History and Future of Particle Number Legislation in Europe

The Particle Measurement Programme (PMP)

Jon Andersson¹, Giorgio Martini², Andreas Mayer³

19th ETH-Conference on Combustion Generated Nanoparticles

ETH Zentrum, Zürich, Switzerland. July 1ˢᵗ 2015

¹ – Ricardo UK
² – JRC, Italy
³ – TTM, Switzerland
History and Future of Particle Number Legislation in Europe

- Setting the Scene
  - Inception and Scope of the Particle Measurement Programme (PMP)
  - The Regulatory Particle
  - Determination of a PN Limit Value
  - Relationship between mass and number
  - Benefits delivered by the particle number metric
  - Future Directions
  - Wrap-up
Drivers for PN Regulations

Setting the Scene: What were the historic drivers for particle number legislation? The 1990’s

- Drivers
  - EPEFE (aromatics & S); Auto Oil II (PM as future issue); COMEAP (and others) concerned about the long term effects of PM air pollution
  - Swiss and EU Govts, led by the UK
    - PM pollution presented the greatest health (and economic) threat
    - Submicron particles from diesels - greatest future health challenge

- Enablers
  - Fuel S just reduced in diesel and gasoline
  - DOC and TWC becoming widespread
  - Peugeot introducing DPF along with necessary engine measures

- Open questions
  - Substantial pressure on the automotive industry to develop some new technology solutions
  - Still some concerns on the relative impacts of fuels and engines (plus aftertreatment)
Setting the Scene: What were the historic drivers for particle number legislation? The 1990’s

- The UK Govt of the time favoured a greener transport agenda, and its DETR co-funded research into PM emissions sources with oil and motor industries
- DETR/SMMT/Concawe Particulate Research Programme was established, which explored key knowledge gaps:
  - Particles & PM chemistry from LDV and HD engines of different technology levels
  - Impacts of aftertreatment, fuels and, to a lesser extent, lubricants
  - Instrumentation and measurement issues, and sampling influences
The variability in the gravimetric PM was as large as the mass measured, so if DPFs were to be mandated, a new PM method, or alternative approach was required.

**Drivers for PN Regulations**

DETR/SMMT/Concawe demonstrated the poor repeatability of PM gravimetry at post-DPF levels, and the potential for a particle metric.
Particle size distribution data showed good resolution between trap-equipped and conventional oxidation catalyst-equipped diesels, *but only at low speeds*…

Drivers for PN Regulations

DETR/SMMT/Concawe demonstrated the poor repeatability of PM gravimetry at post-DPF levels, and the potential for a particle metric

- To ensure resolution of DPF and non-DPF technologies, it seemed necessary to either discriminate by particle size range, or discriminate based on carbon content, or both
History and Future of Particle Number Legislation in Europe

• Setting the Scene

• **Inception and Scope of the Particle Measurement Programme (PMP)**
  • The Regulatory Particle
  • Determination of a PN Limit Value
  • Relationship between mass and number
  • Benefits delivered by the particle number metric
  • Future Directions
  • Wrap-up
The UK Govt reviewed the conclusions of the DETR/SMMT/Concawe programme:
- PN promising, but not (yet) suitable for regulatory use
- Soot health effects proven but size and number impacts uncertain

Previous Euro PM limit values had failed to mandate the use of DPFs

‘Precautionary principle’ invoked – elimination of carbon particles via the use of DPFs was imperative on health grounds
- but this couldn’t be achieved without an appropriate measurement method

UN-ECE Particle Measurement Programme (PMP) was conceived, chaired by the UK

Political will and drive came from the EC, but PMP operated under the auspices of UN-ECE to include Switzerland’s expertise, and other parties

PMP’s AIM was to identify a new method that
- “Complements or replaces existing mass measurement method”
  - Measurement capability for modern diesel engines
    - Pre and post PM aftertreatment [Alternative interpretation – with and without carbon present]
The PMP Comprised Three Phases

examining different candidate measurement systems and measurement metrics
[Substantial Swiss Input]

