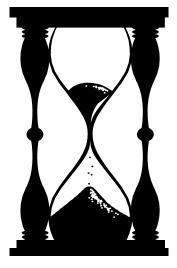


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17th ETH-Conference on Combustion Generated Nanoparticles 23-26 June 2013

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- Introduction: PM, PN and the PMP
- Particulate Mass Vs Particle Number
- The Trouble with Volatiles...
- Background Issues
- Is there still a role for the mass metric?
- Conclusions

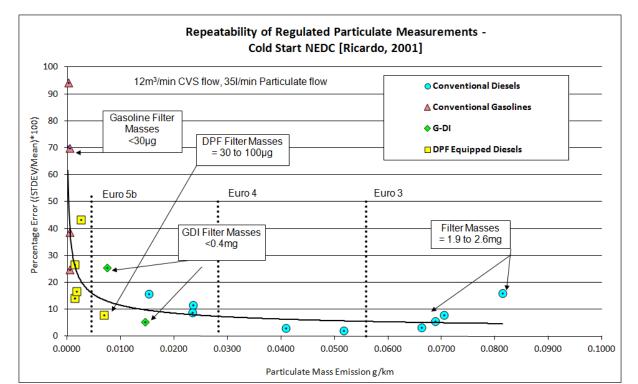


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## PM is no longer the sole metric for controlling IC engines' particle emissions

- The Particle Measurement
  Programme was started in around
  2002 with the objective to
  "complement or replace the (filterbased) mass measurement metric"
  - The primary driver was the inaccuracy and poor repeatability of the filter-based method at low emissions levels and the regulators wished to mandate PM control aftertreatment and realise economic health benefits linked to reduced PM



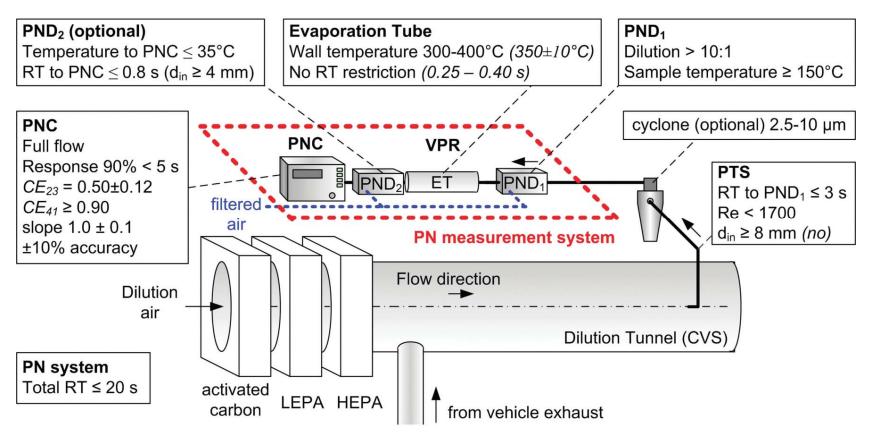
- Several years of investigations led to the eventual identification of the number of nonvolatile, or solid, particles as the new metric.
- The outcome was a method sensitive enough to mandate effective control of carbon particles through a tough particle number limit



#### **PN Measurement Approach**



#### Schematic of PN measurement system



- Now PN applies to light-duty diesel at Euro 5b, gasoline DI during Euro 6, HDD (Euro VI) and possibly will be applied to NRMM at Stage V
- Is there now any need for, or relevance of, filter-based PM?



