

Working Paper No. **WP-9** (Special Informal Meeting of GRPE, 15 September 2003)

# Comments on the recommendations of the government research project for the GRPE Particle Measurement Programme

## 1. Summary

The report<sup>1</sup> provides an overview of government research from France, Germany, Sweden, Switzerland and the UK undertaken as part of Phase II of the Particle Measuring Programme (PMP) and arrives at the conclusion that the following two measuring systems should be considered for future regulatory use:

- ✓ Modified 2007PM (a gravimetric filter based mass measuring system)
- ✓ CVS + thermodiluter + CPC (a number based measuring system)

The type approval of a vehicle or of an engine for exhaust emissions requires a measuring system, which meets the criteria of robustness, repeatability, reproducibility and traceability as defined on page 6 of the report.

According to the report<sup>1</sup>, CPC are robust, sensitive and commercially available instruments. CPC with a thermodiluter is said to have a good repeatability for tests carried out within a day.

However, CPC requires a very diluted sample. The dilution system may prove to be a critical element of the proposed measuring system. With reference to the report<sup>2</sup> published by ACEA on July 2002, the stability of the number measurements with CPC over the duration of the programme was not satisfactory. An average decrease of about 35% in particulate number concentration was observed during the 4 months duration of the programme. As there was no indication that this effect was caused by the vehicles, it is assumed that the dilution system was responsible for this decrease in spite of the rigorous quality control measures taken in this programme.

Reproducibility of exhaust emission measurements in different laboratories and with different instruments requires absolute calibration standards for the instruments being used. Whilst very stringent standards are prescribed for the analysers used to measure gaseous emissions and for the balance used in the gravimetric method, no absolute calibration standards exist for particle number instruments such as the CPC.

<sup>&</sup>lt;sup>1</sup> Report of the GRPE Particle Measurement Programme (PMP), Government Sponsored Work Programmes, July 2003

<sup>&</sup>lt;sup>2</sup> ACEA Programme on Fine Particulate Emissions from Passenger Cars: 2, July 2002

As mentioned on page 13 of the report<sup>1</sup>, currently instruments that measure nonmass based parameters are calibrated against other instruments using an aerosol generation system, such as the Combustion Aerosol Standard (CAST) or atomisers run with a NaCl solution. This approach does not validate the absolute value being measured.

At present only the gravimetric method fulfils the measurement requirements for a technique to be applied for type approval and has proved to have the highest potential for further development<sup>3</sup>. Measured particle number and size distributions can be manipulated with a careful choice of test conditions. The CPC is still far from achieving absolute measurements of particle number and can, as yet, only be used for qualitative assessment of particulate number as a relative comparison.

Furthermore, the ACEA programme confirmed the good correlation between the mass of particulates emitted and number of particles as measured with a CPC + thermodenuder. By setting ambitious limit values based on the gravimetric method both objectives of reducing the mass and the number of the particles emitted by diesel vehicles can be achieved.

Finally, particle number measurements are found to be very time consuming as much time and effort are required to maintain and operate the particulate number instruments and sampling systems.

Including particle number measurements during engine development will not only require substantial investments but also slow down the certification process.

In conclusion, OICA is of the opinion that

- the major development should focus on the further improvement of the gravimetric method, and
- much investigation is still to be carried out before measurements of particle numbers can be introduced in an ECE Regulation/EU Directive.

In addition OICA regrets that the industry report<sup>3</sup> was not commented or discussed by the governments.

# 2. OICA comments on the different chapters of the report on the government research

0. Executive Summary

- The report is only a summary and does not back-up the results with references to the national programme reports or data (information from "attached CD" is not available). Therefore, it gives the impression that selected results are presented.
- "... the overall sensitivity of the measurement ..."

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<sup>&</sup>lt;sup>3</sup> Industry comments on proposed particulate measurement techniques, OICA contribution to PMP, Informal Paper No. 7 to 45<sup>th</sup> GRPE Session (see also: <u>www.oica.net</u>)

Counting the number of particles might lead to a higher sensitivity, but to discuss the limits of a measurement technique the repeatability and the background level must also be considered.