Phase II (2002-2003)
evaluated a range of promising measurement techniques and sample conditioning systems and recommended the most suitable

Phase III (2004-2010)
validated the recommended measurement techniques via inter laboratory test programmes:
Reliability, repeatability and reproducibility, methodology
Golden Engineer to help steer…

- Validation for light duty testing completed in 2006 and reported in 2007
- Validation for heavy-duty testing completed in 2009 and reported in 2010
- PMP working group recently revived and continues into 2015
The Measurement System Defines The “Solid” Particle Measured

- 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 in the range 300°C to 400°C (350°C)
  - Analogous to heated FID hydrocarbon method
- System sufficiently sensitive to determine differences in fill-state of DPF; repeatability as low as 2% with non-DPF
History and Future of Particle Number Legislation in Europe

- Setting the Scene
- Inception and Scope of the Particle Measurement Programme (PMP)

**The Regulatory Particle**

- Determination of a PN Limit Value
- Relationship between mass and number
- Benefits delivered by the particle number metric
- Future Directions
- Wrap-up
What does a European regulatory non-volatile particle look like?

- Calibration was, and remains, a challenge!
  - Particle Concentration Reduction Factor (PCRF) corrects for losses and dilution inaccuracies in the measurements

Non-volatile particles are measured: volatile particles eliminated by dilution and evaporation

Controlled volatility (survives 350°C for 0.25 – 0.4s)

Nominal minimum size (d50 = 23nm)

Nominal maximum size (d50 = 2.5µm)

Minimum size of primary carbon sphere ~20nm

Readily available CNC with d50 at 23nm

Not of critical importance, but parallels AQ measurements; also protects first diluter from particle contamination

Vast majority of Carbonaceous particles are ALWAYS counted
## History and Future of Particle Number Legislation in Europe

<table>
<thead>
<tr>
<th>Setting the Scene</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inception and Scope of the Particle Measurement Programme (PMP)</td>
</tr>
<tr>
<td>The Regulatory Particle</td>
</tr>
<tr>
<td><strong>Determination of a PN Limit Value</strong></td>
</tr>
<tr>
<td>Relationship between mass and number</td>
</tr>
<tr>
<td>Benefits delivered by the particle number metric</td>
</tr>
<tr>
<td>Future Directions</td>
</tr>
<tr>
<td>Wrap-up</td>
</tr>
</tbody>
</table>
Determination of Particle Number Limit Value (Light-duty Diesel) of $6 \times 10^{11}$ particles/km included many factors

- Limit value constructed from several factors
- Aim to be easily complied with by DPF diesel, but above:
  - Non-DPF emissions
  - Partial / open filter efficiencies (~60%)
- Typical non-DPF PN emissions value was $5 \times 10^{13}$ /km
- Filtration efficiency required > 98%
History and Future of Particle Number Legislation in Europe

- Setting the Scene
- Inception and Scope of the Particle Measurement Programme (PMP)
- The Regulatory Particle
- Determination of a PN Limit Value
- **Relationship between mass and number**
- Benefits delivered by the particle number metric
- Future Directions
- Wrap-up
There is no relationship between PM and PN embedded in European regulations.

Several studies have shown that for diesels, 1 mg equates to between $10^{12}$ and $3 \times 10^{12}$ particles.

European PN standards are much tougher than PM standards:
- Euro 6 LDD regulations require $6 \times 10^{11}$ particles and 4.5 mg
- 4.5 mg could equate to $>10^{13}$/km
- Applications with DPFs can deliver $<10^{10}$ particles and well below 0.5 mg

The PN regulation effectively mandates DPFs and has also ensured they have high efficiencies.

