

## **Particle Measurement Programme**

### ***Overview, conclusions and outlook from the UN-ECE GRPE inter-laboratory exercise for heavy-duty engines***

J. Andersson<sup>1</sup>, A. Mamakos<sup>2</sup>, G. Martini<sup>2</sup>

14th ETH Conference on Combustion Generated Nanoparticles

1st - 4th August 2010

<sup>1</sup> Ricardo UK; <sup>2</sup>JRC IES Ispra. Italy

After 2 years, 5 laboratories, 6 sets of tests and a wide-range of additional investigative work, the Heavy-duty Engine Inter-laboratory Validation Exercise has completed. During this work particle number and mass methodologies, developed during previous phases of the PMP, have been evaluated for repeatability and reproducibility, and the emissions levels from a DPF-equipped diesel engine have been studied in detail.

The test engine was an Iveco Cursor 8 (7.8 litres, 6 cylinders) in Euro III build, retrofitted with a CRT-type Pt-based DOC – cordierite wall-flow DPF. All tests were conducted using RF06-03 fuel and the OEM recommended lubricant (BP Vanellus E8) which was a fully synthetic 5W/30 oil with <0.2% sulphur. Test cycles included at least 8 repetitions of ETC, ESC, WHTC and WHSC at 5 different test labs, with one lab testing twice.

The particle number methodology employs a condensation nucleus counter with a carefully defined counting efficiency inlet, but uses sample pre-conditioning to eliminate the most volatile particles which may contribute significantly to variability. The regulatory 'solid particles' are defined by the measurement equipment in the size range ~23nm to 2.5µm and able to survive evaporation after heating to between 300 °C and 400 °C.

Two Golden Particle Measurement Systems (GPMS; Horiba SPCS) were circulated between laboratories, one measuring from a full flow dilution system (FFDS) and the other from a partial flow dilution system (PFDS). Staff from the European Commission, and the Golden Engineer, supported testing at each laboratory.

The particulate mass methodology drew substantially on the approach used in the US since 2007, validated in earlier phases of the PMP, and is not very different to that present in current (Euro V) and future (GTR) procedures.

PM levels from all test cycles were generally < 6 mg/kWh, with no obvious difference between cold and hot tests. However, high and variable background levels in the FFDS from 2 labs led to elevated PM. The PFDS showed consistently low and stable PM backgrounds from all labs, and repeatability from PFDS (~20%) was superior to that of the FFDS studied (~50%). Reproducibility levels of PM from the two dilution systems were similar (~40%). Notably background levels of ~2mg/kWh were similar to the emissions levels from most hot start cycles in both dilution systems.

Additional experiments conducted during this work indicated that PM emissions strongly depend on sampling approach and filter media: With the TX40 medium, repeatability can be seen to be optimized for filter face velocities in the ~70 to 100 cm/s range. Pre-baking of filters has no beneficial effect (no significant residual HCs). It is also clear that with TX40, relative to Teflon membrane filters, there are significant volatile artefacts that result in the mass collected on Teflon membrane filters being on average 70% lower than TX40. Comparing samples taken with a pair of TX40

filters in series (sample and back-up) with a single filter sample, deletion of the back-up filter reduced collected mass by ~30%.

PN emissions from all cycles were less than  $5 \times 10^{11}$ /kWh, with highest emissions from both PFDS and FFDS from the cold-start WHTC. Lowest emissions were seen from the test cycles that do not have substantial periods of passive regeneration (WHTC hot and ETC), but higher levels and increased variability of results were apparent from ESC and WHSC due to passive regeneration reducing the filter cake efficiency. Despite this impact on filtration efficiency, emissions from all hot cycles were still  $< 4 \times 10^{11}$ /kWh and in some cases lower than  $10^9$ /kWh.

As seen in the PM results, background contributions from some FFDS had a major impact on PN levels, and hot start cycles levels could not be discriminated from the background. Notwithstanding, at all labs cold start and ESC cycle results were broadly similar. PN background levels from PFDS were consistently low across labs at between  $10^8$  and  $10^9$ /kWh. This enabled an accurate quantification of the PN emissions of the various cycles to be determined at all labs.

At labs with very low FFDS background levels the emissions levels recorded were highly similar to the levels seen from the corresponding PFDS tests.

Due to the high sensitivity of the PN method, the repeatability of the measurement varied from cycle-to-cycle: best from high emissions, relatively cool cycles such as the WHTC (~20%) and poorest from the lower emissions hot, passively regenerating WHSC (~60%). Reproducibility ranged from ~40% with the cold WHTC to ~80% (WHSC).

Direct comparisons of PN with PM at all facilities, including both low and high background CVS systems, did not indicate any correlation between mass and number. This is clear evidence that the two methods quantify the presence of different chemical materials. The volatiles collected by PM filter medium and particulate matter are the major difference. In addition, while PN varied by 3 orders of magnitude across the cycles tested, mass varied by less than a factor of 10. This hundred-fold difference in sensitivity illustrates the key advantage of PN in determining emissions levels rather than indicating a regulatory pass/fail.

A comparison of PCRf calibrated alternative (systems compliant with PMP requirements but from other manufacturers) and additional PN systems (those adhering to the same preconditioning principles but not necessarily containing all PMP system elements) with the GPMS indicated agreements of  $\pm 15\%$  over many orders of magnitude. Application of the PCRf approach does appear to converge different systems' results, but does not result in complete alignment.

The high-level conclusions of the work show that the PMP particle number measurement approach system enables an accurate quantification of the true particle number emissions of the engine, and that this methodology is well suited to regulatory use. The particulate mass method remains capable of discriminating mass emissions constituting pass and fail in a regulatory test.

A number of open issues still exist for consideration in the regulatory approach for PM and PN. Research into these should include:

- Background level reduction to enable improved PM discrimination

At present only one PFDS system was able to discriminate emissions from more than one of four emissions cycles from the background PM levels in that system. Cleaning

and preconditioning approaches are required to enable a more accurate determination of PM emissions to be made.

- Dilution air filtration of alternative PN systems

A noteworthy contribution to the background PN levels measured in some PN measurement systems comes from the particles present in the dilution air. To minimize the contribution of particles from this source, the practicality of using dilution air filtration of better than 99.999% should be explored.

- VPR calibration

At present the difference between PN systems has been converged to ~15% using the Particle Concentration Reduction Factor (PCRF) calibration approach. It is possible that variations between the approaches used by instrument manufacturers and their interpretations of the methods employed are contributing to this variability. A round-robin exercise, coordinated by JRC, is currently underway to investigate the possibility of further improving the accuracy of PN measurements.

- <23nm solid particles

Different engine technologies, combustion systems and calibration approaches may lead to emissions of solid particles that are below the current  $d_{50}$  of the PMP system (~23nm). The feasibility of reducing the lower detection limit below 23 nm without introducing the risk of interference from volatile particles should be explored.

