Correlation Between PM, Soot and Airborne Particle Mass Emitted from Diesel Heavy Duty Engines at PM Levels Ranging from Euro V to Post-DPF

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A number of epidemiological studies[1] have associated exposure to ambient Particulate Matter (PM) with a number of adverse health effects. Internal combustion engines, and particularly compression ignition diesels, were recognized as a major contributor to ambient particulate matter and as such have been subject to regulation world-wide. Automotive exhaust particulate emissions were traditionally regulated in terms of mass, but the increasingly tighter emission limits raised concerns regarding the sensitivity of the gravimetric procedure. Based on the findings of the Clean Air For Europe (CAFE) programme[2], the European Commission requested in 2005[3] a significant reduction of the PM emission standards to a level that would necessitate the installation of very efficient wall flow Diesel Particulate Filters (DPF) to all diesel vehicles but at the same time be capable of identifying low efficiency flow through designs. In response, a particle number limit, based on the procedures developed in the framework of the Particulate Measurement Programme (PMP)[4,5], was recently introduced for light duty vehicles[6] and will soon follow for Heavy Duty Engines[7].

The particle number limit only addresses the "non-volatile" fraction of the exhaust aerosol which is what the DPF controls. As such it complements rather than replaces the PM emission standard which further controls the volatile fraction of PM. It is therefore not surprising that a number of studies have found a poor correlation between PM and "non-volatile" Particle Number (PN) emissions for low emitting diesel engines[4,5,8]. The observed discrepancies could point towards a limited sensitivity of the gravimetric procedure and/or a change of the physicochemical properties of emitted PM, highlighting the need for a more detailed investigation of the properties of PM at such low emission levels.

In this study, the particulate emissions of two heavy duty diesel engines were characterized at PM levels ranging from EURO V (20 mg/kWh) to post-DPF. The different emission levels were simulated by means of a DPF/bypass configuration. The engines were tested under the World Harmonized Transient Cycle (WHTC) and selected steady state modes. Filter samples were taken from a CVS tunnel, using TX40, Teflon and Quartz Fiber filters. Measured properties included PM, PN, soot mass (measured with a photoacoustic sensor), thermo-gravimetrically determined elemental/organic carbon, number weighted mobility size distributions and effective particle density. Dedicated tests were also conducted in order to investigate the influence of sampling duration and filter media upon the total collected mass, and the contribution of background at the different emission levels examined.

The main objective of the study was to calculate the mass of the airborne "non-volatile" particles counted by the PMP compliant system and quantitatively compare it with that of PM and soot at different emission levels. The calculation of airborne particle mass from the measured number concentrations requires information on the underlying size distribution and the effective particle density profile of the exhaust aerosol[9].

A TSI's 3936L10 Scanning Mobility Particle Sizer was employed over the steady state tests, sampling directly from the CVS tunnel. The distributions obtained were generally bimodal in shape with a distinct mode peaking at a size smaller than the lower detection limit of the SMPS employed (<8 nm). These nano-particles could be volatile material nucleating as the exhaust dilutes and cools down[10] or metallic oxide particles originating from oil and/or fuel additives[11]. In either case, they are not detected by the PMP system, owing to the large cut-off size of the CPC employed (23 nm). In that respect, a bimodal lognormal fit was applied to the measured size distributions and the accumulation mode was employed for the number to mass conversion. The SMPS was also employed during transient testing, operating at fixed voltages and thus measuring in real time the concentration of monodisperse samples at selected mobility diameters. Convolution of the traces at different sizes over repeated test cycles suggested that the size distribution remained relatively constant, at least for the two engines examined.

The effective particle density of the exhaust aerosol was characterized employing a tandem Differential Mobility Analyzer (DMA) –Dekati's Mass Monitor (DMM) setup, in line with the approach followed by Maricq et al.[12]. The measured effective densities were found to exhibit a power law dependence on the mobility diameter, decreasing from approximately 1 g/cm^3 at 60 nm to 0.6 g/cm^3 at 130 nm. Tests at low (25%) and high (75%) load conditions gave similar results, with a power law fit suggesting a fractal dimension of 2.35, in good agreement with published data[12].

The fitted effective density profiles and lognormal distributions provided the means of calculating the mass of airborne "non-volatile" particles from the PN results. The calculated airborne particle mass was found to be in good quantitative agreement with the mass of EC determined thermo-gravimetrically and the mass of soot measured with the photo-acoustic sensor. The relative difference was on average smaller than ±15% for both engines and at all emission levels examined.

The calculated airborne mass was found to be systematically lower than the mass collected on the filter, as the later also collects some volatile material. The relative differences were found to depend on the filter media employed, the emission levels and the sampling duration.