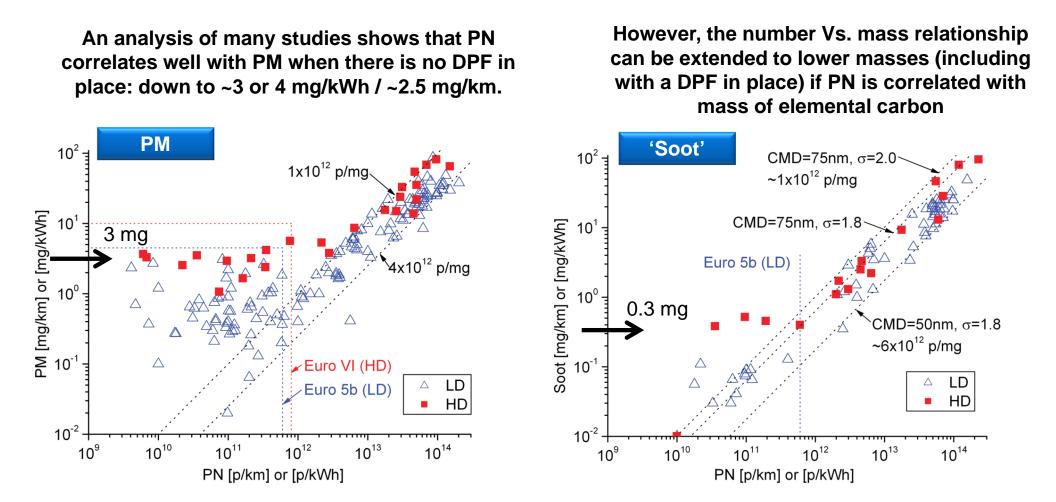
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**Particulate Mass Vs Particle Number** 

#### **Particle Mass Vs Particle Number (1)**



 The PN metric enables legislators to implement controls on particle emissions below the detection limit of the gravimetric procedure



Measurement of Automotive Nonvolatile Particle Number Emissions within the European Legislative Framework: A Review; Aerosol Science and Technology; Volume 46, Issue 7, 2012; Giechaskiel, Mamakos, Andersson, Dilara, Martini, Schindler & Bergmann

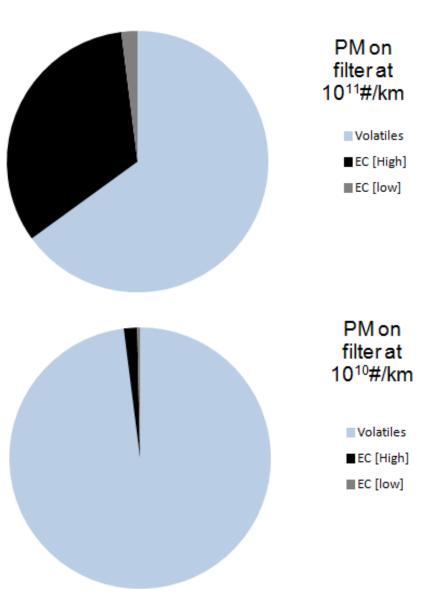
#### Particulate Mass Vs Particle Number

#### **Particle Mass Vs Particle Number (2)**

- The main differences in the correlations between PN and EC and between PN and PM originate from the volatile fraction
  - At 10<sup>11</sup> particles/km: carbon fraction represents 2% to 35% of the corresponding PM mass
  - At 10<sup>10</sup> particles/km: carbon fraction has decreased but the measured PM mass remains the same, so the carbon fraction can be below 0.5% of the PM total
- The remaining 65% to 99.5% of the PM is mostly volatile and comprises:
  - HCs, sulphates, nitrates and water, perhaps some SCR by-products too...
  - Also some ash, but generally very low levels
- So why does the volatile fraction cause a problem?

In both cases filter mass =  $\sim 0.8$  mg/km







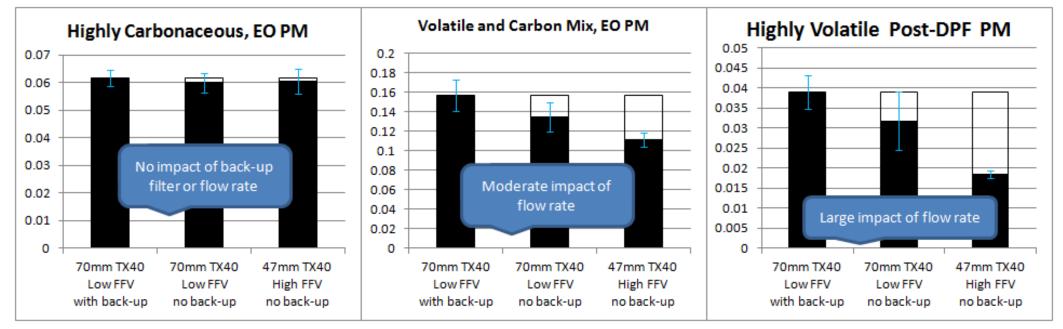
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The Trouble with Volatiles...