# Particle Measurement Programme

*Overview, conclusions and outlook from the UN-ECE GRPE inter-laboratory exercise for heavy-duty engines*

J. Andersson<sup>1</sup>, A. Mamakos<sup>2</sup>, G. Martini<sup>2</sup>  
14th ETH Conference on Combustion Generated Nanoparticles  
1<sup>st</sup> - 4<sup>th</sup> August 2010

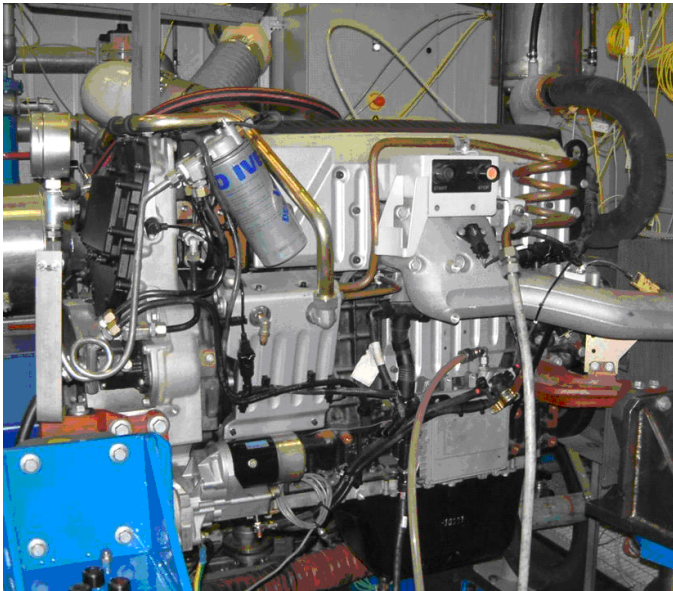
<sup>1</sup> Ricardo UK; <sup>2</sup>JRC IES Ispra. Italy

- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- Conclusions
- Outlook and Open Issues

- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- Conclusions
- Outlook and Open Issues

## What's the PMP All About?

- Inter-governmental research programme under the auspices of UNECE GRPE
  - Mandate was to develop techniques to replace or complement the particulate mass measurement method
  - applicable to Light Duty Vehicle & Heavy Duty Engine type approval testing
- PMP also to deliver
  - Emissions levels from vehicle and engine technologies
  - Validated measurement methodologies
  - Written procedures ready for regulatory use
- Light-duty procedures now present in R83 for Euro 5b/Euro 6
- Heavy-duty procedure validation reported here...



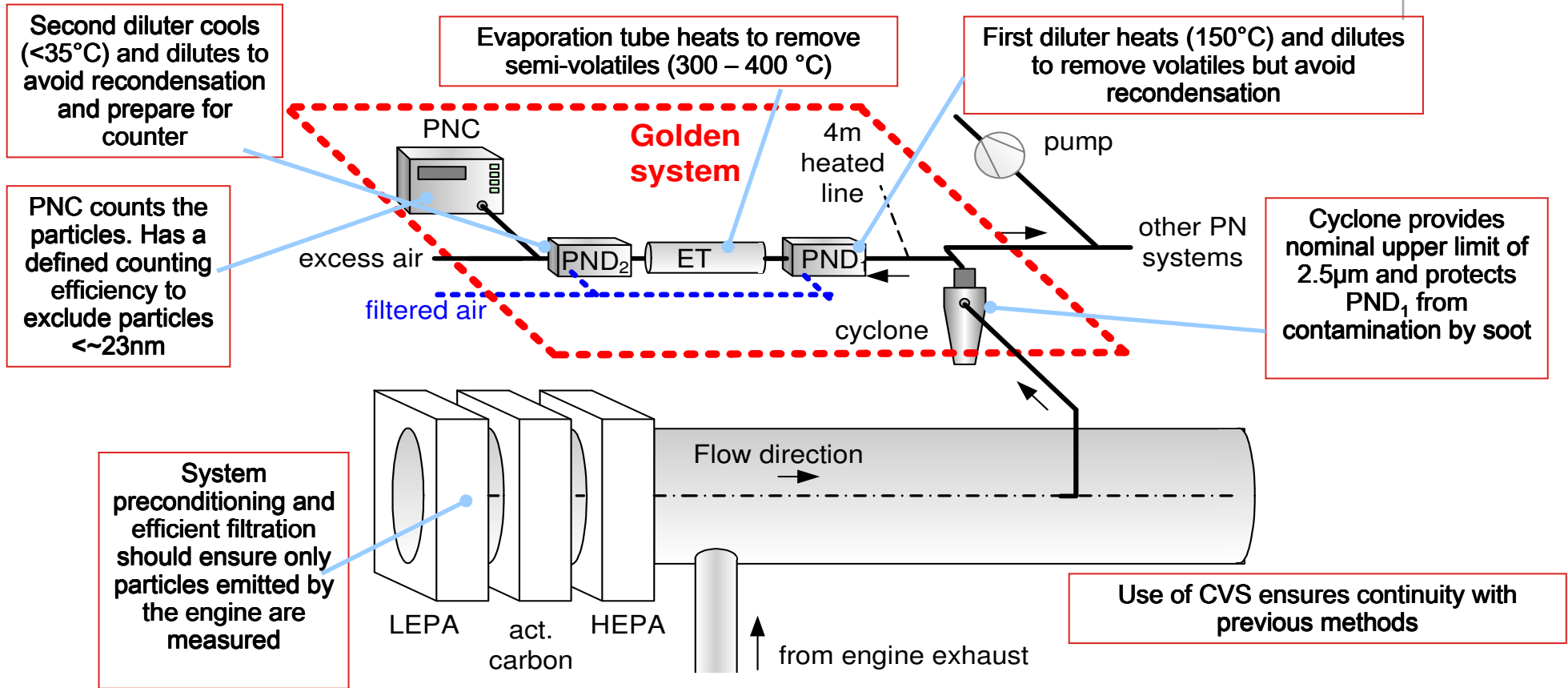
- 7.8 lt – 6 cylinder **Euro III IVECO Cursor 8**
- **Retrofit DPF**
  - CRT: Pt-based oxidation catalyst (4.25 lt) & cordierite wall flow filter (~24 lt)
- **Reference Fuel**
  - RF06-03 fuel (<10 ppm S)
- **OEM Lubricant**
  - BP Vanellus E8 fully synthetic 5W/30 lubricant (<0.2% S)
- Test Matrix addressed replicate European and World Cycles
  - **ETC, ESC, WHTC, WHSC**
- At least **8 repetitions** of each test cycle at each lab
- 5 labs, one (JRC) testing twice = **6 sets**

Previous lab	Day 0	Days 1-7	Day 8
	oil change	IFV	IFV
	2h ESC Mode 10	cold WHTC	cold WHTC
	3 x ETC	10 minute soak	10 minute soak
		hot WHTC	hot WHTC
		10 minutes at WHSC mode 9	10 minutes at WHSC mode 9
		WHSC	WHSC
		CP	CP
		ETC	ETC
		CP	CP
		ESC	ESC
*2 hours at ESC Mode 10	Precon	Precon	*2 hours at ESC Mode 10

ESC - European Steady State Cycle for emissions measurement [30 min]  
 ETC - European Transient Cycle for emissions measurement [30 min]  
 WHTC - World Harmonised Steady State Cycle for emissions measurement [30 min]  
 WHTC - World Harmonised Transient Cycle for emissions measurement [30 min]  
 IFV - Instrument Functional Verification  
 CP - Continuity Protocol  
 Precon - 15 minutes ESC mode 10, 30 minutes ESC mode 7  
 \* DPF regeneration only required if oil change and conditioning not performed



