The Abdus Salam International Centre for Theoretical Physics

United Nations Educational, Scientific and Cultural Organization

IAEA International Atomic Energy Agency

2256-3

Workshop on Aerosol Impact in the Environment: from Air Pollution to Climate Change

8 - 12 August 2011

Emission processes and regulation - combustion engines

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Joint Research Centre (JRC)

Particulate Emissions from Vehicles

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Contents

- Background information
- PMP work
- Late investigations

Two main sources:

– Combustion by-products emitted in the exhaust
  - Primary emissions
  - Secondary aerosol

– Non-exhaust emissions

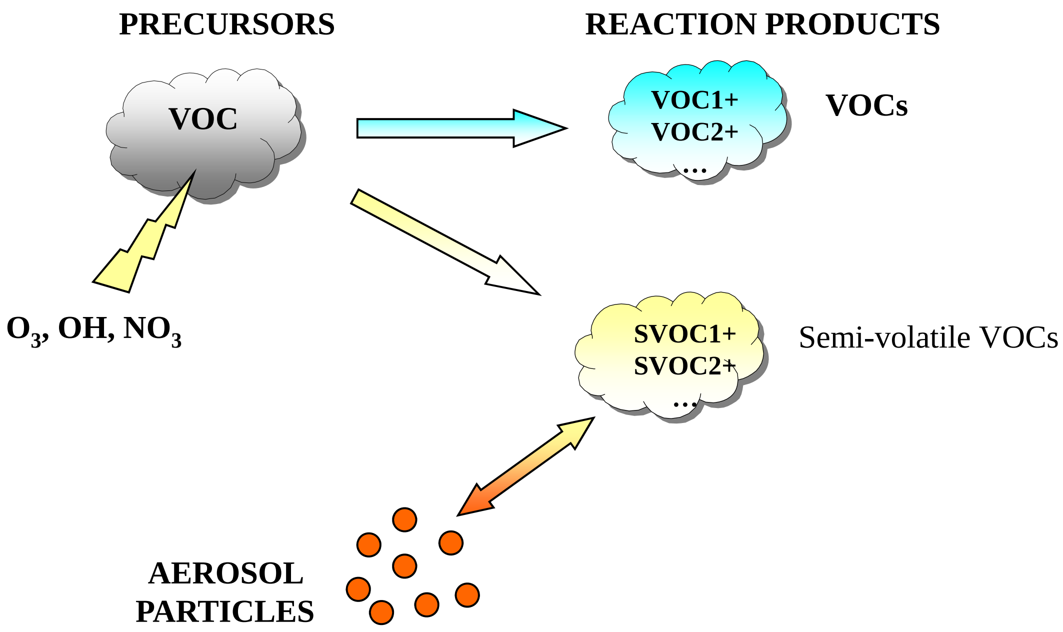
Vehicles are identified as a major contributor to ambient PM

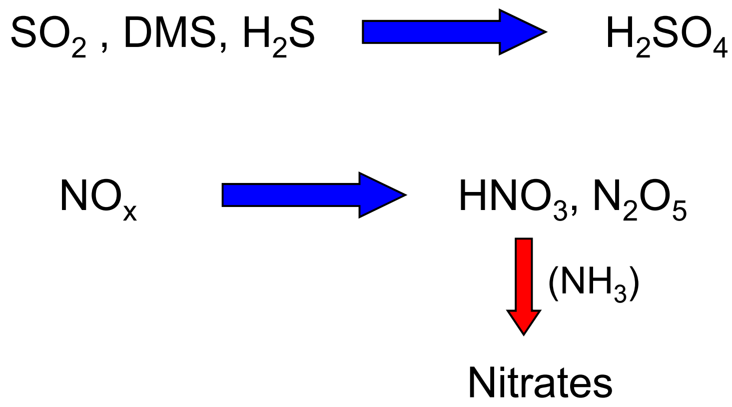
– Combustion by-products emitted in the exhaust:

- Directly (Primary emissions):
  - Solid particles forming due to incomplete combustion (soot, metallic ash)
- Indirectly (Secondary Aerosol):
  - Secondary particles forming from nucleation/condensation of gaseous precursors as the exhaust cools down and dilutes or through more complex chemistry (hydrocarbons, nitrates, sulphates)

Secondary aerosol

- Secondary aerosol originates from gas-to-particle conversion processes
  - Natural or anthropogenic depending on the precursors
  - Inorganic (sulphates, nitrates) or organic (VOCs oxidation)





Secondary aerosol

- Characteristics
  - Ultrafine particles
  - Long residence time
  - Very often the largest fraction of ambient PM during high concentration episodes

	Northern Hemisphere	Southern Hemisphere	Global
Sulphates:	145	55	200
Anthropogenic	106	15	122
Biogenic	25	32	57
Volcanic	14	7	21
Nitrates:			
Antropogenic	12.4	1.8	14.2
Natural	2.2	1.7	3.9
VOCs:			
Anthropogenic	0.15	0.45	0.6
Biogenic	8.2	7.4	16

Intergovernmental Panel on Climate Change (Tg/year)

Vehicles are identified as a major contributor to ambient PM

– **Non-exhaust emissions**: Mostly found in coarse mode (larger than ~ 1 μm)

- Brake wear: metal particles
- Tyre wear
- Resuspension/road wear: strongly depend on the type of pavement and tyres (e.g. studded tyres)

- COPERT IV model gives a figure of 14 mg/km PM10, of which 7 mg/km are PM2.5. For heavy duty vehicles the figures are 59 mg/km PM_{10} and 31 mg/km $PM_{2.5}$

Vehicles are identified as a major contributor to ambient PM

Non-exhaust emissions

- Brake wear emissions peak at ~ 6 μm (mass weighted) while tyre wear can be even larger (some studies by EMPA suggesting that all of them are above PM10).
- Of course an important parameter is the driving conditions. Emissions are expected to be higher in urban environments where drivers use the brake more often
- The chemistry of the particles depends strongly on the formulation employed which shows large variations from manufacturer to manufacturer but also change in different regions of the world (in US they do not use Fe that is employed in Europe). In general copper is identified as a main component of the emitted brake wear.
- Some late studies have also found nanosized particles emitted in very high quantities, which are presumably formed through evaporation/condensation occurring at the high temperatures developing during braking (could be well in excess of 600 C, at the contact points

Primary particulate emissions

- Do all the vehicles/engines emit particles?
- Are size and composition of particulates the same for all the vehicles/engines?

Primary particulate emissions

- The combustion process plays a fundamental role in the formation of particles

  − Petrol engines
    - Two-stroke engines (a mixture of petrol and lubricant is burned)
    - Four-stroke engines (only petrol)
    - Carburettor, Port Fuel Injection and Direct Injection (stoichiometric or lean burn)

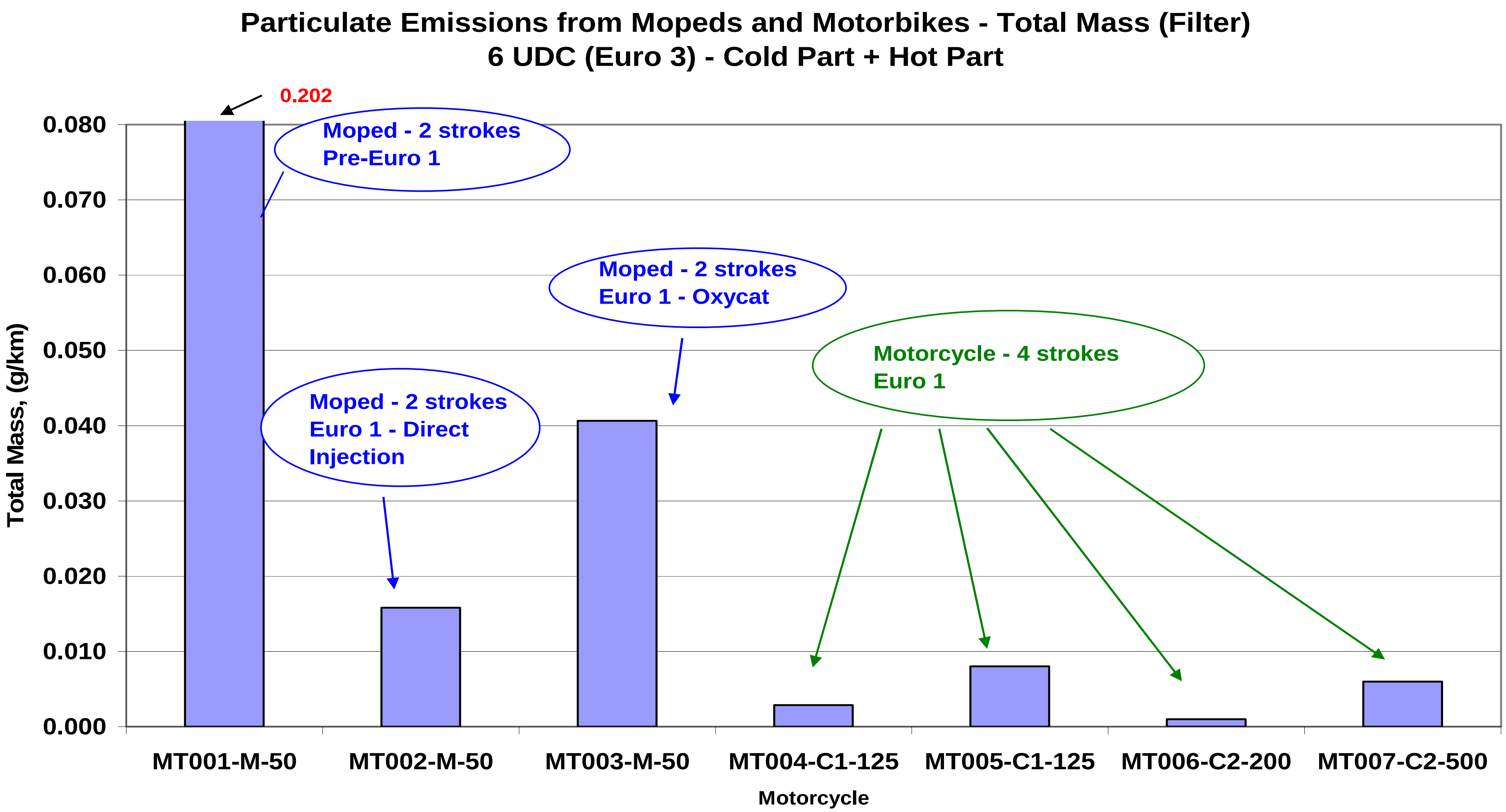
  − Diesel engines (gasoil)
    - Lean combustion with injection of fuel only towards the end of the compression phase

  Main critical factor:
  − Homogeneous combustion (Petrol PFI and GDI stoichiometric): In general no or very low PM emissions
  − Non-homogeneous combustion (Diesel and GDI lean burn): In general high PM emissions