In particular, the total carbon determined from thermogravimetric analysis of Quartz filters was systematically higher than that determined from TX40 filters which in turn yielded higher masses compared to Teflon filters. At simulated Euro V and Euro VI PM emission levels (i.e. when portion of the exhaust bypassed the DPF), the calculated airborne mass was found to constitute 50% to 80% of the mass collected on Teflon filters. TX40 samples suggested that 20% to 80% of the collected mass was "non-volatile" airborne particles, with their contribution decreasing sharply with decreasing emission levels. The total carbon determined thermogravimetrically suggested an even lower contribution of suspended "non-volatile" particles of 20% to 50%. At DPF out levels the calculated airborne particle mass was around 1% of the collected PM for all filter media.

The mass collected on the TX40 and the total carbon in the Quartz samples did not scale up with the sampling time. The results obtained when a single filter was employed over three consecutive WHTC were 20% to 60% (with the discrepancies increasing with decreasing emission levels) lower compared to those derived from samples collected over a single WHTC. Teflon filters yielded more consistent results. All these findings point

towards a gaseous adsorption artefact which is known to be more pronounced in TX40 and especially in Quartz filters[13,14].
The very low emission levels downstream of a DPF constitute also challenging the discrimination of the exhaust PM from the CVS tunnel background. The CVS background was found to be at a level of 2 mg/kWh which is just about the post-DPF emission levels. Thermogravimetric analysis of Quartz samples suggested that this background is entirely volatile in nature but it was not possible to identify in the present study whether it originates from the dilution air or the tunnel.

References:

[1] Dockery et al. 1993. An Association Between Air Pollution and Mortality in Six U.S. Cities. New England Journal of Medicine 329: 1753-1759.
[2] COM(2005) 446 final
[3] Preliminary draft proposal for a Regulation of the European Parliament and of the Council relating to emissions of atmospheric pollutants from motor vehicles (Euro 5)
[4] Anderson et al. 2007. EUR 22775 EN
[5] Anderson et al. 2010. EUR 24561 EN
[6] Regulation (EC) No 692/2008 (18/07/2008)
[7] Regulation (EC) No 595/200 (18/06/2009)
[8] Ntziachristos et al. 2004. SAE technical paper 2004-01-1985
[9] Maricq et al. 2006. Aerosol Science and Technology 40, 68-79.
[10] Kittelson D. 1998. Journal of Aerosol Science, 29:575-588.
[11] De Filippo et al. 2008. Environmental Science and Technology, 42: 7957–7962.
[12] Maricq et al. 2004. Journal of Aerosol Science, 35: 1251 – 1274.
[13] Chase et al. 2004. SAE Technical Paper 2004-01-0967
[14] Mader et al. 2001. Environmental Science and Technology, 35:3422-3432

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Engine 1

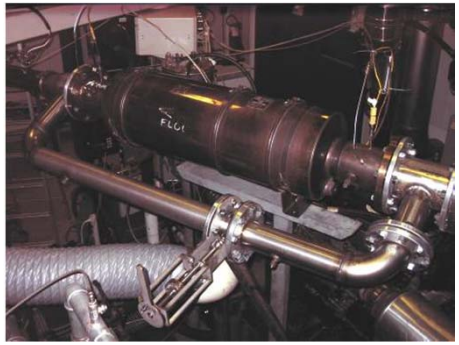
IVECO Cursor 8 (PMP Golden)

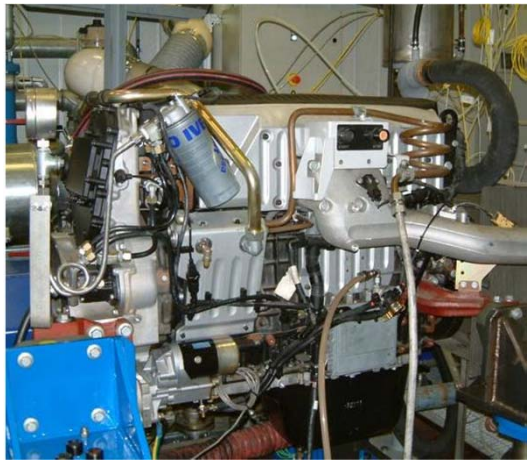
- Euro III, 7.8 l, 6 cylinders
- 260 kW @ 1900-2400 rpm
- 1280 Nm @ 1000-1900 rpm