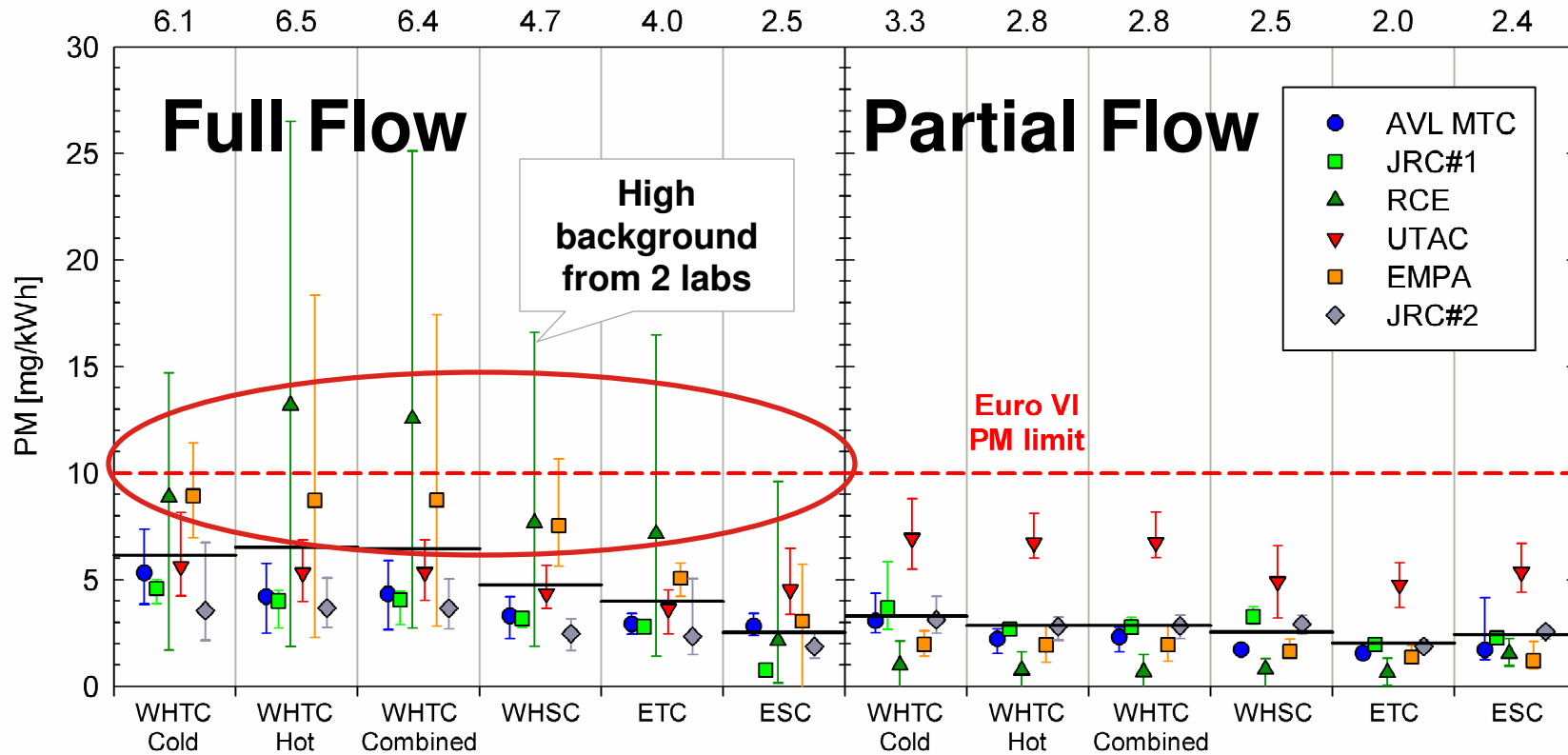
## Particle Number Measurement Approach



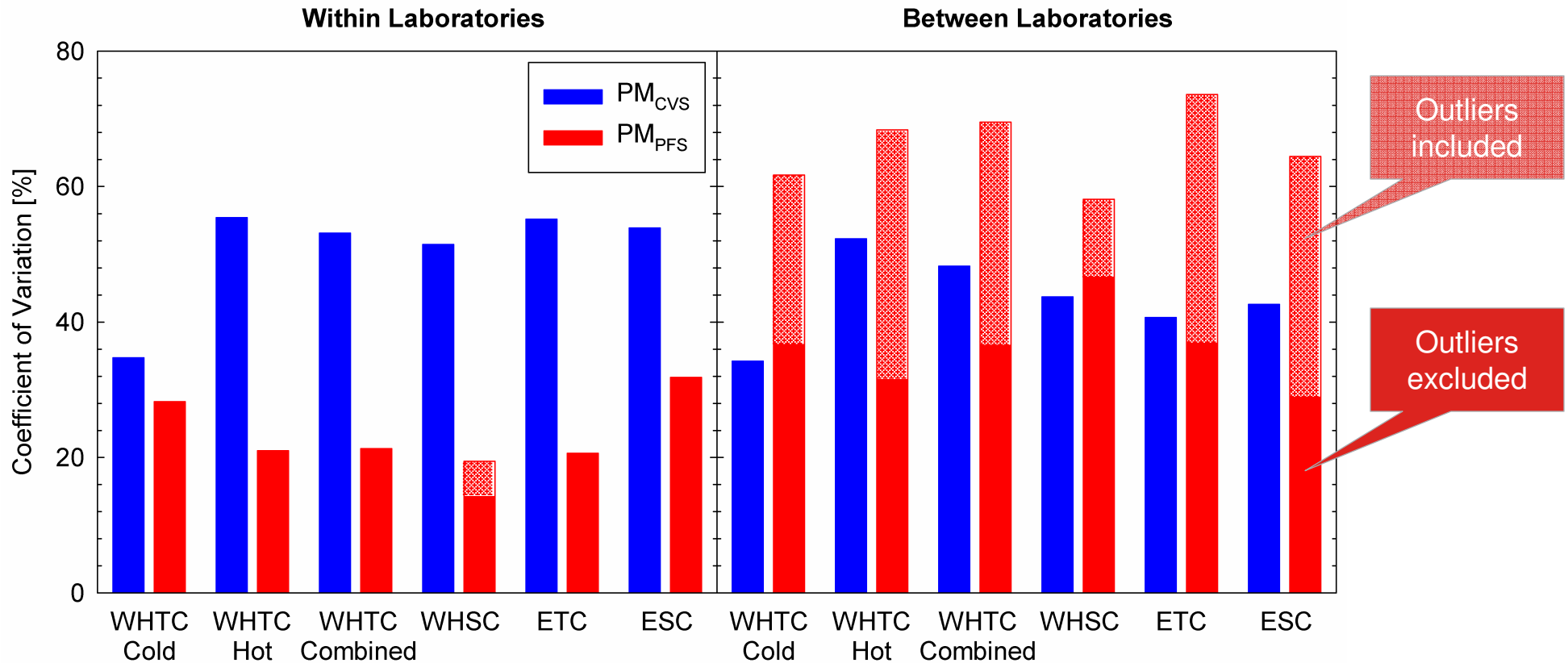
- Measurement employs a condensation nucleus counter, but uses sample pre-conditioning to eliminate the most volatile particles which may contribute significantly to variability
- Solid particles defined by the measurement equipment
  - ~23nm to 2.5µm and surviving evaporation at (or above) 300°C
  - Analogous to heated FID hydrocarbon method

- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- Conclusions
- Outlook and Open Issues