## **PM Sampling for Regulatory Drive Cycles – volatile levels are influenced by dilution systems and sampling parameters (1)**



- PM is sampled from either full flow dilution systems, where the whole exhaust of the vehicle is diluted (always for light-duty vehicles, sometimes for HD), or partial flow systems, where a fixed proportion of the exhaust is diluted (the rest of the time for HD)
- Some filter media can collect gas-phase volatile hydrocarbons that are (probably) already quantified by the FID
- The volatile fraction of PM is susceptible to sampling conditions, meaning that collected mass varies with different sampling parameters such as flow / face velocity



UK Particulate Measurement Programme (PMP): A Near US 2007 Approach to Heavy Duty Diesel Particulate Measurements - Comparison with the Standard European Method; SAE 2004-01-1990; Andersson, Clarke, Watson

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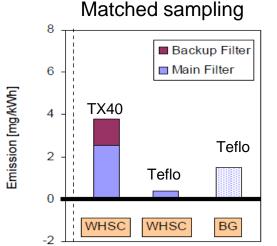
The Trouble with Volatiles...

#### **PM Sampling for Regulatory Drive Cycles – volatile levels are** influenced by dilution systems and sampling parameters (2)

- Filter medium effects
  - Fibrous elements in PM filters (principally glass fibre) have a high affinity for volatile components of exhaust
  - Switching to a Teflon membrane (not a 'filter') can limit the capture of these volatiles, but other issues, such as accumulation of static charges exist with these membranes
- Work in the PMP programme showed that, in extreme cases, dilution systems can become contaminated with volatile and semi-volatile materials
  - These materials can bleed out of the dilution system during sampling, adding mass to PM samples
  - Experience has shown that CVS systems tend to have greater contamination than partial flow systems
    - This may be due to age CVS systems are generally older and therefore have been historically used with older, higher emitting engines

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- Partial flow systems are also easier to dismantle and clean
- What impact does this legacy of volatiles in the dilution system have on PM and PN?







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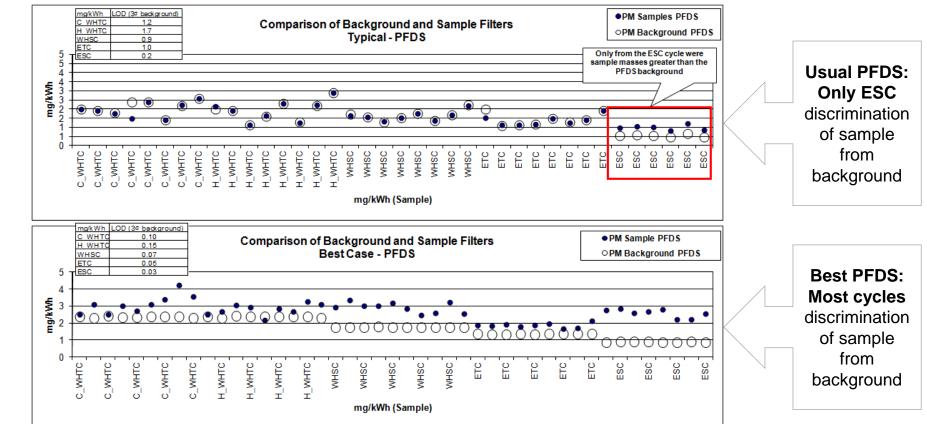
**Background Issues** 

results

### Challenging to discriminate sample PM from background PM on DPF–equipped HDD Engines



 From partial flow systems measurements, all systems could discriminate the ESC cycles' results and one system could discriminate most cycles. Only 1 of 5 full-flow CVS could even discriminate ESC