- 1.1. Introduction Background
- "... carbonaceous particles ..." The definition is not clear: are these solid particles?
- "... test should be based on transient cycles ..." For heavy duty engines also steady state tests were performed.
- "The aim of the programme is not to recommend specific measurement equipment but instead to specify performance parameters ..." OICA fully supports this approach – but we recognize that in the report<sup>1</sup> only instruments and samplings are discussed, there is no reference to performance parameters.

1.2 Introduction - Health impacts

- The independent HEI<sup>4</sup> states<sup>5</sup> that the results of an extensive study "indicate that epidemiological evidence of PM's effects on morbidity and mortality persist even when the alternative explanations have been largely addressed". Although HEI places special emphasis on the necessity of
  - further research to clarify the impacts of the health effects and
  - caution in the use of these results in the political field

the above statement is often used in isolation and an immediate drastic reduction of particulates is demanded as a precaution.

- The results of the epidemiological studies are not clear enough to make a conclusive statement<sup>6</sup>. Aside from a few exceptions the studies are based on measurements of the exposure in relation to the total particulate mass. This requires further clarification before new legislation can be introduced.
- Furthermore, the HEI<sup>7</sup> commented that both the ultrafine (0.010-0.100 μm) and fine (0.010-2.5 μm) particle fractions have shown associations with human mortality; however 'no clear pattern of association indicates relative or temporal differences between ultrafine and fine particles'. This essentially means that there is currently no clear evidence to preferentially address air quality or emission standards for ultrafine particles, as compared to the existing standards for fine PM.
- The US-EPA<sup>8</sup> currently does not perceive a need for a new metric for PM emission regulation, other than PM mass, due to the lack of health effects evidence. This statement is based on the acknowledged health effects related to

<sup>&</sup>lt;sup>4</sup> HEI = Health Effects Institute, Cambridge MA, USA (www.healtheffects.org)

<sup>&</sup>lt;sup>5</sup> Airborne Particulates and Health: HEI Epidemiological Evidence, HEI Perspectives, Cambridge MA, USA, June 2001

<sup>&</sup>lt;sup>6</sup> M. Spallek: The effects of exhaust emissions on human beings – A never ending story?, VDA Technischer Kongreß, 20./21.09.1999, Frankfurt, S. 201-209

<sup>&</sup>lt;sup>7</sup> HEI Research Report 1998

<sup>&</sup>lt;sup>8</sup> EPA Report: Health Assessment Document for Diesel Engine Exhaust

PM mass, which has led to implementation of ambient PM mass standards and emission inventories. However, the US-EPA remains open to considering alternative methods of determining the PM mass emissions<sup>9</sup>.

• In principle, the automotive industry is of the opinion that the reduction of particulate matter which has already been achieved and which may still be possible in future can be already considered as a health precautionary measure.

# 1.3 Introduction – This report

- The OICA contribution to PMP is referenced but the results of this report<sup>3</sup> are not commented on or discussed by the governments.
- In addition ACEA started a new PM Programme to investigate modifications to the regulatory European test procedure with the aim to increase its repeatability, reproducibility and robustness. Results of this programme will be presented to 47<sup>th</sup> GRPE.
- 2.1 Methodology Introduction
- "... testing protocols ... are based on existing regulatory procedures" The testing protocol is based on a modified regulatory procedure because it is not allowed to take further samples from the dilution tunnel (annex 4, fig. 1b).
- "... suppression of the nucleation mode particles ..." The increase of the repeatability by measuring accumulation mode particles only is supported by the OICA work<sup>3</sup>.

2.2 Methodology – Conditioning systems assessed

- "Heavy duty ..."
  OICA is missing the evaluation of partial flow systems. OICA would like to understand the rationale for this.
- "If the losses are consistent then a correction factor could be applied ..." This type of generic correction factor from characterisation of a device should not be introduced into a regulatory procedure. There are standard corrections for ambient temperature/humidity within the Regulations, however these are known mathematical corrections.
- "... prototype diesel soot separator.." The description is not extensive enough to allow OICA to analyse this device. In addition it is impossible to comment on the results without having more insight into the experiments. Therefore, OICA asks for clarification.