## PM Emission Levels Generally Well below 10mg/kWh When Measured from Full and Partial Flow Dilution Systems

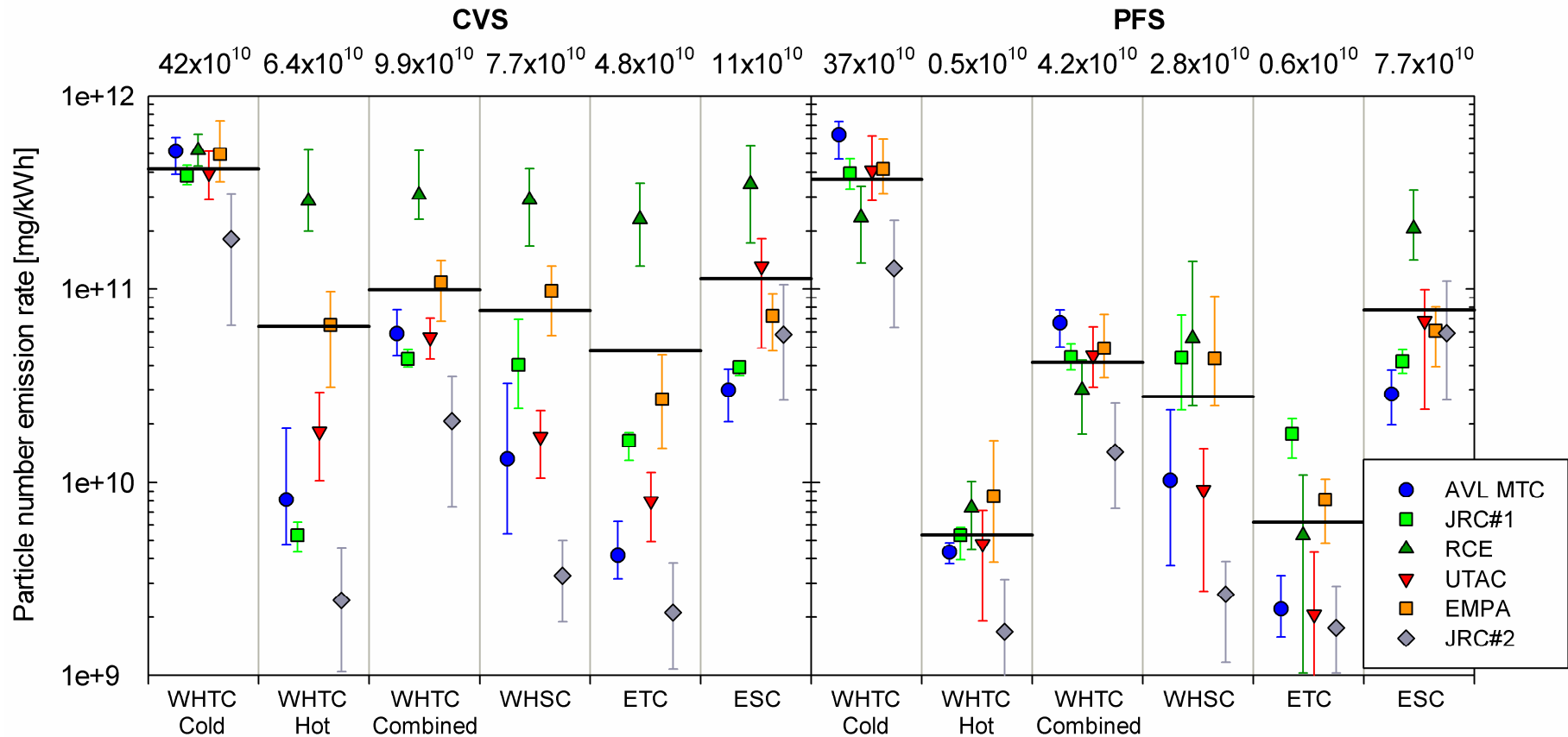


- PM levels generally < 6 mg/kWh, with no obvious difference between cold and hot tests
- High and variable background levels in the CVS tunnel at 2 labs led to elevated PM
- Low and stable backgrounds in PFDS led to more consistent PM

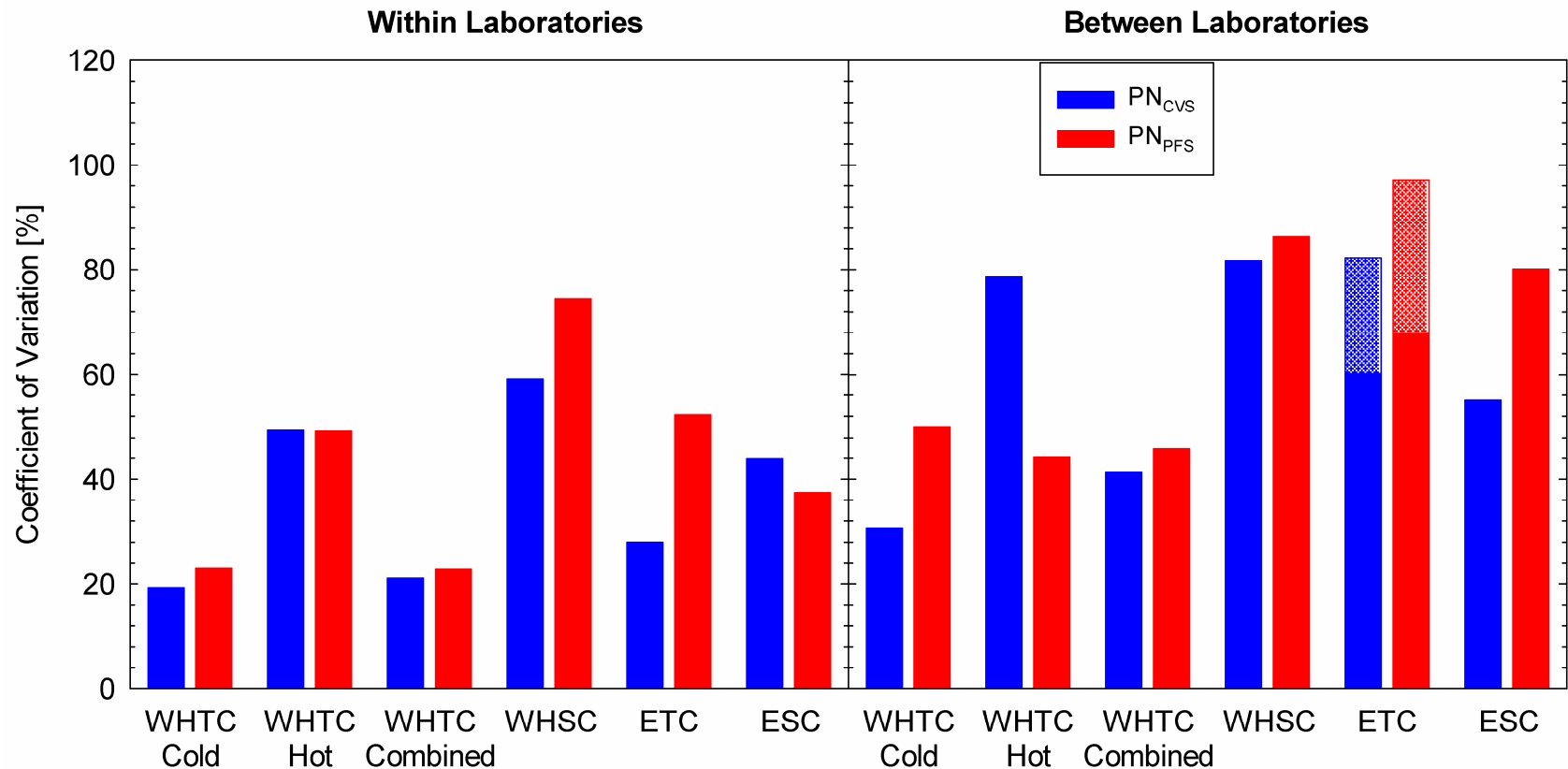


- PM measured at PFS were more repeatable from those determined from the CVS (~20% compared to 50%).
- Reproducibility at ~40% from both systems after excluding outliers (PFS results from UTAC)