CRT & by-pass

- 4.25 l Pt-based oxidation catalyst
- 24 l Johnson-Matthey DPF

10 ppm S fuel





Engine 2

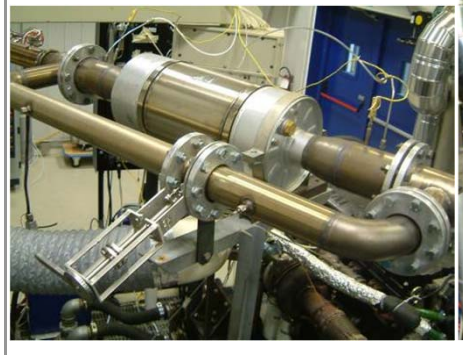
IVECO Cursor 10

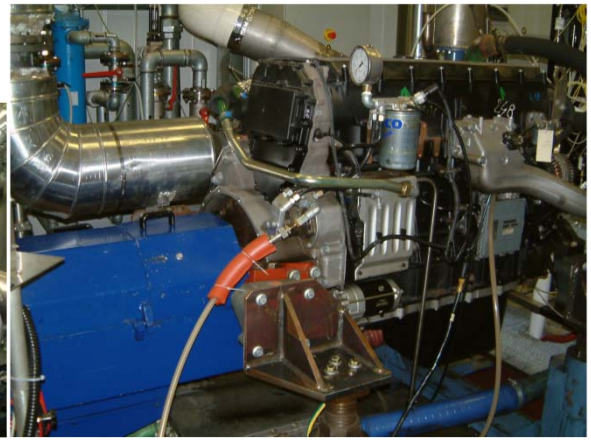
- Euro III, 10.3 l, 6 cylinders
- 316 kW @ 2100 rpm
- 1900 Nm @ 1050-1750 rpm

DPF & by-pass

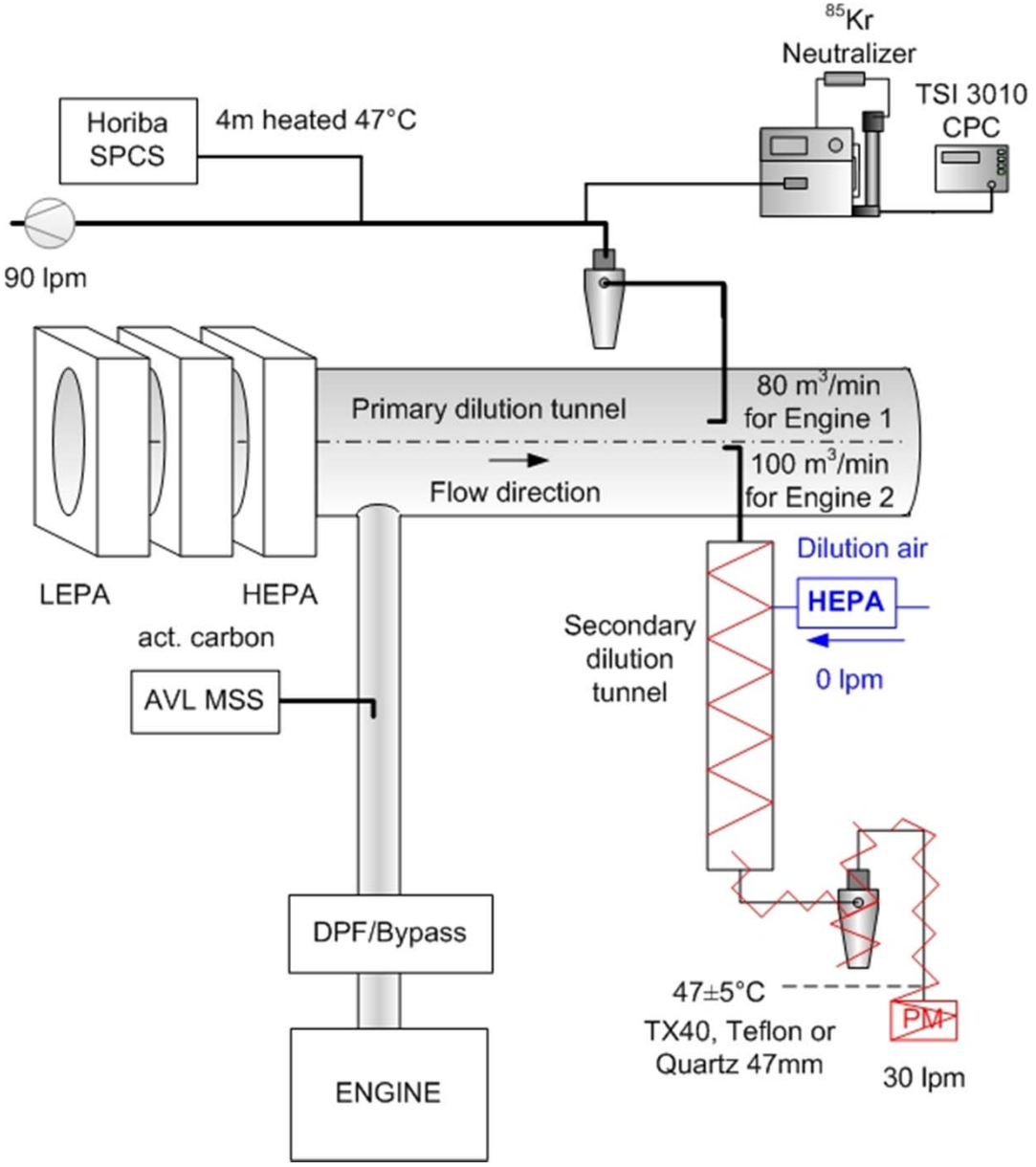
- No oxidation catalyst
- Pirelli Feelpure DPF (SiC 11.24” × 12”)