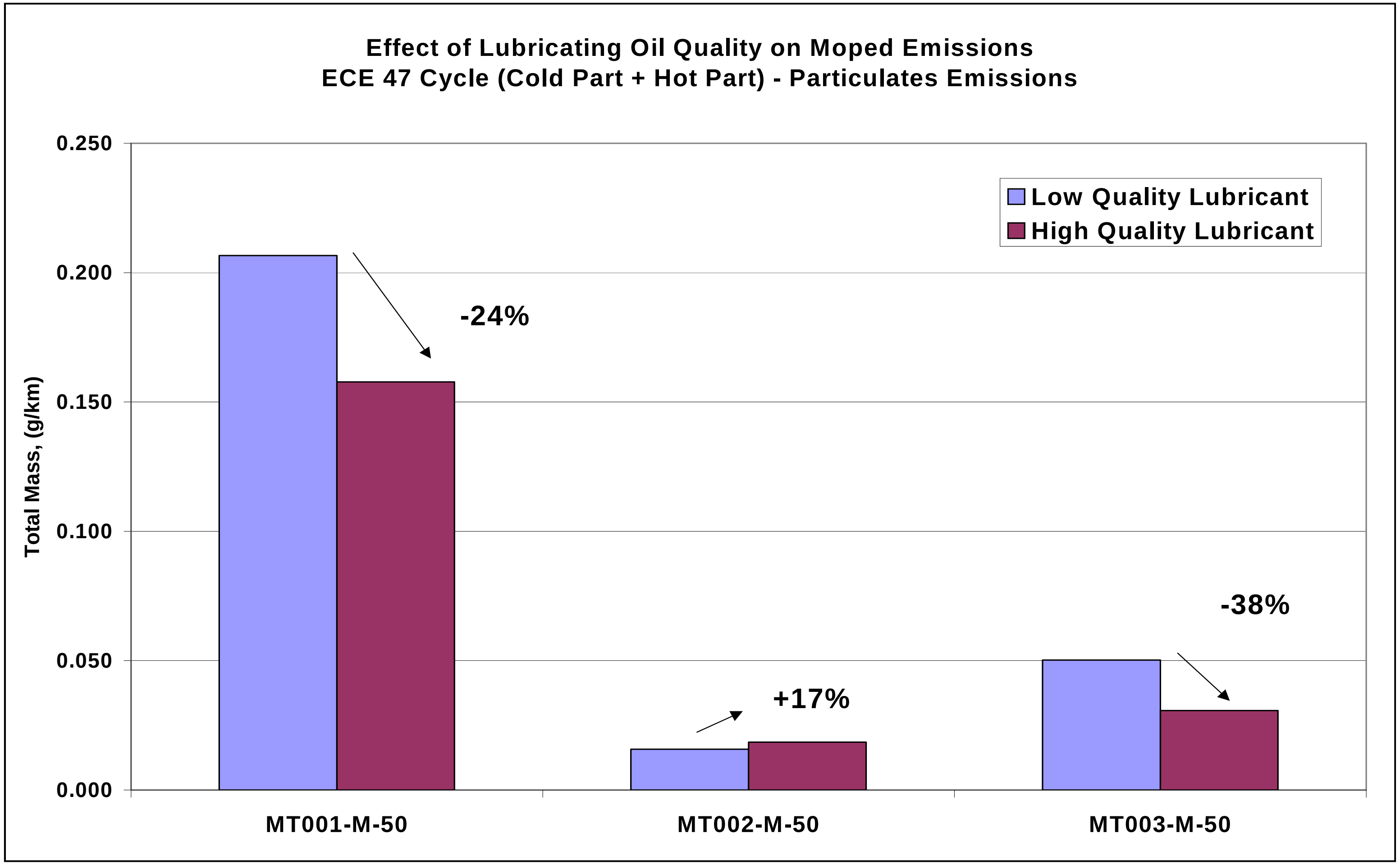
Primary particulate emissions

- Emissions from motorcycles

Particulate Emissions



Particulate Emissions from 2-stroke engines

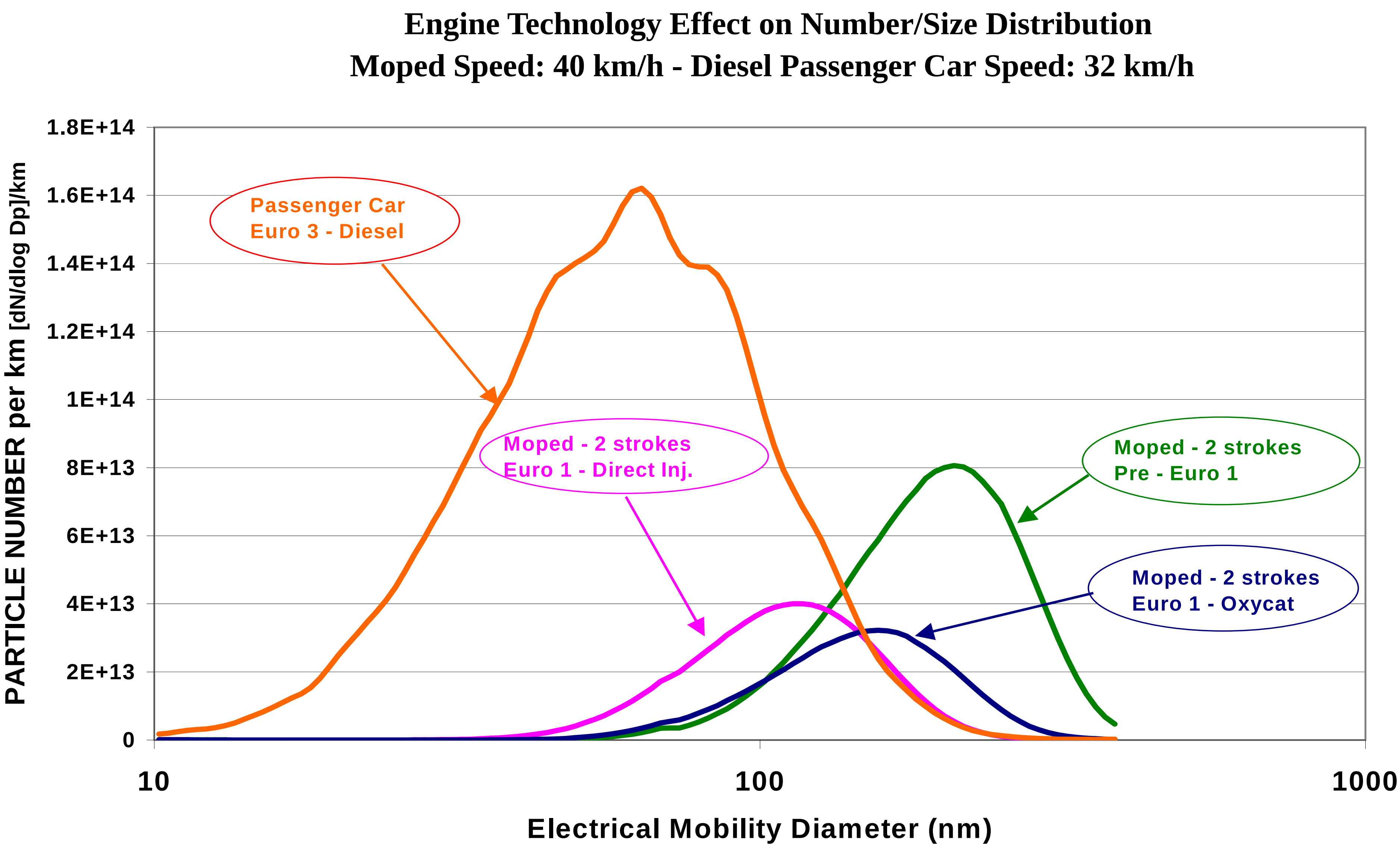


Euro 0 - Carburettor

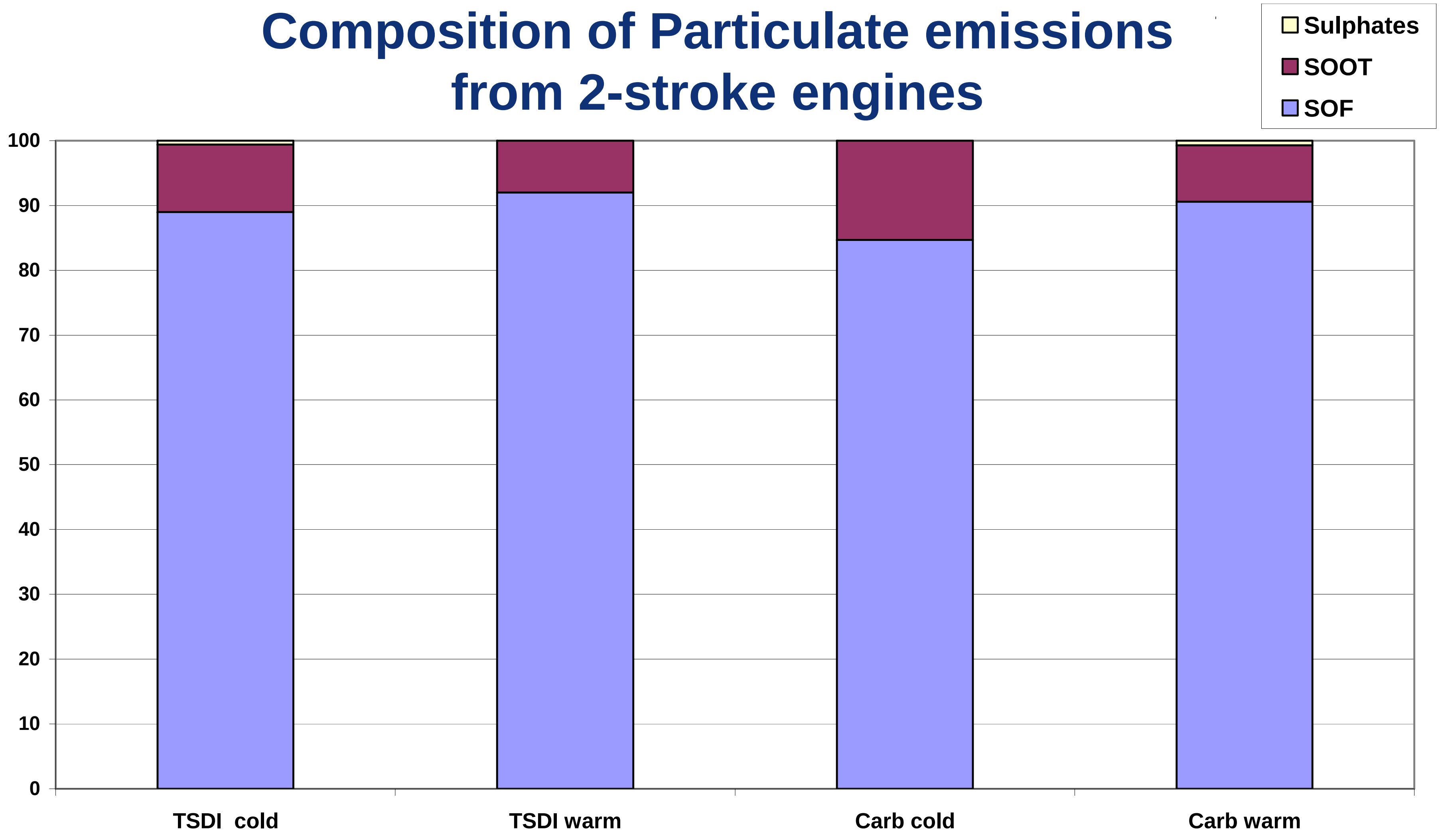
Euro 1 - DI

Euro 1 – Carburettor + OxyCat

Particulate Emissions from 2-stroke engines: Size distribution



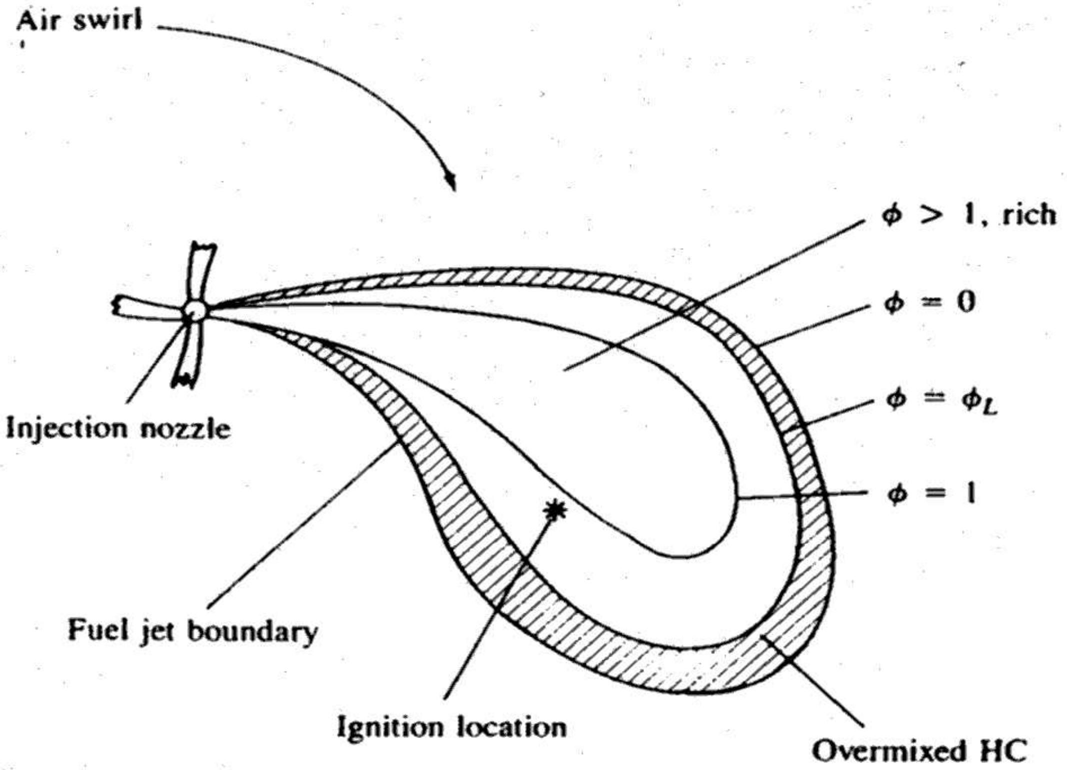
Composition of Particulate emissions from 2-stroke engines



Primary particulate emissions

- Emissions from Diesel engines

PM formation mechanisms



PM formation mechanisms

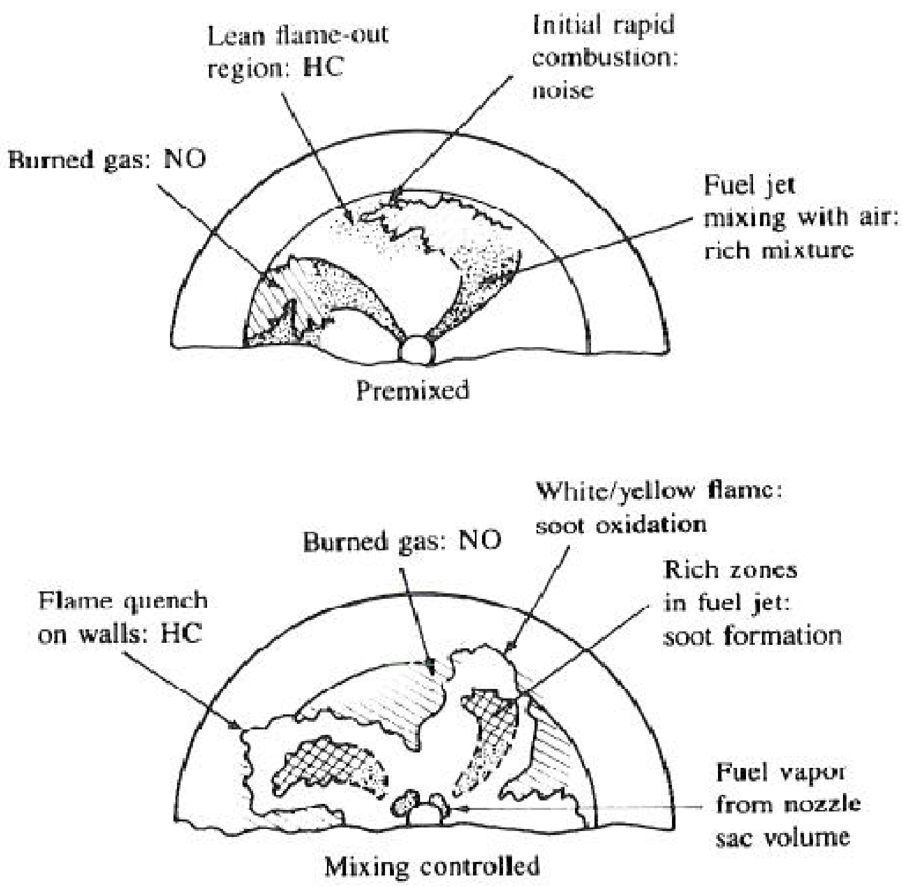


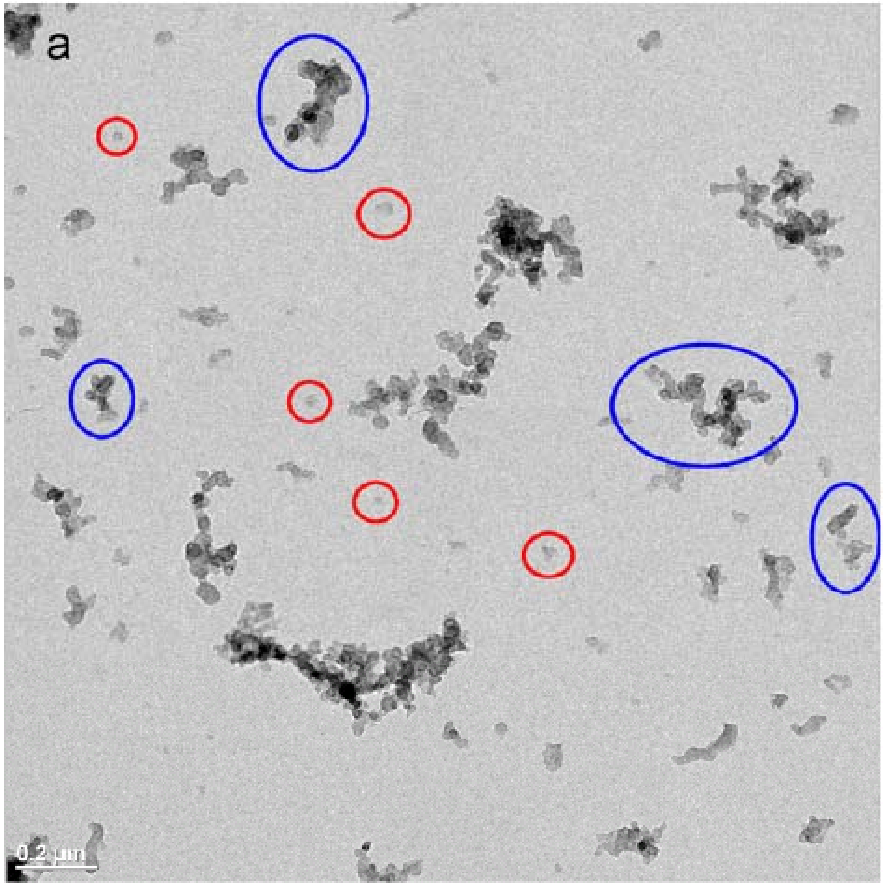
FIGURE 11-3
Summary of pollutant formation mechanisms in a direct-injection compression-ignition engine during "premixed" and "mixing-controlled" combustion phases.

What is Diesel Particulate Matter (DPM)?

Complex mixture of solid (mostly soot) material produced during combustion and volatile organic and inorganic material added as the exhaust gases cool down.