## Greater Confidence in PN Emission Levels Comparability from PFS – due to high background CVS systems

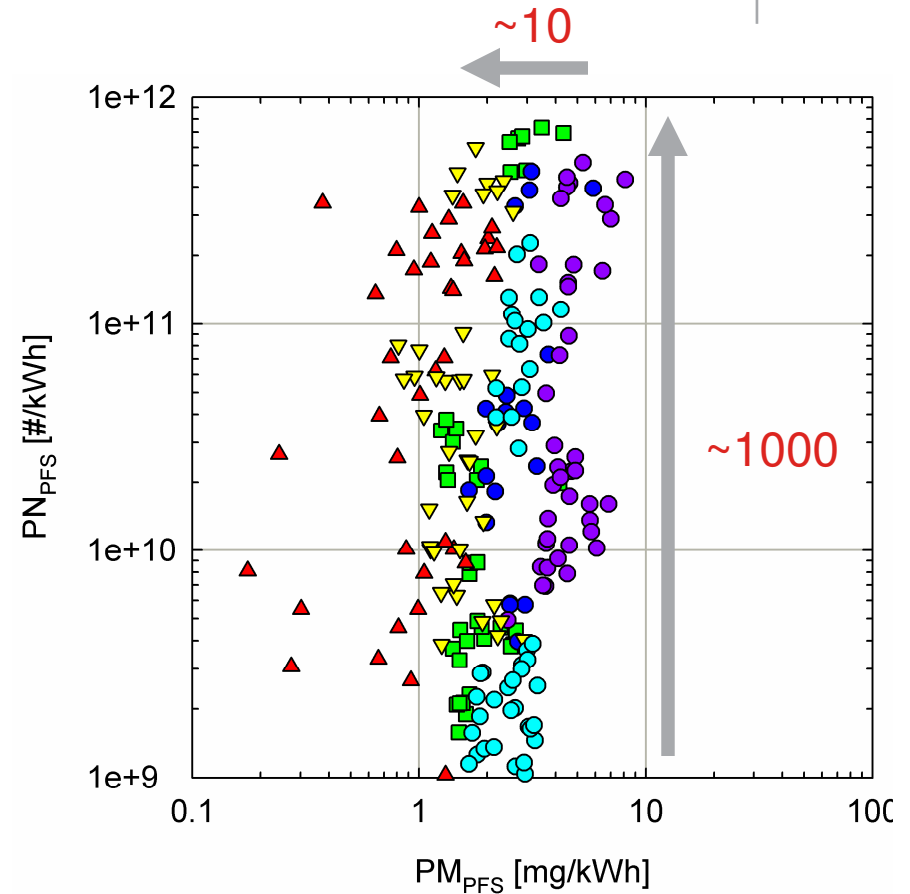
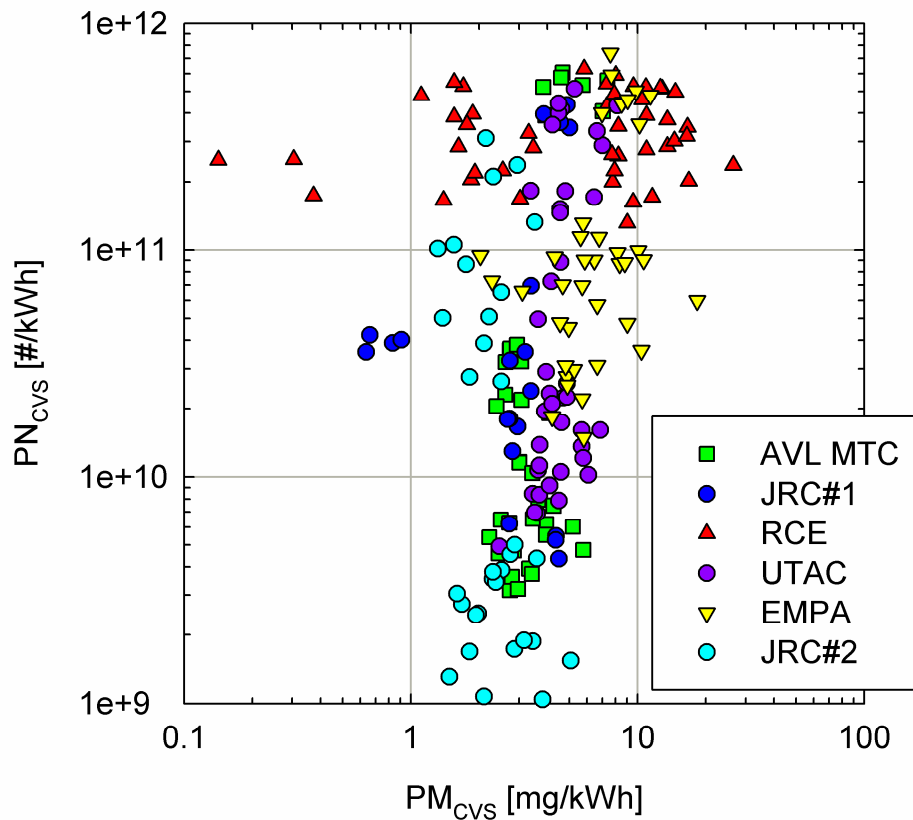


- PN of  $\sim 4 \times 10^{11}$  #/kWh over cold start WHTC
- Lowest emissions from the test cycles that do not have substantial periods of passive regeneration (WHTC hot - ETC)
- Higher levels and increased variability of results from ESC and WHSC due to passive regeneration. But: emission levels below  $4 \times 10^{11}$  #/kWh



- Repeatability ranged between ~20% (over the high emission cold WHTC) and ~60% (over the high temperature WHSC) for both CVS and PFS after removing outliers
- Reproducibility ranged between ~40% (cold WHTC) and 80% (WHSC).

## No Correlation between PN & PM in any Test Facility



- PM does not correlate with PN from either CVS or PFS
- PN method much more sensitive
  - PN levels vary over three orders of magnitude
  - PM varies by less than 1 order of magnitude, even for clean dilution systems

- Based upon repeatability and reproducibility, PM and PN performed similarly
- Cycle emissions' PM levels very close to background, PN levels at least 10x higher