10 ppm S fuel





Experimental - Instrumentation



PM collected on TX40, Teflon and Quartz filters

Horiba Solid Particle Counting System (SPCS)

AVL Micro Soot Sensor (MSS)

TSI Scanning Mobility Particle Sizer (SMPS)

Experimental – Test Cycles

Engine 1

- Blank Tests
- WHTC cold
- WHTC hot
- ESC mode 12
- ESC mode 7

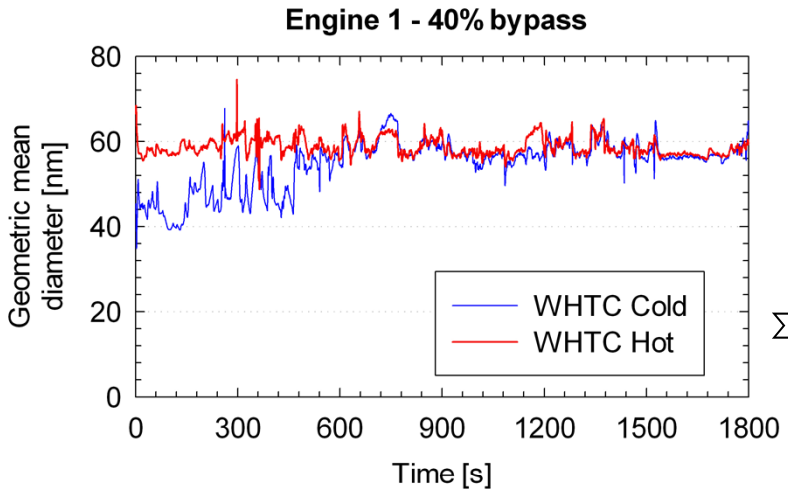
Engine 2

- Engine warm up
- WHTC hot
- WHTC hot
- WHTC hot

Single filter

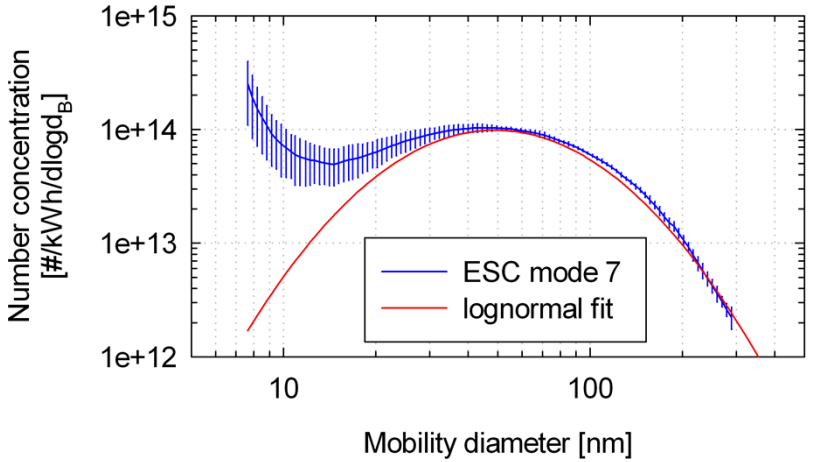