<sup>4</sup> 

<sup>&</sup>lt;sup>9</sup> CRC workshop San Diego, EPA presentation M. Spears

#### 2.3 Methodology – Instruments assessed

- OICA appreciates the task of comparing 15 different types of instruments in different regulatory procedures.
- Fundamental requirements must be the same calibration of the instruments and the same set-up of test equipment, see also the OICA report<sup>3</sup>. OICA could not identify from the report<sup>1</sup> that a quality assurance system was used in the various national programmes. OICA has shown<sup>3</sup> that, without careful specification of experimental conditions, the measured particle number and size distribution can be manipulated with a careful choice of test conditions.

2.5 Methodology – Draft test protocols

- A balance with a resolution or readability of 0,1 µg was not considered in the national programmes. The influence will be evaluated in the ACEA PM Programme No. 3.
- "... LD draft protocol ... using NEDC/FTP ..."
  From the report<sup>1</sup> OICA cannot determine if there are differences in the results for the European and the US procedures. OICA asks for additional information.
- "These protocols ... not all the PMP studies used them" and "laboratories ... applied their own interpretation of the US 2007 test and therefore adopted some different approaches"
  This is one of the major concerns which OICA would like to express. A common

basis for the comparison as required in remark to 2.3 (see above) is not available!

# 2.7 Methodology – Instrument calibration

• "... a primary standard for particle number and other metrics does not yet exist. Currently instruments that measure non-mass based parameters are calibrated against other instruments."

This is another major concern of OICA. The objective of the GRPE PMP is "to develop a new measurement system that could replace or complement the existing particle mass measurement system" (see 1.1 Introduction – Background). Such a system may be widely used in type approval, COP and in-use compliance testing against absolute limit values set by legislation. Therefore it is mandatory that the new system can be calibrated with an absolute standard. Using instruments (which must be calibrated too – but how?) to calibrate other instruments is not acceptable.

 "An aerosol generation system (...CAST) has been tested as calibration device." OICA has demonstrated in its report<sup>3</sup> that particles generated by CAST show a stable average diameter, but a trend towards decreasing number concentration with time. Furthermore, it was shown that a "calibration" by METAS could not be reproduced by a user and by the CAST manufacturer<sup>11</sup>. Therefore, it appears that the CAST instrument is unsuitable as a calibration aid.

<sup>&</sup>lt;sup>11</sup> Carli et al., Influence of Compressed Air Quality on CAST Performance, 7th ETH Conference on Combustion Generated Aerosols, Poster No. 31, Zürich, 18.-20. August 2003

2.8 Methodology – Regulated gases

• It is common practice to measure regulated emissions simultaneously with particles. It allows the verification that the results are as expected.

2.10 Methodology – Inter-laboratory comparison

• It is misleading to refer to these results without further explanation as the PMP test protocol was not used and the equipment was not included in the list of candidate systems.

3.1 Results – Introduction

- "... DPF. These devices effectively remove the carbonaceous particles with a higher proportion of volatile exhaust aerosols being measured post-DPF." Comparing this result with the results of the carbon fraction in post-DPF from section 2.2 (discussion of the prototype diesel soot separator) OICA requests clarification. Do the results give the same message?
- For the sensitivity please refer to the OICA comment on section 0.
- "... emissions from the HD engines appear to be generally more stable ... than those from LD vehicles ..." This reflects not only the differences between chassis and bench dynamometer test and engine management. One could imagine other contributing parameters, e.g. cold start.
- "... the repeatability of the mass and particle number concentrations measured was often of the same order as those obtained for gaseous emissions." Today, it is the experience of OICA that the repeatability of the mass measurement is better than the repeatability for HC and NOx. In addition it is questionable if the discussion on repeatability at such low numbers should be performed with relative numbers and not with absolute ones.