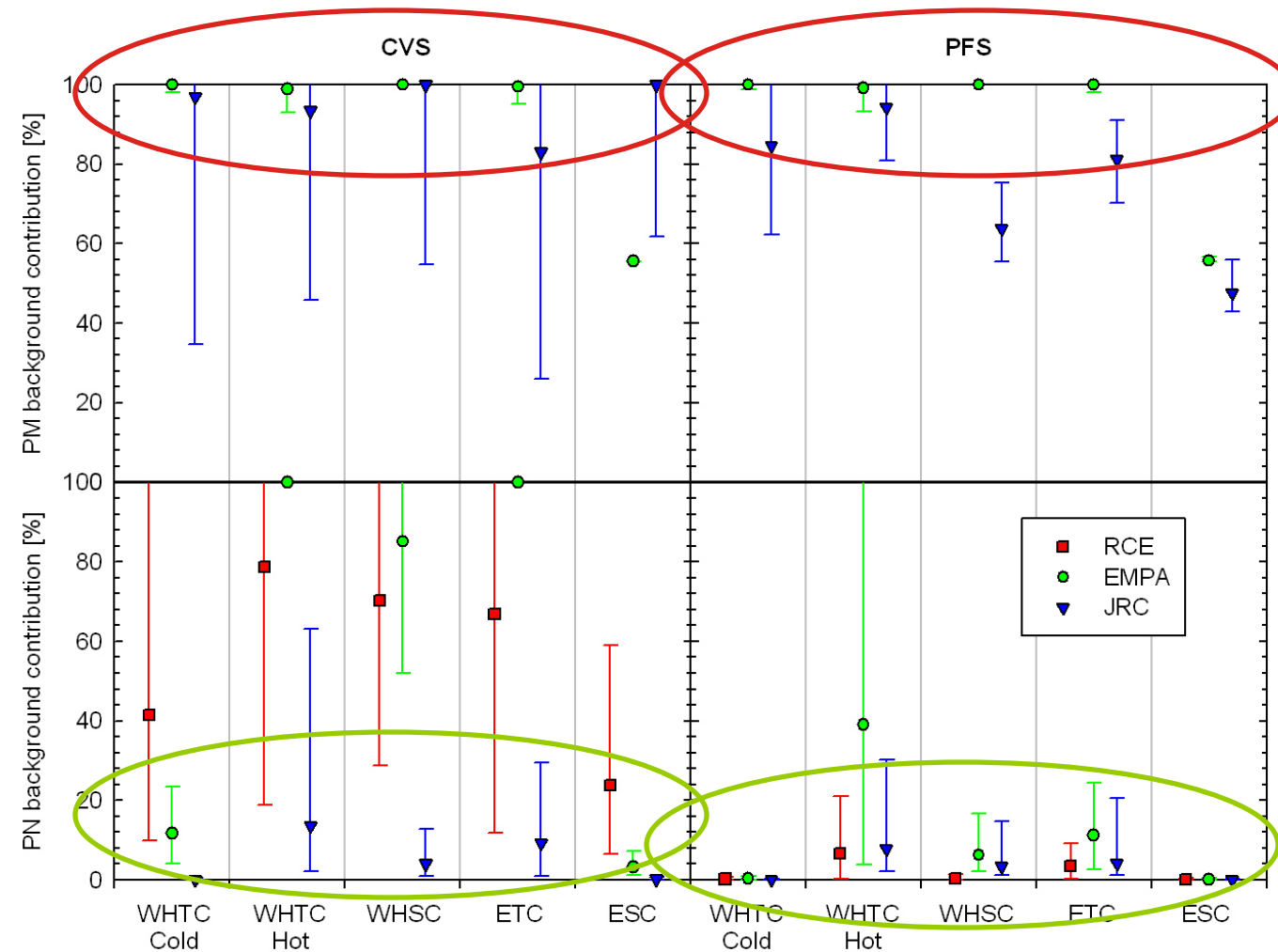
	PM		PN	
	FFDS	PFDS	FFDS	PFDS
<b>Proposed Emissions Limit Values Euro VI</b>	<b>10mg/kWh (WHSC and WHTC)</b>		<b>6 x 10<sup>11</sup>/kWh (WHTC) 8 x 10<sup>11</sup>/kWh (WHSC)</b>	
<b>Limit of detection (3s * background) - 'dirty' dilution system</b>	<b>≥10mg/kWh</b>	<b>~2mg/kWh</b>	<b>~10<sup>11</sup>/kWh</b>	<b>&lt;10<sup>10</sup>/kWh</b>
<b>Limit of detection (3s * background) - 'clean' dilution system</b>	<b>~0.25mg/kWh</b>	<b>~0.15mg/kWh</b>	<b>~10<sup>9</sup>/kWh</b>	<b>~10<sup>9</sup>/kWh</b>
<b>Background - 'clean' dilution system</b>	<b>~2.0mg/kWh</b>	<b>~1.5mg/kWh</b>	<b>~10<sup>9</sup>/kWh</b>	<b>~3x10<sup>8</sup>/kWh</b>
<b>Emissions levels from 'low emitting cycle' in clean dilution system</b>	<b>~2.0mg/kWh</b>	<b>~2.0mg/kWh</b>	<b>&lt;10<sup>10</sup>/kWh</b>	<b>&lt;10<sup>10</sup>/kWh</b>
<b>Interlab Variance CoV (%)</b>	<b>40%</b>	<b>40%</b>	<b>40% to 80%</b>	<b>40% to 80%</b>
<b>Intralab Variance CoV (%)</b>	<b>50%</b>	<b>20%</b>	<b>20% to 60%</b>	<b>20% to 60%</b>



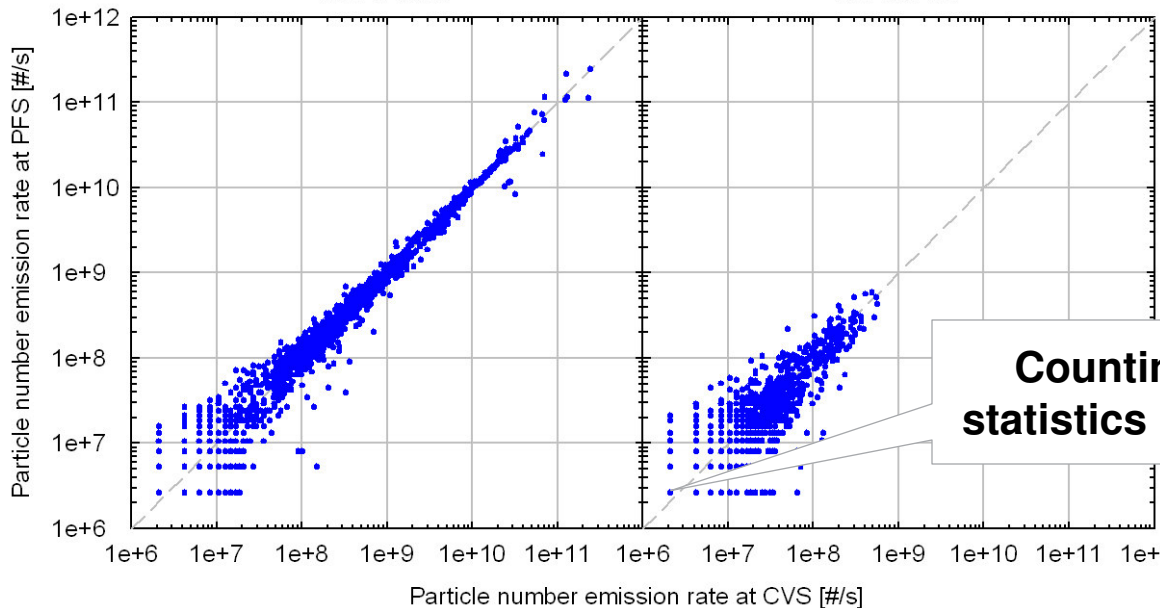
- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- Conclusions
- Outlook and Open Issues