- WHTC hot

Airborne mass calculation

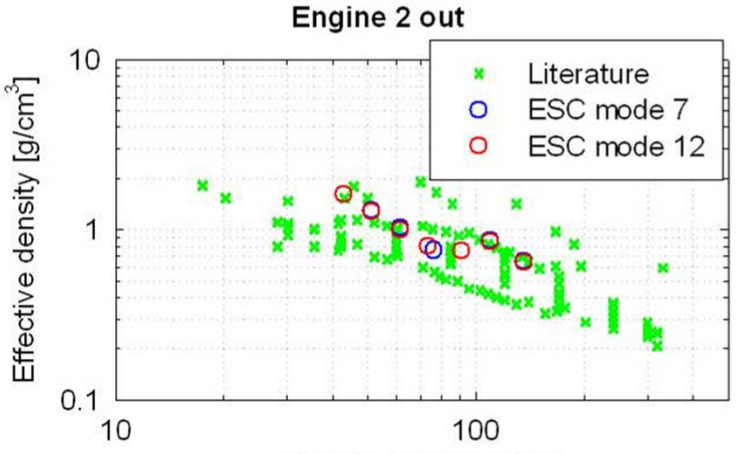


$$\rho_{eff}(d_B) = \rho_0 \left(\frac{d_B}{d_0}\right)^{DF-3} \Longleftarrow$$

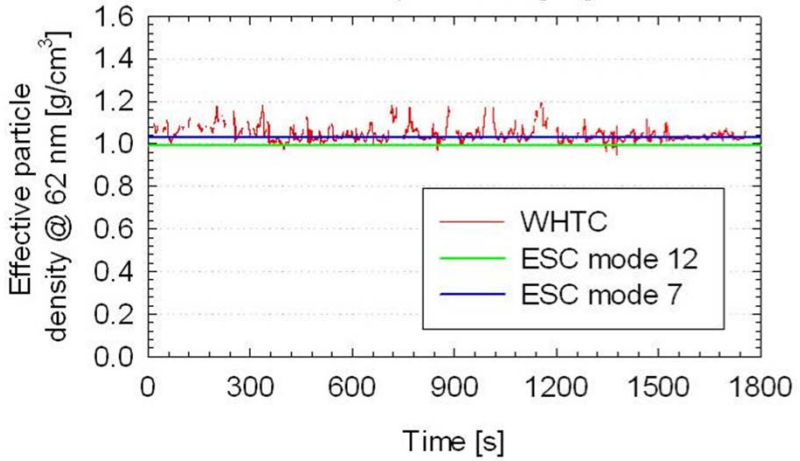
$$\Longrightarrow d_g \approx const.$$



$$\Longrightarrow d_g, \sigma_g$$



$$\rho_{eff} \approx const \Longleftarrow$$



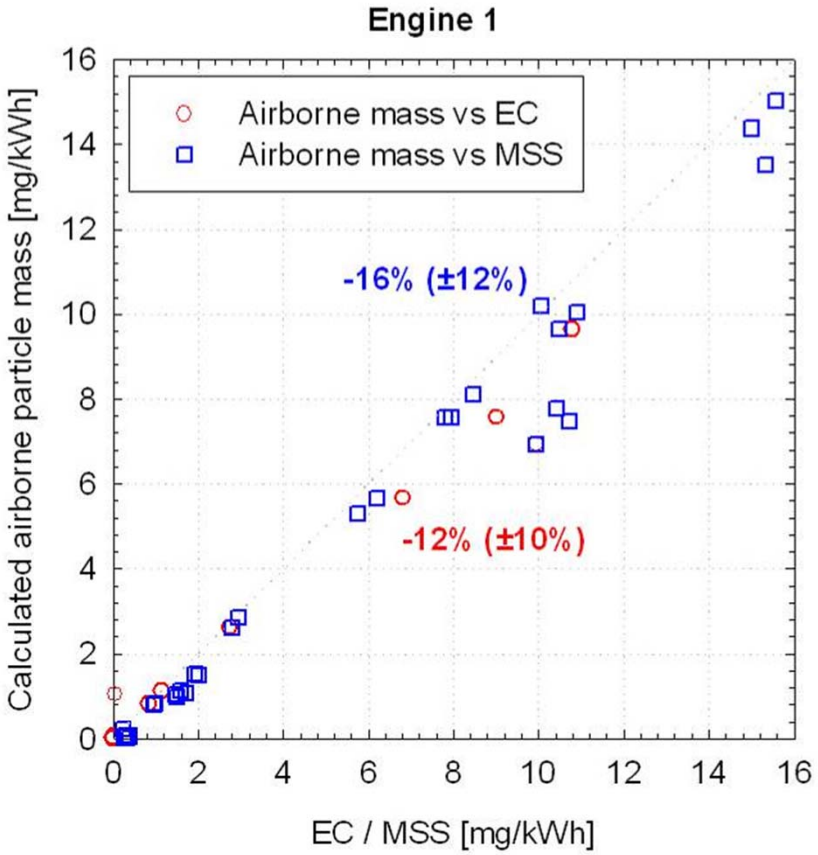
Assumptions:

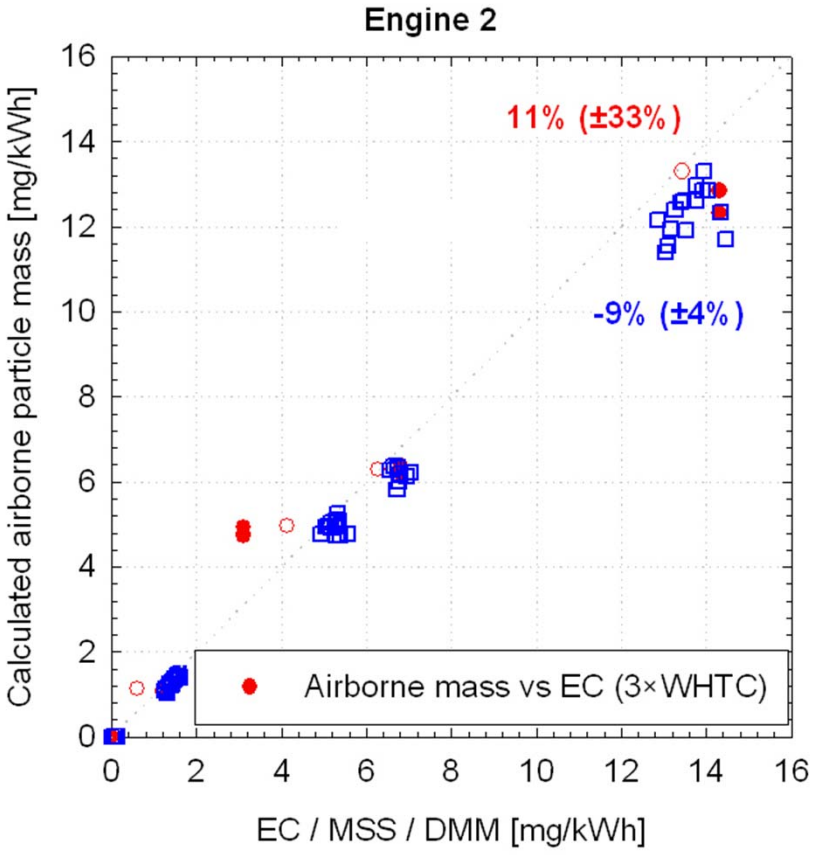
- Lognormal distribution
- Fractal-like structure