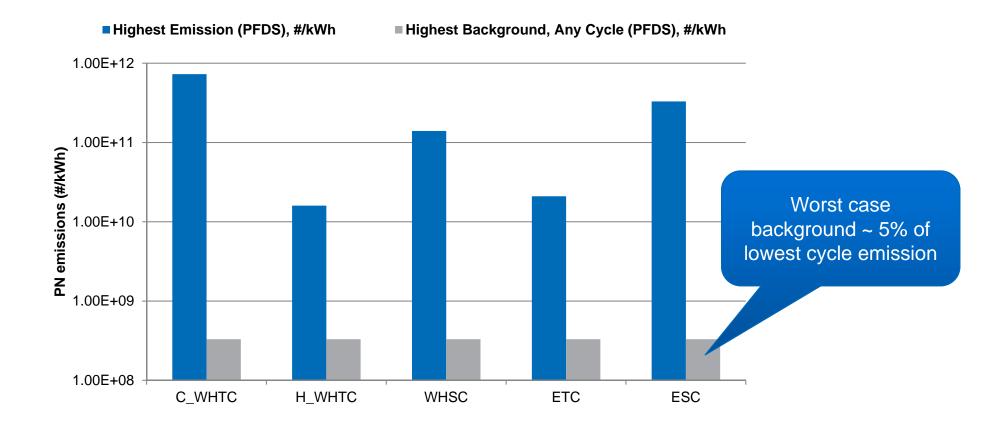


- The ESC results are attributed to two influences:
  - Thermal desorption of volatiles from the exhaust & dilution system increased contribution to PM
  - Passive regeneration at full load condition reduces DPF filtration efficiency- leads increased carbon-based particulate emissions

#### Simple to discriminate sample PN from background PN on DPF– equipped HDD Engines – Partial flow dilution systems



- PN (which doesn't include the volatiles), measured in parallel to the previous PM results, showed clear separation between PN background and emissions cycle results
  - In the worst case, PFDS background levels were 5% of sample levels
    - All cycles' PN results can be discriminated from background





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Is there still a role for the mass metric?

#### Is there still a role for the mass metric?







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## It appears that PM may be unable to distinguish mass emissions from dilution system background levels – so what does this mean for the metric?



- In standard testing, PM appears to be insufficiently sensitive to accurately quantify PM mass emissions at the levels seen post-DPF from HD engines
  - Typically 1 to 3mg/kWh
- Euro VI emissions limits over the WHTC and WHSC are 10mg/kWh
  - PM method is entirely suitable for indicating a pass/fail below 10mg/kWh, so does it merely become a **qualitative** metric, or is it no longer at all relevant?
- Euro VI includes factoring-in the contribution from active regenerations of emissions control systems, so PM here will increase significantly
  - Most of the PM increase, compared with a non-regenerating test, will come from volatile contributions that are true PM emissions
    - Has to be measured gravimetrically, as the particle number method will not quantify these components; levels expected to be high enough to be quantifiable in all dilution systems
- Euro VI solutions usually place the SCR system downstream of the DPF and there is the possibility that unreacted urea and by-products of urea hydrolysis/ side reactions may produce semi-volatile materials that would be legitimate contributors to PM
  - These too will not be adequately quantified by PN, and will need to be controlled using the PM approach



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#### Conclusions



- The PN method has entered the European legislative framework and it enables regulators to mandate emissions control systems that reduce elemental carbon (& PM) emissions to very low levels
- The PM metric enables a reliable pass/fail measurement to be made for Euro VI PM levels (10mg/kWh), but accurate quantification is difficult due to sampling issues
- However...
  - Active DPF regenerations are likely to increase particulate mass levels through contribution of volatile and semi-volatile materials
  - Incomplete decomposition, and side reactions, of reductants used for SCR may lead to contributions to PM that are not detected by the PN method
- Quantified PM emissions from Euro VI engines may well be higher than from DPFequipped Euro V engines, and the mass metric will be required to consider the contribution from semi-volatile materials
- The PM method still has relevance at the next regulatory stage. It's not dead. Yet...



#### Wrap-up



 Thanks to my co-authors, and also to PMP colleagues across Europe, Asia and the US too numerous to mention...



# Thanks for listening!

• Any questions?