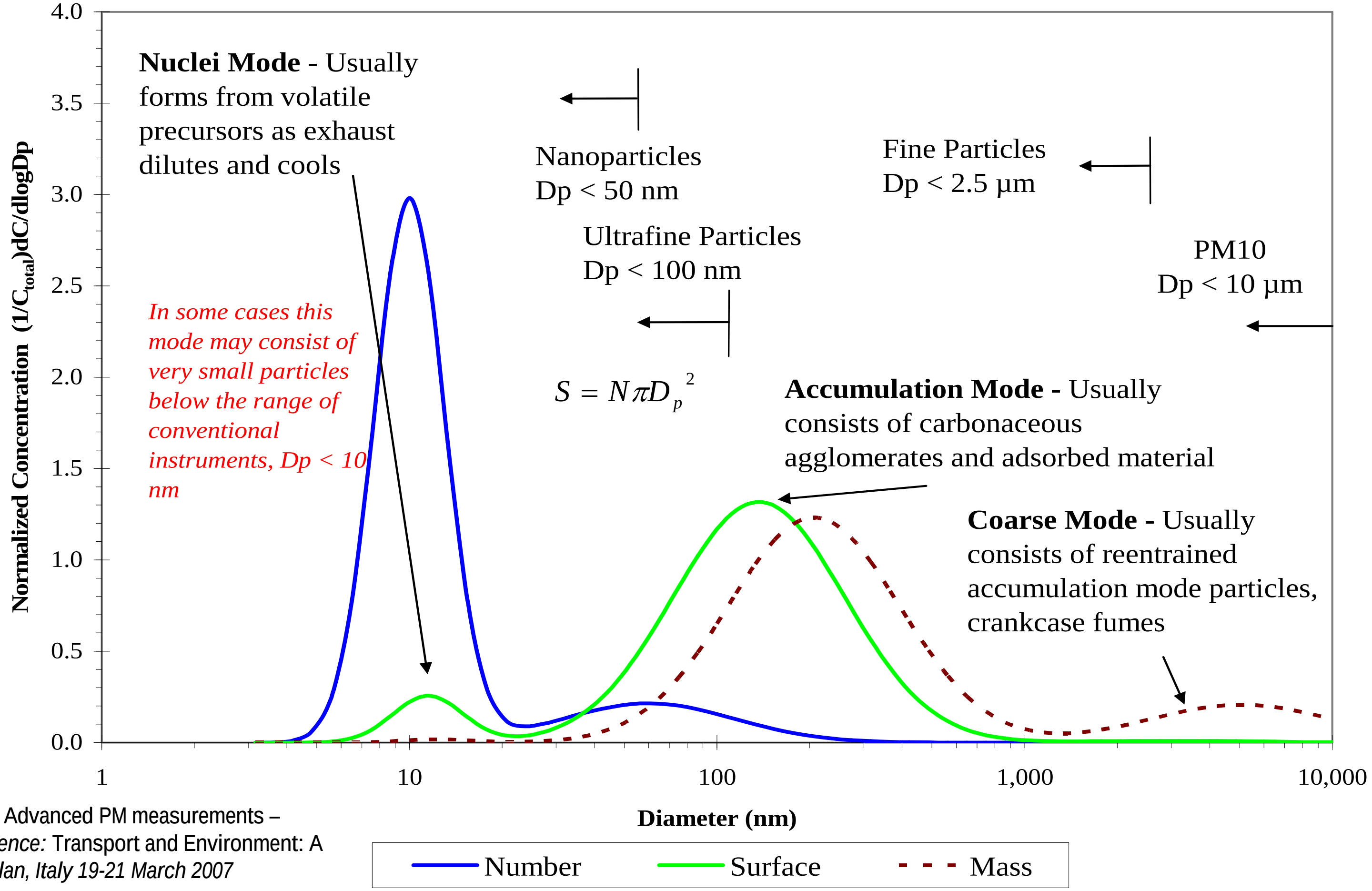
TEM images suggest the presence of highly agglomerated soot particles and volatile material internally and externally mixed.

Due to the complexity in DPM nature there does not exists a deterministic definition according to its chemical of physical properties.



Source: Kirchner et al. 2009, Journal of Aerosol Science, 40:55-64

Typical engine exhaust particle size distribution by mass, number and surface area



Source: Prof. Kittelson D.B. - Advanced PM measurements – *International Conference:* Transport and Environment: A global challenge - *Milan, Italy 19-21 March 2007*

Health effects of DPM

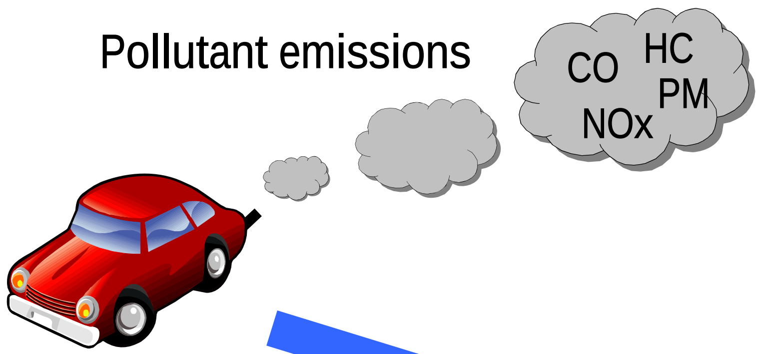
Several organizations have reviewed the health effects of DPM characterizing it as:

- Likely to be carcinogenic:
  - *National Institute for Occupational Safety and Health (1988). Carcinogenic Effects of Exposure to Diesel Exhaust.*
  - *International Agency for Research on Cancer (1989). Diesel and Gasoline Engine Exhausts and Some Nitroarenes. IARC Monographs of the Evaluation of Carcinogenic Risks to Humans.*
  - *U.S. Department of Health and Human Services. National Toxicology Program (2005). Report on Carcinogens, Eleventh Edition.*

- Carcinogenic:
  - *California EPA (1998). Part B: Health Risk Assessment for Diesel Exhaust. In: Proposed Identification of Diesel Exhaust as an Air Toxic Contaminant*

Emission Standards for Light Duty Vehicles

Pollutant emissions



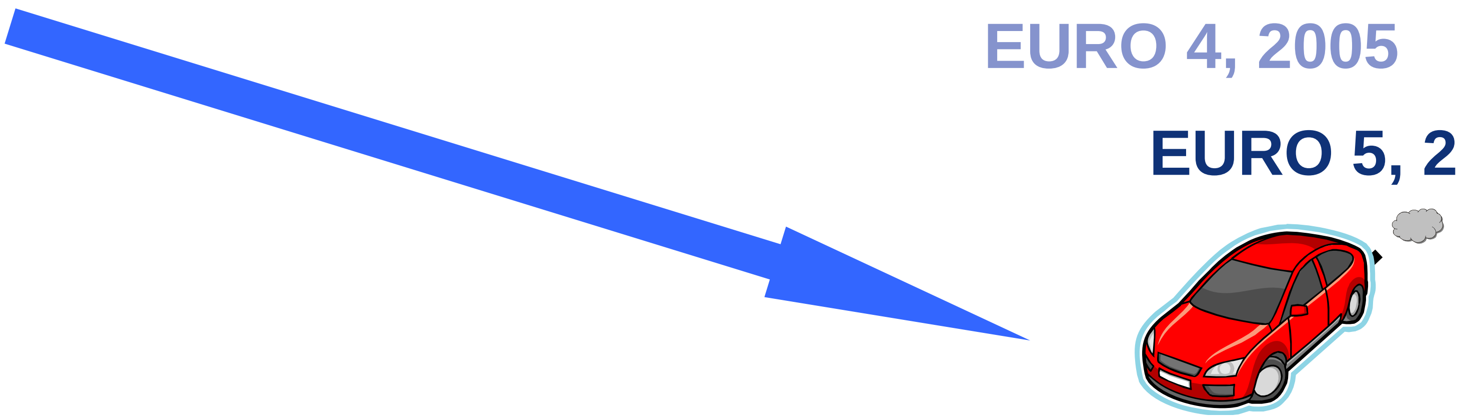
EURO 1, 1992

EURO 2, 1996

EURO 3, 2000

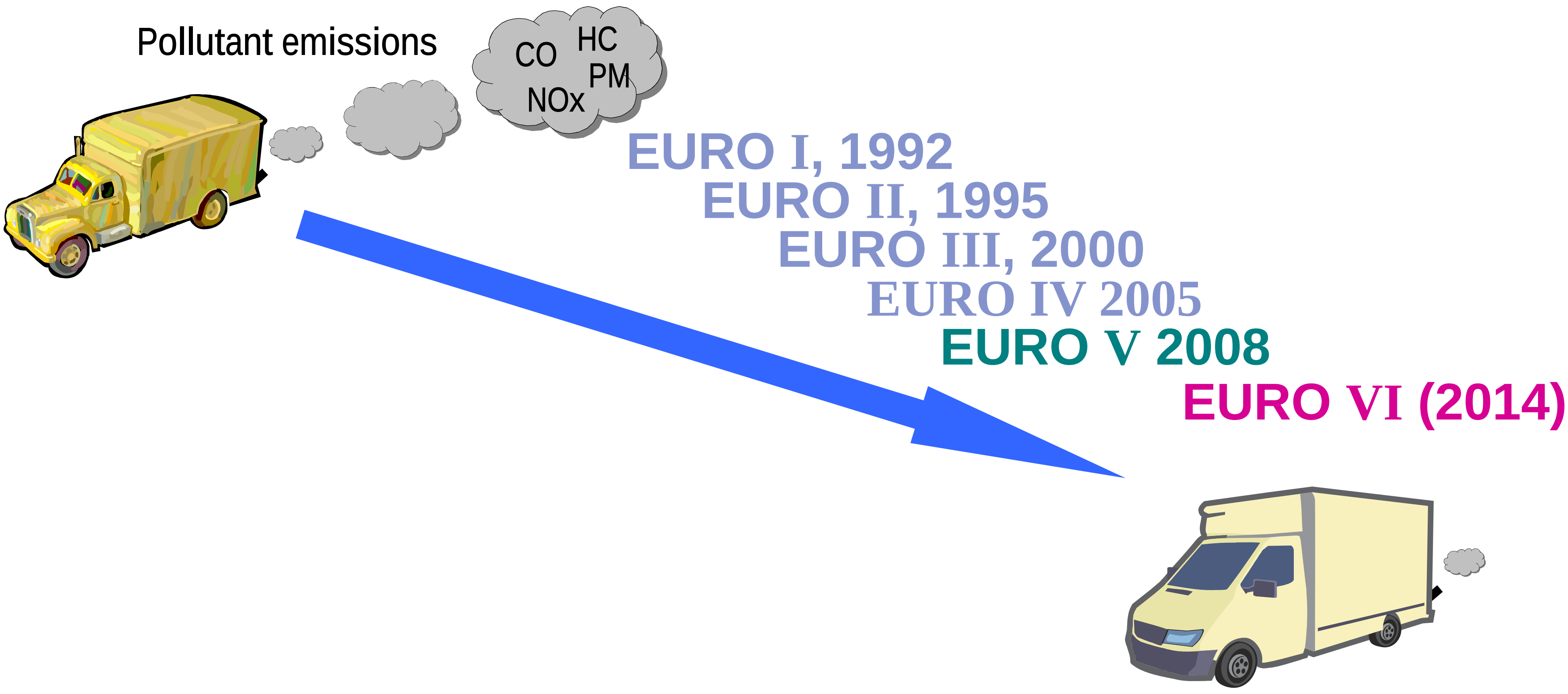
EURO 4, 2005

EURO 5, 2009



EURO 6 (2014)

Emission Standards for HD Engines

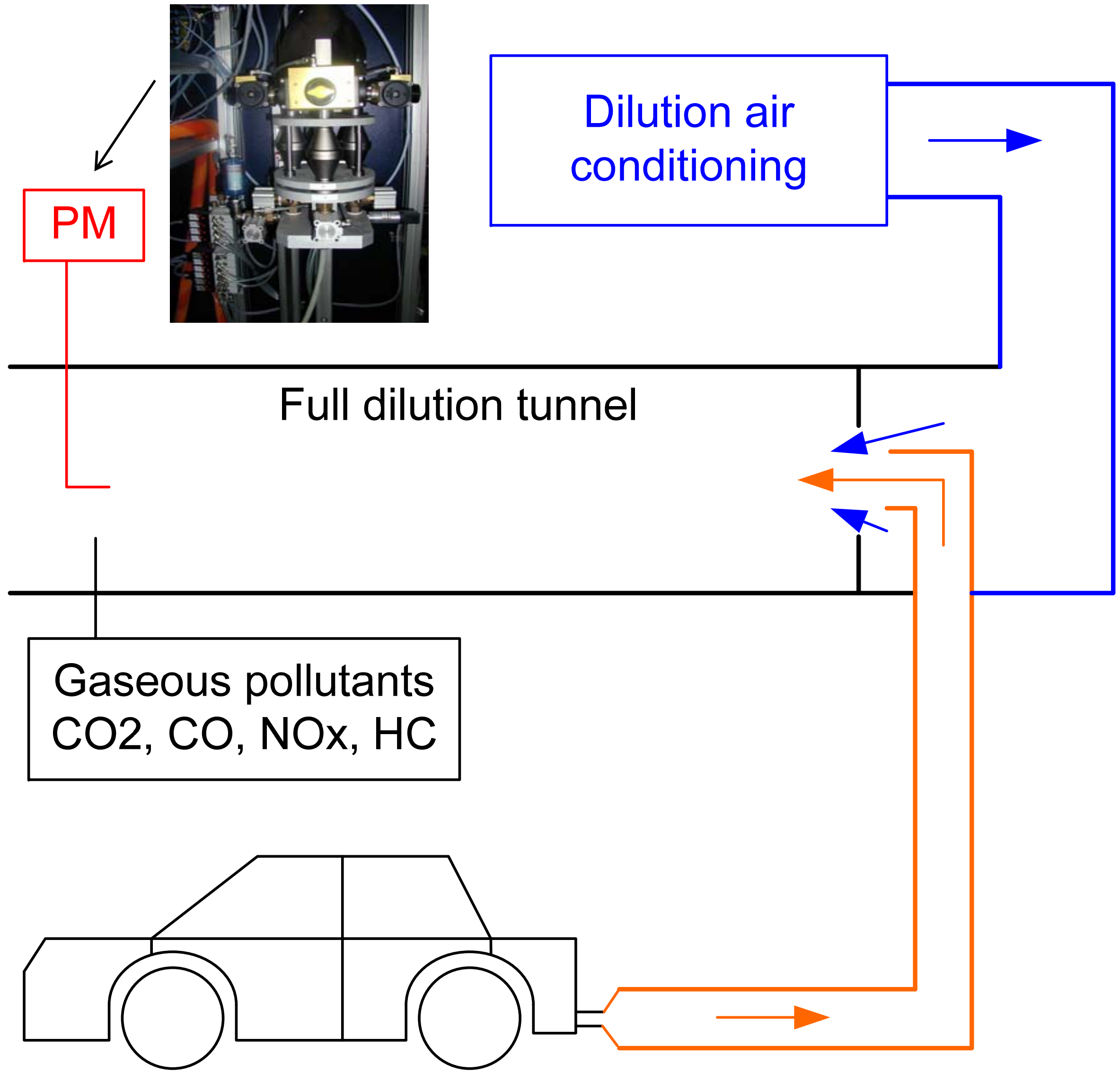


How was DPM measured?

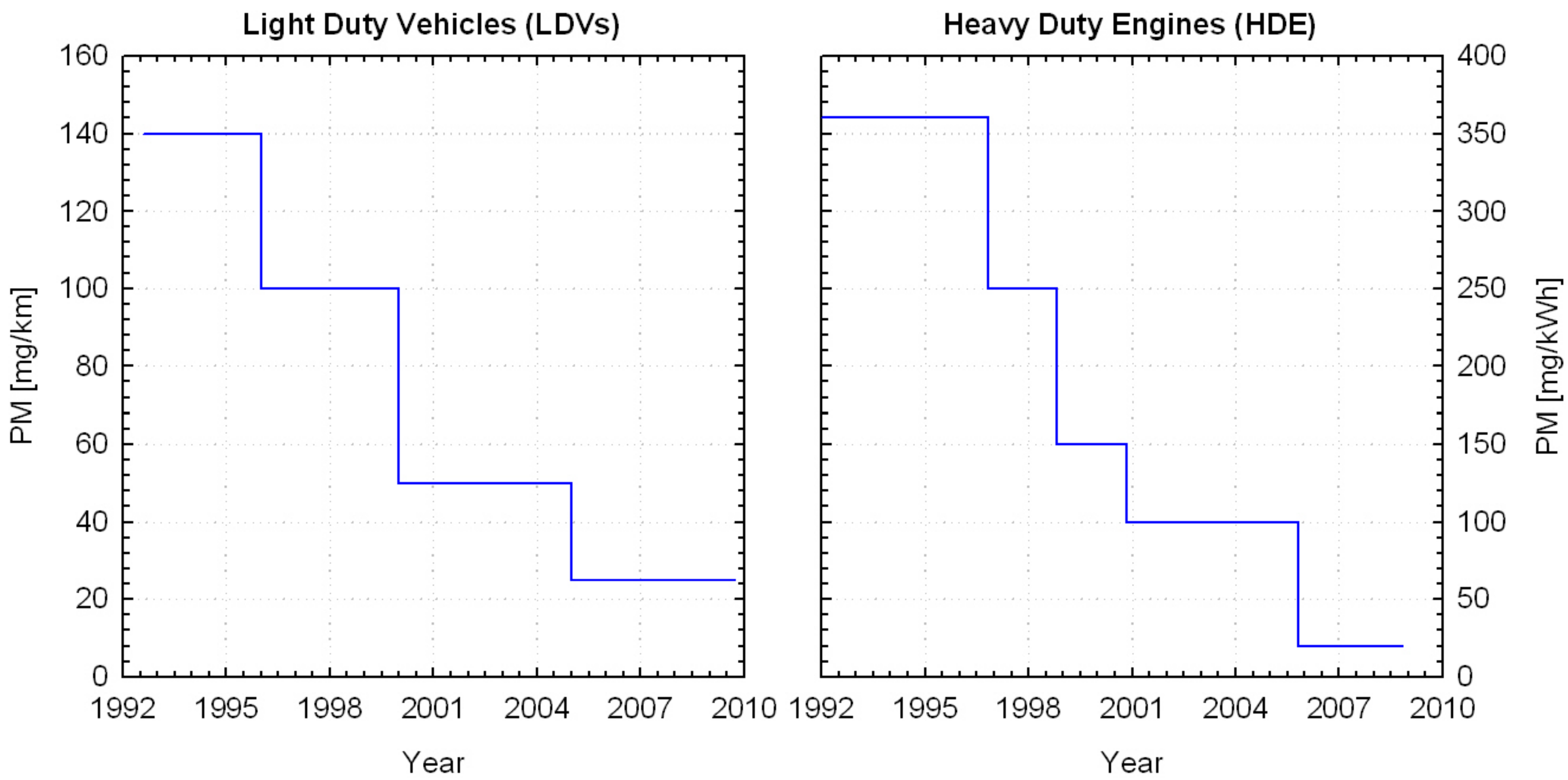
Traditionally, DPM was defined as anything collected on a filter after exhaust sampling and dilution at a T<52 °C.