3.2 Results – Assessment of the enhanced gravimetric mass measurement method

- "For some LD vehicles the repeatability was poorer with the modified 2007 PM method than the conventional method. This is perhaps unsurprising ..."
  The information in the report<sup>1</sup> is not sufficient to comment on the numerical results. More details about the statistics of the experiments must be known. In its PM Programme 3 ACEA will analyse the different steps towards an improvement of the gravimetric method. First results show already that also for LD vehicles the repeatability can be improved.
- "... the limits of detection of the modified 2007 PM method may be about 3 times better than that for the conventional PM method."
  The report<sup>1</sup> does not show any results which underline this statement. For such a discussion at the very low PM levels the background level has to be considered, too. OICA refers to the results of the ACEA PM Programme 3 which will be available for the 47<sup>th</sup> GRPE.

#### 3.3 Results – Other mass measurement systems

- For mass measurement only the gravimetric method can be directly calibrated<sup>3</sup>, • however, other mass based systems can be indirectly calibrated using gravimetric or coulometric procedures. But any new measurement methodology should fulfil the same quality criteria as the current gravimetric technique, which still holds potential for future development.
- "...unlikely that LII will have a role ... unless cost is reduced significantly ..." This is true for LD vehicles but not for HD engines where LII replaces the CVS.

### 3.4 Results – Number measurement systems

- The OICA report<sup>3</sup> summarizes the experience with the CPC: It is very sensitive to • nucleation effects. Due to lack of size resolved information, these effects are not identifiable. The operation range is limited (variable high dilution needed) and the repeatability is low compared to state of the art PM mass method. Additionally, no absolute particle number calibration standard is available for system verification.
- "For LD the repeatability is poorer than for HD but is similar to that found for LD with the modified 2007 method." Please note comments to section 3.2.
- "This requires a very dilute sample" • Consequence of different dilution steps is the need for calibration of each step.
- "A detection limit of 60 particles/cm<sup>3</sup> was found with one model in one study, • although higher detection limits were found with other models ..." OICA asks for clarification of the consequences of this statement.
- "It is medium priced, but this may drop as the technology matures and the • demand increases." This is true for all instruments and not only for the CPC.

3.5 Results – Calibration

- "Any measurement system used for regulatory purposes must be capable of • robust calibration. ... calibration remains an important issue for the development of a regulatory test procedure." Fully supported by OICA<sup>3</sup> and see also OICA comments to section 2.7.
- "For regulatory purposes it will be essential to define this to establish calibration intervals." Long-term stability was not addressed by the report<sup>1</sup>, but is a very important issue<sup>3</sup>.
- "... neither a reasonably fast calibration method ... nor procedures traceable to a primary standard are available ..." As commented to section 2.7 this is a major concern of OICA.
- "... a CPC ... was calibrated using NaCl aerosol". • The procedure needs a further clarification, e.g. question of local equilibrium of the particles. Furthermore, there is no information about the calibration of the

DMA which was used. It appears that this procedure cannot be seen as an absolute calibration procedure.

• OICA has commented on the CAST in section 2.7.

3.6 Results – Removal of volatile material by sample treatment

- Concerning the thermodenuder, OICA agrees with the assessment of the report<sup>1</sup> and refers to the results of the OICA report<sup>3</sup>: Because of the many parameters that must be kept constant and verified by regular calibration, a thermodenuder is in principle not suitable for regulatory measurements.
- "... thermodiluters show virtually no loss of particles ..." By comparing the figures in annex 6 it appears that there are losses for the thermodenuder as well as for the thermodiluter. The losses for the thermodiluter are smaller but still considerably high.
- 3.7 Results Inter-lab comparison
- "... the results for this one vehicle look promising ..." The results are not relevant for the selected system (CPC + thermodiluter).

#### 4. Conclusion

- The new or improved measuring methods must fulfil all requirements for a procedure which might be included in ECE regulations and EU directives.
- OICA continues to question the need for counting particles.
- A performance description for the procedure, measurement and sampling equipment is necessary.
- A robust calibration procedure to a primary traceable standard is prerequisite.
- A robust test protocol is necessary.
- 5. Recommendation
- Beside the technical feasibility of a new instrument the cost-effectiveness of the introduction of a new measurement method must be investigated.