## Background contribution to PM and PN emissions

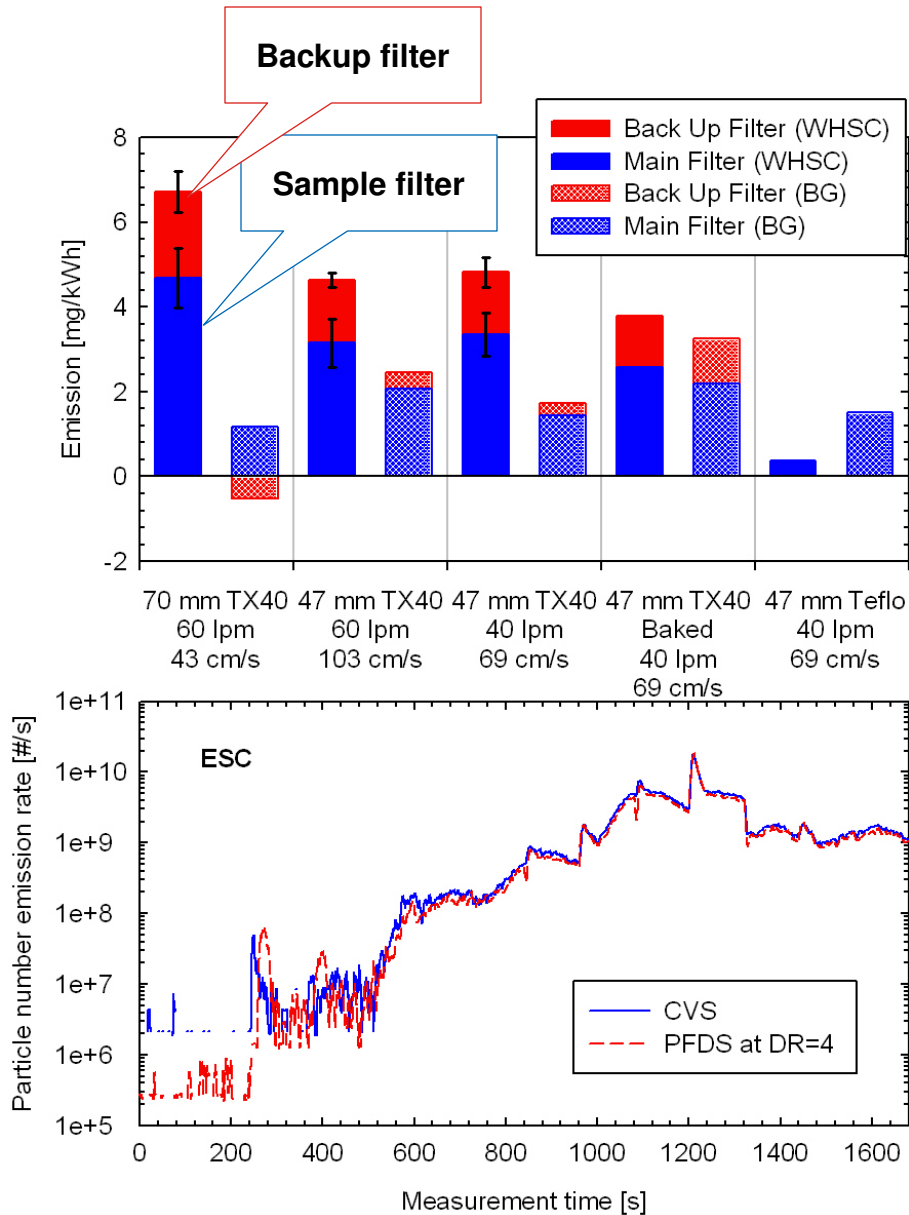
- **PM**
- CVS results show no discrimination of PM from background except EMPA over ESC (low volatility HCs and sample time effects)
- PFS system results at JRC suggest that it is just possible to resolve PM emissions from background in a new, very clean PFS
- **PN**
- Some labs (EMPA and RCE) suffered from high PN background levels in the CVS but this was low compared to WHTC cold and ESC emissions levels
- The contribution of background in PFS systems was lower than 20%



- PM levels from CVS and PFS broadly similar ( $\pm 50\%$ ) for labs with low background.
  - Likely demonstrates similarity between background levels
    - the agreement does improve with increasing emission levels
- PN agreement was better than  $\pm 20\%$  at emission levels above the background

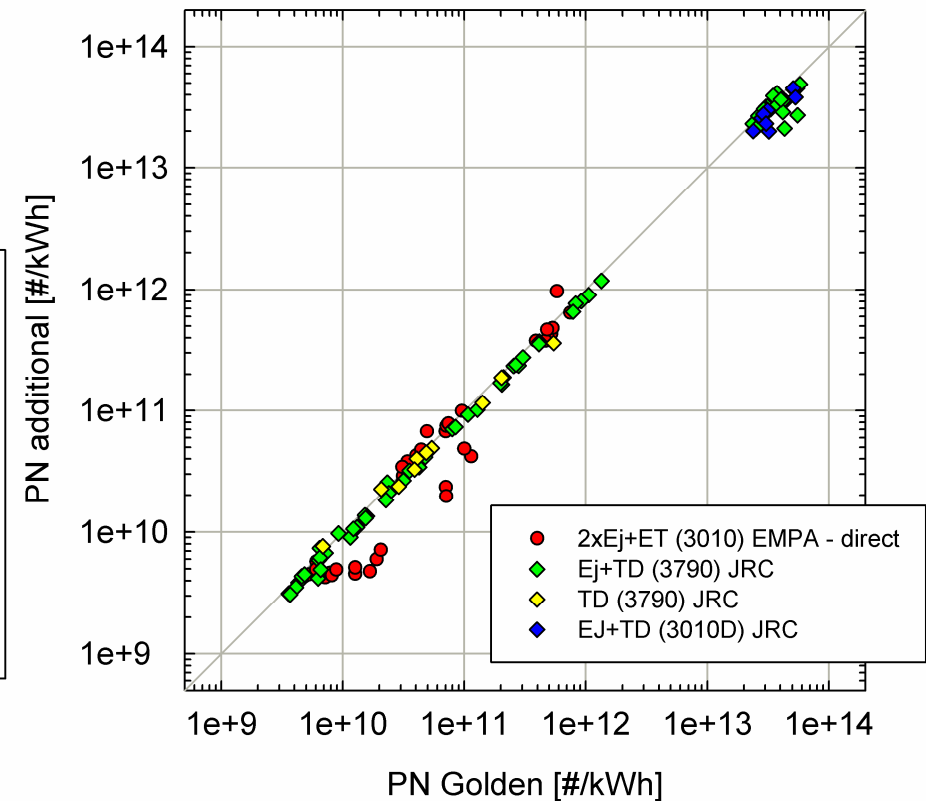
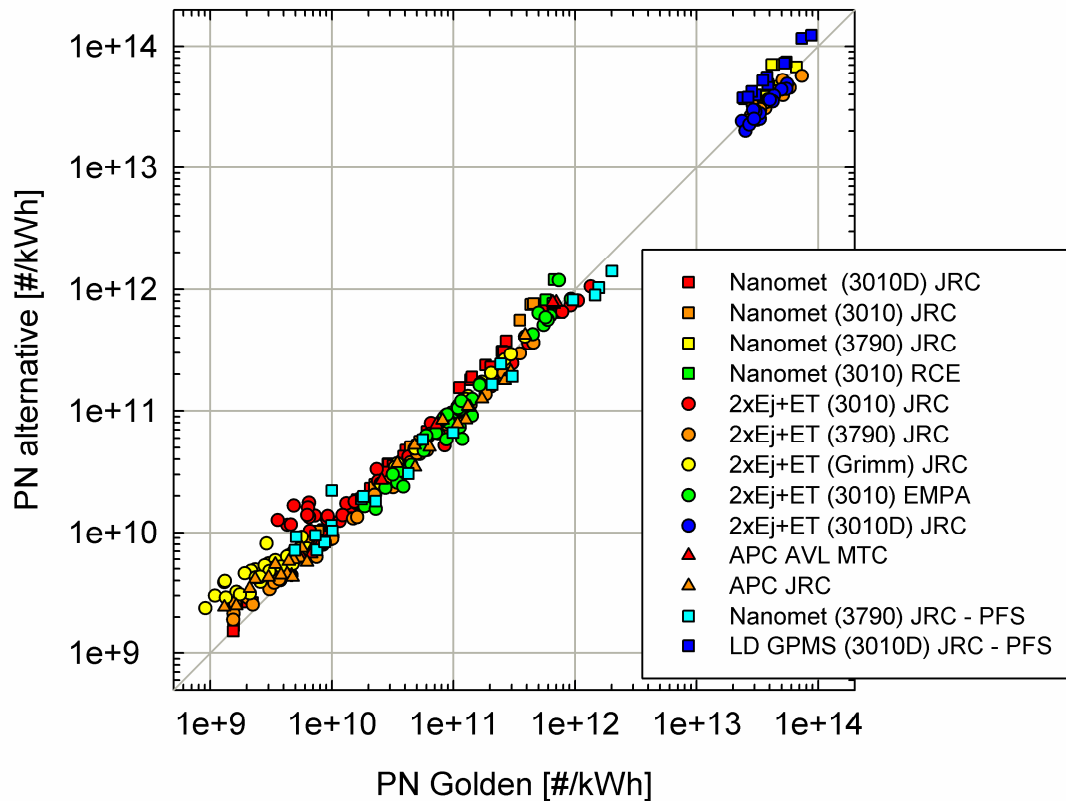