Reasoning behind the Constant Volume Sampling (CVS) approach:

- Reduced risk of water condensation.
- Stabilization of the exhaust to reduce pressure and temperature fluctuations.
- Precise control of the constant flowrate established → Straightforward calculation of distance average emissions.



Legislation tackling DPM in Europe up to 2005



DPM emission limits at Euro 4 stage (applicable since 01/2005 for LDVs, 10/2005 for HDEs) at ~20% (LDVs) and 5% (HDEs) of the Euro 1 standards. Is this enough?

Clean Air For Europe (CAFE) Programme

Objective: "*Development of a thematic strategy on air pollution in order to attain levels for air quality that do not give raise to significant negative impacts on, and risks to human health and the environment.*"

Programme started in 2001 and finished in 2005 [COM(2005) 446 final], concluding that:

- "*Significant negative impacts will persist even with effective implementation of current legislation*":
  - *"5.5 months loss in statistical life in the EU in 2020 due to PM2.5, which is equivalent to 2.5 million life year lost or 272000 premature deaths."*
  - *"No safe level of exposure to PM exists."*

Further steps

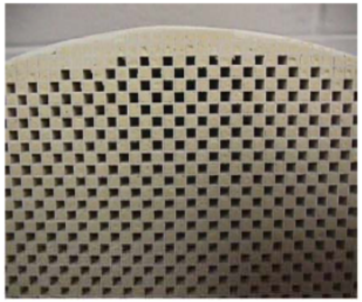
In order to achieve a significant improvement of air quality, a stricter limit of 5 mg/km (corresponding to a further 80% reduction) forcing de-facto the use of the best available technology to control PM emissions from light duty vehicles, was deemed necessary and proposed by the European Commission [Preliminary Draft Proposal for Euro 5].

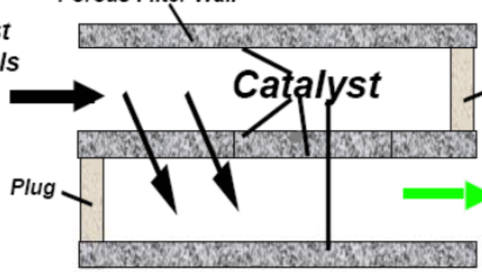
Most importantly, the proposal stated that:

- "*To prevent the possibility that in the future open filters are developed that meet the new particulate mass limit but enable a high number of ultra fine particles to pass, it is foreseen to introduce at a later stage a new standard limiting the number of particles that can be emitted.*"

- "*Once the results of the UN/ECE Particulate Measurement Programme (PMP) are available*":
  - *"a number standard will be implemented"*
  - *"a new PM measurement procedure will be implemented, and the proposed PM limit recalibrated"*

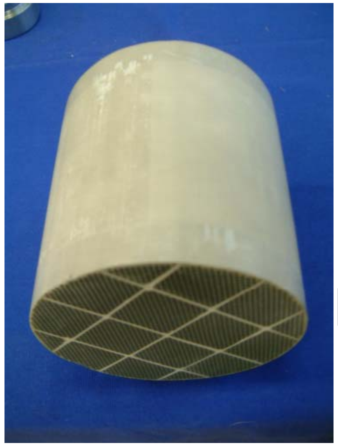
Catalysed - DPF





Efficiency of Diesel Particulate Filters

PM in



Diesel Particle Filter (DPF)

PM out

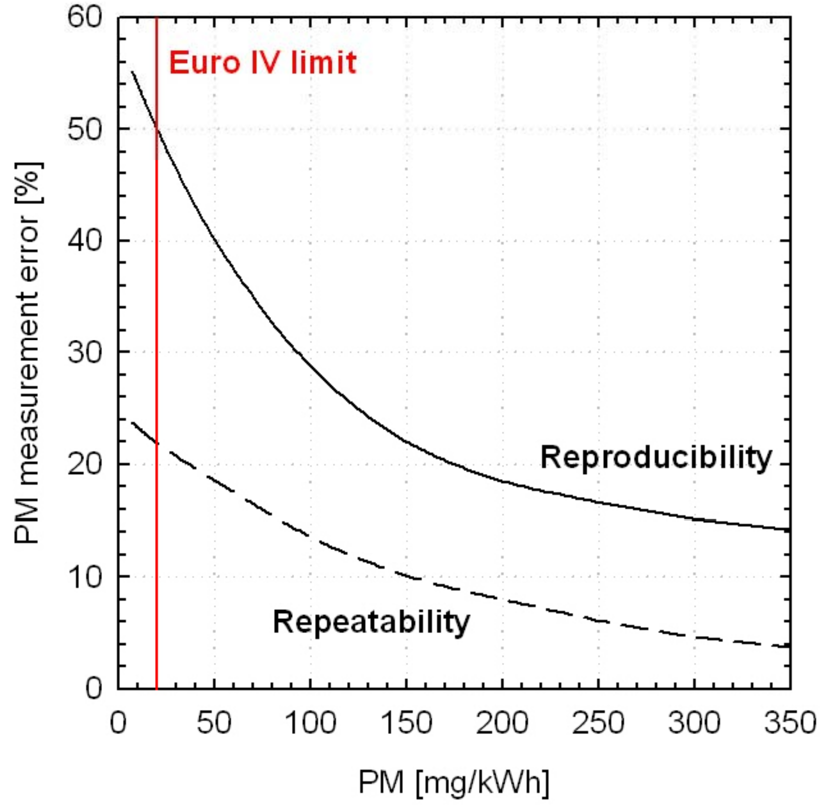
Tailpipe of a DPF equipped Euro 4 diesel vehicle after 60000 km

Limitations of gravimetric DPM procedure

Sources of variability:

- The means of transporting the exhaust to the CVS tunnel
  - Wall to particle interactions:
    - Inertial impaction
    - Thermophoresis
    - Diffusionphoresis
    - Reentrainment
  - Particle to particle interactions (coagulation/agglomeration - not affecting PM though)
  - Gas to particle interactions:
    - Condensation / evaporation
    - Adsorption / desorption
- Tunnel operating conditions:
  - Dilution ratio
  - Dilution air specifications (temperature, humidity, background concentration)
  - Collection temperature

Is the gravimetric procedure sensitive enough?

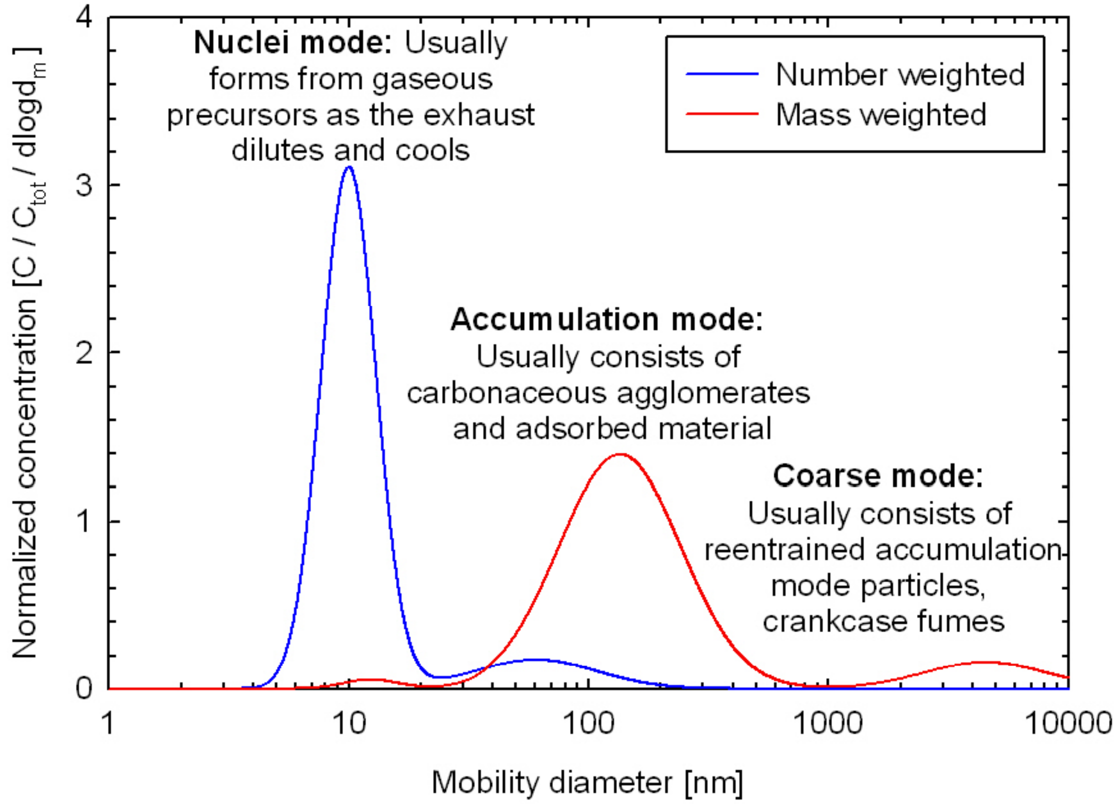


Source: ACEA report 99000524. Results based on experimental data collected in 6 inter-laboratory studies.

Typical size distributions of DPM

- Main challenge in particle number characterization is the sensitivity of the nucleation mode on the sampling conditions that are left uncontrolled in the CVS tunnel.

- The Diesel Particulate filters (DPFs) are very efficient in reducing the solid accumulation mode particles. High nucleation rates were reported downstream of a DPF. These were attributed to the absence of the solid core, on which gaseous precursors can condense, thus enhancing nucleation.



Source: Pr. D. Kittelson - Advanced PM measurements – International Conference: Transport and Environment: A global challenge - Milan, Italy 19-21 March 2007

PMP Working Group

Governments:

France, Germany, Greece, Japan, Korea, Sweden, Switzerland & UK

European Commission DG JRC

Industrial Association:

AECC, CONCAWE, OICA

Participating Laboratories:

AEA, EMPA, JRC, LAT, MTC, NMVERL, NTSEL, Ricardo, RWTUV, Shell, UTAC

PMP Mandate

- Development of new techniques to replace or complement particulate mass measurement for Light & Heavy Duty type approval
  - Simple and robust procedure suitable also for conformity of production testing
  - Good repeatability and reproducibility
  - Limited investments in terms of measuring equipment
- To provide data on the performance of different technologies, including DPF equipped vehicles, according to the new measurement procedures in order to set reasonable particulate emission limits

Investigation of:

Sampling & Conditioning Systems

- CVS (+ secondary dilution)
- CVS + thermodenuder or thermodiluter
- Rotary dilution
- Raw exhaust

Measurement Techniques

- Gravimetric (modified US 2007)
- Filter + chemical analysis
- TEOM
- Laser Induced Incandescence
- QCM
- Photoacoustic absorption
- Coulometric
- Photoelectric charging
- Light extinction
- Laser Light Scattering
- Differential Mobility Spectrometer
- Optical counter (CPC)
- Electrical Mobility
- ELPI
- Diffusion battery
- Diffusion charger

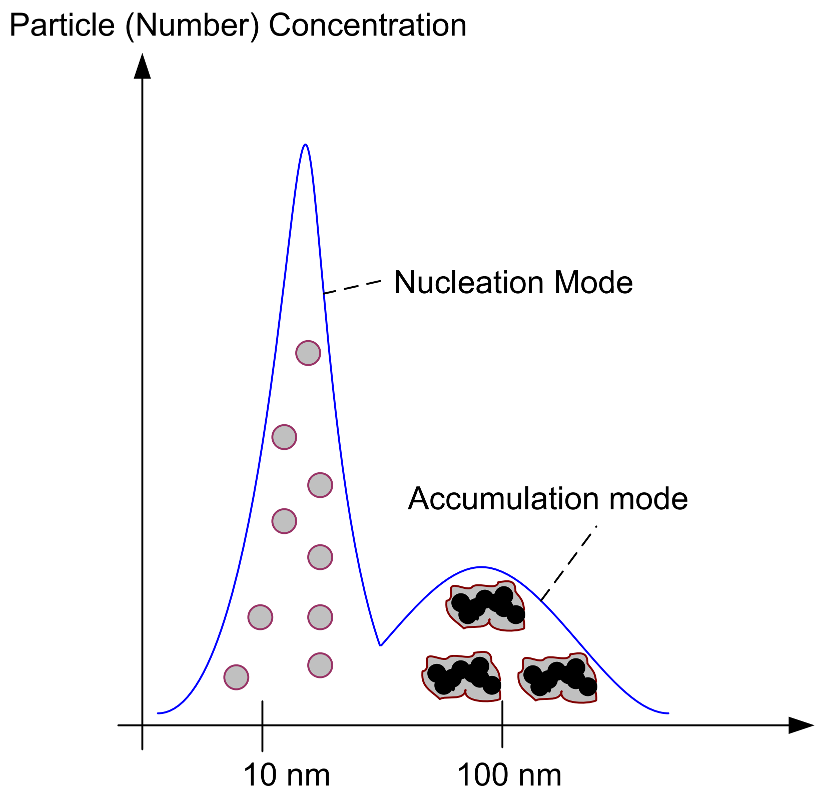
PMP Phase I & II: Conclusions

1. Improved Particulate Mass Measurement
   - Dilute exhaust sampling with HEPA & HC filtered dilution air
   - Cyclone pre-classifier
   - Improved sample temperature control 47 ±5°C
   - Deletion of back-up filter