$$m = N_0 \frac{\pi}{6} \rho_0 d_0^{(3-DF)} d_g^{DF} e^{\frac{DF^2 (\ln \sigma_g)^2}{2}}$$

Maricq et al., JAS, 35:1251-1274
Maricq et al., AST, 40:68-79

Results – Airborne mass vs EC, MSS, DMM



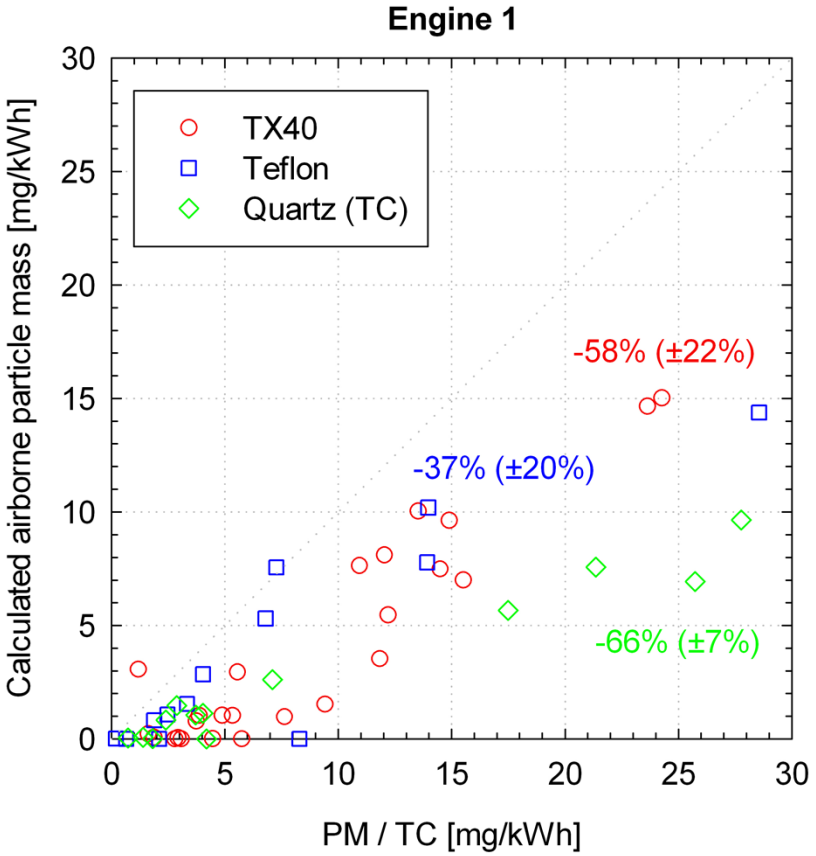


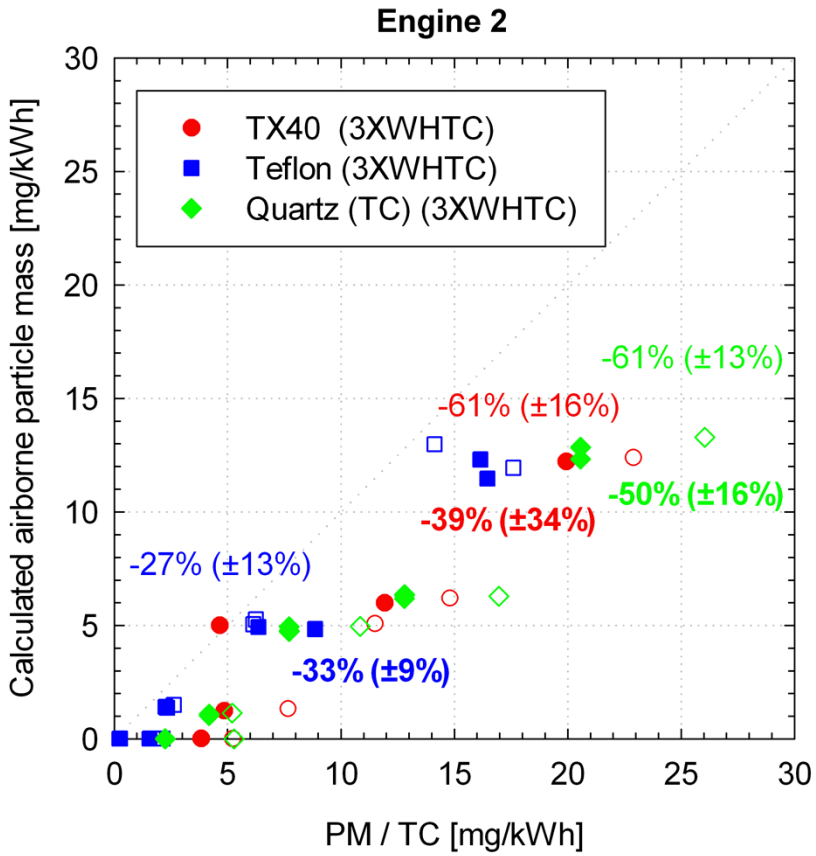
Very good agreement between the calculated “non-volatile” airborne mass and the mass of soot.