PM remains part of the regulation:
- Volatiles are still controlled.
History and Future of Particle Number Legislation in Europe

- Setting the Scene
- Inception and Scope of the Particle Measurement Programme (PMP)
- The Regulatory Particle
- Determination of a PN Limit Value
- Relationship between mass and number

**Benefits delivered by the particle number metric**

- Future Directions
- Wrap-up
Benefits of the PN legislation

**What the Particle Number Approach has Delivered**

- Proven effectiveness of mandating DPF fitment to all LD and HD diesel on-road engines
  - PN widely applied to diesel certification across Europe
  - High efficiency DPFs mandated and ‘effective’ PM emissions reduced to ≤ 0.2 mg/kWh / 0.2 mg/km (mass metric cannot force this)
  - Primary PM$_{10}$ / PM$_{2.5}$ reductions in European cities
  - Better filters for low back pressure and high efficiency developed

- Number of applications equipped with DPFs in Europe is at least 30 million
  - At average mileage of 10,000 km / year, PN emissions at the limit value and engine-out emissions at $5 \times 10^{13}$/km, total PN emissions reduced by $>10^{25}$/year

- Reductions in urban PM are delivering health benefits
  - Post-DPF PN vehicle emissions levels are often lower than in the intake air
  - Combined DPF and SCR systems now promise low PM with low NO$_2$ in the urban area
History and Future of Particle Number Legislation in Europe

- Setting the Scene
- Inception and Scope of the Particle Measurement Programme (PMP)
- The Regulatory Particle
- Determination of a PN Limit Value
- Relationship between mass and number
- Benefits delivered by the particle number metric

**Future Directions**

- Wrap-up
Future Regulatory Directions – Near Term

• DISI PN at Euro 6c will align with light-duty diesel (6x10^{11} particles/km)
  • Possibly achievable without a gasoline particle filter, but throughout vehicle life?
  • GPF applications are already being developed for certification and safe in-use compliance
  • Suitability of the current PN method for this SI engines must be proven

• Real Driving Emissions (RDE) requirements are leading research into the development of PN-PEMS for certification purposes

• Widened application (relative to Swiss Ordinance) of PN controls to NRMM at Stage V to include:
  • Wider power-bands; some spark ignition as well as diesel; new application types
  • PN from active regenerations and open engine breather vent systems
Future Regulatory Directions – Later?

- The current PN regulation was developed for diesel, so is restricted to the size range above which carbonaceous particles are to be found (> ~20nm), and works well!
  - With low carbon emitting engines and vehicles: SI liquid, SI gas, diesels with DPFs, it is possible that numbers of particles equivalent to those seen from non-DPF applications are being emitted below the measurement range of the current system
  - Recent research has shown that
    - Smaller non-volatile particles do exist
    - DPFs capture these smaller particles with high efficiency
    - Emissions from SI applications may be very high, especially without GPFs
  - With some modifications, the existing measurement approach could be adapted to a lower size threshold of ~10nm, but no lower
    - Calibration challenges increase hugely with lower d50 than 10nm
    - Complete elimination of volatile particles may need catalytic approach
    - Change to 10nm d50 could be achieved if it is proven necessary
History and Future of Particle Number Legislation in Europe

- Setting the Scene
- Inception and Scope of the Particle Measurement Programme (PMP)
- The Regulatory Particle
- Determination of a PN Limit Value
- Relationship between mass and number
- Benefits delivered by the particle number metric
- Future Directions

**Wrap-up**
Political will, technological maturity in engine control and aftertreatment, the right fuel, strong health evidence for reducing PM and common ground between stakeholders – as well as the need for improved measurements - drove the development of a new measurement metric.

PMP took ~10 years to investigate and develop the PN metric
- But for LDD the mechanism of legislation was in place after only 3 years.

PN delivers increased sensitivity and accuracy in quantifying diesel particle emissions.

A PN limit was developed that forced diesel emissions to be reduced by >98%: a reduction readily achievable by existing emissions control technologies.

The twin aims of developing a new metric and forcing technologies that eliminate carbon particle emissions from diesel engine exhausts were fully achieved.

Future application of the PN approach to spark-ignition engines and challenging exhaust chemistries, plus the simplification of calibration processes, are the subject of on-going research.

Finally, a question: PN emissions will soon be regulated for on- and off road, CI and SI applications, is it time to measure success with a number-based AQ method?
Thanks for listening

“Not everything that counts can be counted, and not everything that can be counted counts…”

“but counting particles counts!”