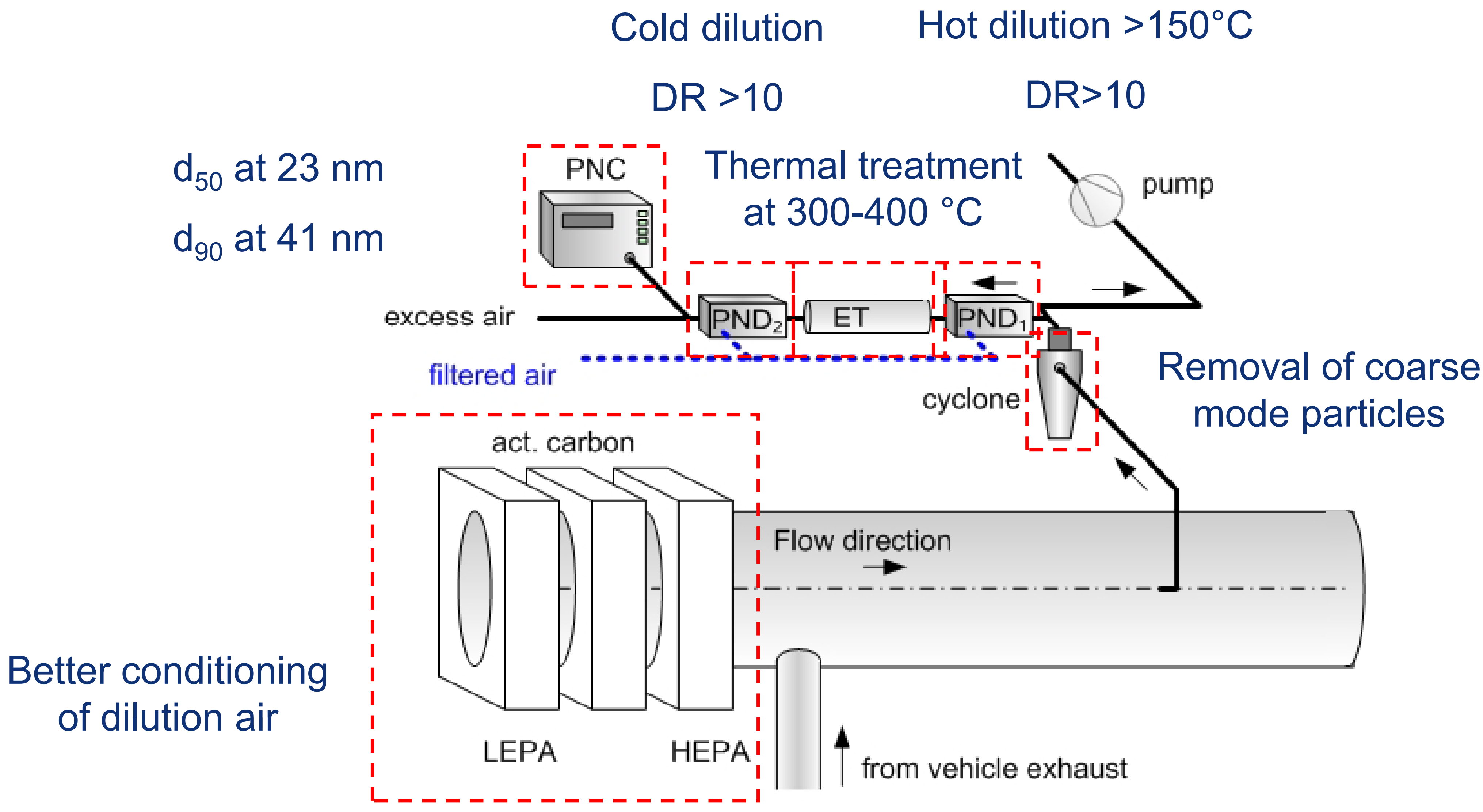
2. Solid Particle Number Count
   - Dilute exhaust sampling with HEPA & HC filtered dilution air
   - Cyclone Pre-classifier
   - Sample thermal conditioning: heated dilution, evaporation tube, dilution
   - Condensation Particle Counter 23nm (50%) cut point

PMP procedure focuses on solid particles with a diameter >23 nm

- **Accumulation mode very stable and not depending on sampling conditions**
- **Nuclei mode too sensitive to sampling conditions – too high variability**



PMP particle number measurement system



PMP Phase III - Validation

Objectives

- To assess repeatability and lab to lab reproducibility of proposed PM & PN techniques
- To assess comparability of available PN measurement systems
- To assess performance levels of different engine/vehicle technologies

Separate Light & Heavy Duty Inter-Laboratory Correlation Exercises (ILCE)

PMP Light Duty ILCE

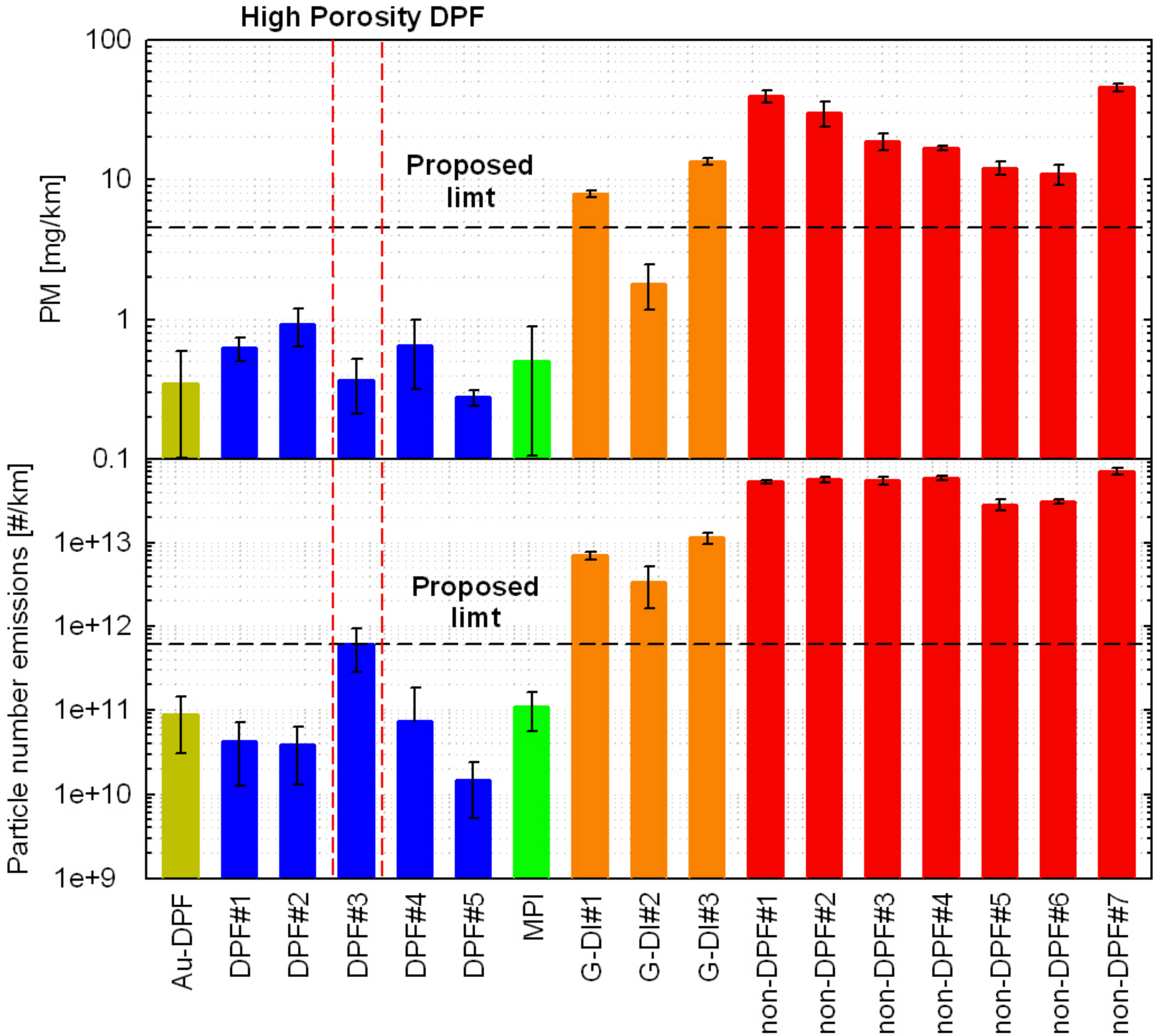
- JRC was asked by PMP Chair to manage Phase III, in order to guarantee impartiality to participants and industry
- Programme run mid 2004- mid 2007 with participation of 10 international labs (Japan, Korea, US, UK, Germany, France, Sweden, Greece, JRC)
- 'Golden Vehicle' (low emissions vehicle with particle filter DPF) + 'Golden' particle number measurement equipment
- 'Golden Engineer' + two of JRC staff to ensure best and reproducible testing practice
- Golden vehicle tested at least 5 times in each lab with the golden and alternative instrumentation
- 15 additional vehicles were tested by the different labs, using the golden instrumentation:
  - 5× DPF diesels
  - 6× Conventional diesels
  - 3× Direct injection gasolines
  - 1× Port fuel injection gasoline

Light Duty ILCE – PM/PN emission levels

Both PM and PN methodologies capable of quantifying the different emission performance of DPF diesels, non-DPF diesels and G-DIs

Only PN method sensitive enough to identify the diesel vehicle equipped with a high porosity (low filtration efficiency) DPF

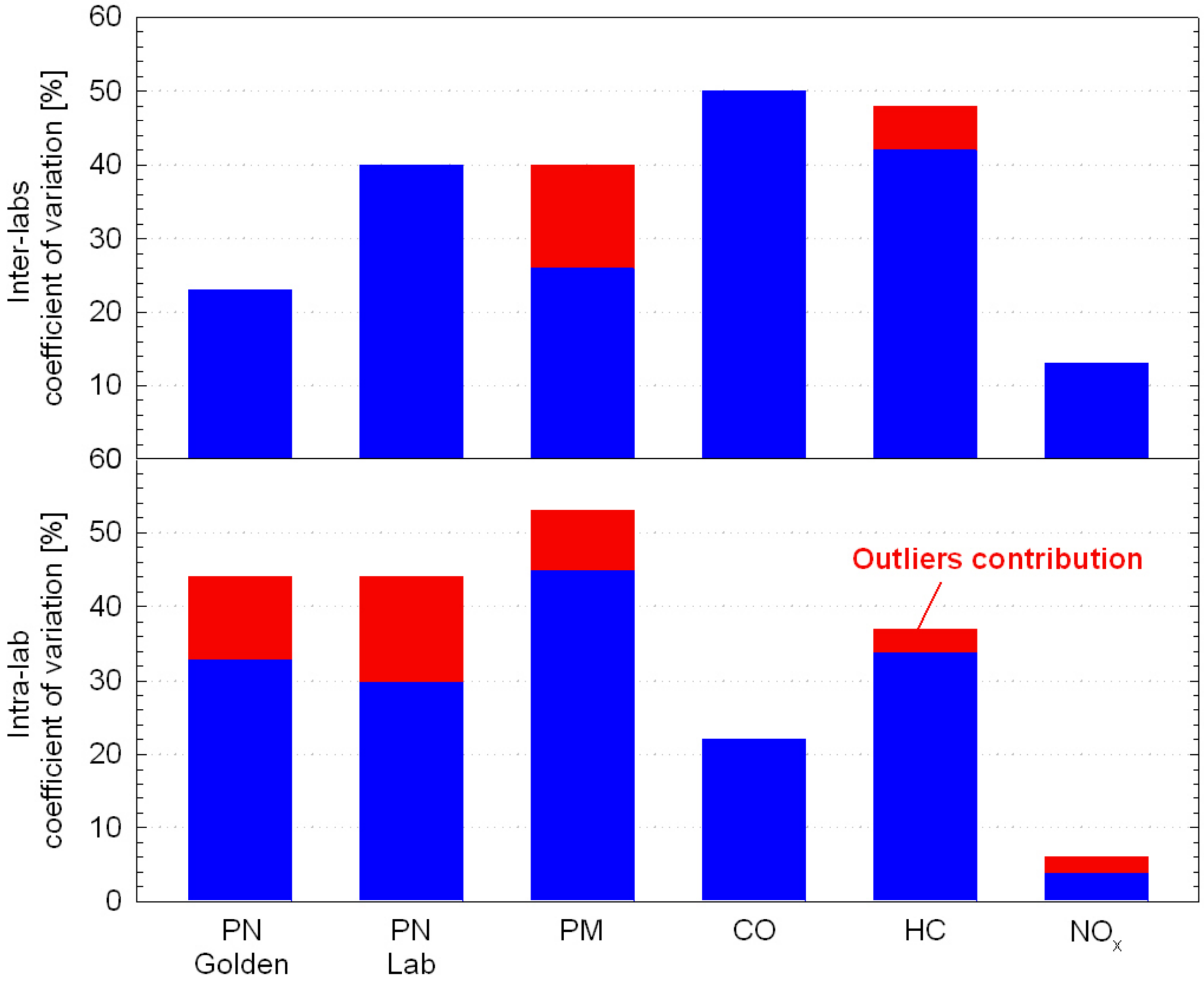
Particulate emissions from G-DI vehicles become an emerging issue



Light Duty ILCE – Repeatability/Reproducibility

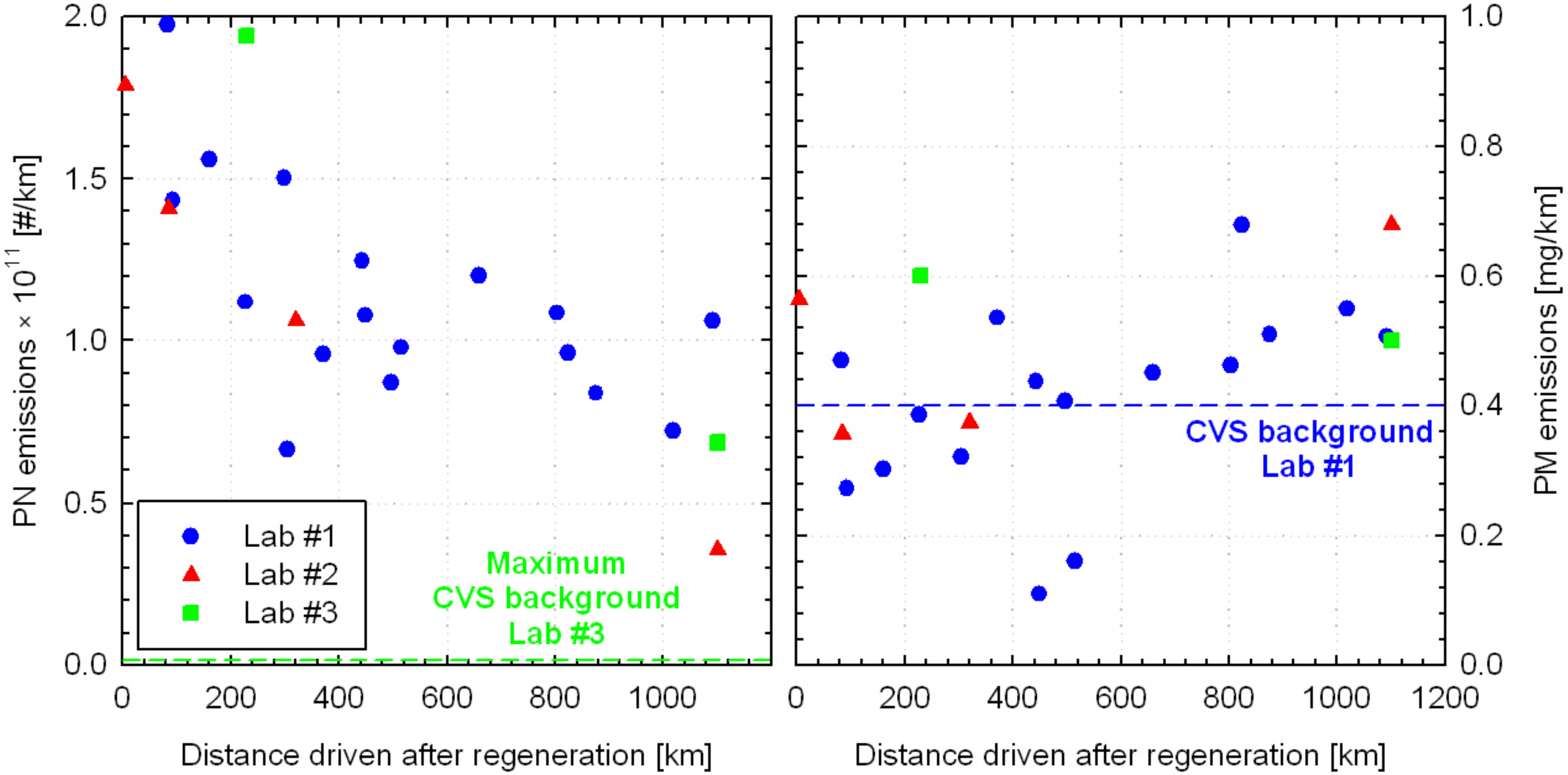
Repeatability and reproducibility of the PMP methodology were quantified following the ASTM E691-99 standard.