TM1 Remove DMM from the chart

Thanasis Mamakos; 15.06.2011

Results – Airborne mass vs PM





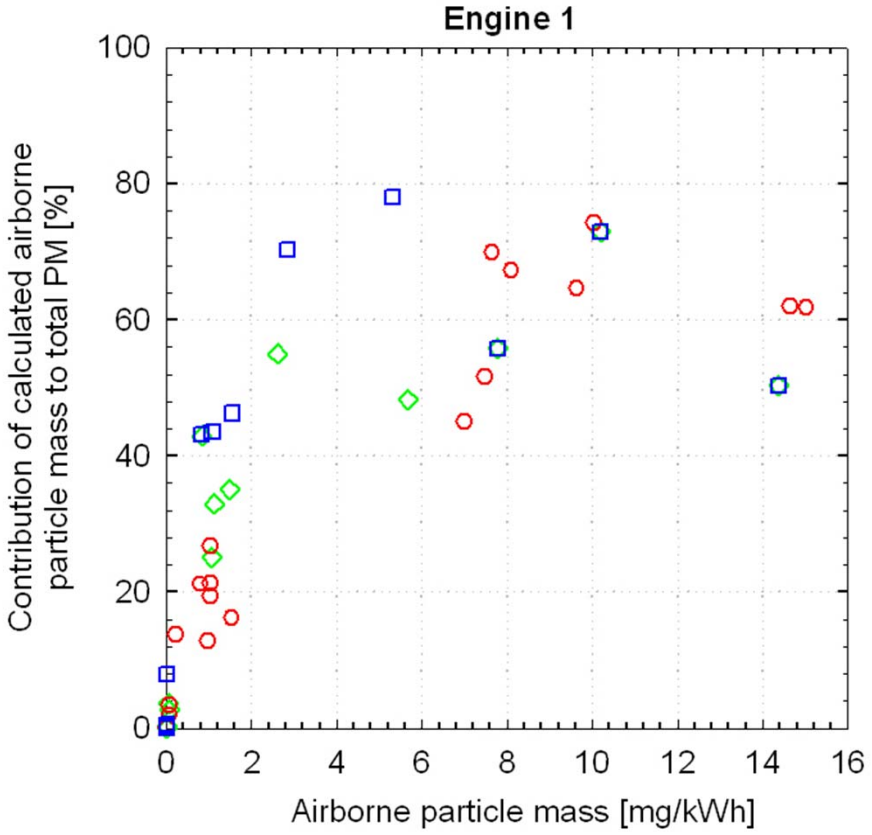
Airborne particle mass is a fraction of PM emissions due to the volatile compounds collected on filter.

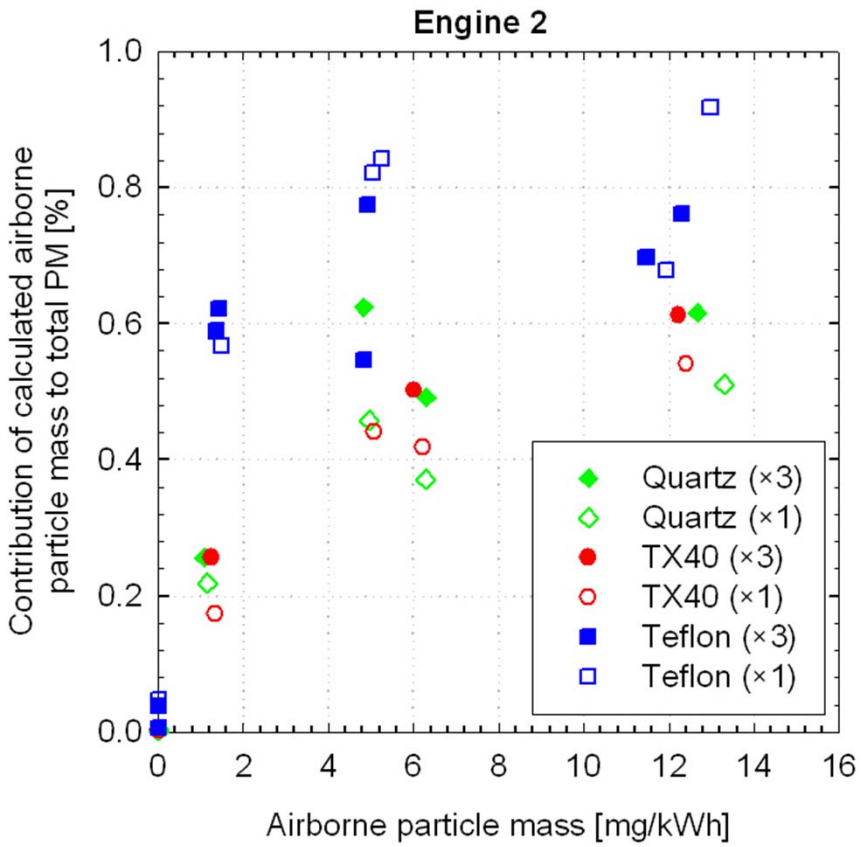
The volatile content of the PM filter mass is found to depend on the filter media and (particularly for TX40 and Quartz filters) on the sampling time.