- Real time responses of the PFS and CVS also correlate very well over 5 orders of magnitude



- PM emissions strongly depend on sampling approach and filter media
  - Repeatability improves for filter face velocities in the ~70 to 100 cm/s range
  - Pre-baking of filters has no beneficial effect (no significant residual HCs)
  - Significant volatile artefacts
    - Mass collected on Teflo filters 63% to 81% lower (volatile artefact) than TX40
    - Back-up filters collect ~30% of primary filter mass
- PN methodology is much more robust.
  - Acceptable accuracy (better than 15%) was observed even at an extreme setting of a constant DR~4

## Alternative & Additional PN systems agree to +/-15% with GPMS (with PCRf retrospectively applied)



- Calibrated alternative VPR systems generally agreed with GPMS within  $\pm 15\%$ 
  - PCRf does converge results from different systems but not complete alignment
- The agreement holds over a range of 5 orders of magnitude but weakens at low concentrations due to background effects.

- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- **Conclusions**
- Outlook and Open Issues

- PM:
  - PM emissions were generally  $<6$  mg/kWh.
  - However, background levels were equivalent to drive cycle emissions levels.
- Particle number:
  - PN emission levels over cold WHTC were determined to be  $\sim 4 \times 10^{11}$  #/kWh with both CVS and PFS systems. At these emission levels, the background effect is insignificant.
  - PN emission levels over hot start WHTC and ETC cycles were  $< 2 \times 10^{10}$  #/kWh. Passive regeneration occurring over the WHSC and ESC cycles results in an increase of the emissions up to  $6 \times 10^{10}$  #/kWh
    - Background in some labs was a substantial influence from these cycles

- Particle Number#2:
  - Repeatability and reproducibility levels for the CVS and PFS were similar, ranging from:
    - ~20% and ~40%, respectively, over cold WHTC
    - ~70% and ~80%, respectively, over WHSC, due to passive regeneration effects
  - PFS systems showed lower backgrounds than CVS systems, but when the two systems had similar backgrounds, the correlation between PN emission levels was excellent.
  - Particle number emissions do not correlate with PM results, as the latter are almost entirely volatile material.
- Alternative Systems:
  - The majority of the alternative systems correlated closely with the GPMS, the difference being on average smaller than  $\pm 15\%$  after accounting for the PCRf values and the slopes of the CPCs.



## Conclusions#3: PM and PN for Euro VI Legislation

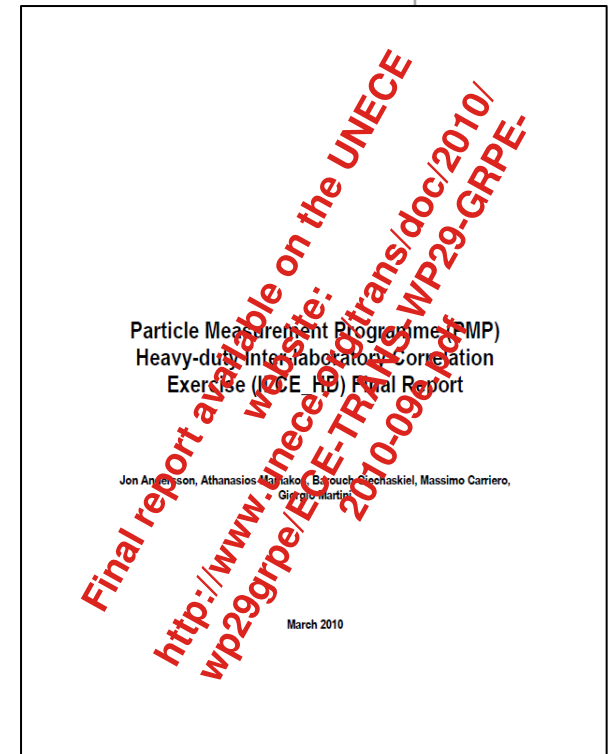
- The PM method is suitable for discriminating PM emissions on a pass / fail basis at 10mg/kWh from both full and partial flow dilution systems
  - At efficient wallflow DPF emissions levels (c. 2mg/kWh), even clean dilution systems have background PM levels similar to sample levels
- The PN method provides accurate quantification of particle emissions from engines with efficient DPF systems and is suited to regulatory use
  - Clean dilution system backgrounds are typically <10% of sample levels from engines with efficient wallflow DPFs

- Programme Outline
- PM and PN Methodologies
- Major Impacts on Measurements
- Conclusions
- Outlook and Open Issues

- Outlook
  - Final Report published on UN website, DG JRC to issue hardcopies
  - Updated R49 procedure available as draft
    - <http://www.unece.org/trans/doc/2010/wp29grpe/ECE-TRANS-WP29-GRPE-2010-07e.pdf>
  - PN limit values to include contribution from active regeneration; and limit values to be agreed imminently
- Further Areas of Study
  - Background:
    - Possibilities to reduce PFS background making PM resolvable at low levels
    - Dilution air filtration of alternative PN systems (HEPA or ULPA?)
  - VPR calibration
    - Round robin exercise currently underway investigating the possibility of further improving the accuracy of PN measurements.
  - <23nm solid particles
    - Feasibility of reducing the lower detection limit below 23 nm without introducing the risk of interference from volatile particles



Thank you for  
your attention!



## Acknowledgements

Chris Parkin, Jonathan Hall, Tim Pearson, Simon de Vries, Carl Jemma, Massimo Carriero, Barouch Giechaskiel, Jan Gaste, Gigy di Bernardo, Daniel Schreiber, Silke Weimer, Stefano Alessandrini, Fausto Forni, Francois Montigny, Dominique Lesueur, Maarten Kieft and many others...