The proposed PN method exhibited similar intra- and inter-lab variability with PM, CO and HC.



Reference: Giechaskiel et al. (2008) Aerosol Science and Technology, 42:528-543

Light Duty ILCE – Method Sensitivity



PN method was sensitive enough to identify the improved filtration efficiency as soot cake accumulates on the DPF.

PM emissions were found to be at the background levels

Heavy Duty ILCE

Both coordinated by the JRC

PMP HD Validation Exercise

- Golden Engine-1
- Golden Systems
- Golden Engineer
- Same fuel and lubricant
- Both full flow system and Partial Flow Dilution System (PFDS)
- Participants: JRC, UTAC, AVL MTC, EMPA, Ricardo

Concluded – Final Report published: ISSN 1018-5593

PMP HD Round Robin exercise

- Golden Engine-2
- Instrument chosen by each lab
- No Golden Engineer
- Standard ref. fuel/lubricant
- Both full flow system and partial flow system
- 10 laboratories from EU, Japan Korea, Canada

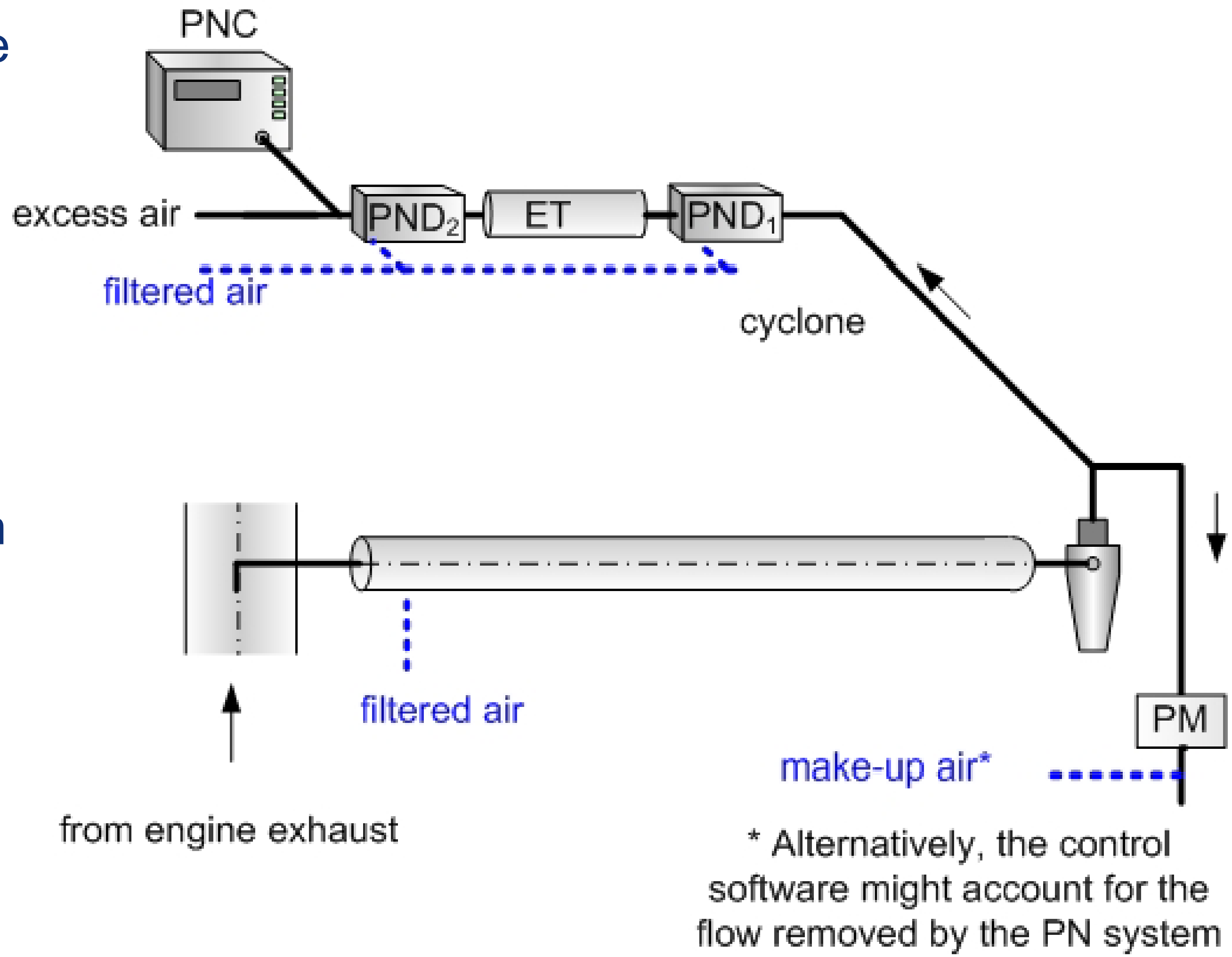
Underway – Testing expected to finish this autumn

PMP PN system in PFDS

In PFDS, a small portion of the exhaust is sampled and used for the determination of the PM emission rate. PFDSs overcome the cost and size problems of the CVS.

Initially only allowed over steady state testing (European Stationary Cycle - ESC).

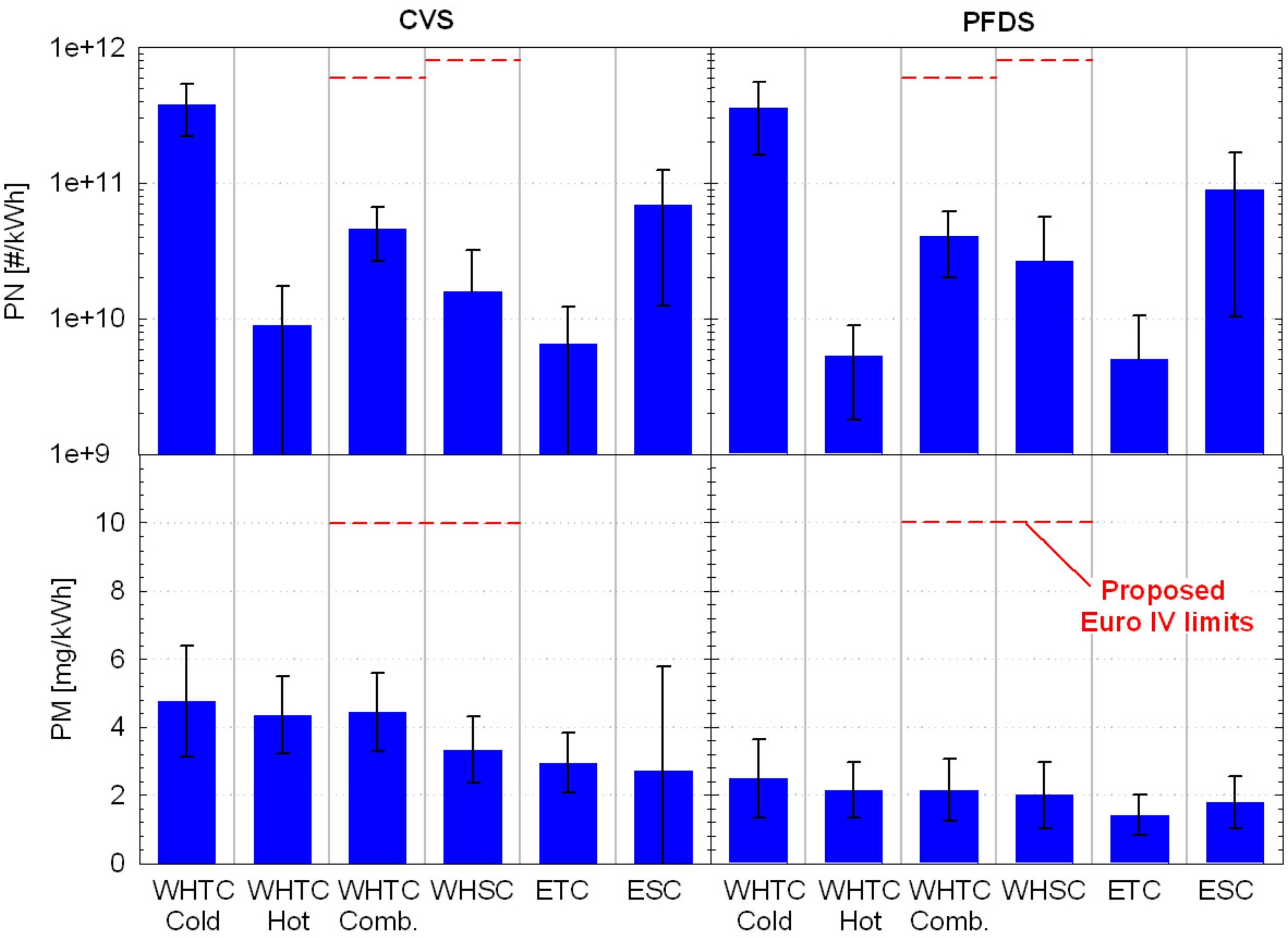
Directive 2005/55/EC introduced provisions for PFDS (compliant with the ISO 16183:2002) to be employed over transient test cycles also.



HD Validation Exercise - Results

Cold start operation resulted in 1½ to 2 orders of magnitude increase in PN. The PM methodology was not sensitive enough to detect this.

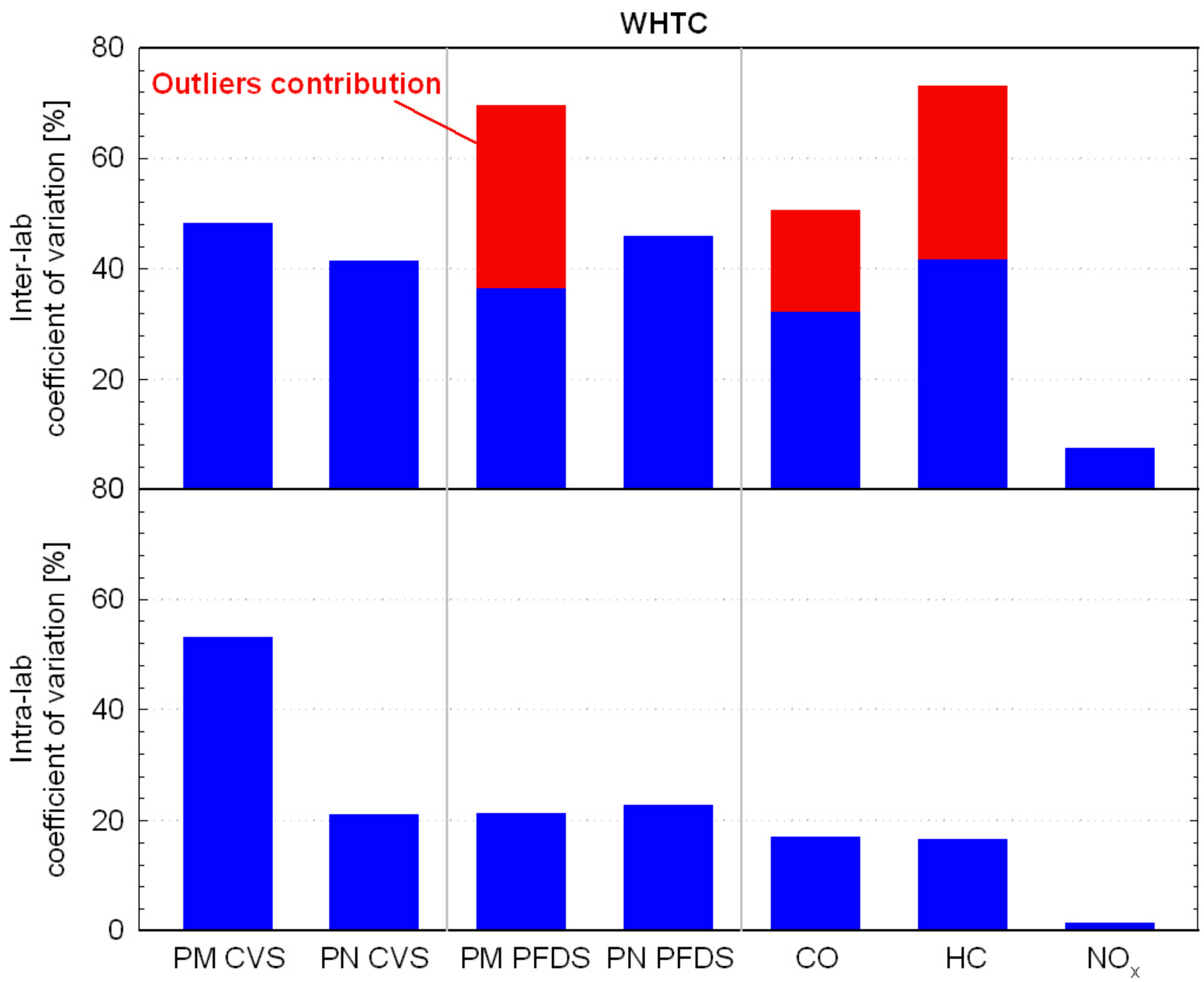
PFDS exhibited similar performance with CVS both in terms of PM and PN



HD Validation Exercise – Repeatability/Reproducibility

The proposed PN method exhibited similar intra- and inter-lab variability with PM, CO and HC.

The accuracy of PFDS was similar to that of CVS



Introduction of PN limits in the European legislation

- Light Duty Vehicles:
  - Regulation (EC) No 715/200 (20/06/2007) empowered the Commission to introduce a number based limit the latest upon entry into force of the Euro 6 stage, which will also be applicable to gasoline-powered vehicles.
  - Regulation (EC) No 692/2008 (18/07/2008) introduced a PN limit of 6×10^{11} #/km applicable to diesel vehicles from 09/2011 (Euro 5b). The same regulation requires that a PN limit for Euro 6 certified gasoline vehicles will also be introduced before 09/2014.

- Heavy Duty Engines:
  - Regulation (EC) No 595/200 (18/06/2009) empowered the Commission to introduce a number based limit applicable to all Euro VI certified HDEs.
  - A draft version of the regulation implementing and amending Regulation (EC) No 595/200 is currently scrutinized by the Parliament and the Council and will soon enter into force. The proposed limits are 6×10^{11} #/kWh over WHTC and 8×10^{11} #/kWh over WHSC.