TM3 Add the relative differences as well

Thanasis Mamakos; 15.06.2011

Results – Contribution of airborne mass to PM

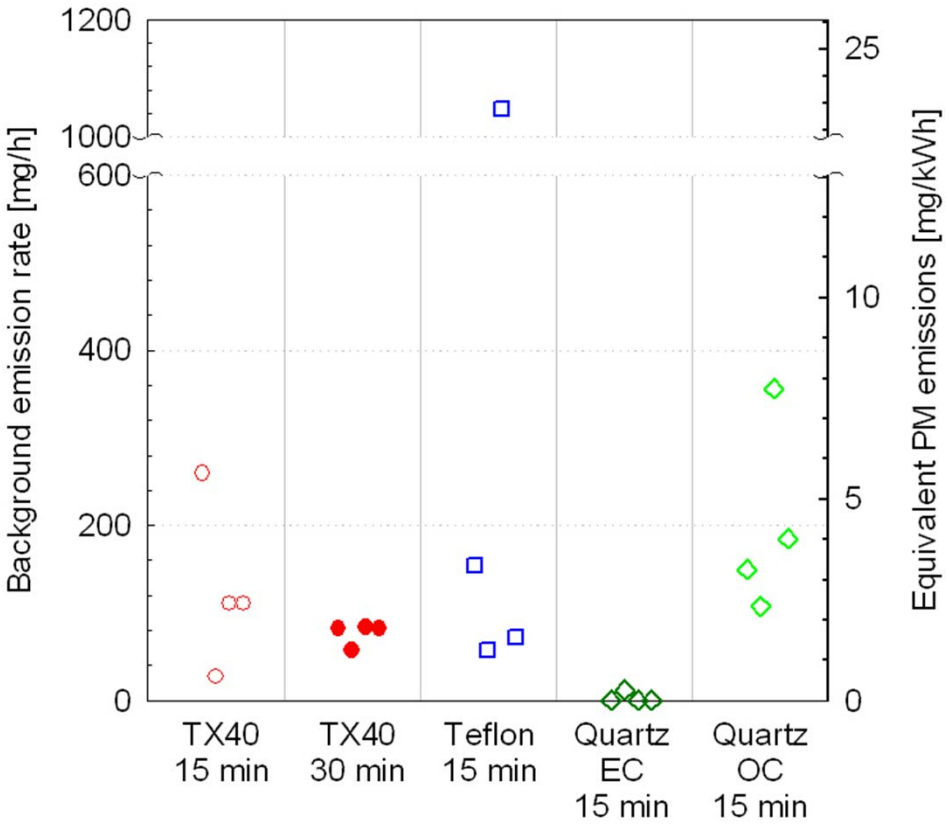




The contribution of airborne particles on the filter mass decreases with decreasing PM levels.

The filter mass did not increase proportionally to sampling time when employing TX40 and Quartz fiber filters.

Results – Blank tests



The mass collected on filters during blank tests was similar to DPF out PM mass: ~ 2 mg/kWh

Thermogravimetric analysis of Quartz filter samples revealed that this background PM was entirely volatile in nature

Prolonged sampling times on TX40 filters resulted in more repeatable results and on average lower PM masses

Conclusions

- **Calculated mass of “non-volatile” particles measured with a PMP compliant system, using information on size distribution and effective density, was found to be in very good agreement with the mass of EC/soot.**
- **The volatile fraction of the filter masses was found to depend on the filter media, with the Quartz and TX40 filters collecting more volatile material.**
- **Teflon filters yielded more consistent results.**
- **The contribution of airborne particles on the PM filter mass decreased with decreasing PM emission levels, constituting around 1% of the collected mass at DPF/CRT out levels.**
- **Background PM was found to be entirely volatile in nature and at levels similar to those measured downstream a DPF**

Thank you for your attention!

More info:

EU-PEMS PM EVALUATION PROGRAM - Second Report - Study on Post DPF PM/PN Emissions

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