust gas temperature T_{exh} at the sampling position are given. All records are synchronized with speed.

Vehicle speed is stabilized very quickly at each measurement point, but not the temperature. Non-volatile particle concentration is almost constant at all speeds. Total particle number concentration at 80 and 90 km/h is almost constant. At 100 km/h total particle concentration starts to increase. These are NM particles as it was shown from SMPS size distributions (Figure 2). Comparison of ELPI and CPC leads us to the conclusion that these NM particles are volatiles.

Results: Prehistory

One parameter that affects the absolute level of the NM is the prehistory of the measurements. It was shown in Figure 3 that the average particle concentration at one speed (e.g. at 100 km/h) is higher when lower speed proceeds (e.g. 90 km/h) than when higher speed (e.g. 110 km/h) proceeds.

Prehistory effect on particle emissions is better described in Figure 4 for 100 km/h, which is the most "sensitive" speed concerning the NM formation. Small changes of the prehistory lead to completely different emissions (no NM when coming from higher speed, increasing NM when coming from lower speed). At other vehicle speeds the tendencies will be the same, the effect of the preconditioning might be smaller however.

Figure 4 shows with thick lines the speed, the temperature and the particle emissions at 100 km/h during the speed ramp test at an arbitrary common time (i.e. the 4 min at 100 km/h during ramping up and down from Figure 3 are plotted at a common time scale). The thin grey line shows a steady state test at 100 km/h of the same vehicle (with the same fuel) that lasted approximately 30 min. A 120 km/h steady state test had proceeded, so this measurement is comparable with the "down" measurement of the speed ramp test. Indeed the temperature profile of the two tests (down and steady state) is similar for the common 4 min. Unfortunately there are no CPC data available. However, there are 3 SMPS scans that show the expected tendency. No NM at the beginning and NM starts to appear 15 min later.

Discussion

The parameters that affect the NM formation are (Figure 5): a) engine, engine mode, fuel, lubricant b) aftertreatment devices c) sampling conditions. Here only the effect of the aftertreatment devices will be discussed.

It is know that exhaust gas temperature affects the SO_2 to SO_3 conversion in the catalyst. At 80 and 90 km/h the conversion ratio is almost zero, so no NM appears. At 120 km/h the conversion ratio is high, so a distinct peak is formed. At 100 km/h (T_{exh} = 200ºC) the conversion ratio is very low (5%), so the production of SO_3 is also low. And in this case the effect of the prehistory of the vehicle is huge.

When higher speed has proceeded, produced sulfates are stored at the catalyst to achieve equilibrium at the new speed (temperature). At 100 km/h the produced SO_3 is low, so all SO_3 is stored in the catalyst. 15 min later the storage mechanism slows down and NM starts to appear. However, when lower speed proceeds, SO_3 has already been stored, so the produced SO_3 is measured at the sampling system. Moreover, the increase of the temperature releases some SO_3 from the catalyst. How long stored SO_3 is going to be released it is not know. Dashed lines in Figure 4 show an estimation that is based on 120 km/h steady state test (not shown). After 2 – 10 min the release reaches a maximum and then starts to decrease for another 20 min. Theoretical calculations that are based on a literature review on the subject showed that this procedure could last from 3.5 to 30 min for the case of the 100 km/h.

Conclusions

❑ From the speed ramp tests it was shown that NM is not an artifact but a true vehicle component that for a Euro 3 vehicle depends on the vehicle speed (exhaust gas temperature).

❑ NM measured depends not only on the sampling conditions but also on the prehistory of the vehicle. It is higher when lower speed precedes. Consequently, for repeatable NM measurements, not only the sampling conditions, but also the preconditioning of the measurement must be standardized.

❑ The aftertreatment devices affect the NM emissions in two ways: i) increased temperature increases the SO_2 to SO_3 conversion leading to more NM precursors. ii) temperature differences during speed changes affect the release/storage of NM precursors (sulfates and hydrocarbons). While the first (i) is actual vehicle emissions, the second (ii) might also include emissions from previous measurements (e.g. lower speeds, previous fuels etc). For steady state tests, the NM needs more than 20 min to stabilize. For transient tests (cycles) the problem is not so intense. However, preconditioning must be standardized to decrease the effect of the prehistory of the vehicle. We believe that a preconditioning that includes the extreme conditions of the test that follows is necessary (e.g. EUDC for NEDC cycle).

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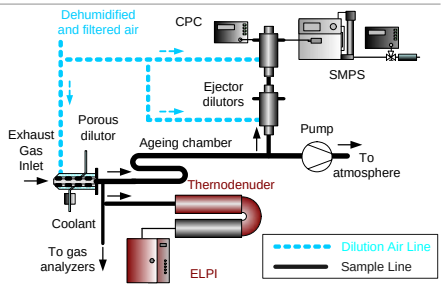

Figure 1: Schematic of the partial flow sampling system.

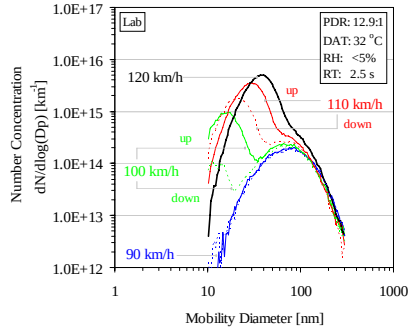

Figure 2: Particle size distributions measured with SMPS at the laboratory. "Up" corresponds to distribution as the vehicle ramps up to higher speed and vice versa for "down" scans.

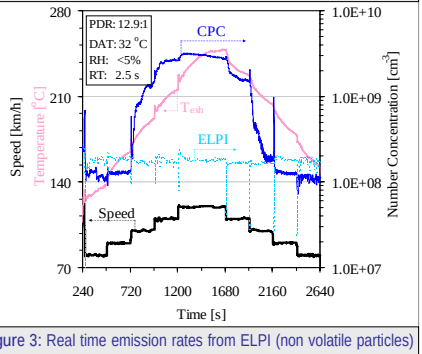

Figure 3: Real time emission rates from ELPI (non volatile particles) and CPC (total particles) for the speed ramp tests.

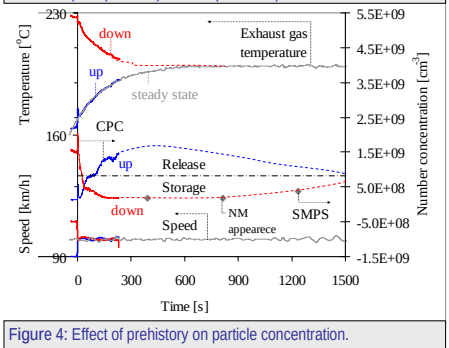

Figure 4: Effect of prehistory on particle concentration.

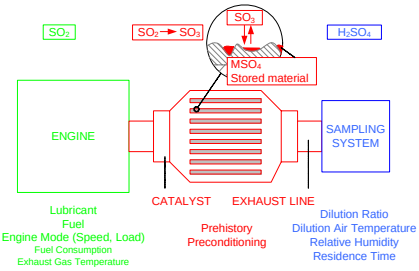

Figure 5: Parameters that affect NM formation and measurement.