Primary particulate emissions

- Emissions from petrol engines

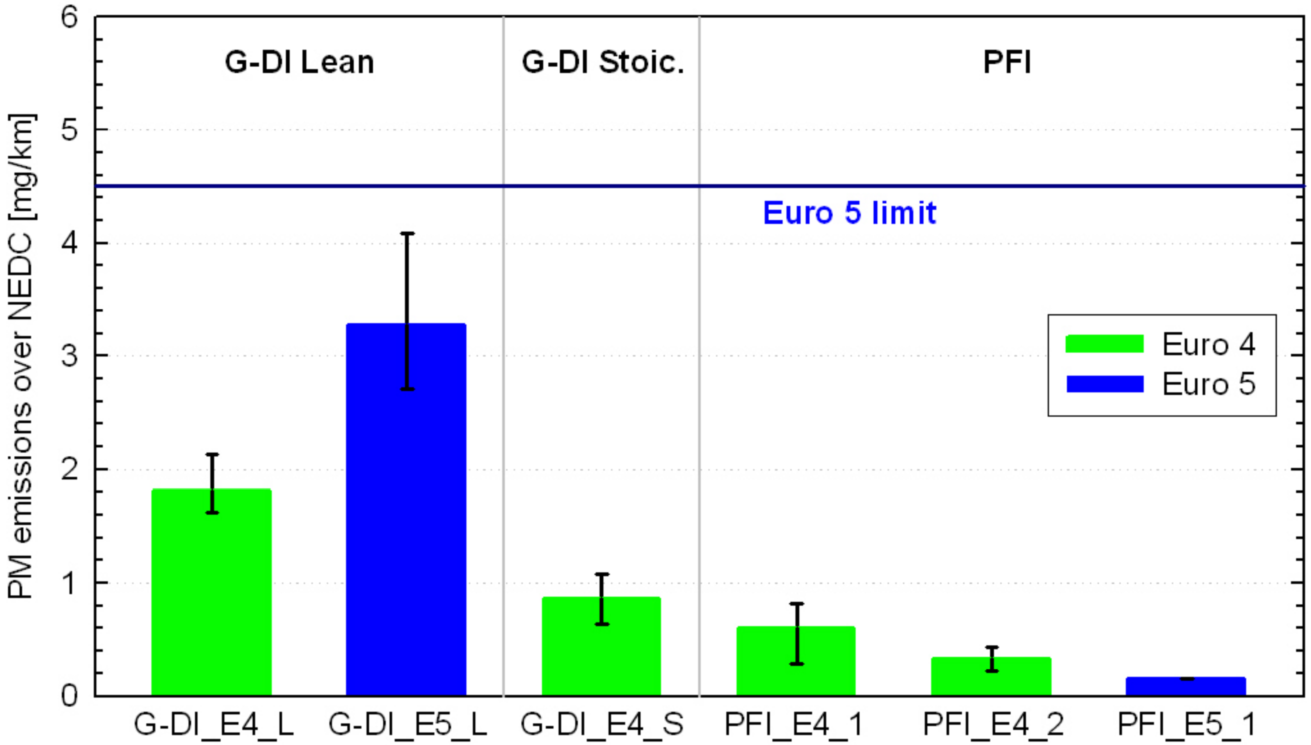
Gasoline particle emissions regulation
Next steps

On December 2010 a first report was submitted addressing the particle emissions of current technology vehicles, based on experimental data collected at JRC and information found in the literature: http://circa.europa.eu/Public/irc/enterprise/automotive/library?l=/mveg_vehicle_emissions/107th_january_2011

By this September a cost benefit analysis study will be prepared addressing the cost effectiveness of introducing Gasoline Particulate Filters (GPF) on Gasoline Direct Injection (G-DI) vehicles.

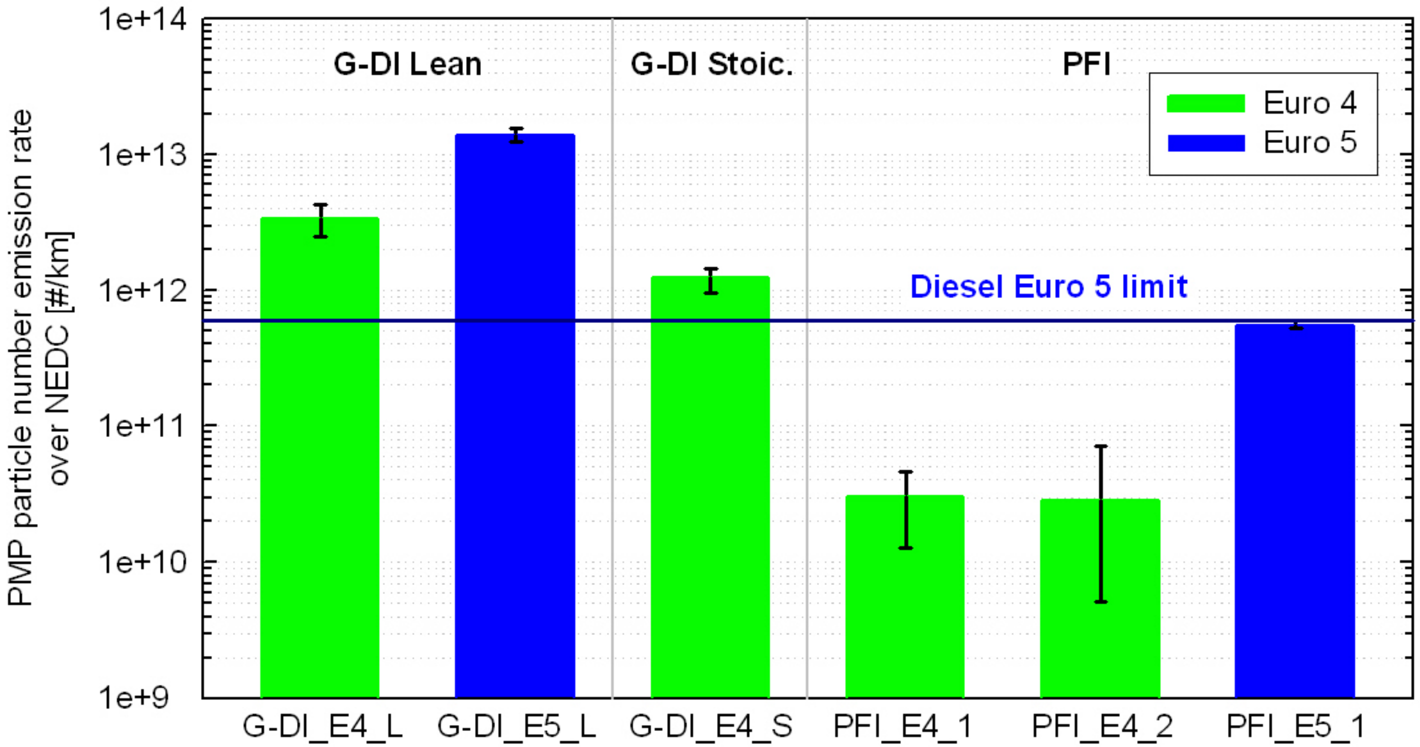
By this December a third report will be submitted addressing the emission performance of late technology gasoline vehicles.

PM emissions of gasoline vehicles



All gasoline vehicles emit well below the PM limit which is already applicable to Euro 5 G-DI vehicles.

Particle number emissions of gasoline vehicles



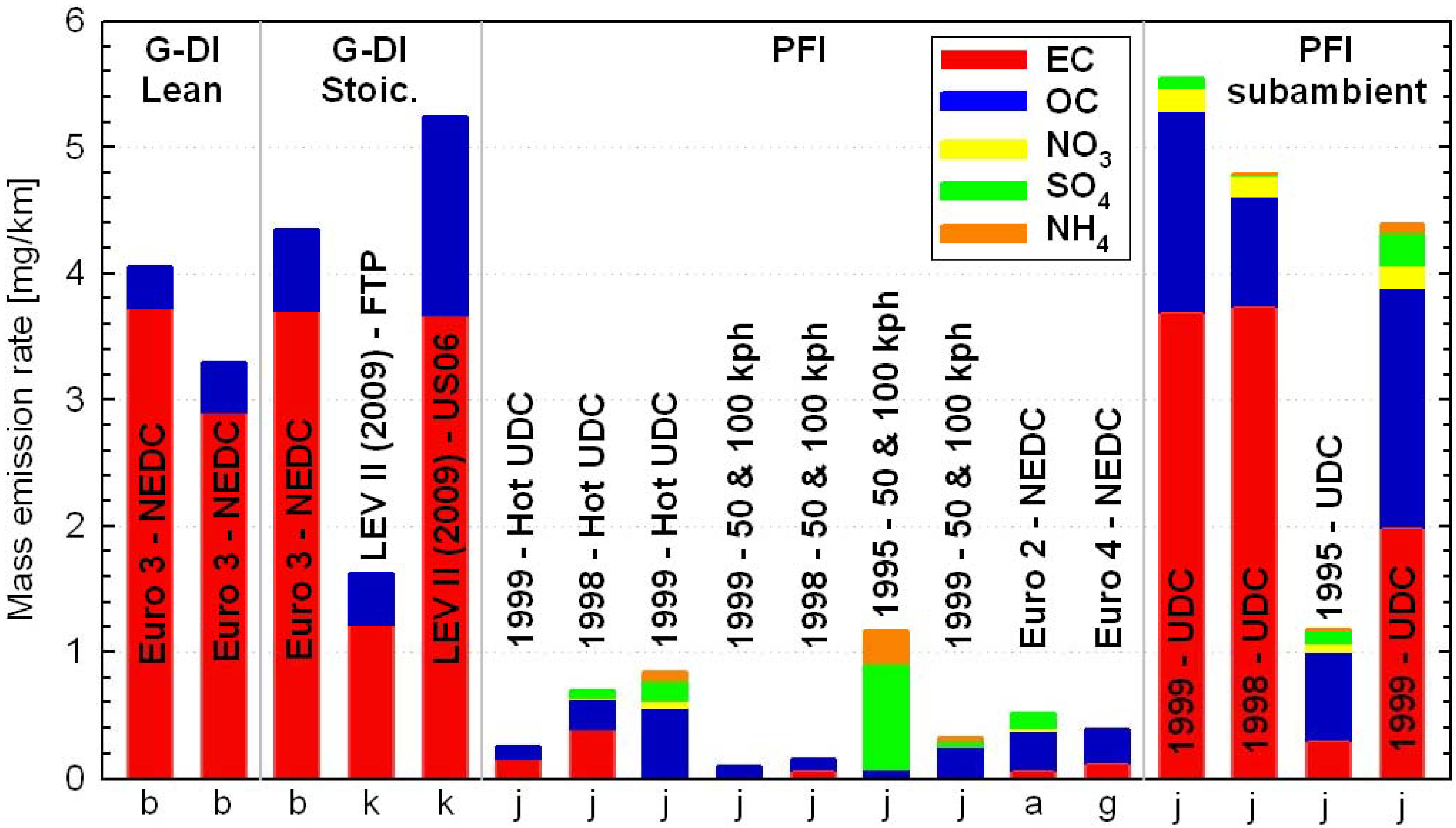
Gasoline Direct Injection (G-DI) vehicles were found to systematically exceed the diesel emission limit.

Conventional Port Fuel Injection (PFI) vehicles emit well below this threshold.

JRC
EUROPEAN COMMISSION

Composition of particulate emissions from gasoline vehicles

Institute for Energy

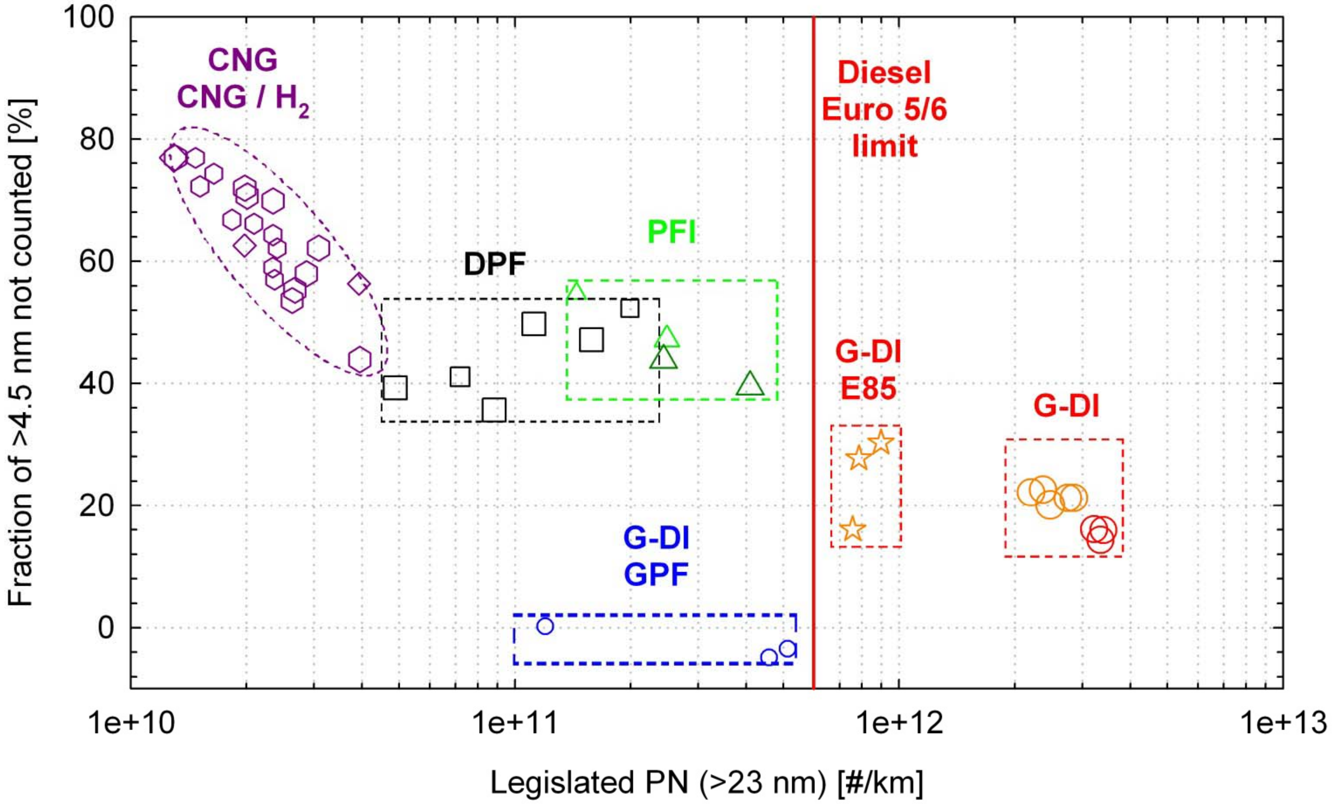


A: Andersson et al. 2001, b: Mohr et al. 2003 ,g: Bosteels et al. 2006, j: Schauer et al. 2008 , k: Khalek et al. 2010

Primary particulate emissions

- Latest investigations

Fraction of sub-23 nm particles not counted



The fraction of “non-volatile” particles >4.5 nm in the exhaust of gasoline vehicles not counted by the PMP CPC is similar or even less from that in the exhaust of the Euro 5 diesel tested.

Gasoline Particulate Filters were proven to be very effective in controlling sub-23 nm particles as well.

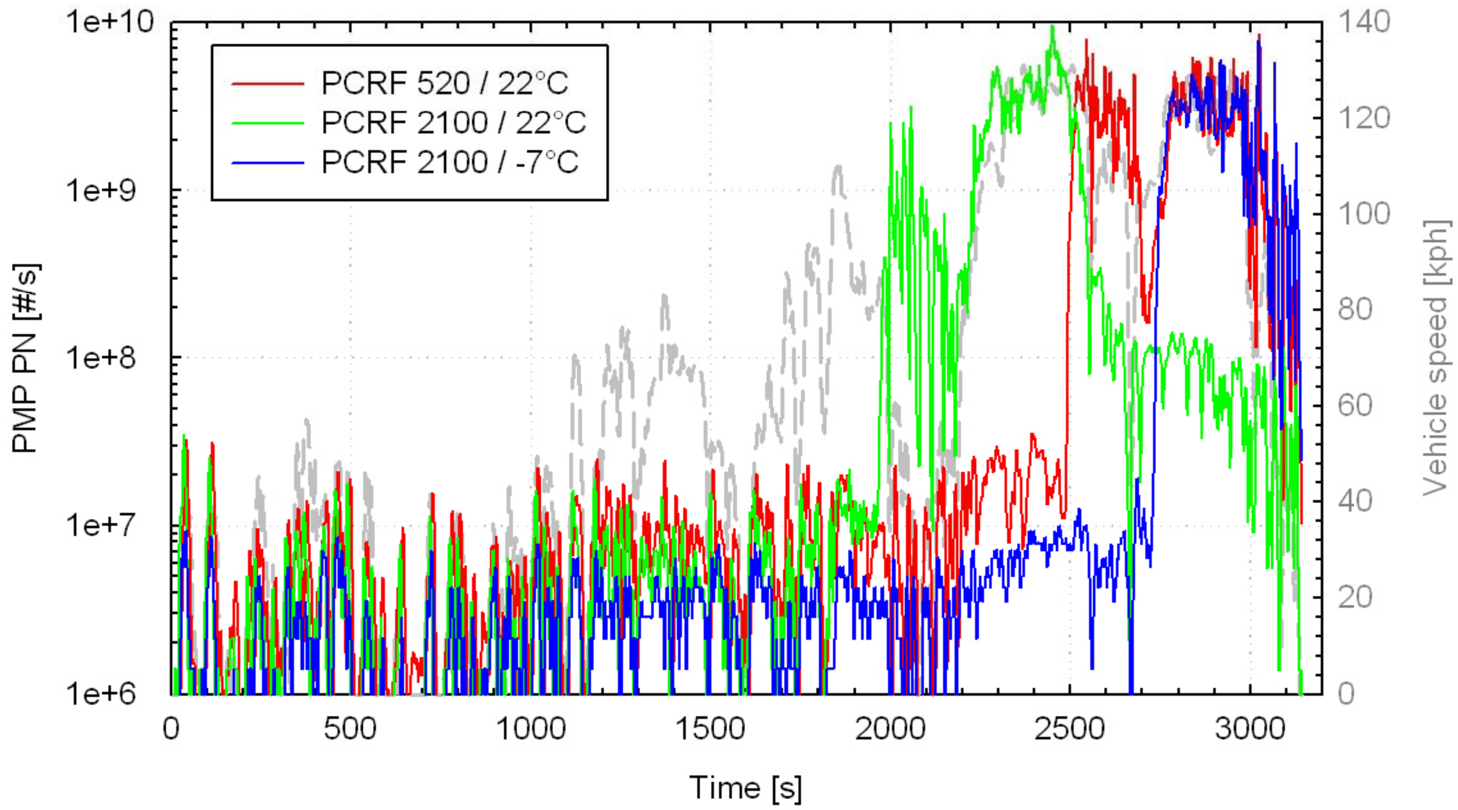
WLTP PM/PN Regeneration

- **In the frame of the WLTP work the need / feasibility / possibility to introduce PN emission measurements during periodic regeneration of DPFs is examined.**

- **Objective:**
  - Understand whether increased "non-volatile" PN emissions occur during regeneration in late technology diesel vehicles.
  - Quantify their contribution over the useful life of the vehicle.
  - Investigate a potential interference from volatile material emitted in high concentrations during regeneration events.

- **Experimental investigation during the validation phase of WLTP test procedures:**
  - A dedicated experimental protocol will be developed (currently under discussion)
  - Starting from October 2011 several vehicles will be tested at different laboratories

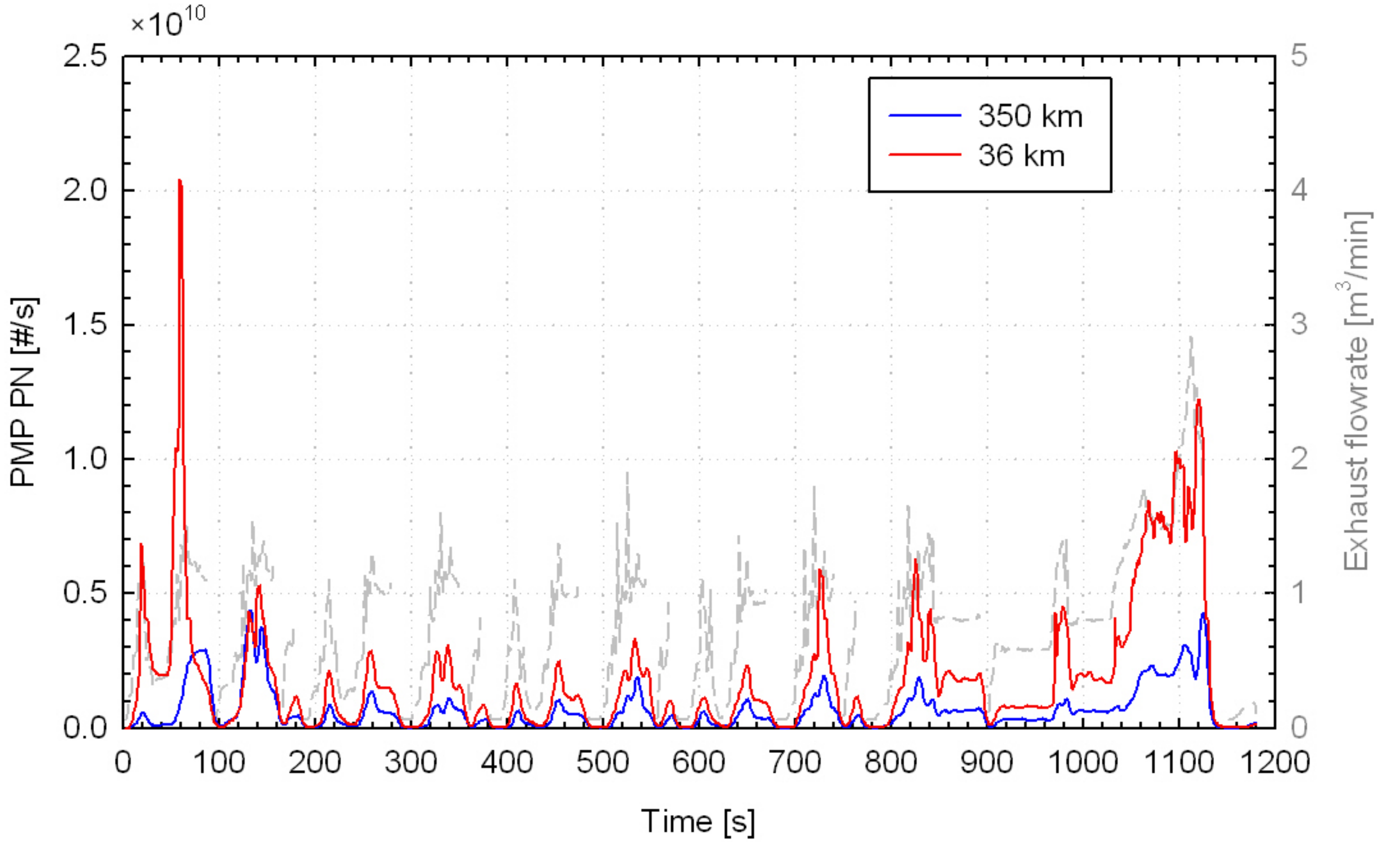
Particle emissions during DPF regeneration

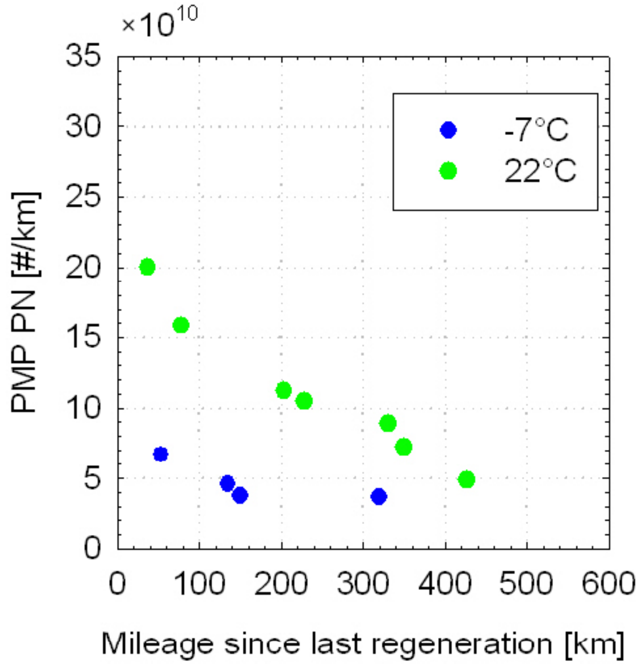


Experiments conducted under driving cycles representative of real world driving conditions (Common Artemis Driving Cycles), suggested two orders of magnitude increase in the particle emissions during active regeneration of the DPF.

For the particular vehicle tested, the active regeneration of the DPF occurred every 6 repetitions of the CADC resulting in a 15-fold increase in the cycle average emissions.

Particle emissions from late technology diesels





Particle number emissions are found to strongly depend on the fill status of the DPF.

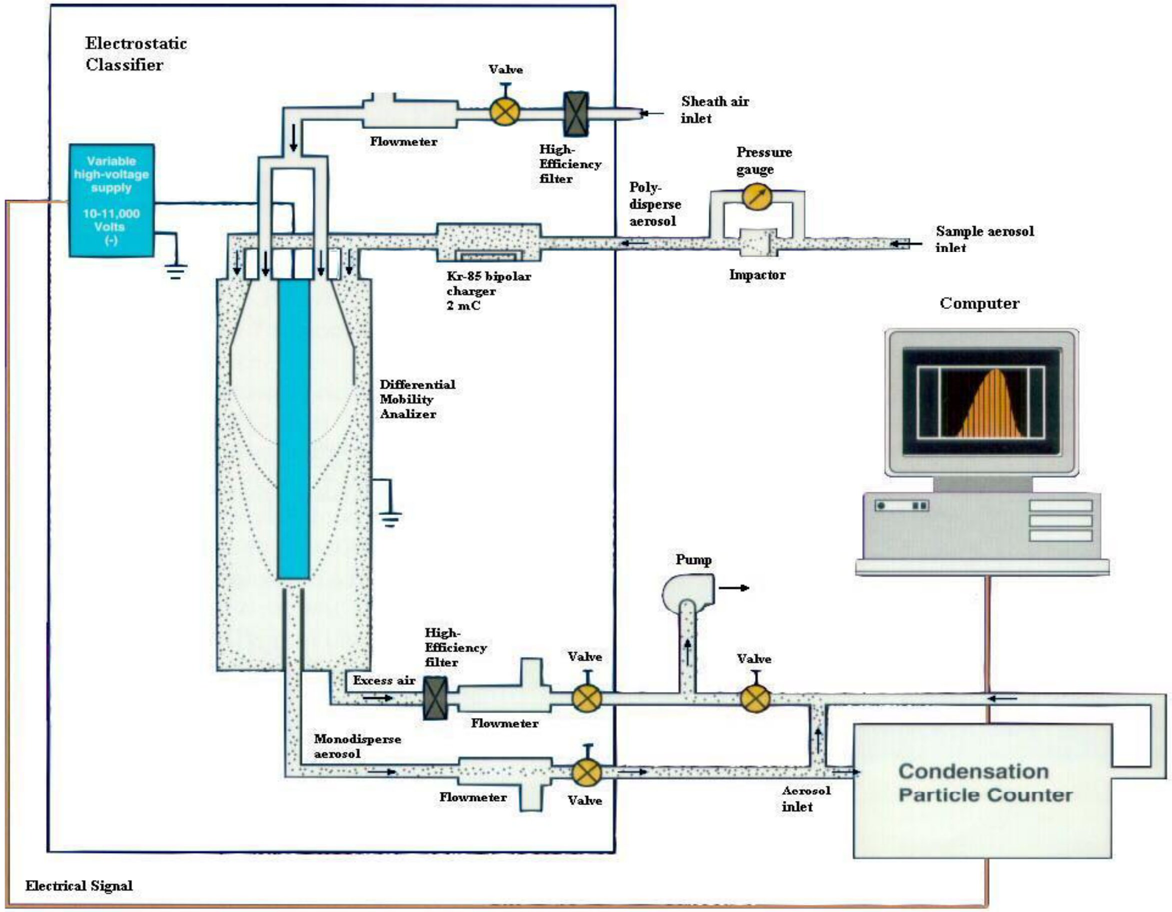
At -7°C ambient temperature, the EGR rate is reduced to avoid condensation of water and HC in the EGR line. This results in higher NOx and lower PN emissions.

Conclusions

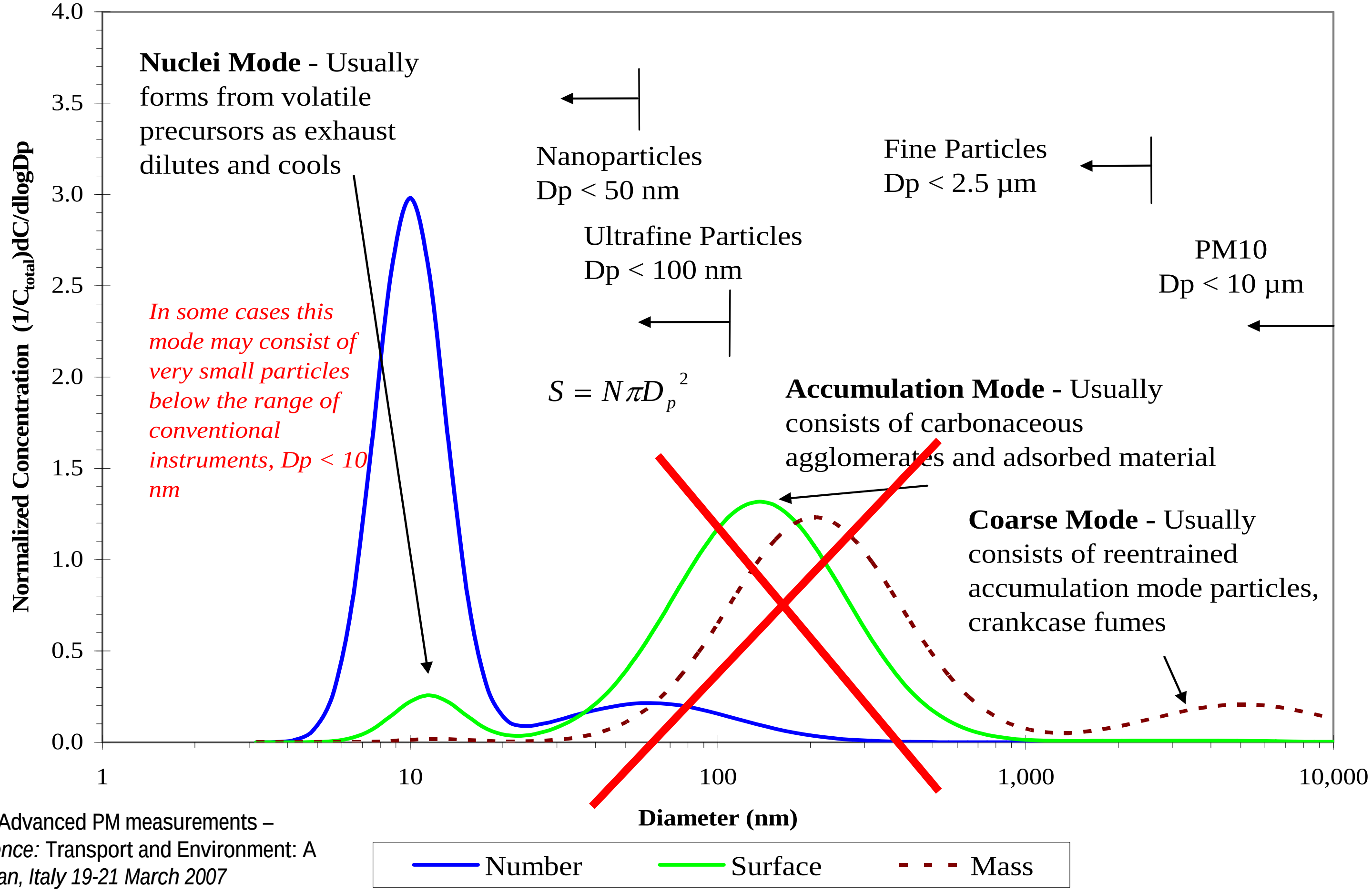
- The PMP project succeeded in developing a new procedure for particle number emission measurements, suitable for type approval purposes.
- The developed procedure was already introduced in the European regulation tackling the emissions of light duty vehicles and will soon be introduced in the heavy duty sector.
- Further investigation is underway to define the procedures and limits for gasoline vehicles.
- Work is also underway to assess the possibility of controlling particle emissions during DPF regeneration.

Thank you for your attention!

SMPS


Electrostatic Classifier
Valve
Sheath air inlet
Flowmeter
High-Efficiency filter
Pressure gauge
Variable high-voltage supply
10-11,000 Volts (-)
Poly-disperse aerosol
Sample aerosol inlet
Kr-85 bipolar charger 2 mC
Impactor
Computer
Differential Mobility Analizer
Pump
High-Efficiency filter
Valve
Valve
Excess air
Flowmeter
Monodisperse aerosol
Condensation Particle Counter
Flowmeter
Valve
Aerosol inlet
Electrical Signal

Typical engine exhaust particle size distribution by mass, number and surface area – with aftertreatment



Source: Prof. Kittelson D.B. - Advanced PM measurements – *International Conference:* Transport and Environment: A global challenge - *Milan, Italy 19-21 March 2007*